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Review

Interaction of toxic chemicals with microplastics: A critical review

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ABSTRACT

Occurrence of microplastics (MPs) in the environment has attracted great attention as it has become a global concern. This review aims to systematically demonstrate the role of marine microplastic as a novel medium for environmental partitioning of chemicals in the ocean, which can cause toxic effects in the ecological environment. This review assimilated and analyzed available data published between 1972 and 2017 on the interaction between MPs and selected chemicals. Firstly, the review analyzes the occurrence of chemicals in MPs and outlines their distribution patterns. Then possible mechanisms of the interaction between MPs and organic chemicals and potential controlling factors were critically studied. Finally, the hazards of MPs and affiliated organic chemicals to marine organisms were shortly summarized.

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1. Introduction

In recent years, marine environment pollution has been attracting increased attention with the boom of marine activities, particularly plastic pollution. As lightweight, strong, versatile, durable and affordable materials (Derraik, 2002; Laist, 1987), plastics have been heavily used as basic constituents for a variety of commercial and consumer products (Andrady, 2011; Bockhorn et al., 1999; Roy et al., 2011; Teuten et al., 2009). However, the disposal of plastic products has caused heavy pollution to the marine environment (Laist, 1987, Pruter, 1987). Plastic is a main component of marine trash, occupying about 60%–80%, in some areas up to 95%, of ocean garbage (Derraik, 2002; Gregory and Ryan, 1997; Moore, 2008). The so-called “microplastics (MPs)” or “plastic pellets” have been detected in various oceanic compartments, including surface seawater, water column and sediment (Claessens et al., 2011; Derraik, 2002; Hammer et al., 2012; Hidalgo-Ruz et al., 2012; Obbard et al., 2014; Van Cauwenberghe et al., 2013), as well as beaches, estuaries, remote islands and polar regions (Barnes et al., 2009; Gregory and Ryan, 1997; Thompson et al., 2004). While their definitions may vary (Andrady, 2011), MPs are generally categorized as plastic particles with effective diameters less than 5 mm, which are usually cylinder or disk shaped (Barnes et al., 2009; Betts, 2008; Dubaish and Liebezeit, 2013; Liebezeit and Dubaish, 2012; Thompson et al., 2009; Turra et al., 2014; Zhang et al., 2015). According to the different shapes of MPs, they can be mainly divided into fragments, foams, granules and fibers (Tsang et al., 2017). Because the degradation rate of MPs is quite low (Andrady, 2011; Bakir et al., 2014; Hidalgo-Ruz et al., 2012), MPs can accumulate and concentrate in the oceans via various routes, such as mechanical, physical/chemical and biological actions (Carpenter et al., 1972; Thompson et al., 2004, 2005).

Microplastics can be derived from primary and/or secondary sources (Andrady, 2011). Primary sources mainly include additives used to increase friction in consumer products, such as cosmetic and facial care products or hand-cleaners and toothpaste (Andrady, 2011; Browne et al., 2011; Carr et al., 2016; Cole et al., 2011; Fendall and Sewell, 2009; Gregory, 1996; Wang et al., 2016; Zitko and Hanlon, 1991), medical supplies, such as grinding polishing agents used in dental teeth and capsules as vectors for inclusion of drugs (Patel et al., 2009), overflowing drilling fluid in oil exploration, industrial abrasives and air-blast cleaning media (Gregory, 1996; Hidalgo-Ruz et al., 2012). Secondary sources are large plastics degraded to small particles through physical, chemical and biological processes (Browne et al., 2007).

Microplastics are ubiquitously distributed in the marine environment, accounting for approximately 92% of global marine plastic debris (Eriksen et al., 2014). Because of their minute sizes, MPs are difficult to identify and can be easily ingested by aquatic organisms. Microplastics have larger specific surface area than large plastic particles, so they may sorb much more organics (Lee et al., 2014). The kinetic of chemical sorption on MP is important for their fate and transportation. Guo et al. (2012) showed that the sorption of four hydrophobic organic compounds on three polymers reached equilibrium within 6 days. In oceans, the equilibrium will changed from time to time, as gyres are still in a kinetic phase. The chemicals in the MPs also exchanged with the surrounding media, as MPs can be transferred with very long distance (such as the Arctic and the North Pacific Gyre) under the action of wind and ocean currents (Rios Mendoza and Jones, 2015; Zarfl and Matthies, 2010). For most case, it was proposed that the equilibration was reached in short time scales when analyzing chemical concentrations in MPs. Moreover, the sorption behavior of chemicals on MPs is related to plastic types, color and other physical and chemical properties (e.g., size and chemical composition) (Endo et al., 2005). Discolored MPs have been reported to adsorb more PCBs than non-discolored ones and polyethylene (PE) generally exhibits a greater sorption capacity to environmental pollutants than other plastic types (Alimi et al., 2017). And the presence of other sorbents is also important. According to Koelmans et al. (2016) and Lohmann (2017), the total organic contaminant fraction absorbed by plastic is much smaller than that adsorbed by other media like dissolved organic carbon and colloids in the ocean. Seidensticker et al. (2017) experimentally found that the concentration of dissolved organic matter could change the distribution of contaminants between the MP and water. In addition, the increase of MP concentration would increase adsorption of chemicals. The sorption of chemicals on MPs may transfer harmful substances such as persistent, bioaccumulative and toxic (PBT) chemicals into the food chain (Bakir et al., 2012; Carpenter et al., 1972; Carpenter and Smith, 1972; Mato et al., 2001; Rochman et al., 2013b; Rockstrom et al., 2009; Rothstein, 1973; Zitko and Hanlon, 1991), which would seriously endanger marine life and ecological environment. However, some critical papers regarding the transfer potential of MP didn't draw the conclusion that MP is an important transport carrier or a vector to marine organism (Beckingham and Ghosh, 2017; Gouin et al., 2011; Koelmans et al., 2013). As the number of studies regarding MPs and affiliated chemicals has considerably increased in recent years, it is useful to examine the sorption mechanisms between MPs and organic chemicals, and identify research gaps and directions.

The objectives of this review are to (1) summarize the types of organic chemicals preferably affiliated to MPs; (2) analyze the occurrence of these chemicals in MPs and outline their distribution patterns; (3) examine possible mechanisms of the interaction between MPs and the organic chemicals and potential controlling factors and (4) discuss the hazards of MPs and affiliated organic chemicals to marine organisms.

