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A review of the potential utilisation of plastic waste as adsorbent for removal of hazardous priority contaminants from aqueous environments

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Abstract

There is growing global awareness of the presence and negative impacts of waste plastic in the marine environment. Risks to wildlife include ingestion and entanglement for macro-plastic (larger than five millimetres in length), alongside food chain transfer for micro-plastics (less than five millimetres in length). Plastics in the marine environment have also been shown to adsorb and accumulate contaminants from seawater, e.g., heavy metals and hydrophobic organic compounds. This means that plastics can additionally act as vectors for transport of contaminants, permitting ecotoxicological risks to be spatially extended. However, the ability of waste plastic to adsorb pollutants also offers potential opportunity, if they can be used for the decontamination of wastewater. Here, we provide an overview of marine plastic types and distribution, and then systematically assess their potential to be repurposed as novel

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adsorbents. Data published in recent years are interrogated to gain an overview of the interaction mechanisms between marine plastics and both organic and inorganic contaminants. In addition, factors that may be exploited to enhance their performance in removal of contaminants are also reviewed and prioritised, e.g., surface modification and activation. This paper highlights the novel potential of repurposing plastic waste for wastewater treatment applications and seeks to identify key knowledge gaps and future research priorities for scientists and engineers.

Keywords: Adsorption; Plastic-based adsorbent; Water treatment; Material characterisation; Interaction mechanisms

1. Introduction

High quality water is fundamental to society and, because conventional water and wastewater treatment is generally not able to remove all emerging contaminants efficiently (such as flame retardants and pharmaceuticals (Geissen et al., 2015)), new and innovative treatment technologies are being sought (Cosgrove and Loucks, 2015). Non-conventional water treatment technologies (utilised by water and wastewater treatment plants (WWTPs)) can be broadly divided into three main groups: phase-changing technologies, modified biological treatments, and advanced oxidation processes (Rodriguez-Narvaez et al., 2017).

Adsorption processes are phase-change technologies that have proven attractive due to their simplicity of design; ease of operation; effective removal of pollutants at low

concentrations (e.g., μg/L or ng/L); and comparatively low cost (5-200 US$/$m^3$ of treated wastewater as compared to 10–450 US$/$m^3$ for advanced oxidation, reverse osmosis, ion exchange or electrolysis (Ali et al., 2012)).

In WWTPs, adsorption techniques are used to remove dissolved contaminants from the aqueous phase. Development and application of new adsorbents is needed as an increasingly diverse range of pollutants are detected in wastewater, e.g., pharmaceuticals, radionuclides, flame retardants, industrial chemicals, etc. (Balarak et al., 2015; Cuerda-Correa et al., 2010; Wang et al., 2019). In recent years, significant attention has been directed at identifying and assessing carbon based materials (Pang et al., 2019), especially adsorbents derived from low-cost and/or waste materials, i.e., those materials abundant in nature, or, a by-product or waste material from another industry (Ali et al., 2012; Bailey et al., 1999). Such adsorbents can have potential for the removal of dissolved pollutants, and since they are reused or sourced from waste, their use is consistent with the principles of 'reuse and recycling' and the 'circular economy'. As such, they represent an alternative to the traditional 'linear economy' (make, use, dispose), ensuring materials are utilised to extract their maximum value, whilst minimising resource consumption (De Gisi et al., 2016; Rae et al., 2019).

These low-cost adsorbents are highly diverse and principally sourced from agricultural and household wastes, industrial by-products, municipal sludge, marine resources, or soil materials (De Gisi et al., 2016). Many low-cost materials, such as crab carapace, apricot and plum kernels, orange peel, etc., have been suggested by researchers to

remove a wide range of organic and inorganic pollutants from aqueous solutions (Chowdhury et al., 2011; Rae et al., 2019; Tran et al., 2016; Turk Sekulić et al., 2018). In parallel, plastic pollution has now become a global environmental issue, as rapidly increasing production overwhelms the world's ability to deal with this waste. A key challenge with plastic is its lack of biodegradability, facilitating environmental persistence for hundreds of years (Lavers and Bond, 2017). In the marine environment, it is recognised that waste plastic can adsorb a wide range of pollutants including toxic hydrophobic, persistent, and bio-accumulative substances such as PAHs, polychlorinated biphenyls (PCBs), dichloro-diphenyl-trichloroethanes (DDTs), heavy metals, etc. (Bakir et al., 2012; Brennecke et al., 2016; Provencher et al., 2018). Adsorption onto plastic can then promote the long range transport of contaminants within the marine environment, extending their spatial presence and providing additional opportunities for food chain transfer and bioaccumulation (Bakir et al., 2014; Ivar Do Sul and Costa, 2014). However, this also opens up the prospect to exploit marine plastic waste as an adsorbent for contaminant removal from polluted water. This approach is particularly attractive given the scale of the marine plastic waste issue. If the natural affinity of marine plastics to adsorb pollutants can be effectively exploited, their reuse (as novel low-cost adsorbents) may also reduce their negative impacts on the marine environment.

This paper provides an overview of plastic waste in the marine environment, including all types of macro- and micro-plastics, and reviews their potential (as adsorbents) for

pollutant removal. The objectives are to identify the potential for the use of plastic waste adsorbents in water remediation; to highlight plastic/adsorbate interaction mechanisms; and, to identify optimal preparation techniques. The paper also seeks to identify key challenges for scientists and water engineers, and to make suggestions for further applied research and innovation.

2. Problems, distribution, and transport of waste plastics in the marine environment

Plastics are synthetic, organic materials polymerised from monomers extracted from gas or oil (Cole et al., 2011; Derraik, 2002; Thompson et al., 2009). The development of plastic began in the middle of the 19th century when English inventor, Alexander Parkes, produced waterproof woven fabrics (Shashoua, 2008). Later, polyvinyl chloride (PVC) was first polymerised by the German chemist Eugen Baumann in 1872, and is now the third most widely produced synthetic plastic polymer after polyethylene (PE) and polypropylene (PP). Meanwhile, Bakelite (patented in 1909) is regarded as the first real synthetic plastic to enter into mass production (Plastic Europe, 2016).

