



Evaluating the potential of microplastics and natural organic matter for sorption of hydrophobic organic contaminants based on selected properties

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Abstract

Interest in microplastic behaviour as vectors for hydrophobic organic pollutants has increased in the literature over recent years, however these materials constitute only one of many organic particulate groups in the environment able to compete for pollutants. This review examines particle characteristics including size, surface area, porosity, and age, and how these may influence natural organic particle and microplastic sorption capacity. The impacts these particles manifest as contaminant vectors to biota are also discussed, along with additional chemical and environmental factors that play a role in favour of each particle type. Microplastics are primarily considered to behave as more ideal sorbent due to their hydrophobic nature and acceptable size range below that considered as "particulate" organic matter, however the abundance of these anthropogenic pollutants in comparison to their natural counterparts and their initial hydrophobicity makes their risk as a vector far less prominent than the latter group.

Keywords: HOC, microplastics, natural organic matter, particulate organic matter, sorption, vector, size, surface area, porosity

Popular Science Summary

Microplastic (MP) particles in aquatic environments are a relatively new organic pollutant group, originating from either intentional design at the 0.1µm-5mm size or as a result from breakdown from larger pieces over time. They have received notable attention in the media and literature because of their effects on animals and food chains, one proposed threat being their capacity to carry hydrophobic organic pollutants (HOCs). These are a group of harmful chemicals including pesticides, polycyclic aromatic hydrocarbons (PAHS), etc. that do not mix well with water and will seek other hydrophobic (water-repelling) solid surfaces. Plastics however are not the only particles within aquatic systems capable of carrying or "sorbing" these pollutants. Most comparable are various forms of natural particulate organic matter (POM) in the 0.45µm-1mm size range such as suspended and bottom sediments, soil, bacteria etc. This review looked at several physical properties (size, surface area, porosity and age) shared by these two organic particle categories to see how they each affect HOC sorption, and how this could later potentially impact living organisms. For this last part, other factors like chemistry and the surrounding environmental conditions are also very important to consider.

Surface area and porosity reflect the availability of sorption sites with which HOCs can interact, so larger values for these properties imply a higher potential for sorption. Size and age play a role in influencing the above two properties, altering the ratio between surface area and volume and state of the particle surface after exposure to elements such as UV radiation and physical abrasion. Overall, natural particles tended towards larger median surface areas and porosities than MPs, even though they have a more restricted size range. This is likely because they possess very uneven surface textures as a result of their diverse chemical composition and the uneven degradation of less resistant materials over time, leaving behind an irregular shape. As MPs are made up repeating subunits, they possess much more smooth surfaces but are also much more resistant to being eroded over time. Based on physical characteristics alone then, it would be assumed that POM materials are better HOC carriers or "vectors". When chemistry is also considered however, MPs are much more favourable because they are more hydrophobic compared with natural particles that both attract and repel water.

In experiments done in laboratory settings, both positive and negative effects were seen on animals when were exposed to different particle and HOC combinations. These can be explained as particles facilitating the transfer of HOCs into their bodies when eaten, or conversely reducing the likelihood of contact if not. In order for HOC transfer to organisms to be of a legitimate concern however, the abundance of the material in question needs to be considered. MP concentrations in the environment are essentially negligible when compared to the plethora of POM materials, so although they are better pollutant carriers their role is insignificant compared with natural food and water sources.

In order to further understand the real threat posed by both MPs and POM in their surrounding ecosystems, much more research is needed. In particular, research reflecting more realistic environmental conditions such as relevant particle concentrations, as well as more characterization of physical and chemical properties of natural particles. In fact, more research over all on natural organic particles in the 0.45-1mm range is required in order to support the validity of the findings in this review, as available information was quite limited. Media coverage of MPs in future should focus more on other threats such as accumulation of these materials rather than their vector capacity, and more interest in natural particles to act in this manner should be emphasized.

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Abbreviations

HOC	Hydrophobic Organic Contaminant
MP	Microplastic
РОМ	Particulate Organic Matter
NOM	Natural Organic Matter
SA	Surface Area

1. Introduction

Human impact on the environment has become increasingly evident over the last few centuries, with accumulated anthropogenic material now being found in geological layers. The extraction, modification, and production of new materials from natural sources has gone so far as to spark debate on whether or not we have entered a new geological era, i.e. the "Anthropocene" (Zalasiewicz et al., 2011). Organic anthropogenic materials include products such as wool, paper, cotton, as well as more recent durable polymers, i.e. "plastics" (Browne et al., 2015). As a material, plastics are favored for their lightweight and erosion resistant properties (Laist, 1987), as well as the relatively low economic cost associated with their production (Thompson et al., 2009). This has led to an exponential increase in plastic manufacturing over the last few decades from <2 million tons in the 1950s (Shen, et al., 2020) to an estimated 359 million tons in 2018 (PlasticsEurope, 2019). Unfortunately, due to a universal lack of proper recycling techniques capable of handling both the abundance and diversity of compositions, only approximately a quarter of plastics are recycled (Nizzeto et al., 2016), with the rest accumulating in landfills and natural environments (Van Cauwenberghe et al. 2013).

This accumulation has been most notable in aquatic domains, with freshwater systems draining material above and below the Earth's surface into water bodies eventually flowing into the sea. The pathways in which these materials enter a particular environment are both point and diffuse, though strong correlations to population density, urbanization, degree of wastewater treatment, and waste management of the surrounding area have been observed (van Emmerick and Schwarz, 2019; Best, 2019; Eriksen et al., 2013). Some more well-known transport routes include runoff from terrestrial surfaces (Horton et al., 2017; Bruge et al., 2018; Lebreton et al., 2017; Magnusson et al., 2016), direct dumping (Rech et al., 2015; van Emmerick and Schwarz, 2019), unintentional spillage from wastewater treatment plants or landfills, (Dris et al., 2015; Horton et al., 2017) and aeolian deposition (Dris et al., 2015). A visual schema of these pathways can be seen below in Figure 1 below taken from Horton and Dixon (2018).



Figure 1. Routes of plastic entry to aquatic environments (taken from Horton and Dixon, 2018)

When exposed to the elements, these materials are slowly broken down through physical and chemical processes such as abrasion by water and sediment, and/or exposure to ultraviolet radiation (Mason et al., 2016). This results in the creation of "microplastics" (MPs), defined as particles between 0.1µm and 5mm in size (Thompson et al. 2009; Bråte et al., 2017) in a variety of shapes (i.e. fibres, fragments, films, foams, etc.). Alternatively, MPs are also the result of intentional design at the 5mm size or less for purposes including creating synthetic textiles, use as exfoliants in personal care products, or for industrial processes that later synthesize smaller pieces into larger materials (i.e. virgin plastic) (Eerkes-Medrano et al. 2015). Upon introduction to the environment, these small pieces are added to an already heterogeneous mixture of pre-existing natural particles (both organic and inorganic), and to an extent become part of the surrounding natural system.

Although a large knowledge gap remains concerning the effects these particles have in a larger ecosystem or global context, various impacts have been documented at laboratory scales. Frequently mentioned is their ingestion by aquatic organisms ranging in size from zooplankton (Wagner et al. 2014) to seabirds (Colabuono et al. 2010, Provencher et al 2015) and inclusively humans (Cox et al., 2019). This can result in insufficient caloric intake and potential bioaccumulation in both benthic and pelagic food chains (Castañeda et al. 2014). Environmental contamination due to leaching of weather-resistant plasticizer reagents has also been reported as a possible concern, particularly should the volume of MPs in aquatic environments continue to increase with time (Sajiki and Yonekubo 2003). For the focus of this review, the potential of these particles to behave as surfaces and vectors for harmful hydrophobic compounds will be considered.

A wide variety of MPs are commonly cited in the literature as capable of sorbing HOCs based on their particular physio-chemical properties (Rochman et al.,

2013). Table 1 below contains a summary of these plastics and some further characteristics. A common trait to all is the presence of non-polar alkyl zones granting HOC sorption potential. The presence of substituted aromatic rings, halogens and/or carbonyl groups within each plastic's monomer structure however will alter the chances or strength of sorption occurring (Mei et al., 2020).