2. Data assimilation and analysis

Peer-reviewed journal articles related to MPs in Web of Science published between 1972 and 2017 were assimilated and analyzed. The reported concentrations of MPs in the environment (river, seawater and sediment) are extracted and summarized in Table S1. Total concentrations of $\sum_{13}$PCB (sum of PCB 66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170 and 206), $\sum$DDT (sum of DDT, DDE and DDD) and $\sum$HCH (sum of α-HCH, β-HCH, γ-HCH and δ-HCH) on plastic pellets were calculated and divided into major cities, small cities, remote areas and remote islands (Fig. 1). Concentrations of main chemicals in cities of Australia and New Zealand were averaged using median concentrations from different sampling sites. Chemical concentrations in cities of Portugal and remote islands were based on reporting median data from two selected articles. Fig. 2 and Fig. S3 were drawn based on the reported concentrations of different chemicals on MPs with different colors (black, aged, white, and colored). Fig. 3 outlined median concentration of some selected congeners or isomers of target chemicals on MPs based on data from three different countries (China, Greece, and Portugal). To study the temporal distribution of chemicals on MPs, data from two related papers were summarized in Fig. 4 to compare changes of chemical levels with time. Table S5 outlined partition coefficients of various chemicals in different types of plastic polymers from 12 selected studies. The correlation between the octanol-water partition coefficient (Log K_{ow}) and the equilibrium partitioning coefficient between polymer and seawater or water (Log $K_{POLYMER\text{-}sw}$/Log $K_{POLYMER\text{-}w}$) of the chemicals through the relevant regression analysis are also demonstrated in Fig. 5. $K_{POLYMER\text{-}sw}$ was calculated using partitioning ratios as follows:

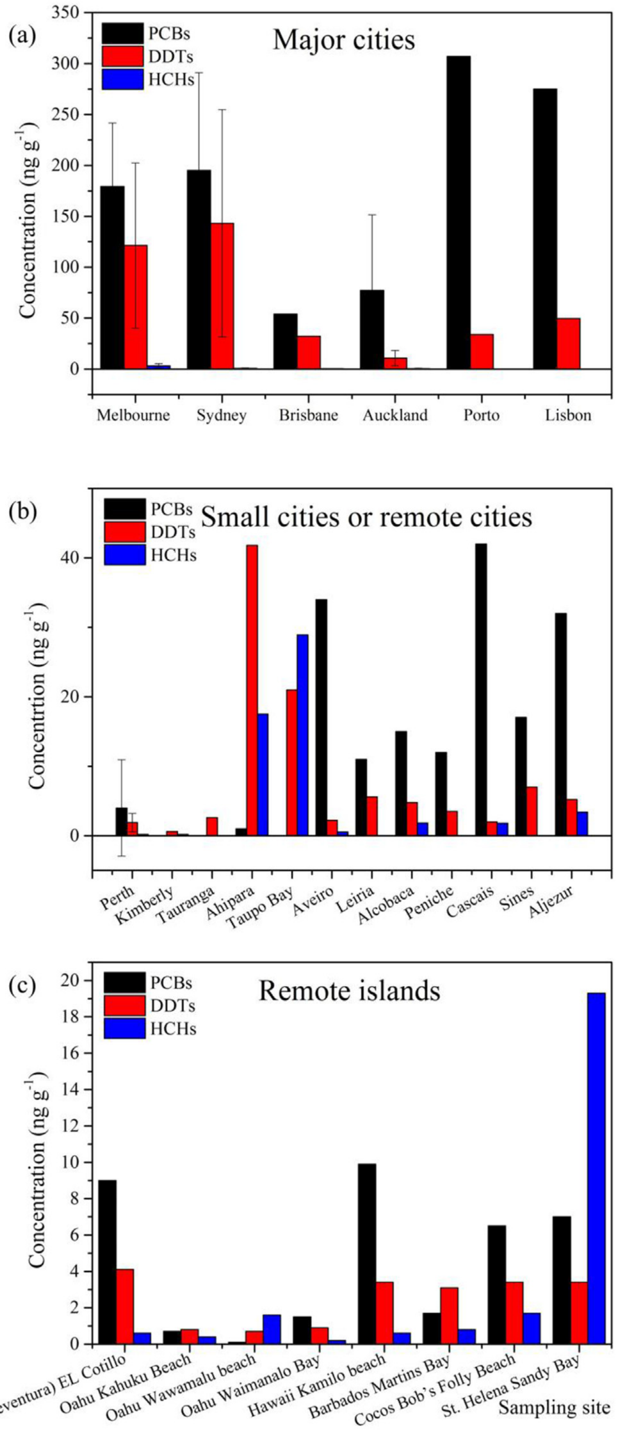


Fig. 1. Concentrations of PCBs, DDTs and HCHs on plastic pellets (ng g^{-1}-pellet) collected from (a) major cities, (b) small cities or remote areas, and (c) remote islands (For studies reporting chemical concentrations on microplastics, mean concentration were calculated and plotted) (Heskett et al., 2012; Mizukawa et al., 2013; Yeo et al., 2015).

$$K_{POLYMER\text{-}sw} = K_{POLYMERsm}/K_{PDMSsm} * K_{PDMSsw} \qquad (1)$$

where $K_{POLYMERsm}$ and K_{PDMSsm} are partition coefficients between polymer and solvent mixture and between PDMS and solvent mixture, respectively.