Global plastic production reached 348 million tons/year in 2017, and was dominated by five main types: PE (17.5%), PP (19.3%), PVC (10%), polystyrene (PS) (6.7%) and polyethylene terephthalate (PET) (7.4%) (PlasticsEurope, 2018). These materials and their diverse characteristics, including being lightweight, durable, inert, and degradation-resistant, have led to significant technological and societal advances (Andrady and Neal, 2009; Cole et al., 2011), i.e., in the fields of food packaging,

102 automotive manufacture, engineering, construction and medical treatment (Zarfl and
103 Matthies, 2010).
104 However, improper disposal/recycling practices associated with increasing global use
105 of plastics has resulted in extensive threats to the natural environment. Plastic waste,
106 can be divided into two categories: municipal and industrial (Lazarevic et al., 2010).
107 Municipal waste includes domestic items (food containers, packaging foam, disposable
108 cups, etc.); agricultural waste (mulch films, feedbags, fertiliser bags, etc.); wire and
109 cable, and automobile wrecking waste. It is normally a mix of PE, PP, PS, PVC and
110 PET. Meanwhile, industrial waste is more homogeneous in nature, resulting from
111 plastic manufacturing and from the automotive, construction and packaging industries
112 (Panda et al., 2010).
113 While some plastics are made from modified degradable polymers (like cellulose
114 acetate; (Mohanty et al., 2003), most plastics are not biodegradable – which ultimately
115 means that their life cycle tends to end at waste disposal points/facilities (Luo et al.,
116 2000). Fig.S1 shows the most common plastic waste management routes in use today.
117 Landfill and incineration are the two most common methods for plastic waste disposal
118 at the moment, but these in turn can cause ground- and surface-water contamination,
119 air, and soil pollution (Fujita et al., 2003; Hopewell et al., 2009).
120 Since the 1960s, there have been ever growing concerns over plastic debris in the
121 marine environment (Derraik, 2002; Fendall and Sewell, 2009; Moore, 2008). It is
122 estimated that plastics make up 85% of marine litter, and that 80% of plastic litter in

123 the marine environment is from terrestrial sources (Bourguignon, 2018). Other sources
124 include marine industry, commercial fishing, boats and ships (Andrady and Neal, 2009;
125 Moore, 2008; Sheavly and Register, 2007), and only 5% of the total plastic material
126 that has entered the marine environment has been recovered (Moore, 2008).
127 Furthermore, due to increasing plastic production, consumer-behaviour and
128 demographics, indications are that plastic loading in the ocean will continue to increase.
129 Some estimates suggest that the oceans will contain one tonne of plastic for every three
130 tonnes of fish by 2025, and by 2050, more plastic than fish (Auta et al., 2017; World
131 Economic Forum et al., 2016).

132 There are various pathways for plastics to reach the marine environment, and rivers
133 have been identified as a major source. Lebreton et al (2017) recently established a
134 model based on waste management practices, population density and hydrological
135 information - and estimated that between 1.15 and 2.14 million tonnes of plastic waste
136 enter the world's oceans per year via rivers. The worst 20 rivers are estimated to account
137 for 67% of the plastic entering the world's oceans (Lebreton et al., 2017); (Table 1).

138 **Table 1**

139 Top 20 rivers carrying plastic waste to the ocean (from Lebreton et al., 2017).

Catchment	Country	Midpoint mass input estimate (t/year)	Total catchment surface area (km^2)
Yangtze	China	3.33×10^5	1.91×10^6
Ganges	India, Bangladesh	1.15×10^5	1.57×10^6

Xi	China	7.39×10^4	3.89×10^5
Huangpu	China	4.08×10^4	2.62×10^4
Cross	Nigeria, Cameroon	4.03×10^4	2.38×10^3
Brantas	Indonesia	3.89×10^4	1.11×10^4
Amazon	Brazil, Peru, Columbia, Ecuador	3.89×10^4	5.91×10^6
Pasig	Philippines	3.88×10^4	4.07×10^3
Irrawaddy	Myanmar	3.53×10^4	3.77×10^5
Solo	Indonesia	3.25×10^4	1.58×10^4
Mekong	Thailand, Cambodia, Laos, China, Myanmar, Vietnam	2.28×10^4	7.74×10^5
Imo	Nigeria	2.15×10^4	7.92×10^3
Dong	China	1.91×10^4	3.33×10^4
Serayu	Indonesia	1.71×10^4	3.71×10^3
Magdalena	Colombia	1.67×10^4	2.61×10^5
Tamsui	Taiwan	1.47×10^4	2.68×10^3
Zhujiang	China	1.36×10^4	4.01×10^3
Hanjiang	China	1.29×10^4	2.95×10^4
Progo	Indonesia	1.28×10^4	2.24×10^3

Kwa Ibo	Nigeria	1.19×10^4	3.63×10^3

In general, WWTPs are regarded as important barriers - preventing plastic waste from reaching the wider water environment (such as rivers, lakes and oceans). But currently, WWTPs can only trap larger plastic (diameter >5 mm), and some smaller plastic debris (within oxidation ponds or sewage sludge). This means that a significant quantity of finer plastic is discharged to waterways through effluent (Fendall and Sewell, 2009). For instance, it is estimated that ~65 million plastic particles (many derived from cosmetics and synthetic clothing fibres) are discharged into the River Clyde via WWTPs in Glasgow on a daily basis (Murphy et al., 2016). It is also estimated that about 60% - 64% of terrestrial plastic litter reaching the ocean enters via storm-water runoff (which flows into watercourses or directly into marine waters) (Cozar et al., 2014). The fishing industry is also an important source of marine plastic, with about 20% (by number) and 70.4% (by weight) of floating macro-debris being fishing gear (Eriksen et al., 2014). Other notable pathways for plastics to directly enter the marine environment are through coastal tourism, marine vessels, and other marine industries (Law, 2017).

The fate of plastic introduced into the marine environment will depend on various properties, such as density, size, and shape. Polymers denser than seawater (like PVC) will sink into sediments, but those with lower density (e.g., PE and PP) will tend to float within the water column (Avio et al., 2017; Dubey et al., 2014). It is estimated that about 50% of marine plastic is buoyant, 62% of which will move beyond coastal waters

and out into the open-ocean (Cozar et al., 2014).

Plastic debris in the ocean can also be categorised in terms of size: i.e., macro- (> 5 mm) and micro- (< 5 mm) plastics (Moore, 2008). Typically, plastics are not fully degraded in the marine environment, but are progressively broken down into smaller and smaller pieces by ultraviolet (UV) light, mechanical abrasion (wave and sand), and the effects of temperature (Song et al., 2017). It is estimated that, by number, about 92% of marine plastic exists as micro-plastic. Further, their presence in the surface ocean is lower than would be expected (given simple estimates of composition) by a factor of 100, which may indicate that most microplastics eventually sink to, or become incorporated within, marine sediments (Eriksen et al., 2014).

In the last two decades, many studies have focussed on the presence of plastic in the marine environment, from beaches to the open ocean. McDermid and McMullen (2004) investigated the presence of small (1-15 mm in size) plastic on Hawaiian Islands beaches and found them to be present in all sediment samples collected; 87% (by weight) were original microplastics, and 11% (by weight) were primary resin pellets. Carson et al. (2011) studied Kamilo Beach (a heavily polluted beach in the Hawaiian Islands) and found plastic fragments occurring in the top 15 cm of most sediment cores. The shoreline and infralittoral sediments of beaches in Slovenia were assessed by Laglbauer et al. (2014), who found that 64% of macro-debris in sediment samples were plastic, and that microplastics were detected in 5 out of 6 samples. Kunz et al. (2016) carried out the first study on the distribution and quantity of plastic on sandy beaches along the

181 northern coast of Taiwan and found 0.32-42.56 particles per dm^3 from eight samples
182 sites, concluding that microplastic was ubiquitous in this coastal region. Kim et al.
183 (2015) explored microplastic on isolated island beaches in a high-tidal coastal region.
184 They found the abundance of microplastic to be 56–285,673 (46,334 ± 71,291) particles
185 per m^2 and polystyrene (PS) was overwhelmingly dominant. Jayasiri et al. (2013)
186 quantified plastic debris on recreational beaches in Mumbai (India) and found that
187 41.85 % (by weight) of plastics were microplastics. But, there was high spatial
188 variability due to beach usage, with land-based activities providing the major source of
189 plastic pollution to these beaches. Lusher et al. (2014) collected 470 samples along a
190 12,700 km transect of the Northeast Atlantic Ocean - and found that 89 % of 2315
191 identified particles were microplastics; and, that the calculated mean plastic abundance
192 in the Northeast Atlantic was 2.46 particles per m^3. These studies act to indicate that
193 marine plastic pollution is clearly pervasive, extensive and merits its place as an issue
194 of global concern.