Plastic Type	Monomer Structure	Properties ^{a,c,d}	Application ^c
Polyethylene (PE) (HDPE and LDPE grouped together)	H H H H H H H	ρ: 0.85-0.90g/cm ³ GTT: 160-239K Crystallinty: semi- crystalline	Toys, household and kitchenware, food wrapping, gas and water pipes, high frequency insulation
Polystyrene (PS)		ρ: 1.05-1.06g/cm ³ GTT: 373-388K Crystallinity: amorphous	Disposable cups, rigid packaging, cosmetic packs and costume jewelry, packing foam
Polyvinyl Chloride (PVC)	H H H H - C - H H - C - H - C - H H H - C - H	ρ: 1.37-1.40g/cm ³ GTT: 345-354K Crystallinity: amorphous	Plumbing needs, cable and wire insulation, automotive interiors and seat coverings, synthetic leather
Polypropylene (PP)	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	p: 0.85-0.88g/cm ³ GTT:258-270K Crystallinity: semi- crystalline or amorphous	Flexible food containers, microwave components
Polyamide (PA, Nylon)*	H-C-H H-C-H H-C-H R H-C-H H-C-H H-C-H	ρ: 1.06-1.08g/cm ³ GTT: 308-324K Crystallinity: semi- crystalline	Textiles, fishing lines, carpets, food packaging
Polyethylene Terephthalate (PET)		p: 1.31-1.43g/cm ³ GTT: 346-361K Crystallinity: semi- crystalline	Fiberfill in winter clothing, exterior components (e.g. window wipers)

Table 1. Popular plastic monomer structures, physio-chemical properties and common uses

*Nylon 6 used for property descriptions, GTT: Glass transition temperature, ^aPolymer Property Database, 2019, ^bCrawford and Quinn, 2017, ^cBritish Plastics Federation, 2020, ^dLi et al., 2018b

The term "hydrophobic organic contaminant" (HOC) is used when referring to chemicals characterized by low aqueous solubilities and high partition coefficients (log Kow > 2) (Elzerman and Coates, 1987). This term describes a compound's hydrophobicity, defined as the ratio to which a compound separates itself between immiscible polar and nonpolar solvents, as can be seen in Equation 1 below. This value is commonly expressed in logarithmic format, with higher values representing a greater affinity for the non-polar solvent (Speight, 2018).

Eq. 1. K_{ow}= [solute]_{octanol}/[solute]_{water}

HOCs encompass but are not limited to categories including polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (Semple et al., 2003), as well as certain pesticides and pharmaceutical products. Being surrounded by polar water molecules causes these compounds to associate with organic-rich phases, making fragile aquatic ecosystems vulnerable to their influence (Jaffé, 1991). The capacity for HOCs to sorb onto particles in the environment is driven by the physical and chemical characteristics of both the sorbate and sorbent (Hüffer and Hofmann, 2016). The term "sorption" in this review will be used to describe the general interaction between HOCs and solid phase particles as used by Mei et al. (2020).

The creation of the above mentioned HOC categories is specific to each pollutant group under consideration. For example, PAHs are compounds with two or more fused benzene rings resulting from the extraction and incomplete combustion of raw organic materials such as coal, gas, or wood (Abdel-Shafy and Mansour, 2016). Conversely, PCBs are a group of 209 highly stable and anthropogenically made congeners, historically produced for use in industrial applications such as electrical equipment and pigments (US EPA, n.d.). Both pharmaceuticals and pesticide groups are also intentionally designed in order to eliminate ailments or undesirable organisms in health and agricultural sectors (e.g. bacteria, fungi, insects) (Swanton et al., 2011). Their introduction to the environment occurs in routes similar to MPs, including leakage from industrial wastewaters and sewage (Moyo et al., 2014), runoff from modified land surfaces (Burant et al., 2018) and atmospheric deposition (Countway et al., 2003; Abdel-Shafy and Mansour, 2016). Depending on their specific physio-chemical properties such as vapor pressure, solubility, resistance to biodegradation and hydrophobicity (Jaffé, 1991), they are more or less persistent in various environmental media. Their presence in the environment has been observed within layers of topsoil (Benoit et al., 2008), surface water and sediment samples (Moyo et al., 2014), and of importance in this review, sorbing to both natural and anthropogenic organic particles. Determining the presence and degree to which

HOCs can be transported in the environment is of great interest to researchers as they are considered hazardous compounds when flora, fauna and humans are exposed. In some instances, the degradation products of HOCs are of greater concern than their parental form (Yu et al., 2020). For various groups, accumulation in organismal fatty tissue has also been documented, one example being the organochlorine pesticide DDT (Bakirhan et al., 2018). The aromatic structures of these and other HOC compounds impart a very high degree of chemical stability, leading to bioaccumulation potential in food webs starting with small filter feeding organisms up to large carnivores and humans (Jayaraj et al., 2016). Vectors carrying HOCs from one location to another therefore enhance the risk of their negative consequences being perpetrated over a larger area.

As alluded to earlier, MPs introduced to natural systems are but one of many particle types found in the environment. Within both aquatic and terrestrial ecosystems there exists a plethora of other natural particles, both mineral and organic, each with their own distinct origins, compositions and degrees of hydrophobicity (Ćosović and Vojvodić, 1989). In many ways, the threats posed by MPs in the environment are the result of their substitution in roles ordinarily occupied by natural organic matter (NOM). For example, both can be consumed as food by different species, or release small fragments while undergoing degradation. As MPs can be considered a form of anthropogenic organic matter, similar characteristics between these particle groups could explain their interchangeability in natural processes, and how both have been documented to behave as vectors for HOCs (Gourlay et al., 2005).

Like those of anthropogenic pollutants, the occurrence of NOM in freshwater ecosystems occurs via multiple pathways but differs in that sources can be allochthonous or autochthonous (Artifon et al., 2019). The former result from the input of terrestrial vegetation (and related components) such as leaves, pollen, and wood, usually in low order streams (Peters et al., 1989) with the volume of fallen material changing on a seasonal basis (Atkinson et al., 1993). Soil may also enter from the erosion of embankments through wind dispersion and runoff after hydrological events (OMAFRA, 2020). As an extremely heterogeneous material, soil composition will vary drastically based on the ratios of its sediment size fractions (Barriuso et al., 2000), as well as the origins of both mineral and organic parent components. For example, forest versus agricultural or grassland areas will contain different C:N ratios and specific phenolic compounds based on what material has been historically contributing to the soils' development (Benoit et al., 2008). This in turn influences the aliphaticity and aromaticity of the soil, and therefore its overall hydrophobic properties (Benoit et al., 2008). Autochthonous material in contrast is the result of living or deceased flora and fauna within aquatic systems, including communities of microscopic organisms such as phytoplankton,

zooplankton, and bacterial communities (Countway et al., 2003). Consumed allochthonous or autochthonous material by organisms who then excrete undigested components as faecal pellets is another example of autochthonous NOM (Bundschuh and McKie, 2015). These materials are present throughout a river's continuum but become the predominant source of NOM in larger downstream tributaries, estuaries and marine areas (Countway et al., 2003).

As NOM begins to decompose, elements and molecules in dissolved form (N, P, C, etc.) are leached out and fragments become colonized by bacteria and fungi. The exposure of more available nutrients increases the palatability of these NOM sources for organisms (Ferreira et al., 2006), further encouraging the breakdown of organic matter into smaller pieces and creating a range of NOM size classes, each with their own characteristics. Fragments >1mm in size are denoted as "coarse particulate matter" (CPOM) (Elosegi and Pozo, 2016), followed by particular organic matter (POM) defined as material that is retained on a 0.45 μ m filter (Artifon et al., 2019). Matter less than 0.45 μ m is then grouped together as dissolved organic matter (DOM). It should be noted that the distinction between these last two classes is not as definite as that of CPOM, for between them there exists a colloid fraction that can easily break down or agglomerate to be grouped within either category (Broman et al., 1991). A visual representation of these size classes by Artifon et al. (2019) is presented in Figure 2 below.



Figure 2. Summary of NOM size class distinctions and examples (taken from Artifon et al., 2019. MW: molecular weight (Da), HAc: humic acid, FAc: fulvic acid, HC: hydrocarbons, CHO: carbohydrates, FA: fatty acids, AA: amino acid.)