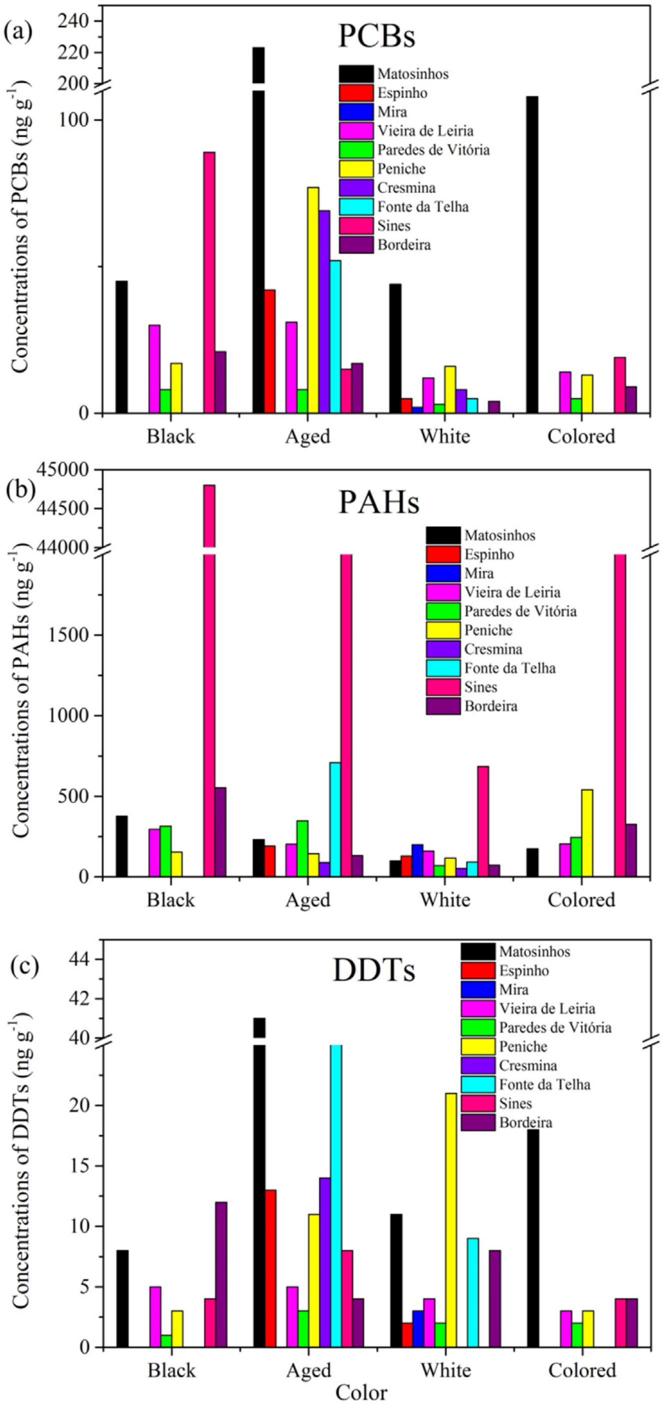


Fig. 2. (a) PCBs concentrations, (b) PAHs concentrations, and (c) DDTs concentrations in different colored plastic pellets (ng g^{-1}-pellet) from ten beaches along the Portuguese coast (For the study reporting chemical concentrations on different colorized microplastics) (Antunes et al., 2013).

3. Microplastics and adhered chemicals

3.1. Spatial variability

Fig. 1 shows concentrations of $\sum_{13}$PCB, $\sum$DDT and $\sum$HCH on plastic pellets from major cities, small cities, remote areas and remote islands. It is clear that concentrations of PCBs, DDTs and HCHs in MPs collected from large cities such as Sydney, Auckland

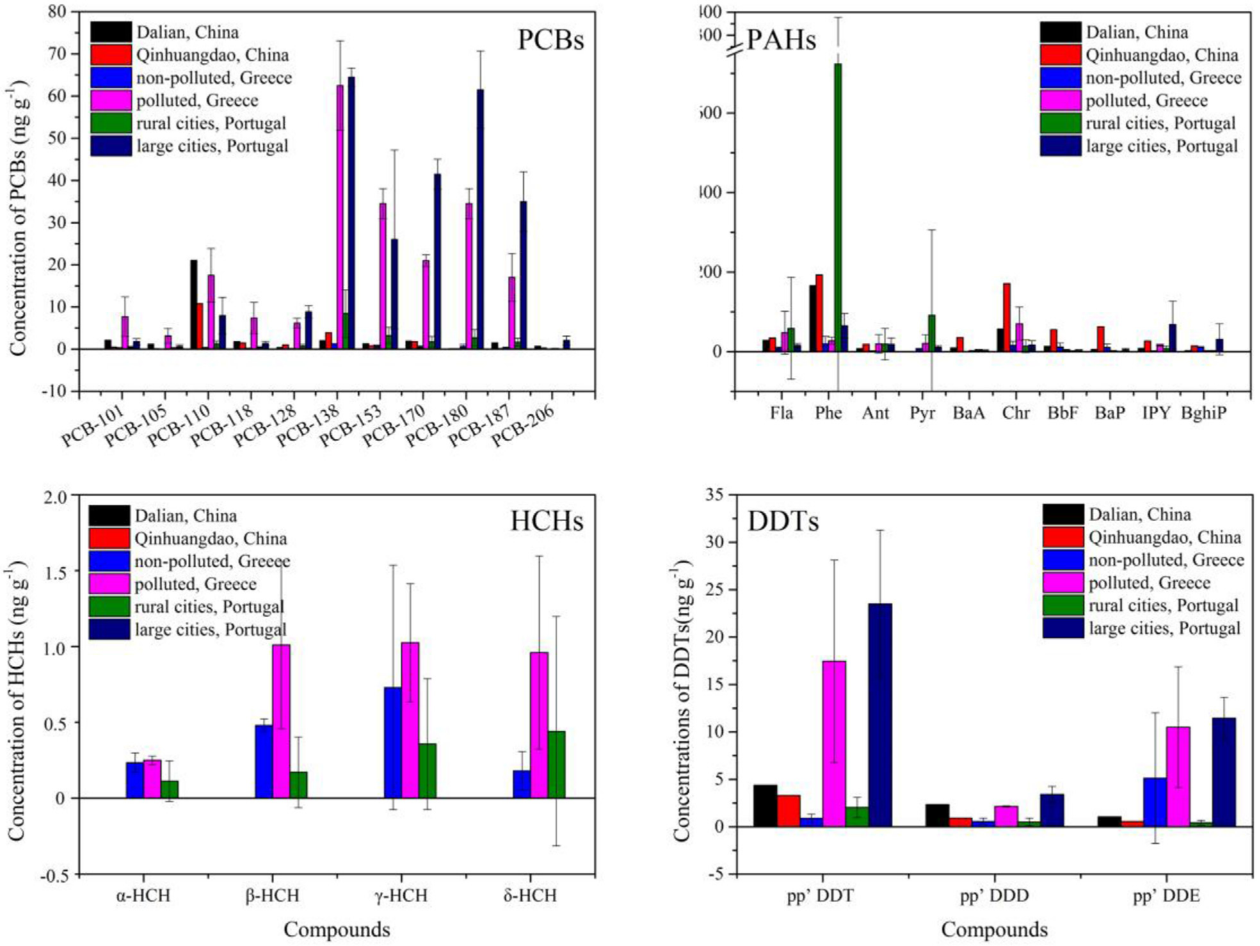


Fig. 3. (a) Concentrations of PCB congeners, (b) concentrations of PAH isomers, (c) concentrations of HCH isomers, and (d) concentrations of DDTs and its metabolites in plastic pellets (ng g^{-1}-pellet) from three studied countries-China (Zhang et al., 2015), Greece (Karapanagioti et al., 2011) and Portugal (Mizukawa et al., 2013). For each study, mean concentrations were calculated and plotted.