195 **3. The impacts of plastic waste in the marine environment**

196 The environmental impact of plastic waste varies according to particle size. For the
197 macro plastics, the main impacts in the ocean include aesthetics and entanglement
198 (Moore, 2008). Plastic debris can accumulate on beaches and cause damage to
199 recreational infrastructure, commercial activities dependent on tourism, and fishery
200 activities (World Health Organization, 2002). Stelfox et al. (2016) also reviewed 76
201 studies and concluded that >5400 marine megafauna from 40 different species were

202 influenced by entanglement due to lost or discarded fishing line and plastic nets (so-
203 called ‘‘ghost nets’’). Moreover, the cost of removing these lines/nets could threaten the
204 economic viability of commercial fishing (Moore, 2008).
205 For microplastics, one of the most serious threats is that of ingestion. Many studies have
206 shown plastic ingestion throughout the food chain, and microplastics can be eaten by
207 lower level consumers (e.g., plankton and small fish) as well as transferred through
208 trophic levels to top predators (Ivar Do Sul and Costa, 2014) (Fig. S2). Hart (1991) and
209 Wilson (1973) reported the ingestion of marine microplastics by zooplankton,
210 invertebrates and echinoderm larvae. In mammals/seabirds (amongst many other
211 species), plastic also presents a significant mechanical hazard – blocking and
212 congesting digestive tracts (Barnes et al., 2009; Fendall and Sewell, 2009; Tourinho et
213 al., 2010; Derraik, 2002; Thompson, 2006).
214 Numerous studies have shown that marine animals have ingested plastic directly or
215 through the process of food chain transfer (Browne et al., 2008; Fendall and Sewell,
216 2009). Plastic fragments were first found in the guts of seabirds in the 1960s (Cole et
217 al., 2011); and almost thirty years ago, Moser and Lee (1992) reported on 1033 seabirds
218 from North Carolina (USA), and identified that 55% had ingested plastic. They
219 concluded that some seabirds (and fish) selected specific plastics, as a result of shape
220 and colour (which was perhaps linked to natural prey item characteristics). Carpenter
221 et al. (1972) studied plastics debris in fish guts, and further confirmed that only white
222 plastic spherules were ingested. Davison and Asch (2011) have since reported that

223 plastic fibres, fragments, and films were found in the stomachs of 13 out of 141
224 mesopelagic fish caught in the North Pacific gyre. In the Clyde Sea, 83% of lobsters
225 (*Nephrops* sp.) had ingested monofilament line and fragments of plastic bags (Murray
226 and Cowie, 2011); while, Connors (1982) concluded that ingestion of plastic may
227 influence long distance migration and reproductive effort in red phalaropes
228 (*Phalaropus fulicarius*).
229 Wilcox et al. (2016) recently carried out a survey about the impacts of different marine
230 debris on marine fauna and elicited information from experts on the ecological threat
231 of entanglement, ingestion and contamination for three major marine taxa: seabirds, sea
232 turtles and marine mammals. This survey used models to quantitatively predict the
233 impacts of marine plastic waste, and pointed out that plastic debris such as fishing gear,
234 balloons and plastic bags posed the greatest threat to marine fauna in terms of
235 entanglement; while plastic bags and plastic utensils posed the highest ingestion risk.
236 However, chemical contamination was not considered as critical as its effect is not
237 immediately lethal (Wilcox et al., 2016).
238 However, the subtle sub-lethal effects of plastic ingestion on marine biota should not
239 be overlooked. Nobre et al. (2015) assessed microplastic toxicity to embryos of
240 Echinoidea (*Lytechinus variegatus*). Researchers exposed embryos to virgin and beach-
241 collected plastic pellets, and both increased anomalous embryonic development (due to
242 additives and the hydrophobic pollutants adsorbed on them). Luís et al. (2015) studied
243 the influence of microplastics on the acute toxicity of chromium (VI) to early juvenile

common goby (*Pomatoschistus microps*). A significant decrease in predatory performance (≤67%) and a significant inhibition of acetylcholinesterase (AChE) activity (≤31%) were found with simultaneous exposure to Cr (VI) and a microplastic mixture. AChE inhibition was not detected in the test with Cr (VI) alone, thus, toxicological interactions between Cr (VI) and the microplastic were indicated. Toxicity of virgin PS and HDPE particles and their leachates were also explored by Martínez-Gómez et al. (2017). In their experiments, adverse effects on fecundity and embryonic development were found. They considered that leached chemicals from the plastic were more toxic than the plastic itself, and that the deleterious effects were more likely caused by chemical toxicity than physical damage. Brandts et al. (2018) studied the adverse effects of PS nanoplastic combined with carbamazepine on Mediterranean mussel (*mytilus galloprovincialis*) at both molecular and biochemical levels. These researchers demonstrated changes in the expression of biotransformation and immune related genes, in the inhibition of cholinesterase activity in haemolymph, and in lipid peroxidation effects in the digestive gland. Lee et al. (2019) further observed that nano-sized PS could readily penetrate the chorion and developing zebrafish embryos, and then accumulated throughout the whole body, especially in lipid-rich areas. In their study, PS nanoparticles posed marginal effects on survival, hatching rate, developmental abnormalities, and cell death in zebrafish embryos.

4. Plastic as an adsorbent for organic and inorganic contaminants

Although the general assumption is that plastics are relatively chemically inert (Holmes

et al., 2014; Roy et al., 2011), some studies have shown that plastic may also interact with contaminants, e.g., with heavy metals and organic pollutants, and as such, it may act as a vector for the long range transport of these contaminants in the marine environment (Table 2).

Table 2

Example studies regarding the binding of pollutants to marine plastics.

Types of plastic	Contaminants considered	Reference
PP resin pellets	Polychlorinated biphenyls (PCBs), DDE, and Nonylphenols (NP)	(Mato et al., 2001)
PE, PP resin pellets	PCBs	(Endo et al., 2005)
PE, PP fragments and pellets	PCBs, DDTs, and Polycyclic aromatic hydrocarbons (PAHs)	(Rios et al., 2007)
Ingested plastics	PCBs	(Provencher et al., 2018)
PE, PP and PS microplastic	PAHs, PCBs and DDTs	(Frias et al., 2010)
PE pellets	Al, Fe, Mn, Cu, Cr, Pb and Zn	(Ashton et al., 2010)
Styrofoam debris	Hg	(Graca et al., 2014)
PP, PE microplastic	Ni, Cd, Pb, Cu, Zn and Ti	(Wang et al., 2017)

Based on such observations, researchers have started to consider the adsorption capacity of marine plastic for organic and inorganic pollutants. In some cases, the plastic substrate has served as a precursor for the development of potentially low cost and efficient adsorbents for use in water treatment (Table 3). This approach would represent the ‘re-use or re-purposing’ of an existing waste stream into a potentially valuable commodity - while at the same time contributing to a reduction in plastic waste

277 in the marine environment.