It follows that organic matter resulting from diverse origins will present a diverse array of physical and chemical properties. Vegetation and soil-sourced NOM for example is composed of a variety of humic and non-humic substances (Artifon et al., 2019). The former are amphiphilic, polar organic acids with assumed polymeric structure, granting them an extremely recalcitrant nature (Piccolo, 2001). Examples of major humic substance groups include humic acids, fulvic acids, and humins. The diversity and overwhelming abundance of these groups causes their exact makeup and the boundaries separating them to remain poorly understood (Piccolo, 2001). Non-humic substances on the other hand are groups of macromoleculesnamely carbohydrates, amino acids, lipids and lignins (Artifon et al., 2019) more available to degradation and assimilation by biota. Together both humic and nonhumic substances generate particles with hydrophobic and hydrophilic regions capable of carrying or repelling HOCs (Pignatello and Xing, 1996). NOM from bacteria and organisms is likewise a mixture of various components including the aforementioned macromolecules (Henderson et al., 2008) as well as additional species-specific mineral components (e.g. silica in diatom shells). These constituents present in NOM together with other physical parameters (i.e. surface area, degree of degradation, etc.) will impact a particles' overall sorption capacity. Competitive sorption by other environmental substances (e.g. pathogens, DOM, etc.) may simultaneously occur, but will be discussed in a later section.

The aim of this review is to examine physical properties shared by MPs and natural particulate organic matter (POM) cited in the literature that favour the sorption of HOCs, as well as where they differ. These include size, surface area (SA), porosity and age. With greater knowledge of these characteristics, this review also aims to compare the biological impacts posed by the two particle types in their capacity to act as vectors for HOCs. To do this, factors such as chemical composition, external environmental factors, and particle abundance must also be considered. With the information gathered, this review will venture to draw preliminary conclusions on the threat posed by MPs and POM as vectors of HOCs. Within this review, focus will be on natural particles >0.45 μ m to the exclusion of the DOM in order to ensure a proper comparison within similar size ranges.

2. Methods

For this literature review, the search engines Web of Science, Scopus and Google Scholar were used between May and June of 2020. The combination and results of the search terms can be seen in Tables 2, 3, and 4 below for both MPs and NOM. For both sets of searches, the main terms "microplastic*" or "natural organic matter + particle" where combined with "hydrophob*" and "chemical". They were then searched as stated (labelled "Plain" in Tables), as well as with the additional key terms "size", "surface area", "residence time", "equilibrium", "bioaccumulation", "fate", "toxicity" and "freshwater" in the given order. Articles with information relevant to this review were found in the majority of the search combinations, however results did not necessarily pertain to the exact key words used (e.g. a search using "surface area" generating a result related to "bioaccumulation"). The subcategories "P", "T" and "K" in the tables respectively represent the number of results produced by search engine, the number of studies taken into consideration (added to Zotero), and those kept for use in the generation of data. The values in the "T" column only include the number of new articles added that were not already found in a previous search or search engine, hence there being many "0" values. It should also be noted that the search order for NOM and microplastics was done in reverse for Scopus as is reflected in Table 3.

Articles that appeared in the search result were initially screened by looking at the title and/or abstract to ensure they had potential to meet the aims of this review. Those with information on soil, sediments, biota, wood, etc. or different MP types were kept for further screening. Articles written as summaries, reviews, or meta-analyses were not included in this selection as the majority did not provide quantitative details on physical/chemical characteristics. Other rejected papers whose themes did not sufficiently align with the aim of this review included those with natural particles outside the 0.45µm-1mm size range, using manufactured geosorbants, or isolated modified organic matter such as biochar, activated carbon, etc. The exacerbated sorption behaviour exhibited by this last group would render comparison beyond the scope of this review. Articles emphasizing particle transport/flow velocity or filtration of WWTP sewage sludge were also excluded.

Research pertaining to freshwater environments was the primary aim of the search, however experiments undertaken in estuarine and soil environments with

applicable supporting material were also taken into consideration. Marine research however was excluded, as the significant differences in environmental conditions that influence sorption would introduce unnecessary uncertainty into any conclusions drawn when compared with the aforementioned systems

	Plain			Size			SA			RT			Equil		
	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K
NOM+particle+H+C	217	19	9	105	3	2	27	0	N/A	2	0	N/A	26	0	N/A
Microplastic+H+C	153	52	31	52	6	3	19	2	0	0	0	N/A	15	1	0
]	Bioac	c	Fate			Toxicity			Fresh			Total		
	R	Т	K	R	Т	K	R	Т	K	R	Т	K	Т		K
NOM+particle+H+C	9	0	N/A	36	2	0	19	0	N/A	20	0	N/A	24		11
Microplastic+H+C	37	0	N/A	41	2	2	83	0	N/A	23	0	N/A	63		36

Table 2. Quantitative search results of articles produced, taken, and kept from Web of Science search engine

Table 3.	Quantitative	search	results of	articles	produced,	taken,	and kept	t from	Scopus	search
engine										

	Plain			Size			SA			RT			Equil		
	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K
Microplastic+H+C	128	9	5	52	1	1	21	0	N/A	1	0	N/A	14	0	N/A
NOM+particle+H+C	138	6	4	73	1	0	17	0	N/A	0	0	N/A	16	1	0
	Bioacc														
	ŀ	Bioa	ee		Fat	e	Г	oxic	city		Free	sh		Tot	al
	P	Bioa T	cc K	P	Fat T	e K	Т Р	Toxic	city K	Р	Free T	sh K	Т	Tot	al K
Microplastic+H+C	Р 22	Bioa T 0	K K N/A	Р 28	Fat <i>T</i> 0	e K N/A	Т Р 30	T	K K N/A	Р 16	Free <i>T</i> 0	sh K N/A	T 10	Tot	al K 6

Table 4. Quantitative search results of articles produced, taken, and kept from Google Scholar search

	Plain			Size			SA			RT			Equil		
	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K
NOM+particle+H+C	-	9	0	-	6	2	-	4	0	-	5	3	-	3	2
Microplastic+H+C	-	11	7	-	3	2	-	5	5	-	1	1	-	2	2
	Bioacc			Fate			Toxicity			Fresh			Total		
	Р	Т	K	Р	Т	K	Р	Т	K	Р	Т	K	Т		K
NOM+particle+H+C	-	10	6	-	2	1	-	0	N/A	-	6	1	45		15
Microplastic+H+C	-	4	4	-	1	1	-	3	3	-	3	3	33		28

Remaining articles were then analyzed for available physical characteristic data including size, shape, SA, porosity and age, and any information on particle

chemistry. Information on combined particle and HOC toxicity to biota were also extracted. While in some cases articles on this theme did not provide comprehensive information on particle characteristics, they were kept if they provided quantitative or qualitative data on particle influence on HOC bioavailability.

Once data on physical characteristics was assimilated, the range of each value for each particle type in the MP and NOM classes were given and the median calculated. In the case of studies who found that certain measurements were below their limit of detection, a 0 value was used in the median calculation. Additionally, those articles who presented values as a range (particularly for size), the mean value of the range was used when calculating the median. In the case of MPs where particle aging was considered, the number of articles confirming the use of virgin or aged particles were compared to the total number of articles with information on age to obtain the percentages.

3. Results

3.1. Particle properties

3.1.1. Size

The size ranges presented in Table 5 from the results of the literature search did not differ dramatically between the various MP polymer types, with nearly all spanning the entire acceptable size range from 0.1μ m-5mm. Their medians however did vary, with PA possessing the smallest value, followed by PVC and PS over double the size of PA, then PE and PP. PET had the largest median at over five times that of PP.