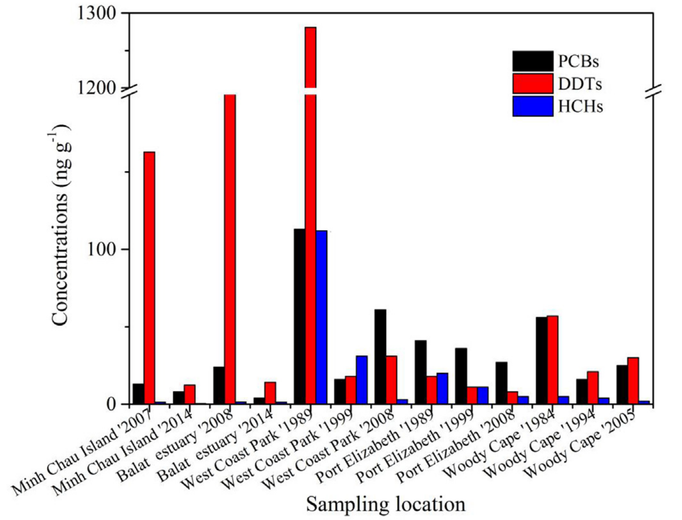
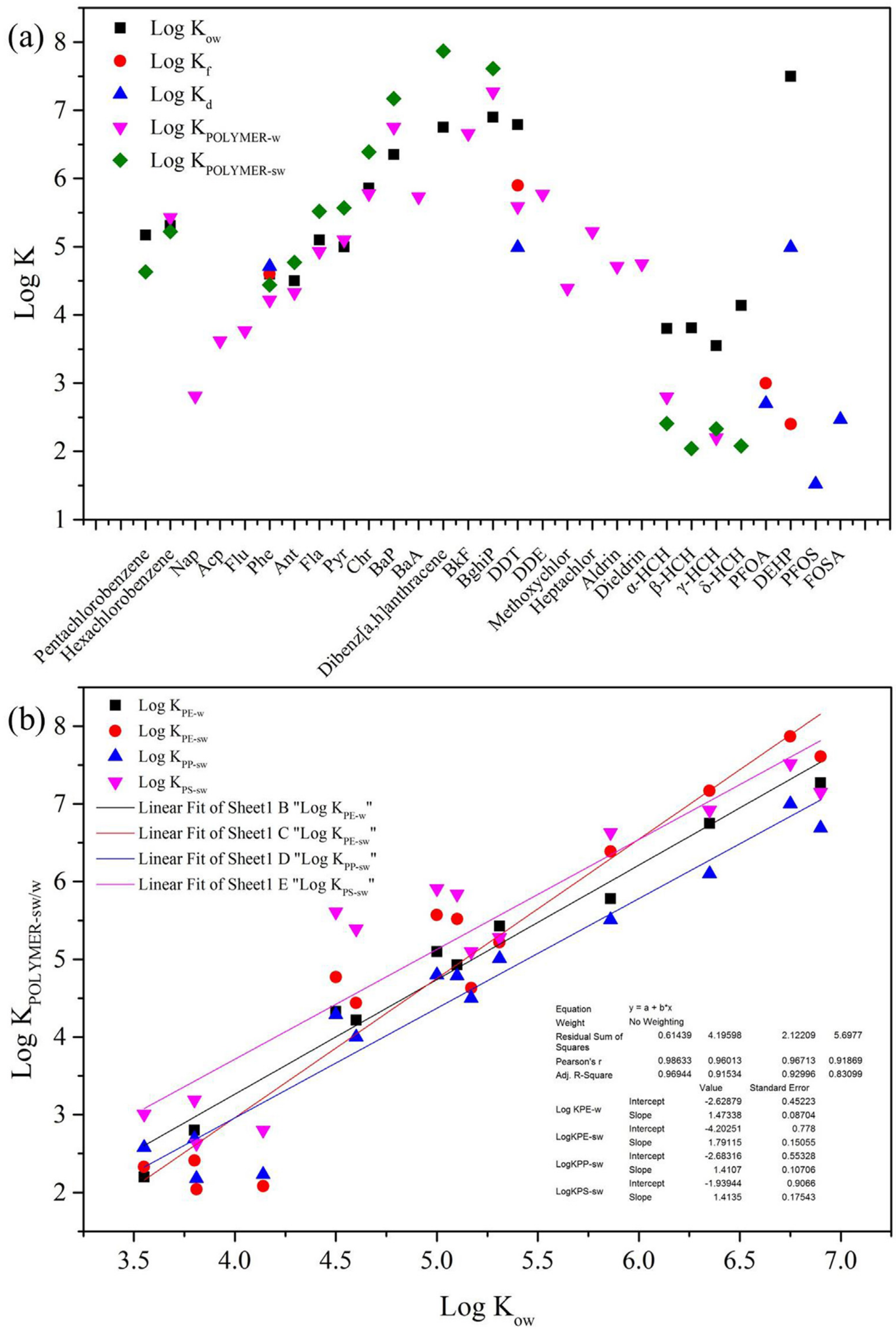


Fig. 4. Temporal changes of PCB, DDT and HCH levels in plastic pellets (ng g^{-1}-pellet) collected from South Africa (Ryan et al., 2012) and Vietnam (Le et al., 2016).

and Lisbon were almost an order of magnitude higher than those from small cities (e.g., Tauranga in New Zealand) or remote areas (e.g., Kimberley in Australia) and were almost two orders of magnitude greater than those on remote islands (e.g., Oahu Kahuku Beach, Oahu Wawamalu beach in US) (Heskett et al., 2012; Mizukawa et al., 2013; Yeo et al., 2015). Fig. S2 shows the concentrations of PCBs, DDTs, HCHs and PAHs in plastic pellets from four different places in Greece (Karapanagioti et al., 2011). The concentrations of all detected chemicals in pellets from Loutropyrgo beach and Aegena Island were much higher than those from Kato Achaia beach and Vatera Island. It is well-known that the Saronikos Gulf is a polluted place, as Loutropyrgo and Aegena are located at the Saronikos Gulf (Gómez-Gutiérrez et al., 2007; Galanopoulou et al., 2005). Kato Achaia and Vatera are located on the non-polluted Patras Gulf (Karapanagioti et al., 2011). Likewise, there are regional differences in the chemical concentrations detected on the MPs. PAHs in MPs found on the sandy beach were also analyzed, and the results showed that PAHs on the MPs sediments in different depths had different concentrations and surface layer had the highest PAH levels (Fisner et al., 2013a, 2013b). Apparently, levels of chemicals adhered to plastic pellets are related to the regional levels of industrialization (Endo et al., 2005; Heskett et al., 2012; Hirai et al., 2011; Hosoda et al., 2014; Karapanagioti et al., 2011;



Equation	y = a + b*x				
Weight	No Weighting				
Residual Sum of Squares		0.61439	4.19598	2.12209	5.6977
Pearson's r		0.98633	0.96013	0.96713	0.91869
Adj. R-Square		0.96944	0.91534	0.92996	0.83099
			Value	Standard Error	
Log KPE-w	Intercept		-2.62879	0.45223	
	Slope		1.47338	0.08704	
LogKPE-sw	Intercept		-4.20251	0.778	
	Slope		1.79115	0.15055	
LogKPP-sw	Intercept		-2.68316	0.55328	
	Slope		1.4107	0.10706	
LogKPS-sw	Intercept		-1.93944	0.9066	
	Slope		1.4135	0.17543	

Fig. 5. (a) The octanol-water partition coefficient (Log K_{ow}) partition coefficients of linear (Log K_d) and Freundlich (Log K_f) models, and polymer-water partitioning coefficients in seawater (Log $K_{POLYMER\text{-}sw}$) and water (Log $K_{POLYMER\text{-}w}$). of different compounds, (b) the correlation between the octanol-water partition coefficient (Log K_{ow}) and the equilibrium partitioning coefficient between polymer and seawater/water (Log $K_{POLYMER\text{-}sw}$/Log $K_{POLYMER\text{-}sw}$) of the compound.