278 **Table 3**

279 Selected studies regarding contaminant adsorption onto plastic polymers

Adsorbents/Condition	Contaminants	Adsorption models	Techniques used for detecting contaminants	References
PP/Extraction of samples	3,3',4,4'-tetrachlorobiphenyl (PCB77)	Langmuir model and Freundlich model	Gas chromatography (GC)	(Zhan et al., 2016)
PVC/Model solution experiments in laboratory	Phenanthrene and 4,4'-DDT	Langmuir and Freundlich model	Liquid scintillation counting (LSC)	(Bakir et al., 2012)
PVC, PE/Model solution experiments in laboratory	Phenanthrene and 4,4'-DDT	Freundlich model	Liquid scintillation counting (LSC)	(Bakir et al., 2014)
Low- and high-density PE/Extraction of samples	Polycyclic aromatic hydrocarbons (PAHs)	n. a.	Gas chromatography–mass spectrometry (GC-MS)	(Fries and Zarfl, 2012)
PE, PS, PA, and PVC/Model solution experiments in laboratory	Seven aliphatic and aromatic organic probe sorbates	Polanyi-Manes, Langmuir model and Freundlich model	GC-MS	(Hüffer and Hofmann, 2016)
PE, PS, PVC, PA and PP/Model solution experiments in laboratory	Antibiotics	Langmuir model and Freundlich model	HPLC	(Li et al., 2018)
PS, PS-COOH and HDPE/Extraction of samples	Perfluoroalkyl substances	Freundlich model	LC-MS/MS	(Llorca et al., 2018)
PS, PVC, PP, PE/Model solution experiments in laboratory	Tylosin	Langmuir model and Freundlich model; Pseudo-second order kinetic model	HPLC	(Guo et al., 2018)
PE, PS, PP/Model solution experiments in laboratory	Tetracycline	Langmuir model	HPLC	(Xu et al., 2018)

PP/Extraction of samples	Synthetic musks	Pseudo-first order kinetic model	GC-MS	(Zhang et al., 2017)
PE and nylon fibers/ Model solution experiments in laboratory	Phenanthrene and phenol	Linear and Freundlich Model; Pseudo-second kinetic model	HPLC	(Wang et al., 2018)
PE/Model solution experiments in laboratory	Carbamazepine (CBZ), 4-methylbenzylidene camphor (4MBC), triclosan (TCS), and 17α-ethinyl estradiol (EE2)	Linear Model	HPLC	(Wu et al., 2016)
PE resin pellets/Model solution experiments in laboratory	Cr, Co, Ni, Cu, Cd and Pb	Langmuir model and Freundlich model.	ICP-OES and ICP-MS	(Holmes et al., 2012)
PE pellets/Model solution experiments in laboratory	Cd, Co, Cr, Cu, Ni and Pb	Langmuir model and Freundlich model	ICP-MS	(Holmes et al., 2014)
PET, PVC, HDPE, LDPE and PP/Extraction of samples	Al, Cr, Mg, Fe, Co, Ni, Zn, Cd and Pb	Pseudo-first order kinetic model	ICP-MS	(Rochman et al., 2014)
PS, PVC/Extraction of samples	Zn, Cu	Pseudo-first order kinetic model	Flame atomic absorption spectrometry (FAAS)	(Brennecke et al., 2016)
PET, PVC, PBAT plastic pieces/Extraction of samples	Si, Al, Fe, Mg, Cr and Ni	n. a.	ICP-MS	(Kedzierski et al., 2018)

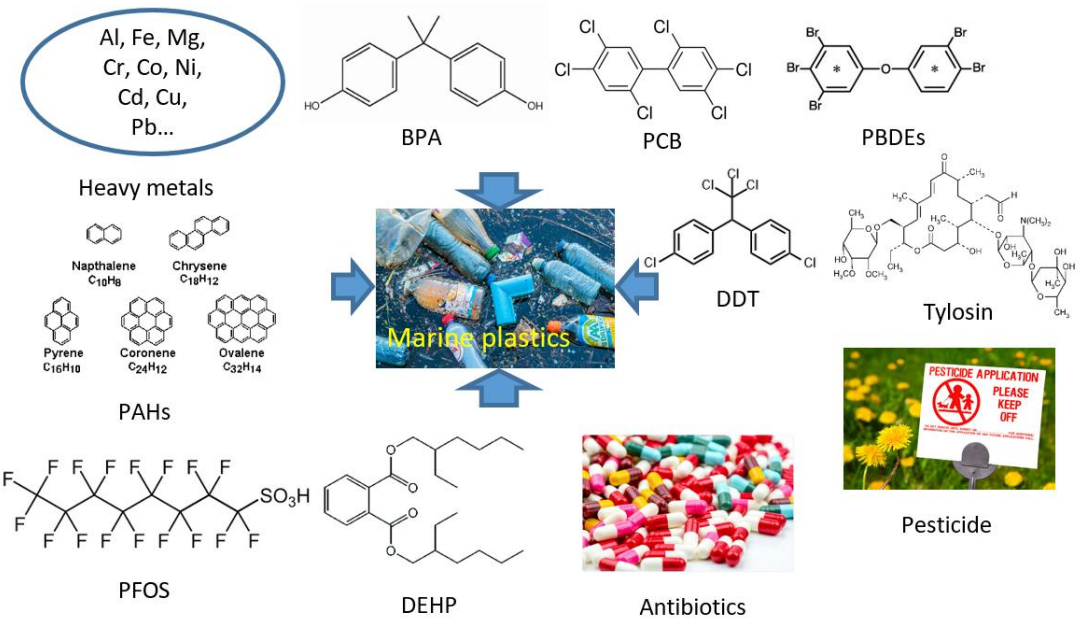

Fig. 1. A cocktail of contaminants that could potentially be associated with marine plastic debris

4.1 Adsorption studies with organic pollutants

A range of organic compounds (including PCBs, PAHs, pesticides and pharmaceuticals) can adsorb to plastic (Fig. 1).