	Si	ze	Surfa	ce Area	Por	osity	Age			
Particle	Range	Median (µm)	Range (m²/g)	Median (m²/g)	Range (mm ³ /g)	Median (mm³/g)	Virgin (%)	Aged (%)	# articles	
PE	1μm- 5mm	180	n.d 7.16	0.35	n.d 4.9	0.29	93.3	11.1	45	
PS	0.1µm- <5mm	125	n.d 15.62	0.87	n.d 20	1.15	88.6	18.2	44	
PVC	<1µm- <5mm	112.5	n.d 9.77	0.86	n.d 1.24	0.48	94.7	15.8	19	
РР	20µm- 5mm	275	4 ⁻⁴ - 0.96	0.34	n.d 1.72	0.92	88.9	16.7	18	
РА	10μm- 4mm	57,64	1.7 ⁻³ - 0.86	0.43	0.13- 0.46	0.29	90.9	9.1	11	
PET	<15µm- <5mm	1575	0.49- 0.86	0.68	0.42- 1.83	1.13	85.7	14.3	7	

Table 5. Physical Characteristics, age and concentration of environmentally relevant studies for different MPs extracted from literature search

In the case of NOM materials, suspended sediments, soil, and bottom sediments possessed the most similar and greatest range in particle size. The median for suspended sediment however was close to double the value of the latter two materials. The range from the literature search of these three NOM categories all overlapped into both the fine and coarse particulate size definitions (i.e. >1mm and <45 μ m), though the medians all remained in the POM size class. The range for the biota category was much smaller remaining below 100 μ m, though the median leaned towards the smaller end of the range at 0.7 μ m in the DOM size range. Wood as the last category had only one article which cited it specifically, with the material being <63 μ m.

	Size		Surface Area		Porosity		
Particle	Range	Median	Range (m²/g)	Median (m²/g)	Range (mm³/g)	Median (mm³/g)	Origin
Suspended Sediment	0.02μm- 2.8mm	200.5	1.7	1.7	N/A	N/A	N/A
Biota	0.2-80µm	0.7	N/A	N/A	N/A	N/A	e.g. Mixed estuarine plankton, Selenastrum <i>capricornutum</i> algae, bacteria
Soil	0.7µm-2mm	124	1.6- 11.49	6.58	10-18	10	e.g. grassland/ forest, "topsoil vegetation", biological settlement from flood periods, fine roots
Bottom Sediment	3µm-<2mm	93.75	0.26- 59.7	5.82	N/A	N/A	e.g. wetland plants, marsh plants, coal, faecal pellets, vegetation, bacteria, algae
Wood	<63µm	63	0.2	0.2	N/A	N/A	Driftwood

Table 6. Size, surface area, porosity and origin of different natural POM extracted from literature search

3.1.2. Surface area

The SA range for MP materials was largest for PE, PS and PVC, from being undetectable or explicitly non-porous to near or over $10m^2/g$. The ranges of PP and PA were much smaller below $1m^2/g$ though still detectable. PET had the smallest range of all MPs of only $0.37m^2/g$ between values, though it also remained below $1m^2/g$. The median values therefore followed the order of PS> PVC>PET> PA> PE> PP, a trend which did not reflect the expected relationship to median MPs sizes of smaller particles having larger SAs (size being PET> PP> PE> PS> PVC> PA). Some degree of relationship however could be observed if the orders are divided into the first and last 3 plastics. PS and PVC found in the middle/smaller end of the size range possessed larger SAs, and PP and PE in the middle/larger size of the MPs had the smallest. The exceptions lied with PET possessing the largest median size and a larger SA, and PA having the smallest size but one the smaller three SA values.

SA for NOM materials was a characteristic far less reported than for MPs. Only one article examined this characteristic for both suspended sediment and wood producing the two smallest values, whereas no information was provided for biota. The ranges for bottom sediments and soil differed by $\sim 1m^2/g$ on the lower ends, but the former far outweighed the latter on the larger end by a factor or nearly 5 (59.7 m²/g versus 11.49 m²/g). The median of these two however was larger for soil, which also had a larger median size compared with bottom sediments. The resulting order for median SA therefore followed the order of soil> bottom sediment> suspended sediment> wood.

3.1.3. Porosity

The median porosity (i.e. pore volume) for plastics followed the order of PS> PET> PP> PVC> PE=PA. Interestingly, though the difference between them was only 0.02mm³/g, the range for PS far exceeded that of PET, particularly on the larger end by nearly ten times. Similarly for the smallest median porosities of PE and PA, the range of PE was greater by over ten times on the larger end. Some similarities between porosity and SA were observed, with PS possessing the greatest value of both, and PE being in the smallest range for both characteristics. For other plastics no clear relationship was observed.

Porosity was not a physical characteristic described for NOM with the exception of soil, ranging between 10-18mm³/g with median 10mm³/g. Data for the other categories for this review therefore remains unknown.

3.1.4. Particle age and source

Plastic age was categorized as either "virgin" or "aged" based on the experimental details given by authors. Virgin plastics were either bought from manufacturers or isolated from purchased products (e.g. personal care products like a facial scrub), while aged plastics included those that were taken from the field or artificially aged using UV light/chemical oxidation. Over 80% of articles for all six plastic types used virgin materials.

Depending on the style of experiment conducted and aims of the study, identifying the source of the various POM categories was either extremely specific or not done at all, though the different POM sources for each category were generally far more diverse than those of MPs. The origin or identification of suspended sediment was identified in one of five articles, while specific algal and bacterial species were given for biota. Soil experiments provided the most information on the source of POM, with samples being taken from various geographic land types (e.g. farm field, grassland, forest, etc).

3.2. Data availability for selected properties

A total of 100 articles were found for this literature review, however many articles reported the use of multiple MPs and/or NOM types in the article. Values in Figure 3 below therefore depict the number of articles including individual particle types, but not the total number of articles found in this search. The use of different plastics was generally far greater than for NOM materials in the articles found, with the exception of PA and PET which were the least frequently used MPs. Size was reported for all articles as it pertained to the selection criteria of this review. PE and PS were the most commonly used MPs with 45 and 44 articles citing them respectively, after which the number of articles then decreased by more than 50% for the other plastics. Natural particles were also far less commonly used than PS and PE, bottom sediment as the most cited with only a quarter the number of articles at 12 total, and wood having only one instance of use.

SA was the second most highly reported characteristic for all particles, though for only \sim 50% or fewer papers. For already underrepresented particles this could reflect a bias in the ranges and median values for the chosen characteristics and explain a lack of visible trends. Porosity was the least reported characteristic among all articles, with approximately half the number of papers as SA, and in the case of NOM only in 2 articles.



Figure 3. Number of articles reporting on each particle type and physical characteristics

3.3. Biological impacts

The values in Figure 4 below depict a tally of positive/neutral and negative effects on biota observed in the articles of this review after exposure to either MPs or POM in combination with one or many HOCs. Positive effects were judged as those resulting in reduced mortality or consequential health effects with the presence of MP/POM particles compared with their absence, while the opposite was true for negative effects. Neutral effects were those where the authors observed no change in HOC availability and toxicity with particles present compared with their absence. If both positive and negative effects were observed within an article they were independently accounted for, hence the number of cited impacts being smaller or larger than the number of articles in certain cases.



Figure 4. Counts of positive/neutral and negative effects relative to control groups observed for different particle types from the literature search

A total 24 positive/neutral changes and 27 negative changes occurred as a result of particle presence in combination with HOC exposure to biota. MPs had 20 positive and 18 negative changes, while POM accounted for 4 positive and 9 negative. The tally of positive effects was larger than or equal than negative for MPs with the exception of PE. Following the trend in Tables 5 and 6, literature using plastic was more abundant than NOM, in the order of PE > PS> PP=PVC> PA. In the case of POM, bottom sediments had the highest number of cited negative incidents for the POM category, followed by soil. Biota and suspended sediment on the other hand recorded 2 and 1 incidents of positive/neutral effects with no negative counts in the two articles found.

4. Discussion

The following section will more closely examine the physical properties of POM and MPs obtained from the literature to determine their similarities and differences. The sorption of hydrophobic pollutants onto particles in the environment is highly dependent on the characteristics of both sorbent and sorbate (Tourinho et al., 2019), however for the sake of simplicity particle characteristics alone will be examined and in a broader sense. Properties outlined here will also be discussed somewhat separately, however it should be emphasized that they vary heavily together. Other characteristics not retrieved from the literature and some influential external environmental factors will later be introduced, and together an effort to interpret these finding will be made.

4.1. Influence of selected particle properties and biological impacts

4.1.1. Size

The articles of this literature review were chosen if particles therein ranged between the lower and upper limits of the size classifications for both particulate NOM $(0.45\mu\text{m}-1\text{mm})$ and MPs $(0.1\mu\text{m}-5\text{mm})$. As these ranges were part of the screening criteria, a value for this characteristic was consistently given for all articles regardless of particle type. In the case of MPs, size was determined either through given manufacturer labels, the use of different microscopes (SEM microscopy in the case of smaller particles), or occasionally the use of sieves to isolate a certain size range. POM sizes on the other hand were most frequently determined through sieving to separate particles of different sizes.