Mato et al., 2001, 2002; Mizukawa et al., 2013; Yeo et al., 2015). The concentration of pollutants is also related to the abundance of MPs.

3.2. Color difference

Plastic pellets are divided into four categories: black, aged, white and colored. Fig. 2 summarizes the levels of PCBs, PAHs and DDTs on plastic pellets with different colors from ten beaches along the Portuguese coast (Matosinhos, Espinho, Mira, Vieira de Leiria, Paredes de Vitoria, Peniche, Cresmina, Fonte da Telha, Sines and Bordei) (Antunes et al., 2013). Fig. S3 shows the total concentrations of PCBs, PAHs and DDTs in each color fraction in Alcobaça, Portugal (Frias et al., 2013). Aged and black pellets contained higher PCB and PAH concentrations than colored and white ones (Fig. 2(a)-2(b) and

S3). On the other hand, DDT concentrations were similar in black, colored and white pellets, but not aged ones (Fig. 2(c) and S3). Overall, aged and black pellets had higher concentrations of PCBs and PAHs than white and colored pellets, especially yellowing pellets (Antunes et al., 2013; Frias et al., 2010, 2013; Rios et al., 2007).

Aged pellets are yellowing ones that have been present for a long time in the oceans and with a high state of degradation. Sorption of hydrophobic organic pollutants to microplastic includes not only adsorption to surfaces but also absorption into particles. Their long residence time would increase the sorption concentration of chemicals on MPs, while degradation would increase their specific surface areas, further increasing their sorption capacities (Antunes et al., 2013; Endo et al., 2005; Frias et al., 2013; Karapanagioti and Klontza, 2008; Mato et al., 2001; Ogata et al., 2009). And longer residence time allows diffusion of hydrophobic organic pollutants deeper into plastic particles from contaminated water. These are the main reasons why aged pellets can sorb much more chemicals than other tinctorial pellets. Some researchers hold the opposite view that originally absorbed chemicals might be released to less polluted water during the migration and hence the concentration may decrease. Thus, the effect of prolonged stay in the surrounding water environment on the sorption and desorption of pollutants by aged microplastic is dependent on the chemical levels of the water environment. Another reason is that black granules basically consist of polystyrene (PS) and polypropylene (PP) materials, while aging particles are usually made up of PP and PE materials (Cavani and Trifirò, 1995; Frias et al., 2010). With regards to plastic materials, PE usually has higher sorption affinities than other plastic polymers towards organic chemicals, followed by PP and PS (Karapanagioti and Klontza, 2007, 2008; Karapanagioti et al., 2010; Teuten et al., 2007). There are also some other potential reasons. For example, polyurethane (PU) was identified as a constituent of black pellets occasionally, which may increase their ability to sorb chemicals (Antunes et al., 2013; Frias et al., 2013; Zia et al., 2007). Pigmented particles may also contain some colored additives, such as pigments, that make it easier to sorb chemicals from surrounding seawater (Frias et al., 2013). The reason for pigments favour sorption is the organic contents taken from plants and animals can promote sorption capacity. Compared to black and aged particles, white pellets sorb significantly less amounts of chemicals, due to their translucent characteristic (Frias et al., 2013). White pellets are often present in seawater for a short time, resulting in low sorption and small specific surface areas (Endo et al., 2005; Frias et al., 2010; Ogata et al., 2009).

The above results explain why black and aged particles can sorb higher concentrations of chemicals than colored and white particles. However, Endo et al. (2005) suggested that the degree of aging in particles had no association with sorption of PCBs. They argued that plastic aging occurs after stranding on the beach rather than in seawater, while plastic sorption of chemicals occurs in seawater (Endo et al., 2005; Gregory, 1983).

3.3. Congeners or isomers

In addition to the total concentrations of the target chemicals in MPs presented above, these chemicals also include different congeners or isomers. Fig. 3 depicts levels of PCB congeners, PAH isomers, HCH isomers and DDT metabolites in plastic pellets (ng g^{-1}-pellet) from three different countries, China, Greece and Portugal (Karapanagioti et al., 2011; Mizukawa et al., 2013; Zhang et al., 2015). Generally, concentrations of PCBs, HCHs and DDTs on plastic pellets from China were lower than those from Greece and Portugal, while PAH concentrations in China were higher. The results also showed that PCB 110 was the most abundant congener among all PCBs in China, followed by PCB 138 and PCB 170, while PCB-138, PCB-153, PCB-170, PCB-180, and PCB-187 were predominant in Greece and Portugal (Fig. 3(a)). As many countries began to reduce or stop PCB production after 1973, the occurrence of PCBs in these three countries may be a result of past pollution. To be specific, in the past these chemicals were released into water and accumulated in the water column and sediment and was later released to the environment. However, PCB concentrations on plastics were much higher in polluted areas and large cities, which were probably related to industrial activities from their surroundings. In addition, heavily-chlorinated congeners were enriched in MPs, especially hexa-chlorinated PCBs (PCB 138 and PCB 153) and hepta-chlorinated PCBs (PCB 187, 180 and 170). This indicates that either heavily-chlorinated congener has high ambient concentrations in the environment or plastic particles have strong tendency to sorb such homologues. This question requires further study.

For PAH isomers, the concentrations of phenanthrene (Phe) and chrysene (Chr) were dominant in all three countries, especially in China. In rural cities from Portugal, fluoranthene (Fla), Phe and pyrene (Pyr) were the most prevalent PAH components, while Phe, indeno(1,2,3-cd)pyrene (IPY) and benzo[*ghi*]perylene (BghiP) were dominant in large cities (Fig. 3(b)). Budzinski et al. (1997) and Boonyatumanond et al. (2006) suggested that high-ring (4–6 rings) PAHs were derived mainly from fossil fuels and incomplete combustion of organic matter. In contrast, low-ring PAHs (2–3 rings) are derived mostly from petroleum products, such as gasoline and lubricant oils. Apparently PAHs on plastic pellets may have derived from both combustion of fossil fuels and petroleum residues. Due to the proximity to large oil and gas electric power plants, rural cities of Portugal mainly contained low-ring PAHs (Mizukawa et al., 2013). And it can clearly see from Fig. 3(b) that the concentration of high-ring PAHs on MPs in Qinhuangdao is much higher than that in Dalian, which is because there is a China's largest transportation port of coal near the sampling site. The main source of PAHs may have been the combustion of fossil fuels at other sampling sites.