Zhan et al. (2016) examined adsorption of 3,3',4,4'-tetrachlorobiphenyl (PCB77) by PP and indicated a maximum predicted adsorption capacity of 350 μg/g (of PP; with plastic particle size 0.425–0.85 mm) in simulated seawater. Also, that factors which could significantly influence adsorption included reaction time, particle size, temperature (which affects the surface tension of the solution and solubility), and the solution itself (e.g., salinity). Fries and Zarfl (2012) studied the adsorption of PAHs onto low (0.919 g/cm) and high (0.957 g/cm) density microplastic (PE), and observed that polymer density was an important factor in PAH adsorption (and that adsorption

294 equilibrium time could be shorter with a lower density PE). The adsorption of seven
295 aliphatic and aromatic organic probe adsorbates (n-Hexane, cyclohexane, benzene,
296 toluene, chlorobenzene, ethylbenzoate and naphthalene) by four polymers (PE, PS, PA,
297 and PVC) was investigated by Hüffer and Hofmann (2016). They reported that the
298 adsorption capacity of PS was highest, and PA was the lowest. Linear isotherms for PE
299 suggested that adsorbate uptake was due to absorption into the bulk polymer, while
300 non-linear isotherms for PS, PA, and PVC suggested a predominance of adsorption onto
301 the polymer surface. Personal care products and pharmaceuticals are also 'emerging
302 contaminants' that are increasingly being detected in freshwater resources. The
303 adsorption behaviour of carbamazepine (CBZ), 4-methylbenzylidene camphor (4MBC),
304 triclosan (TCS) and 17α-ethinyl estradiol (EE2) to PE debris (250 to 280 μm) was
305 conducted by Wu et al. (2016). They indicated that the sorption isotherms best fitted a
306 linear model and that the sorption coefficients (K_d) were 191.4, 311.5, 5140, and 53,225
307 L/kg for CBZ, EE2, TCS, and 4MBC. Sorption capacity was highly affected by the
308 hydrophobicity of each chemical, with higher adsorption rates for more hydrophobic
309 compounds. Bakir et al. (2012) studied the competitive adsorption of phenanthrene and
310 4,4'-DDT onto PVC and showed that DDT appeared to interfere with the adsorption of
311 phenanthrene onto plastic (indicating an antagonistic effect). As a result, they suggested
312 that the highly variable "cocktail of contaminants" present in the environment would
313 likely influence the adsorption capacity of plastics (Fig. 1). Recently, Li et al. (2018)
314 considered the adsorption of 5 antibiotics (sulfadiazine (SDZ), amoxicillin (AMX),

315 tetracycline (TC), ciprofloxacin (CIP) and trimethoprim (TMP)) on 5 types of
316 microplastics (PE, PS, PVC, PA and PP) in both freshwater and seawater environments.
317 They found that PA had the highest adsorption capacity for four of the antibiotics (CIP,
318 TMP, AMX, and TC) in freshwater systems, and proposed that this was due to the
319 higher porous structure of PA (and hydrogen bonds formed between amide groups on
320 the PA and the carbonyl groups of the AMX, TC and CIP). However, in seawater
321 systems, all adsorption capacities decreased significantly, with no adsorption of CIP
322 and AMX. Accordingly, they concluded that plastic particles would be a carrier of
323 antibiotics within the freshwater environment.
324 Llorca et al. 2018 considered the adsorption of perfluoroalkyl substances (PFASs) onto
325 microplastics. They analysed 18 PFASs and 3 polymers (HDPE, PS and polystyrene
326 carboxylate (PS-COOH)). PS and PS-COOH had a higher overall adsorption capacity
327 than HDPE. This was considered to be due to the formation of micelles and the
328 functional groups of the PFASs. Finally, Guo et al. (2018) studied the adsorption of
329 tylosin (TYL) onto four different plastics (PVC, PP, PE and PS), and found the
330 adsorption capacity order was PE < PP < PS < PVC. They also suggested that pH and
331 ionic strength were important factors in the adsorption process.

332 *4.2 Adsorption studies with heavy metals*

333 In addition to organic pollutants, the adsorption of inorganic pollutants onto marine
334 plastics has also been demonstrated. Holmes et al. (2012) studied plastic pellets (PE)

collected from beaches in southwest England, and tested their metal loading (of Cr, Co, Ni, Cu, Zn, Cd, and Pb). They found that virgin PE pellets could adsorb trace metals rapidly, but that aged beach pellets had a much higher equilibrium partition coefficients, e.g., up to 225 mL/g for Cr. The difference in equilibrium partition coefficients was attributed to the role of polymer and coatings on the virgin material and adherence of small metal-bearing mineral particles to the aged pellet surface (Holmes et al., 2014, 2012). The same authors also considered adsorption to virgin and beached (aged) pellets (PE) in an estuarine environment - and found that the adsorption rate for Cd, Co, Ni and Pb decreased with increasing salinity and deceasing pH. Also, Cr (VI) removal increased with increasing salinity and decreasing pH; while Cu was not significantly affected by these factors.

Brennecke et al. (2016) examined the adsorption of Cu and Zn onto virgin PS beads and aged PVC fragments (collected from seawater). The partition coefficients were found to be 650 and 850 mL/g for Cu onto PS and PVC, respectively. Also, the adsorption of Cu was significantly greater in PVC fragments than in PS, a difference attributed to different polarities and surface area.

The longer-term adsorption of metals onto plastics is also of interest to researchers, due to the importance of this in influencing the accumulation and long range transport/effects of metals in the ocean. For example, Rochman et al. (2014) analysed the accumulation of nine metals (Al, Cr, Mg, Fe, Co, Ni, Zn, Cd, and Pb) onto five plastics (PET, HDPE, LDPE, PVC, and PP) and found plastics accumulated metals over

356 time. Kedzierski et al. (2018) immersed PVC and PET (together with a biodegradable
357 plastic, poly-butylene adipate coterephtalate (PBAT)), into the bay of Lorient (France),
358 for 520 days. They found that the aged PVC could adsorb large amounts of heavy metals,
359 but the other two materials were far less contaminated. They also noted that PBAT and
360 aged PVC released estrogenic compounds into the seawater during the study, the first
361 report of its kind.

362 **5. Modification of plastic wastes**

363 In recent years, there has been growing interest in the concept that adsorbents can be
364 engineered from waste (i.e., into activated carbon), including from plastic waste. For
365 example, El Essawy et al. (2017) synthesised graphene from recycled PET bottles to
366 remove dyes from solution; and Brunauer-Emmet-Teller (BET) analysis showed that
367 the surface area increased from 2 m^2/g (raw PET) to 721.7 m^2/g (synthesised graphene)
368 during processing (the graphene pore size was ~2 nm). The maximum adsorption
369 capacity found for methylene blue and acid blue 25 was 761.1 and 642.9 mg/g
370 (respectively), using the Langmuir model. Mendoza-Carrasco et al. (2016) also utilised
371 PET waste to produce activated carbon, using this to adsorb p-nitrophenol (PNP) and
372 Fe from water. Adsorption isotherms demonstrated that the activated carbon had a high
373 affinity for PNP (adsorption capacity up to 639 mg/g), but that removal of Fe was
374 limited (attributed to blockage of pores in the presence of PNP). Rai and Singh (2018)
375 also converted PET waste into activated carbon (using pyrolysis with a carbon dioxide

376 flow), and then used a 0.5M $FeCl_3$ solution to additionally magnetise it. The resultant
377 material was tested for removal of the antibiotic cephalexin (CEX) from water, and a
378 maximum adsorption capacity of 71.42 mg/g was achieved.