The premeditated size selection criteria contributed to MPs and NOM being comparable for other characteristics, but as a consequence somewhat limits discussion of this characteristic itself within the confines of this review. The exception lies in that MP size boundaries extend beyond those of POM for both its upper and lower limits, seen in Tables 5 and 6 with all plastic types possessing a larger range than all NOM categories. While still acceptable within the definition of a MP up to 0.1µm, below the 0.45µm size NOM is categorized as being in a dissolved state (DOM) or the more ambiguous colloidal size range. On the other end of the spectrum, POM above 1mm in size is classified as a "coarse" particle. DOM has been well characterized as material with high sorptive capacity for environmental pollutants (Pan et al., 2007; Ma and Yates, 2018), but is inherently different to its "particulate" counterparts (Loh et al., 2006). Along with being smaller in size, it differs in the ratio of molecular components (e.g. macromolecules and humic substances) after undergoing a greater degree of degradation, in turn imparting a highly recalcitrant nature compared to POM (Gourlay et al., 2005). It is generally observed in comparing two particles of the same material that the smaller will have a greater sorption capacity due to it possessing a larger specific SA (Ma et al., 2019; Liu et al., 2020d). When comparing different materials however, it becomes more difficult to draw this same conclusion as alterations in other characteristics equally important for sorption must be considered. As it is the intention of this review to compare particulate NOM and MPs however (to the exclusion of DOM), the inclusion of results from MPs between 0.1µm and 45µm could result in the appearance of them having a larger sorption capacity compared with POM. The same is true for the opposite end of the size spectrum, where comparing larger sized MPs to smaller POM material can shift the appearance in favour of POM. This is not likely to be the case in this review however, as information on NOM in the coarse size range was much less available

Interestingly and in somewhat of a contrast to the possible situations outlined above, the median values of the two particle classes were comparable for many categories despite the fact that their ranges differed greatly. Examples of similar values are PE and PP being comparable to suspended sediments, PS and PVC to soil, PVC also to bottom sediment, and even PA with wood. These results are in part the due to the number of articles contributing to each particle types and should not be taken as a reflection of all reality in the environments, however they demonstrate that there is likely a large overlap between MP and POM sizes occurring within natural systems. Considering a large portion of MP based articles used materials in the in the range of $100-500\mu m$, it could be suggested that many primary or virgin microplastics entering the environment are already in at a comparable size status as POM.

An additional similarity between natural and anthropogenic OM is the change in size and shape that occurs to both particle groups through degradation or weathering over time. With greater exposure in the environment, particles are increasingly subjected to UV radiation, physical battering, bacterial degradation, etc. (Hartmann et al., 2017). In experiments using artificial weathering with UV light or chemical agents, MP particle surfaces were altered by the breaking of bonds in their matrix and ultimately allowing the loss of small pieces of material over time (Mei et la.,

2020) and changing their size. While these physical and chemical processes are also influential for POM, the primary action responsible for the changes in size of natural materials is bacterial degration (Hu et al., 2018). The time required for the breakdown of each material is also significantly different. Plastic materials are created with the express intention of being durable, and as such require extremely long periods of time to degrade (Barnes et al., 2009), suggesting that plastics are likely to remain in their 0.1-5mm class for a very long period of time. The breakdown of POM however is highly variable between and within materials, for example comparing labile macromolecules in plant materials broken down in a matter of days with complex lignin and cellulose in requiring much longer (Elosegi and Pozo, 2016). Different species of biota can also vary, for example gram positive and negative bacteria being resistant to degradation owing to the peptidoglycan in their cell walls (Jørgensen et al., 2003), but the amount of this protein varies between species. These differences between NOM categories shows that some materials with more resistant constituents will remain in the POM size range for longer periods of time, yet compared to the decades required by MPs are far less recalcitrant as a whole. A caveat in this resistance to degradation is the contrast in material influx of each group to the environment. Whereas individual MP particles will have a longer lifetime before reaching nano sizes, the constant input or cycling of natural materials ensures that there will be plenty of particles in the POM size available in the same media to compete for HOCs.

4.1.2. Surface area

Surface area (SA) as a property describes the external area available for HOCs to make contact, thereby dictating the quantity of pollutants can be sorbed to its surface. From the results of this review, SA values for POM were far greater than those of their MP counterparts by several orders of magnitude. For natural materials, the order was soil> bottom sediment > suspended sediment> wood, with articles using biota not providing information, while MPs as a group followed the order PS> PVC> PET> PA> PE> PP.

Particles of the same size but with more irregularly textured surfaces possess larger SA values reflecting available sorption sites (Fang et al., 2019) and greater sorption potential (Elizalde-Velázquez et al., 2020). Descriptions of particle shape or texture were only given in the case of MPs, the most common shapes being smooth "microspheres/microbeads" for primary plastics or "fragments/granules" from mechanical grinding to uniform size or extraction from field samples. While no direct descriptions were given for NOM classes, it is understood in the case of many groups (e.g. soil, sediments) that samples are heterogeneous mixtures with components of various shapes. OM alone within this mix is observed to have an amorphous shape due to the wide array of complex molecular structures found

within (Filella, 2009; Schaumann, 2005). The quicker degradation of more labile components such as polysaccharides, proteins, etc. that leave behind more resistant components (organic acids, lignin) forms sub-nanometer sized pores responsible for a rough texture (Ran et al. 2013). The uneven distribution of these fractions and pores within a particle can further contribute to increased surface irregularity. In addition to these pores, further enlargement of available area can occur when previously terrestrial OM components are exposed to water, causing aggregates to alter their configuration through swelling (Weber et al., 1998) and the unfolding of previously compact structures (Vialykh et al., 2019). These changes in structure and the highly amorphous nature of OM in comparison to MP's ordered and repeating lattice structures (Li et al., 2020) can therefore help explain the results of POM having dramatically larger median SA values compared with plastics (Mai et al., 2018).

When comparing the entire range of SA for the two particle categories, certain plastic groups can be seen to overlap with the NOM groups. Linked to changes in size with increased weathering, MP shape and texture can also be altered with exposure to environmental elements over time. Like NOM, surfaces will develop irregular folds, dents, larger pore spaces and voids, expanding usable sorption space (Ding et al., 2020; Atugoda et al., 2020; Vockenberg et al., 2020). With sufficient exposure time to develop a greater degree of irregularity (Wang et al., 2020b), MPs such as PS, PVC and PE can obtain SAs that match their amorphous natural counterparts (Li et al., 2012). The abundance of the two materials must of be considered if considering the impact of larger SAs on vector capacity in the context of real systems, however the results of this review demonstrate that MPs and POM on an individual scale may have the same HOC sorption potential with regards to SA after varying degrees of time and weathering.

In addition to surface texture and shape above, particle size heavily influences SA. For example, MPs with the same polymer composition but smaller size possess a greater sorption capacity as the result of their greater SA to volume ratio (Liu et al., 2017; Elizalde-Velázquez et al., 2020). The overlap in POM and MP sizes was previously mentioned, but it should be noted that the difference between the upper limits of each particle's range is far greater than the difference found on the smaller end. The use of larger MPs with smaller SA to volume ratios in certain articles could therefore be a contributing factor to MPs as a group possessing smaller median SAs.

Another explanation for the gap between the median values of the two material groups is sample heterogeneity. In addition to the diverse makeup of NOM from its internal structure, the heterogeneity of the entire sample being used can contribute to greater observed SA values (Karapanagioti et al., 2000; Zhao et al., 2012).

Organic matter is the major factor responsible for sorption capacity in soil/sediment samples (Zhai et al., 2020), however they will also contain inorganic mineral components with varying SAs themselves (i.e. clay has a much higher SA than sand or gravel) (Li et al., 2012). Though attempted to be excluded from the selected literature, other organic carbonaceous material (e.g. kerogen and coal materials, black carbon from burning) are likely also present in the majority of samples (Moyo et al., 2014; Golding et. al, 2005; Ran et al., 2013). These materials are well known to possess large SAs that would contribute to a sample's higher range or average (Karapanagioti et al., 2000).