The concentrations of HCHs and DDTs in MPs were relatively higher at polluted sites or large cities, except for HCHs in Portugal. In Greece, γ-HCH was the most abundant HCH isomer, followed by δ-HCH, while γ-HCH and δ-HCH were the main isomers in Portugal (Fig. 3(c)). HCHs are commonly used legacy pesticides. The γ isomer (also known as lindane) has the only insecticidal effect and the most rapid degradation rate, followed by α and δ isomers, with the β isomer extremely recalcitrant (Willett et al., 1998). This is an important reason why γ-HCH is the main isomer found in MPs. HCHs, especially γ-HCH, were mainly used for insect control on fruits and vegetables, rice and seed treatment, which is also the main reason why HCHs mostly occurred in rural areas. Another possibility is that plastic particles have higher affinity with γ-HCH than β-HCH or δ-HCH (Ziccardi et al., 2016). Experimentally measured partition coefficients of γ-HCH are consistently greater than those of β- and δ-HCH using PE, PP, and PS debris (Lee et al., 2014), which also supports such argument. The DDT products have played a significant role in preventing agricultural diseases and pests, and reducing malaria, typhoid fever, and other mosquito-borne diseases for the first half of the 20th century. The major DDT compounds, *p,p*'-DDT, *p,p*'-DDD and *p,p*'-DDE, have been detected in MPs from both rural and urban areas, while urban areas generally have higher DDT concentrations than rural areas (Mizukawa et al., 2013) (Fig. 3(d)). The use of DDTs as pesticides in nearby catchments may be attributed to the higher concentrations of DDTs in cities.

3.4. Temporal trends

Fig. 4 shows temporal characteristics of various chemicals in plastic particles collected from South Africa and Vietnam. From Fig. 4, it is clear that plastic particles-associated chemical concentrations in these places have overall declined. More specifically, PCB and DDT concentrations in plastic pellets decreased markedly from 2007 to 2014 in the Minh Chau Island (Vietnam) and from 2008 to 2014 in the Balat estuary, while HCH levels had a slight decrease (Le et al., 2016). The same trend was also observed in plastic samples collected from South African beaches (Ryan et al., 2012). However, a slight increase of PCB and DDT concentrations occurred at West Coast Park in the Western Cape Province of South Africa from 1999 to 2008 and at Woody Cape in the Eastern Cape Province from 1994 to 2005.

Concentrations of chemicals in the environment will theoretically be reduced over time if the source is controlled. Chemicals in the environment will gradually degrade with time, although the degradation process can be very slow. The decreasing trends in Vietnam from 2007 to 2014 and on three South African beaches from 1984 to 2005 and from 1989 to 2008 are consistent with this assumption. The main reason is the ban of PCB and DDT usage since 1970, which has been effective to reduce environmental sources of these chemicals (Ryan et al., 2012). However, the slight increase of PCB and DDT levels at West Coast Park from 1999 to 2008 and Woody Cape from 1994 to 2005 may be due to analytical uncertainties as well as the roles of contaminant reservoir (such as: contaminated sediments). According to Ryan et al. (2012), the increase may be due to the underestimation of the PCB concentrations on plastic samples in 1999 and by re-suspension of sediment during industrial activities.

4. The interaction between microplastics and chemicals

Microplastics are known to sorb PBT compounds from surrounding water, which may further act as carriers or vectors to transport these chemicals to biota (Bakir et al., 2012, 2014; Bowmer and Kershaw, 2010; Brennecke et al., 2016; Endo et al., 2005; Hirai et al., 2011; Holmes et al., 2014; Mato et al., 2001; Rios et al., 2007; Teuten et al., 2007, 2009). Nevertheless, Beckingham and Ghosh (2017), Gouin et al. (2011) and Koelmans et al. (2013) draw the inverse conclusion that MP is not an important transport vector of chemicals. Recent study of Chen et al. (2018) showed that plastic might have a role to play in the transfer of chemicals to certain marine organisms if the gradient of plastic to predators is present. Furthermore, chemical concentration in MPs might be even up to 10^5 to 10^6 times greater than that in ambient seawater. (Mato et al., 2001; Rios et al., 2010; Takada, 2006; Wurl and Obbard, 2004; Ziccardi et al., 2016). However, fugacity gradient is proportional to ($C_{POLYMER}/K_{POLYMER/sw}$ - C_w). Thus, the debated results lead to comprehensive summary of current study on the interaction between chemicals and MPs.

4.1. The effects of hydrophobicity (K_{ow})

Table S5 shows partition coefficients of different chemicals derived from linear (K_d) and Freundlich (K_f) models, octanol-water partition coefficients (K_{ow}), and polymer-water partitioning coefficients in seawater ($K_{POLYMER\text{-}sw}$) and water ($K_{POLYMER\text{-}w}$).

The equation for the linear model is as follows:

$$K_d = [q_e]_{solid} / [C_e]_{aq} \quad (2)$$

where q_e is the amount of contaminant sorbed onto plastic (ug kg^{-1}) at equilibrium and C_e is the contaminant concentration in the aqueous phase at equilibrium (ug L^{-1}).

The equation for the Freundlich model is as follows:

$$\log q_e = \log K_f + 1 / n_f \log C_e \quad (3)$$

where q_e (ug kg^{-1}) is the concentration of the contaminant on the solid phase at equilibrium, C_e (ug L^{-1}) contaminant concentration in the aqueous phase at equilibrium, K_f (L kg^{-1}) is the multilayer adsorption capacity and $1/n_f$ is the Freundlich exponent and an indicator of the site energy distribution of a sorbent (ie sorbent heterogeneity increases as n decreases).

The equation for polymer-water partitioning coefficients in seawater (or water) is as follows:

$$K_{POLYMER\text{-}sw} = K_{POL\ YMERsm} / K_{PDMSsm} {}^{*} K_{PDMSsw} \quad (4)$$

where $K_{POLYMERsm}$ and K_{PDMSsm} are partition coefficients between polymer and solvent mixture and between PDMS and solvent mixture, respectively, K_{PDMSsw} is partition coefficient between PDMS and seawater.

From Fig. 5(a), the hydrophobicity (K_{ow}) is generally in the order of HCHs < PCBs < DDTs < PAHs. It is clear that the log K_{ow}, log K_d or log K_f of DDT (log $K_{ow} = 6.79$; log $K_d = 4.99 \pm 4.31$; log $K_f = 5.9$) on PE is generally higher than that of Phe (log $K_{ow} = 4.6$; log $K_d = 4.71 \pm 4.08$; $K_f = 4.6 \pm 0.12$). Similarly, the value of log K_{ow}, log K_d or log K_f of DDT (6.79, 5.02 ± 4.18, 5.4) on poly(vinyl chloride) (PVC) is also greater than that of Phe (4.6, 3.36 ± 2.81, 3.3) (Table S5). Fig. 5(b) shows that the partition coefficients ($K_{POLYMER\text{-}sw}$ and $K_{POLYMER\text{-}w}$) of chemicals on different types of MPs have a strong relationship with the K_{ow} of chemicals, which is in accordance with Ziccardi et al. (2016). For the fitting curves in Fig. 5 (b), the values of Adj. R-Square of Log$K_{PE\text{-}w}$, Log$K_{PE\text{-}sw}$, Log$K_{PP\text{-}sw}$ and Log$K_{PS\text{-}sw}$ with Log K_{ow} are 0.97, 0.92, 0.93 and 0.83, respectively. The fitting results of Log$K_{PE\text{-}w}$ and K_{ow} are the best, followed by Log$K_{PP\text{-}sw}$ and Log$K_{PE\text{-}sw}$, and the worst is Log$K_{PS\text{-}sw}$. Generally, the fitting result can successfully indicate the relationship between Log K and Log $K_{ow.}$ However, the log K_f of DEHP is lower than that of DDT while the log K_{ow} and log K_d of the former chemical is higher. This may indicate that the sorption of DEHP is more consistent with the Freundlich isotherm model. In Fig. 5(a), the log K_d (or log K_f) values of DEHP and DDT were all higher than those of Phe and perfluorooctanoic acid (PFOA), which suggests the same plastic materials sorb more DEHP and DDT than Phe and PFOA. It needs to point out that PFOA usually carries ionic charges in aquatic environment, which is also the reason of lower sorption.