379 Meanwhile, Choma et al. (2015) prepared activated carbon from waste CDs and DVDs,
380 and highlighted its potential as an adsorbent for volatile organic compounds, CO_2
381 capture, and hydrogen storage. Chen et al. (2018) used a template carbonisation method
382 (with zinc powder as an efficient hard template) to convert halogen-containing plastic
383 waste (polytetrafluoroethene) into nanoporous carbon. The product had a large BET
384 surface area (of 800.5 m^2/g) and a high total pore volume (of 1.59 cm^3/g), which
385 indicated its potential as an adsorbent.

386 In addition to activated carbons, other adsorbents have also been prepared from waste
387 plastic. Biochar is another common material that can be made from plastic waste, the
388 physicochemical properties and diversified functionality of which may offer significant
389 potential in wastewater treatment (Huang et al., 2019). Jamradloedluk and
390 Lertsatitthanakorn (2014) made char from fast pyrolysis of HDPE waste. Further, a
391 novel carbon-metal double layered oxide nano-adsorbent was synthesised by Miandad
392 et al. (2018), for the adsorption of Congo red dye, using carbon derived from the
393 pyrolysis of PS waste. Yang Xu et al. (2018) also prepared brominated biochar (Br-
394 biochar) using plastic waste - for the removal of elemental mercury (Hg^0).

395 Currently, the modification of waste plastic to an adsorbent normally requires a

combustion step: i.e., incineration, which not only generates energy/heat (a potential positive), but, may also cause air pollution. Typical contaminants created will include n-alkanes, PAHs (including triphenylbenzenes), acids (e.g., terephthalic and 4-hydroxybenzoic), and phthalates (Lazarevic et al., 2010; Simoneit et al., 2005). Cost is another key consideration when proposing plastic incineration (Hopewell et al., 2009; Piasecki et al., 1998), and hence, large-scale production of adsorbents from waste plastic is (at present) severely constrained.

6. Plastic properties related to adsorption

There are several key physical and chemical factors that can influence the adsorption capacity of plastic derived adsorbents. Fries and Zarfl (2012) considered the difference in adsorption capacity of low- and high-density polyethylene (LDPE, HDPE) to PAH's, indicating that a lower density would shorten the equilibrium time during the adsorption process. Previous studies have also shown that concentrations of various chemicals (such as PCBs and PAHs) adsorbed onto white, coloured, black and aged plastic pellets (PE, PP, and PS) were significantly different (Frias et al., 2010). Generally, coloured pellets adsorbed higher concentrations (than white plastics), which may imply that colour could be a factor influencing the adsorption capacity of certain plastic materials (Antunes et al., 2013; Frias et al., 2010).

Surface properties have also been proven to play an important role during adsorption processes - including surface area, porosity and the presence of functional groups.

Llorca et al. (2018) studied the adsorption of perfluoroalkyl substances onto plastic under various environmental conditions, and found that aged plastic had a higher adsorption capacity due to its increased surface area. The adsorption of trace metals onto plastic resin pellets in the marine environment was investigated by Holmes et al. (2014); whose results also indicated that the adsorption capacity of beach-weathered pellets was higher than for virgin pellets. This research highlighted that differences (in adsorption capacity) were due to changes in surface morphology and functional groups (Brennecke et al., 2016; Holmes et al., 2012; Llorca et al., 2018). The adsorption of phenanthrene (a PAH) on plastic (collected in Xiangshan Bay, China), was considered by Wang et al. (2018). Therein, surface functionality was considered to influence adsorption, as did particle size and surface roughness.

Crystallinity is another factor which may play a role. For example, the adsorption of four hydrophobic organic contaminants (phenanthrene, naphthalene, lindane and 1-naphthol) by three polymers (PE, PS, and polyphenyleneoxide (PPO)) was examined by Guo et al. (2012). Results showed that the organic carbon content-normalised adsorption coefficients (K_{oc}) for phenanthrene, lindane, and naphthalene onto PE increased with their crystallinity reduction. As a result, Guo et al. (2012) suggested that the mobility and abundance of rubbery domains within polymers would regulate hydrophobic organic contaminant adsorption.

7. Possible mechanisms of pollutant adsorption onto plastic materials

The adsorption mechanisms proposed to act between organic and inorganic

437 contaminants and marine plastics are summarised in Table 4.

438 **Table 4**

439 Possible mechanisms relevant to contaminants adsorbing to plastic

Adsorbent	Contaminant	Reaction based mechanisms	Reference
PS, PVC, PP, PE	Tylosin	Electrostatic interactions, surface complexation and hydrophobic interactions.	(Guo et al., 2018)
Thermoplastic resin pellets (mainly PP, PE)	Persistent organic pollutants (PCB, PAH, DDT)	Lipophilic interaction	(Rios et al., 2007)
PP, PE, PS	Tetracycline	Polar interactions and π-π EDA interactions, hydrophobic interactions and electrostatic interactions	(Xu et al., 2018)
PE, PS, PA, and PVC	Seven aliphatic and aromatic organic probe sorbates	Hydrophobic interaction and van der Waals interactions	(Hüffer and Hofmann, 2016)
PP, PE, PS	8 polycyclic aromatic hydrocarbons (PAHs), 4 hexachlorocyclohexanes (HCHs) and 2 chlorinated benzenes (CBs)	Polar interaction	(Lee et al., 2014)
PS, PS-COOH and HDPE	Perfluoroalkyl substances	Hydrophobic interactions	(Llorca et al., 2018)
PA, PE, PS, PP, and PVC	Antibiotics	Hydrophobic interaction, van der Waals interactions, electrostatic interaction and hydrogen bonding	(Li et al., 2018)
PE	Sulfamethoxazole	Hydrophobic interaction and van der Waals interactions	(Xu et al., 2018)
Plastic pellets (PE)	Trace metals (Cr, Co, Ni, Cu, Zn, Cd and Pb)	Coulombic interaction, and via non-specific interactions between neutral metal-organic complexes and the hydrophobic surface.	(Holmes et al., 2012)

Plastic pellets (PE)	Trace metals (Cr, Co, Ni, Cu, Zn, Cd and Pb)	Coulombic interaction.	(Holmes et al., 2014)
Plastic pellets (PE)	Al, Fe, Mn, Cu, Zn, Pb, Ag, Cd, Co, Cr, Mo, Sb, Sn and U	Coulombic interaction	(Ashton et al., 2010)

Organic compounds

Hüffer and Hofmann (2016) suggested that hydrophobic interactions were the main mechanism for adsorption of non-polar organic compounds (n-Hexane, cyclohexane, benzene, toluene, chlorobenzene, ethylbenzoate and naphthalene) onto plastics. However, adsorption capacity could not be entirely explained by hydrophobic interactions alone. For polyamide, the authors suggested roles for polar carbonyl and amine groups, which allowed additional interactions, such as hydrogen bonding, to take place. In addition, they suggested that further investigation was necessary, with a focus on interactions between individual molecules (rather than overall adsorption onto plastics). Xu et al. (2018) considered the adsorption of tetracycline onto three types of microplastics (PE, PP and PS) in batch adsorption experiments. Adsorption isotherms were well fitted to the Langmuir model, which indicated monolayer adsorption and suggested not only hydrophobic interactions, but also other interactions (i.e., electrostatic attraction). In their study, PS also had the maximum adsorption capacity (up to 167 (±7.74) μg/g with the Langmuir model), which they believed could be attributed to polar interactions and π-π EDA interactions; while PE and PP interacted with organics via relatively weak van der Waals interactions. The work also indicated that pH and dissolved organic matter could influence adsorption capacity significantly.