While unlikely to greatly skew the comparison between natural and anthropogenic materials, it is important to note the discrepancy in the number of articles supporting the median values for each of the two categories, which almost certainly had a great impact on the ordering of individual materials therein. In the MP category for example, PE was observed to cover a large range of SAs supported by 19 articles, having some of the highest values (Wang and Wang, 2018b; Hüffer and Hofman, 2016; Liu et al, 2019a), but also had instances of undetectable or extremely low values due to very smooth surfaces (Wang et al., 2015; Xu et al., 2019; Ziajahromi et al., 2019). These variations could be the result of aging or differences in their production process, such as in the case of HDPE versus LDPE (Vockenberg et al., 2020), but together show the breadth of SA measurements available for these materials. PET on the other hand had only 3 articles measuring SA, all of which provided large values relative to other plastics (Liu et al. 2019a; Liu et al., 2019b; Liu et al., 2020b). This could perhaps be a true reflection of the material having a large surface but could equally be an inaccurate representation of the true nature of PET. For natural materials the number of articles were even fewer than for plastics, the exception being bottom sediment with 4 articles. Reasoning as to why these materials would possess have higher SAs is available as seen above, however to substantiate the true magnitude of each material's SA much more data is required.

4.1.3. Porosity

Porosity (i.e. pore volume) as a physical characteristic is dictated by the structural composition of the material under consideration. It influences the type of sorptive action that occurs on a particle surface (Liu et al., 2019c) and the likelihood of partitioning into the particle matrix (Hu et al., 2018). Pore size is also important in these sorptive actions, with smaller pores favouring the formation of HOC monolayers on a particle surface while larger pores encourage multiple layers to form (Tourhino et al., 2019). Porosity was occasionally determined for plastics in the articles of this review, while for POM soil was the only material in which this property was given with 2 studies choosing to quantify it (Xiang et al., 2019; Chen et al., 2021). In comparison to plastics however, the values given far exceeded those

of MPs, ranging between 10 and $18 \text{mm}^3/\text{g}$ (median $10 \text{mm}^3/\text{g}$) compared with the highest median porosity of PS at $1.15 \text{mm}^3/\text{g}$.

The large pore volume in soil originates from NOM matrices within the organic fraction (Ran et al., 2013) and represent both the quantity of organic matter present and degree of aggregation. Like size, shape, and SA before it, porosity is subject to change with exposure to environmental influences. The humic acids present in soil and other POMs rich in decomposing plant materials have large micropore diameters but are not particularly stable when interacting with water. The aforementioned swelling of POM and HAs in particular is largely the result of pore size increasing with the introduction of water molecules (Weber et al., 1998). More rigid materials such as kerogen on the other hand will retain their shape (Weber et al., 1998). These microporous behaviours exhibited by NOM are important in understanding sorption processes (Ran et al., 2013), yet much that remains unknown and conjectured. Until further knowledge is ascertained, it could be speculated that POM material mixtures offering both rigid and "flexible" material could potentially offer access to more stable porous sorption sites than those of more rigid MPs alone.

Certain plastics such as PS can be intentionally designed with high porosity for items like packing foam where the greater proportion of air within the pores absorb impacts, hence it possessing the largest median value is not surprising. Its pores like those of certain soil OM fractions can also be reshaped when exposed to the environment. Zhang et al. (2018) observed that MP foams retrieved from the environment had larger pore sizes compared with virgin MPs, however that pore volume decreased with weathering and predicted this to be the result of reshaping due to erosion. Though not representative of the median results for this review, this decrease in pore volume with increased weathering could explain why certain MPs with the largest SA medians are not always accompanied by a higher median porosity value and vice versa.

A more likely for the divide between MPs and POM is the fact that certain microplastics are designed as initially nonporous or with very low porosity as in the case of PE, PVC, PP and also PS. This initial lack of pore volume alone is sufficient to create a very large difference in the median porosity values between each of the different plastic groups as well as between MPs and POM materials. Plastic's resistance to degradation and the longer period of time required for changes to present themselves in a MP surface or matrix would also slow down pore enlargement, further contributing to the large disparity in porosity between the two materials. From the results of this review it would therefore be assumed that "POM" possesses a larger porosity than MPs, however the small number of supporting

articles for these findings mean that this cannot be confirmed and requires more in depth research on this characteristic alone.

4.1.4. Particle influence on HOC bioavailability

Arguably one of the principal reasons for research on particle vector behaviour is to better understand the impacts on aquatic biota and their respective food webs. The results from this review in Figure 4 are evidence that both natural and anthropogenic particles are capable of producing harmful and neutral/beneficial results when exposed to biota in combination with HOCs. As the quantitative results presented here only correspond to counts or "incidents" of observed effects, they should not be considered a true reflection of the magnitude and range of effects these particles have in a real-world setting. However, the nearly even divide of positive and negative effects in both particle categories is accurate in demonstrating the variety of results obtained by researchers, and the lack of consensus on the magnitude of biological impacts. A further in-depth analysis of experimental set ups and resulting impacts for different particle types would help better decipher the mixed results currently found in the literature.

Though not embodied in the results, whether or not particles will enhance the bioavailability of contaminants or reduce their risk is still linked to physical and chemical properties that favour sorption, as well as external environmental elements. Of the previously discussed characteristics, particle size and the influence of age/weathering have the greatest impact on bioavailability from MPs (Ma et al., 2020), dictating both HOC carrying capacity and likelihood of particle ingestion as a food source. POM in the 45µm-1mm range is a natural food source for a variety of biota (Bundschuh & McKie, 2015), therefore it is reasonable then to assume that MPs in this size range are also targeted for consumption by these same organisms, though feeding strategy will dictate the exact sizes (Kukkonen and Landrum, 1996). Due to their larger SA:V ratio, smaller particles of the same material possess a greater HOC carrying capacity and as such are more likely to increase delivery of HOCs to biota (Ma et al., 2016). Smaller MPs <125µm pose further HOC uptake risk as they are capable of being assimilated into organs, thereby transferring HOCs directly into tissues (Rainieri et al., 2018). As POM is normally consumed in real world settings along its entire size range by different organisms, the risks associated with size are less of a concern than particle composition and age dictating carrying capacity (Ahrens et al., 2020).

Particle age and degree of degradation can impact bioavailability (via ingestion) by determining how well the material in question is likely to sorb HOCs. For POM, "younger" and larger materials possess more hydrophobic domains (Frankki et al., 2007) from a greater abundance of labile macromolecules rich in more neutral

functional groups compared with more recalcitrant and polar humic fractions after degradation (Schulten et al., 1991; Verrengia Guerrero et al., 2003). Digestion and absorption of these constituents facilitated by digestive surfactants that solubilize hydrophobic components can help in the uptake of HOCs upon consumption (Ahrens et al., 2001). Virgin plastics also have a more hydrophobic nature prior to weathering (Li et al., 2018a), but the introduction of polar surface functional groups with oxidation over time similarly reduces HOC sorption in favour of more polar contaminants (Liu et al., 2019d). Zhang et al. (2018) for example found that aged/oxidized PS foams sorbed double the concentration of polar pollutant oxytetracycline compared with virgin PS, while Müller et al (2018) observed decreased sorption of hydrophobic benzene, toluene, etc. on PS as a result of increased surface polarity. For both particles then, older and weathered particles can carry a lower risk of transferring HOCs as a food source than their "younger", more hydrophobic versions.

Positive/neutral results observed in labs are those that occur when particles reduce or do not alter HOC exposure to biota. These can occur for the same reason as above if particles that favour the sorption of HOCs remove available pollutants from the environment (Ma et al., 2020) and are not consumed. This removal from samples could be due to particles not being fully equilibrated with the surrounding environment (Mohamed Nor and Koelmans, 2019) or them possessing a greater sorption capacity than more favourably consumed media. Wang et al. (2019a) for example observed that increasing concentrations of PE microplastics in soil decreased HOC accumulation in E. fetida, though which of the mentioned processes is responsible remains unclear. Another possibility could be that as organisms magnify HOC concentrations to a higher degree than the aquatic environment, therefore equilibrium reverses sorption of HOCs from intestinal media to ingested particle surfaces upon digestion (Koelmans et al., 2013). This possibility requires further investigation to understand its relevance in the environment (Ma et al., 2020). As an example, Gourlay et al. (2005) proposed that the lack of HOC bioaccumulation in D. magna by HOC carrying bacteria and algae was due to the strong affinity of the HOC for the biogenic material later excreted in faecal pellets. Whether particles sorbed previously ingested HOCs or previously attached contaminants were not released remains unclear.