The log K_d (or log K_f) values of Phe, DDT, PFOA, and DEHP on PE are apparently higher than on those on PVC. This suggests that PE has a higher affinity with most chemicals than PVC. For example, Teuten et al. (2007) showed that PE particles can sorb and accumulate more organic chemicals than other plastics like PP and PVC pellets. From Fig. 5(a), the values of log $K_{PE\text{-}W}$ for PAH isomers follow the order: BghiP > benzo[*a*]pyrene (BaP) > benz[*k*]fluoranthene (BkF) > Chr > benz[*a*]anthracene (BaA) > pyrene (Pyr) > Fla > anthracene (Ant) > fluorene (Flu) > acenaphthene (Acp) > naphthalene (Nap). Such a finding illustrates that PE sorbs more high-ring (4–6 rings) PAHs than low-ring PAHs (2–3 rings) in aquatic environment as larger (high-ring) PAHs are more hydrophobic. Similarly, PP and PS show higher affinities towards high-ring PAHs than with low-ring PAHs. The log K_d (or log K_f) for PCBs, PAHs, HCHs and DDTs on PE in water are in the range of 4.19–6.88, 2.81–7.27, 2.2–2.8 and 4.22–5.77 and the log K_{ow} for PCBs, PAHs, HCHs and DDTs are as followed 5.11–7.88, 3.00–6.35, 3.55–4.14 and 6.79 (DDT) (Table S5). These findings indicate that PAHs, PCBs and DDTs can partition to MPs more readily than HCHs (Frias et al., 2013; Hirai et al., 2011; Karapanagioti et al., 2011). As for log $K_{PE\text{-}sw}$, log $K_{PP\text{-}sw}$ or log $K_{PS\text{-}sw}$, the value for α-HCH is greater than that of other HCH isomers, followed by γ-HCH, δ-HCH and β-HCH (Fig. 5(a)). This suggests plastic has a higher affinity with α-HCH and γ-HCH than δ-HCH and β-HCH in seawater. When comparing log $K_{PE\text{-}sw}$, log $K_{PP\text{-}sw}$ and log $K_{PS\text{-}sw}$ of HCH isomers, it is clear that HCH could partition more on PS than on PP and PE. For PAH isomers, the values of log $K_{PE\text{-}sw}$ and log $K_{PS\text{-}sw}$ are similar while the log $K_{PP\text{-}sw}$ is relatively low. Generally, log K_d (or log K_f) values in seawater are higher than in water, such as the log $K_{PE\text{-}sw}$ of PAHs is greater than their log $K_{PE\text{-}w}$. However, the log $K_{PE\text{-}sw}$ (5.22) of hexachlorobenzene is slightly lower than log $K_{PE\text{-}w}$ (5.43) (Table S5). It suggests that salinity is an important factor influencing the partition behavior (Bakir et al., 2014), as discussed further below.

4.2. The materials of microplastics

It is well known that MPs have different types, colors, sizes and compositions. The most common types of MPs in the marine environment are PE and PP, PS, and PVC (Endo et al., 2005). It was reported that organic chemicals were easily sorbed on MPs due to their hydrophobic nature (Takada, 2006). Many other studies observed that chemicals usually had higher diffusion coefficients on low-density MPs than high-density MPs (Fries and Zarfl, 2012; Karapanagioti and Klontza, 2008; Lee et al., 2018; Mato et al., 2001, 2002; Teuten et al., 2007). It should be attributed to the slower diffusion rate of chemicals on high-density MPs. For example, the sorption of perfluorooctane sulphonate (PFOS) and perfluorooctanesulfonamide (FOSA) on PE and PVC was reported to be greater than on PS (Wang et al., 2015), while the partition coefficient of Phe on PE was an order of magnitude higher than that of PP, followed by PVC (Teuten et al., 2007). Polyethylene has generally been reported to exhibit greater adsorption capacity than other types of plastics (Alimi et al., 2017). The distribution order of chemical substances on various types of plastic were LDPE ≈ HDPE ≥ PP > PVC ≈ PS (O'Connor et al., 2016), where PVC and PS are glassy polymers with the glass transition temperature (T_g) at > 80 °C is higher than normal ambient temperature, resulting in low mobility of the polymer and low diffusivity of the penetrant. These findings showed the sorption of chemicals was related to the type of plastics (Lee et al., 2014; Rochman et al., 2013a).

Previous studies showed concentrations of chemicals (such as PCBs and PAHs) on black and aged plastic pellets were much higher than those on white and colored ones (Antunes et al., 2013; Frias et al., 2010, 2013; Rios et al., 2007). Black plastic pellets may have more additives like PU than white pellets, which may enhance the sorption level (Frias et al., 2013). The size of plastic particles is also an important factor. Table S6 shows some physical characteristics and partition coefficients for sorption of Phe on various polymers (Teuten et al., 2007). It is clear that PE, PP and PVC of the same size, ranging from 200 μm to 250 μm, had different BET surface areas and distribution coefficients. Table S6 showed that the log K_d of PE (log $K_d = 4.58 \pm 3.75$) was greater than PP (log $K_d = 3.34 \pm 2.23$) and PVC (log $K_d = 3.22 \pm 2.30$), which was attributed to the larger surface area of PE than PP and PVC. Plastic particles with smaller sizes had a higher surface area for sorption than larger particles, and larger one also had longer distance for chemical diffusion than smaller one when the equilibrium is reached (Lee et al., 2014; Seidensticker et al., 2017). For example, the distribution coefficients of nanoplastics (log $K_d = 5.82 \pm 5.23$) were much higher than that of MPs (log $K_d = 4.23 \pm 3.04$) (Ma et al., 2016). Such findings indicated that the particle size of plastics was not only crucial for organic chemical sorption but also for equilibration times (Velzeboer et al., 2014).