458 The highest adsorption rate was at pH = 6, and owing to the predominance of
459 electrostatic repulsion, the adsorption capacity decreased at lower or higher pH's. As a
460 result, the adsorption capacity of microplastic for tetracycline in non-neutral
461 environments was low.
462 Lee et al. (2014) studied the adsorption of 8 PAHs, 4 hexachlorocyclohexanes (HCHs)
463 and 2 chlorinated benzenes (CBs) onto three microplastics (PP, PE and PS), and
464 measured partition coefficients between plastics and seawater. They deduced that
465 polarity was the main factor affecting the adsorption rate, and therefore, that polar
466 interactions (dipole–dipole intermolecular forces and hydrogen bonds) were likely key
467 mechanisms. Llorca et al. (2018) studied adsorption onto three plastics (HDPE, PS and
468 PS-COOH) with 18 perfluoroalkyl substances (PFASs; including carboxylic acids,
469 sulphonates and one sulphonamide) in both freshwater and seawater environments.
470 Beside electrostatic interactions, the researchers believed that hydrophobic interactions
471 (which can increase adsorption affinity for longer-chain compounds) were the major
472 factor influencing the adsorption of perfluoroalkyl substances. Xu et al. (2018) also
473 tested the adsorption of sulfamethoxazole with PE and found that their data closely
474 fitted a pseudo-second order model (R^2 = 0.98) and a linear model (R^2 = 0.99). As a
475 result, they concluded that the main binding mechanism could be weak van der Waals
476 interactions (for hydrophilic sulfamethoxazole onto hydrophobic PE microplastics).
477 ***Metals***
478 There are fewer studies examining interactions between metals and plastics in the

aquatic environment. Holmes et al. (2012) studied interactions in estuarine conditions and observed greater metal adsorption (at least an order of magnitude greater) on beached pellets vs virgin pellets. Ageing and erosion of pellets and the development of a more heterogeneous and reactive surface were considered important, alongside the formation of various surface functional groups, and the attrition and adsorption–precipitation of different charged minerals and organic matter. They also found an increase in the adsorption of Cd, Co, Ni, and Pb, with increasing pH. The proposed adsorption mechanism was interaction with divalent cations linked with functional groups of natural organic matter - with the acid–base properties of the latter being important (Holmes et al., 2014). Ashton et al. (2010) considered that the possible mechanisms for metal adsorption were due to the direct adsorption of cations (or complexes) onto charged sites or neutral regions of the plastic surface. They also concluded that heavy metals have a high affinity for plastics since plastics are composed of organic polymers.

8. Characterisation methods applied to plastic based adsorbents

To understand the key functional properties of any adsorbent, several approaches are required; i.e., testing surface chemistry and morphology, textural properties, thermal stability, crystalline structure, physicochemical properties and hydrophilicity analysis (Tran et al., 2017) (Fig. S3 and Table 5). The information gleaned from such tests can then be used to elucidate possible adsorption mechanisms.

Table 5

500 Characterisation techniques used to consider plastic-based adsorbent properties.

Adsorbents	Analysis methods	Properties	References
Plastic pellets	SEM-EDX, FTIR	Surface morphology, functional groups	(Ashton et al., 2010)
PVC, PET, PBAT	SEM-EDX	Surface morphology	(Kedzierski et al., 2018)
PP, PE, Copolymer	SEM-EDX, FTIR	Surface morphology and functional groups	(Wang et al., 2017)
PE	FE-SEM, BET, PMT	Surface morphology, surface area, pore volume, pH_{pzc} and zeta potential	(Xu et al., 2018)
PS, PP, PE, PVC	SEM, BET, PMT, FTIR	Surface morphology, surface area, pore volume, zeta potential, functional groups	(Guo et al., 2018)
PP, PE	SEM, equiped with EDX, WDS and Cryotrans; BET; PMT, FTIR	Surface morphology, surface area, pore volume, pH_{pzc}, functional groups	(Fotopoulou and Karapanagioti, 2012)
PE, PS, PA, PVC	BET	Surface area	(Hüffer and Hofmann, 2016)
PE	FTIR	Functional groups	(Holmes et al., 2012)
PE, PA, PS, PP, PVC	SEM, XRD	Surface morphology and crystallinity	(Li et al., 2018)

501 For example, Ashton et al. (2010) used scanning electron microscopy–energy
502 dispersive X-ray spectroscopy (SEM-EDX) to examine the surface characteristics of
503 plastic pellets collected from the coast of Devon (Southwest England). They found
504 higher degrees of cracking, fissuring and chalking on the surface of aged vs virgin
505 pellets, and the aged pellets had many chemical and biological precipitates on their
506 surface. The same technique was utilised by Kedzierski et al. (2018) and Wang et al.

(2017) to study the surface properties of different types of plastic and metal accumulation on those plastics. Meanwhile, field emission scanning electron microscopy (FE-SEM) was also used by Xu et al. (2018) to observe the surface morphology of plastics.

Functional groups present on the surface of plastic can have a major influence on adsorption behaviour. FTIR is most commonly used to identify the surface functionality of polymers (Holmes et al., 2012; Wang et al., 2017). Ionisable dimethylamine groups were found on the surface of PP, PS, PVC and PE by Guo et al. (2018). Fotopoulou and Karapanagioti (2012) utilised FTIR to study PE and found that for virgin PE, the main interactions were C and H bonds (CH_2 rocking, CH_2 bending and CH_2 stretching); but for eroded PE, the main bonds were ester carbonyl (–COO–) and ketone (C═O) based. Adsorbent specific surface area and pore size distribution are also important factors in any adsorption process. Xu et al. (2018) used Brunauer–Emmett–Teller (BET) analysis to determine the surface area of virgin PE, PP and PS (0.2341, 0.0365 and 0.0596 m^2/g, respectively) and the pore volume of PE (0.1914 mm^3/g). Hüffer and Hofmann (2016) reported the surface area of virgin PE, PS, PA and PVC as 0.308, 0.338, 0.156 and 0.317 m^2/g (respectively) using the same method. Fotopoulou and Karapanagioti (2012) measured surface area and porosity of virgin and marine eroded PE and PP. The surface areas for: virgin PE, eroded PE, virgin PP, eroded PP were 0.13 ± 0.005, 0.17 ± 0.02, 0.11 ± 0.009 and 0.15 ± 0.006 m^2/g (respectively). Porosity volumes were also 0.6 ± 0.08, 0.5 ± 0.04, 0.3 ± 0.04 and 0.6 ± 0.1 mm^3/g, respectively. Guo et al. (2018) also

used BET on eroded microplastics and recorded values of 0.173 m^2/g for PE, 0.348 m^2/g for PP, 0.508 m^2/g for PS and 0.836 m^2/g for PVC.