Though outside the scope of this review, other factors must be considered when interpreting results on HOC-particle interactions with biota. If ingestion is the main contact route for organisms as mentioned above, the concentration of contaminated particles available to organisms and time required for digestion needs to be considered when interpreting lab results (Kukkonen and Landrum, 1996). Intestinal retention time can range between hours and days (Mohamed Nor and Koelmans, 2019), changing within and between trophic levels. In this case there may be

insufficient time for HOC desorption in intestinal tracts to occur (Mäenpää et al., 2003) and as such this factor should be clarified in the experimental design process to ensure better interpretation of observed results. Considering other exposure routes is also important in understanding how particles can alter HOC-associated impact. Rodrigues et al. (2019) describes additional routes such as particles contacting external organism surfaces (e.g. skin, exoskeletons), and indirect exposure from dissolved HOCs in aqueous solutions contacting external or internal surfaces (e.g. gut linings). In these cases as well as for ingested particles, the likelihood of particle-organism encounters and duration of contact should be taken into account, dictated by species' habitat (i.e. benthic or pelagic zones), the concentration of said particle in the environment, and its behaviour such as if they sink or float. Evidence for the role of MPs as a vector in experimental modelling studies has suggested that the role of MPs as a legitimate source of HOCs to biota is negligible in comparison with POM food sources and aqueous routes (Rehse et al., 2018; Gerdes et al., 2019; Ašmonaité et al., 2020). The higher concentration of natural food sources and other POM in the environment reduces the chances of encounters seen in at concentrations used in lab studies (Mohamed Nor and Koelmans, 2019). It is possible then that the contradiction between papers claiming more harmful effects is the result of authors using plastics of different shapes, sizes, and compositions, and more notably the reported concentrations used in experiment conditions (Yang et al., 2020b)

4.2. Additional particle characteristics and influential environmental conditions

In addition to the physical characteristics discussed above, other properties and environmental conditions for which no quantitative data were obtained in this review also play a significant role in determining the efficacy of particles as vectors. The following section will touch upon these factors in order to provide further insight on the behavioural similarities and differences between MP and NOM, though further research into these specific characteristics and conditions is required for reliable conclusions to be drawn.

4.2.1. Crystallinity

Crystallinity is the term used to describe the degree of organization within the lattice structure of MP materials (Mei et al., 2020). This characterization is not normally constant throughout a polymer, with them often containing both highly ordered "crystalline" and loosely arranged "amorphous" domains (some standard classifications are visible in Table 1.) Amorphous regions are unique in that they exhibit different behaviour with a specific temperature (glass transition temperature, GTT in Table 1) (Tourinho et al., 2019), below which the region will be in a "rubbery" state (movement around the central carbon chain is possible), while above the GTT in a "glassy" state movement is severely restricted (Endo and Koelmans, 2016). As the majority of HOC sorption occurs in these regions (Teuten et al., 2009), "rubbery" plastics at room temperature like PE and PP can encourage greater sorption as they can more easily accommodate HOCs on or into the polymer structure (Gong et al., 2019a), while the opposite holds true for "glassy" plastics such as PVC or PA (Wang et al., 2015; Hartmann et al., 2017; Liu et al., 2019b). Although there no exact comparison for the above terms in NOM, according to Ran et al., (2013), natural particles that include condensed organic matter (e.g. humic substances, kerogen, and coal) can behave equivalently to glassy sorbents, and conversely black carbon as a nanoporous amorphous sorbent. Depending on the concentration and ratio of these materials within different classes of POM, certain plastics such as PE and PP could provide a more favourable sorptive surface that more glassy natural materials, while oppositely POM could have a slightly greater sorption capacity compared with PVC or PA depending on environmental temperature.

4.2.2. Chemical composition

Together with physical characteristics, the chemistry within and on a particle's surface will govern how well a material may sorb contaminants from its surroundings. Chemical structure and polarity driving hydrophobicity are determined by functional groups, in some cases having the greatest influence on sorption behaviour (Mei et al., 2020). Table 1 gives a sample monomer structure for each plastic group- those with long stretches of aliphatic carbon chains (PE) or with substituted methyl groups (PP) or aromatic rings (PS) endow a non or weakly polar characteristic compatible with hydrophobic pollutants. Other functional groups such as chlorine (PVC), amino groups (PA) or ethylene terephthalate (PET) bestow a more strongly polar nature that opposes sorption by non-polar HOCs. With environmental exposure and weathering, plastic polarity will also increase though the introduction of oxygen rich functional groups (e.g. C=O) (Zhang et al., 2018), again decreasing the affinity for more hydrophobic pollutants. Like MPs, the composition of NOM materials is variable depending on the source under consideration (Ran et al., 2013), but decaying plant material common to most POM groups can be used as an example. Various macromolecules from different plant structure (e.g. lipids, polysaccharides, amino acids) are composed of long carbon chains, phosphate groups (-PO4) and hydroxyl groups (-OH). More complex structures such as the earlier mentioned humic and fulvic acids groups are also abundant, and though the exact composition of these groups is less well understood, they are primarily composed of polar carboxylic acids (-COOH), hydroxyls and carbonyls (Schulten et al., 1991; Verrengia Guerrero et al., 2003). The abundance

of polar groups in more degraded POM lends particles a slightly negative surface charge (Bradford and Horowitz, 1988) which can oppose the sorption of more hydrophobic pollutants in favour of plastics with more non-polar surface regions. In this regard, MP are generally more favourable for HOC sorptive action than their natural counterparts. To make a true judgement on sorption capacity however, the makeup of the pollutant in question is important to consider (Tourinho et al., 2019), as affinity will change between chemical species and certain specific functional groups are capable of stronger sorptive interactions not driven by hydrophobicity alone

4.2.3. Intermolecular interactions and sorption pathways

Interactions between particles and HOCs can be divided into hydrophobic partitioning and surface sorption mechanisms. The former is a dynamic process of mass transfer from surrounding aqueous media to a solid phase, reaching equilibrium between the two phases over a period of time (Tourinho et al., 2019) while the latter occurs through multiple attractive forces between individual molecules. Though capable of participating in both sorptive routes, the most important mechanism of HOC association for NOM materials (particularly sediments and soil) is hydrophobic partitioning in hydrocarbon rich regions (Verrengia Guerrero et al., 2003; Frankki et al., 2007). As these materials are often rich in polar acids mentioned above capable of repelling highly hydrophobic pollutants, it is reasonable that partitioning out from highly polar aqueous media and away from polar material regions is the dominant sorption mechanism rather than attractive forces. Compared with NOM, MPs are generally less abundant in these highly charged groups, and interactions are greatly influenced by electrostatic and van der Waals forces (Hüffer et al., 2019; Li et al., 2018b). As stated before, the composition of each material and the HOC under consideration impact the strength of interactions that occur, in many cases there being an overlap between natural and anthropogenic particles. For example, the plastics PA, PET, and weathered plastics groups can undergo H-bonding because of the presence of their polar oxygen containing groups, and likewise for polar POM groups (Guo et al., 2019; Liu et al., 2020b; Isaacson and Frink, 1984). Others such as PS and PET rely heavily on π - π stacking interactions between their aromatic rings (Hüffer and Hofmann, 2016; Liu et la., 2020d) (a form of non-colvalent attraction resulting from their π orbitals), as do more aromatic fulvic acids. For both material classes the presence of these groups increases their affinity for aromatic HOCs (Zhang et al., 2018). The strength of sorption between MPs and NOM categories is therefore highly dependent on the makeup of each sorbent and sorbate, however younger, more hydrophobic particles as a generalized group are a more favourable surface for HOCs than their hydrophobic POM counterparts.

4.2.4. Environmental Conditions

The environmental conditions surrounding particles and pollutants can alter the strength and possibility for sorption between materials, therefore understanding true vector behaviour requires that these conditions be taken into account. One example of these conditions is the presence and formation of biofilms that form an integral part of diverse aqueous systems (Wicke et al., 2008). Up to 50% of materials in the environment are colonized by organisms (Barnes 2002; Ma et al., 2020), and it has been suggested that their presence may concentrate HOCs to a greater extent than bare particle surfaces alone (Wicke et al., 2008). Their growth is slower on more hydrophobic surfaces (Karlsson et al., 1988) than their more hydrophilic components (Wu et al., 2016) as available in POM, and as such could potentially raise the sorption capacity of POM to match that of younger hydrophobic MPs. DOM is another environmental constituent capable of both increasing and decreasing particle sorption capacity. The former can occur if DOM complexes with HOCs and then sorbs to a particle surface (Liu et al., 2020a; Zhang et al., 2018), while the latter can be the result of either DOM competing for HOCs and reducing the concentration available prior to an encounter with larger particles (Wu et al., 2016), or alternatively DOM competing for sorption sites on MP or POM particle surfaces. When DOM is excluded from experimental design, the results of particle sorption seen in laboratory results can exceed those occurring in real aqueous systems and as such should not be neglected. As a final example, environmental pH dictates particle surface charge and potential for electrostatic forces critical in HOC-particle interaction (Fang et al., 2019). Increases in pH can encouraging H+ dissociation from neutral surfaces to become negative and hydrophilic (reducing hydrophobic interactions) (Liu et al., 2019a), or promote greater π - π interactions (Xiong et al., 2018). Many MPs can be slightly negatively charged in more alkaline solutions (Guo et al., 2018) and conversely positive for acidic conditions, therefore the electrostatic interactions that occur with pollutants will change with differing environmental conditions to favour or lessen sorption activity. Likewise for NOM, alkaline solutions promote greater dissociation of acidic functional groups, causing the structure of OM to "open up" and increase HOC access to previously internal sorption sites (Isaacson and Frink, 1984).

4.3. Preliminary risk assessment

From the results of this review and the additional background knowledge, it is apparent that MPs and POM in aqueous environments share various physical and chemical properties, imparting them both the capacity to sorb hydrophobic contaminants from their surroundings. They also have differences that can alter the magnitude of this behaviour in favour of one group over another. In a broad sense, NOM characteristically has more amorphous shapes, larger SAs and pore volumes, but is less resistant to degradation resulting in shorter lifespans at the defined 0.45µm-1mm range. MPs in comparison possess a larger size range, a wide range of SAs and pore volumes, and are very recalcitrant by design. They are however initially much more hydrophobic than POM particles prior to weathering, and for both natural and anthropogenic particles this trait is deemed of high importance regarding sorption capacity (Kördel et al., 1997; Li et al., 2018a). There is some disagreement between authors as to which particle type possesses greater vector capacity based on the specific characteristics of the chosen particles, experimental conditions, and choice of HOC, however the majority observed MPs as being the more favourable sorbent (e.g. Mai et al., 2018; Wang and Wang, 2018a).

The level of plastic pollution entering aquatic systems has been increasing with each passing year (Bussolaro et al., 2019), higher concentrations in urban environments and near wastewater effluent streams being a common observation (Peng et al., 2017). Though treatment plants can eliminate up to 99% of MPs, the remaining 1% is thought to account for 0.21-1.5 particles/L (Ziajahromi et al., 2017; Vockenberg et al., 2020). In large urban centers this quickly adds up to large outputs, and in more rural and developing zones that do not receive treatment the MP output could be far greater than 1%. Heavy rain events leading to sewage spill overs or increased runoff from urban and agricultural areas can also increase the concentration of plastics in the environment. However, the abundance of naturally occurring OM in the environment far exceeds that of MPs (Ašmonaité et al., 2020) making their overall impact to natural systems as vectors comparably negligible (Wang and Wang, 2018a). In both fresh and marine waters, the ratio of plankton alone to MPs is thought to be greater by at minimum a factor of 10 (Frydkjær et al., 2017), or that of zooplankton between 20 times and four orders of magnitude larger (Gerdes et al., 2019). The highest concentration of MPs reported in the literature to date is $\sim 1.5 \mu g/L$ (Fang et al., 2019), and though this value encompasses the entire extent of the water column (which can vary in concentration from the surface to bottom floor), it is miniscule by comparison. Considering POM can also see flash events that greatly increase particle concentration above normal background levels, such as floods or algal blooms the disparity is greater still.

The risks posed by MPs should not be discounted if the trend in volume continues to rise as it has over the last decades, but the danger of MPs as vectors for HOCs is typically only observed at extremely high concentrations still far above those observed in environmental conditions (Beckingham and Ghosh, 2017). Studies conducted in natural systems more often find that natural food and water are the main sources of pollutants to biota because of the low, relative concentration of MPs compared to NOM and sediment particles (Beckingham and Ghosh 2017; Koelmans et al. 2016; Ma et al., 2020). Though the results of studies at lab

concentration are still noteworthy, other MP behaviour such as particle accumulation itself poses a larger problem (Bussolaro et al., 2019). Future research on transport of chemical contaminants in environmental systems should therefore emphasize the role of natural particles as vectors (both organic and mineral), particularly as there appears to be a lack of data on this subject in the literature.

4.4. Observations from literature review process

4.4.1. Keyword search

During the course of searching for relevant literature, it was noted that many results did not match the key words used as defined in this review. In particular, searches using "natural organic matter" and "particle" together produced many results pertaining to DOM rather than POM. It is understood that this is because there is no consistent size class definition for natural organic particles outside the field of stream ecology. Defining DOM as a "natural organic particle" is therefore a reasonable outcome, as is labelling mineral particles as natural particles. The lack of clarification on this separation between DOM and POM however generated far more results with promising titles and abstracts which did not reflect the reality of the particles used in the studies. Another somewhat surprising find from the search results was the large amount of MP centered studies that presented themselves when using the aforementioned search terms.

A further observation upon reading selected articles was the lack of information on the constituents of POM used in different experiments. When particles of the appropriate size were used, oftentimes their origin and composition was not clarified or researched, especially with reference to both bottom and suspended sediments. Though generalizations can be made using decaying plant material and biota, further analysis into the source of the OM under consideration could potentially reveal sorption trends for specific NOM classes over others that currently remain unclear. Also, greater insight into OM properties could help identify the magnitude of this fraction's role alone compared with that of mineral fractions also present within samples. OM is not truly isolated once incorporated into soils and sediment in the environment so results extracted from the articles used here are still somewhat insightful into the sorption capacity of these mixtures, but as the aim of this review to compare two organic materials the results cannot truly be fully attributed to OM alone. Considering the above points together, generally more in-depth research is needed in the area of larger natural organic particles as vectors (not just DOM) and their respective properties that are responsible for HOC transmission in the environment.

4.4.2. Experimental conditions in published articles

As mentioned in the above section, the experimental set up of the articles used in this review did not reflect true environmental conditions. By isolating specific compounds and particles in stable media, understanding what properties influence sorption strength and rate can be achieved for fixed particle-HOC combinations. Due to the simplicity of these conditions however, the complexity of real freshwater (or marine) environments is greatly overlooked. Environmental conditions like pH, salinity, DOM in solution etc. are continuously in flux based on climate, nutrient inputs and biological activity. Once mechanisms are understood for certain HOCparticle combinations under strict conditions, further research needs to expand on translating these results to more realistic conditions.

The variety of plastic shapes and age were also different than those most likely found in real world settings and should be researched in the future. As an example, in this review there were only two articles involving the use of fibres (Akindele et al., 2019; Guo and Wang, 2019) when it has been documented that MP fibres are among the most abundant plastic types in the environment (Wang and Wang 2020b). Insight into their behaviours as sorbents could provide more relevant information for urban water where these shapes are bountiful. Similarly, over 80% of each plastic type used in the selected articles were virgin material unexposed to environmental weathering which does not reflect the reality of items accumulating over extended periods of time. POM samples were taken from the field, and as such had all undergone some degree of environmental exposure though no dating was done to determine sample ages. The lack of relevant data in this area could further have contributed to the differences in quantitative values obtained for this review. Fortunately, there is progress being made with more recent articles examining the effects of weathering through artificial aging of particles, and if combined with research at more environmentally relevant concentrations a clearer picture of true environmental processes will be made increasingly clear.

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