Table 6 compares surface area, pore volume and adsorption capacity data for plastic materials alongside other low-cost adsorbents. It clearly indicates that the surface area and pore volume of plastic materials are generally much lower than for other materials. Marine plastic has a slightly higher surface area and pore volume (than virgin material) due to weathering in the ocean environment. Further, it also has a higher adsorption capacity for pollutants. Consequently, processes that increase surface area and pore volume would act to improve the potential for plastic to be used as a practical adsorbent. However, there are obviously limitations here - unless biochar or activated carbons are produced (by combustion). Techniques that could modify surface chemistry may be one option, with a view to changing the interaction between the plastic surface and hydrophobic contaminants. In addition, adsorption capacity is also dependant on the type of contaminant to be targeted. In this sense, more work on contaminant adsorption behaviour would be worthwhile, especially considering hydrophobic emerging compounds, such as flame retardants and certain pesticides.

Table 6

Comparison of surface area and pore volume properties between plastic adsorbents and other adsorbents

Materials	Surface area (m^2/g)	Pore volume (mm^3/g)	Adsorption capacity (mg/g)	Reference

Virgin PE, PP, and PS microplastic	0.2341 (PE) 0.0365 (PP) 0.0596 (PS)	0.1914 (PE) N/A (PP and PS)	109 ± 3.62 (PE) 113 ± 4.45 (PP) 167 ± 7.74 (PS) for tetracycline	(Xu et al., 2018)
Virgin PE, PS, PA, and PVC powders	0.308 (PE) 0.338 (PS) 0.156 (PA) 0.317 (PVC)	-	n.d.	(Hüffer and Hofmann, 2016)
Virgin and eroded PP, PE	0.11-0.13 (virgin) 0.15-0.17 (eroded)	0.033 (virgin PE) 0.039 (eroded PE) 0.038 (virgin PP) 0.051 (eroded PP)	n.d.	(Fotopoulou and Karapanagioti, 2012)
Eroded PE, PS, PP, and PVC	0.173 (PE) 0.348 (PP) 0.508 (PS) 0.836 (PVC)	-	0.639 (PE) 0.837 (PP) 1.346 (PS) 1.543 (PVC) for tylosin	(Guo et al., 2018)
Citric acid modified rubber leaf powder (CARL) and monosodium glutamate modified rubber leaf powder (MGRL)	0.44 (CARL) 0.33 (MGRL)	0. 474 (CARL) 0.457 (MGRL)	97.19 (CARL); 109.95 (MGRL) for Pb	(Fadzil et al., 2016)
Alkali-modified spent tea leaves	7.2	7	7.813 for Cu	(Ghosh et al., 2015)
Biochar from plum and apricots	146.6 (plum) 85.6 (apricot)	60 (plum) 40 (apricot)	Plums: 11.405 for Pb; 8.07 for Cr	(Pap et al., 2018)

			Apricots: 12.263 for Pb; 8.07 for Cr	
Peat	1.40	10	61.27 for Ni; 82.31 for Pb	(Bartczak et al., 2018)
35% H_2O_2 solution modified macadamia nutshell powder	1.0019	2.717	17.50 for Cr	(Maremeni et al., 2018)
Biochar from garden waste	8.89	18.6	7.25 for diclofenac 2.08 for trimethoprim	(Li et al., 2019)
Basic treated leaf residues from the extraction of essential oils	1.87	1.3	15 for Cd; 45 for Pb; 5 for Ni	(Feisther et al., 2019)
Fly ash	1.87	-	67.2 for phenol; 20.16 for 3-chloropheno; 22.17 for 2,4-dichlorophenol	(Akgerman and Zardkoohi, 1996)

Point of zero charge (pH_{pzc}) and zeta potential are two other factors studied for adsorbents. Xu et al. (2018) measured pH_{pzc} for new plastics (in their solid state). The pH_{pzc} of PE, PP and PS was at pH 4.30, 4.26 and 3.96 (respectively). Zeta potentials for PE, PP and PS were also measured by Guo et al. (2018). They indicated that zeta potentials decreased with increasing pH, and that the zeta potential was negative at pH 3 to 11. Moreover, Fotopoulou and Karapanagioti (2012) noted that for virgin PE, virgin PP and eroded PP zeta potential was neutral, while the overall surface charge of eroded

PE was negative at the pH of seawater (pH~8).

X-Ray Diffraction (XRD) is commonly used to measure crystallinity, and in some cases, it has been applied to plastics. For example, Li et al. (2018) determined the degree of crystallinity of plastic particles (PE, PA, PS, PP and PVC) through XRD, and indicated the degree of crystallinity followed the order: PE > PP > PA ≈ PS > PVC.

9. Future perspectives and challenges

Plastic waste is abundant in the ocean, and research should consider not only how to prevent this, how to remove it, but also new ways to reuse and repurpose this waste. To utilise marine plastic as a low-cost adsorbent for water or wastewater treatment is one possible option. Further research into marine plastic based adsorbents should be carried out - investigating a wider range of contaminants, with their potential for removal based on existing data. Hydrophobic pollutants (such as some pesticides, industrial chemicals and flame-retardants) should be included in these studies. Some of these compounds may be regarded as 'emerging contaminants', which can have negative impacts in the environment even at low concentrations (e.g., ppt to ppb range) (Geissen et al., 2015; Stuart et al., 2012). Due to their specific characteristics, plastic adsorbents may elicit selective adsorption behaviour. Currently, bonding mechanisms of organic and inorganic contaminants to plastics are not particularly well classified; and there is a need for suitably detailed research and experiments. One key area requiring further investigation regards the characterisation of plastic absorbents before and after pollutant adsorption (to aid understanding of the adsorption processes). In order to

improve the adsorption capacity and efficiency of plastic adsorbents, surface modification and functionalisation should also be explored in more detail. Current modification methods are largely based on combustion, which are not a particularly "sustainable" or "green" approach. More sustainable modification or functionalisation approaches will be needed if real applications are to be exploited. As there are also very few studies looking at plastic as an adsorbent in aqueous systems, it will also be necessary to understand the potential for release of pollutants from these materials. Desorption and regeneration of plastics could also be investigated, which would be helpful to understand the longer term use potential for such materials. Finally, the implementation of plastic based adsorbents in a real setting (industry and/or wastewater system), perhaps in combination with other types of adsorbent (e.g., biochar, activated carbon) merits further study.

10. Conclusions

This review considers the potential to utilise plastic-derived adsorbents for the removal of hazardous priority contaminants from an aqueous environment. Given the research discussed, it is evident that marine plastics have the potential to be used as adsorbent materials, for hydrophobic organic and inorganic pollutants. This potentially inexpensive and locally available resource (waste) could be used in wastewater treatment, given the caveats highlighted here, and, given the right setting/application. However, it is also clear that more detailed investigations regarding the modification, optimisation, interaction mechanisms involved, and possible field applications of

plastic based adsorbents are needed to prove if this is a viable and sustainable concept.

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Appendix A. Supplementary document

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