The four common types of plastics (PE, PP, PVC, and PS) at a given environmental temperature can be divided into two categories: rubbery plastics (PE and PP) and glassy plastics (PVC and PS), which are also connected to the T_g (Alimi et al., 2017; Kurtz, 2004; Teuten et al., 2009). Teuten et al. (2009) reported that a certain polymer can transit from a rubbery state to a glassy state when it cools below its T_g. Some studies showed that rubbery plastics such as PE and PP had a higher affinity with chemicals than glassy plastics such as polyethylene terephthalate (PET) and PVC (Guo et al., 2012; Rochman et al., 2013a; Wu et al., 2001). George and Thomas (2001) proposed that glassy polymers have a more dense structure with little void space and present higher cohesive forces while rubbery polymers exhibit a large amount of free volume between molecules and present greater mobility and flexibility. Glassy polymers have long-lived and closed internal nanoscale pores due to their rigidity that act as adsorption sites. Such structures lead to a higher sorption level of chemicals and slower diffusion rate in glassy polymers than rubbery plastics (Teuten et al., 2009). The phenomenon is reflected in the intrapolymer diffusion coefficients as well. Specifically, the diffusion coefficients of organic compounds in PS, PVC are lower than in PE (Chen et al., 2018; Seidensticker et al., 2017). In general, rubbery plastics, small particulate plastic particles and black plastic pellets likely sorb more chemicals.

4.3. The degree of weathering/aging effect

As mentioned in the previous section, the aging of plastics increased its ability to sorb chemicals (Endo et al., 2005; Ogata et al., 2009; Rios et al., 2007). Aged plastic has a greater state of degradation mainly due to long-term photooxidation, seawater corrosion, friction or other processes (Brennecke et al., 2016; Eriksson and Burton, 2003). Photooxidation and friction made large plastic pieces exposed to UV light decompose into MPs, thereby increasing specific surface areas and surface roughness of MPs, which further increase their sorption capacities. Furthermore, Pérez et al. (2010) indicated that the aging process led to a decrease in the molecular weight of the polymer, which indirectly affects its sorbent properties. Another phenomenon was to use beach plastic particles as passive samplers to monitor the concentrations of chemicals in marine environment (Ogata et al., 2009; Takada, 2006). Rummel et al. (2017) showed that biofilm-plastic interactions have the ability to affect the fate and impact of MPs by increasing the specific surface area and equilibration times. In summary, the effect of weathering/aging on plastics would enhance the sorption capacity of chemicals due to a higher specific surface area, the surface coverage of organic matter, and so on.

4.4. The influence of salinity

As shown in Table S5, the log $K_{PE\text{-}sw}$ and log $K_{PE\text{-}w}$ of the same chemical are slightly different. For most PAHs, the log $K_{PE\text{-}sw}$ is slightly greater than log $K_{PE\text{-}w}$, while log $K_{PE\text{-}sw}$ of hexachlorobenzene and α-HCH is lower than log $K_{PE\text{-}w}$. While it would be expected that the salting-out effect would provide a greater fugacity to drive dissolved neutral organic compounds to sorb to MPs, this does not explain the latter observed decreases. Studies exist the effect of salinity on sorption and desorption between plastics and chemicals (Adams et al., 2007; Bakir et al., 2014; Velzeboer et al., 2014; Wang et al., 2015), but further investigations are needed to elucidate such an effect. Table S7 shows the corresponding log K_f for the sorption of Phe and DDT onto PVC and PE over a series of salinity gradients and desorption rate (k, $\text{day}^{-1} \pm \text{SD}$) in pure MilliQ water (0 psμ) and in seawater (35 psμ), adopted from Bakir et al. (2014). Salinity for test included 0, 8.8, 17.5, 26.3 and 35 psμ (practical salinity scale unit), corresponding to

0%, 25%, 50%, 75%, 100% of seawater, respectively. There was no significant change in the partition coefficient log K_f of PVC-Phe, PE-Phe, PVC-DDT and PE-DDT with the increase in salinity. It can be seen that the desorption rate between plastic and chemical in MilliQ water was a bit inferior to that in seawater. The desorption rate between plastic and DDT is distinctly less than that of plastic-Phe. According to Wang et al. (2015), salinity increase would increase the sorption capacity of PFOS on PE, but had no evident impact on FOSA sorption. The same study also found that PFOS in pure water cannot be sorbed on PS pellets, but sorbed on PS particles at high salt concentrations. These observations all indicate that the sorption of PFOS on MPs was much easier in seawater than in water. For the effects of salt on partition coefficients, Lohmann et al. (2012) used the Setschenow equation to calculate the effect of dissolved salts on the K_{sw} of nonpolar compounds. The equation was as follows:

$$\log K_{sw} = \log K_{sw},0 + K_s I \quad (5)$$

where K_{sw} is the sampler—water partition coefficient, I is the ionic strength (in moles per liter), K_s is the Setschenow constant (in liters per mole) and K_{sw},0 is the sampler—water partition coefficient at an ionic strength of 0.

4.5. The impacts of pH and temperature

At present, there are a few reports about the impact of solution chemistry on the chemical sorption on MPs. The pH of solution should theoretically affect ionic chemicals sorption on MP due to the electrostatic interaction. The study of Wang et al. (2015) showed that the sorption of PFOS on PE and PS particles increases with a decrease in pH, while neither had an effect on the sorption of neutral FOSA on these plastics. Such findings may suggest that properties of MPs and chemicals strongly influence the sorption process.

Table S8 shows log $K_{PE\text{-}w}$ values of some chemicals at different temperatures. The log $K_{PE\text{-}w}$ of Phe increases with increasing temperature from 18 to 24 °C (4.2 at 18 °C; 4.23 ± 0.02 at 23 °C and 4.3 ± 0.1 at 24 °C), but was slightly lower at 30 °C (4.16 ± 0.02) than at 24 °C. Pyr presents a similar trend as Phe with a log $K_{PE\text{-}w}$ value of 4.62 at 16.4—18.7 °C, which is smaller than those at 23 °C (5.02 ± 0.03) and 24 °C (5.0 ± 0.1). However, it slightly decreased when temperature reached 30 °C (4.90 ± 0.01). For most HOCs (e.g., Phe, Fla, Pyr, Chr and benzo(e)pyrene (BeP)), log $K_{PE\text{-}w}$ values at 30 °C were less than those at 24 °C. PCB-52 is an exception, with log $K_{PE\text{-}w}$ at 30 °C being slightly higher than that at 23 °C. The study of Lohmann et al. (2012) showed that partition coefficient (K_{sw}) for chlorobenzenes, PCBs, PAHs and DDE on plastics appears to be essentially temperature independent, possibly with the exception of Phe.

5. Microplastics and marine organisms

At present, MPs had been found in a large number of marine organisms (Gall and Thompson, 2015), as they mistake and swallow tiny MPs as food and transfer them to their food chains (Cole et al., 2013; Lusher et al., 2013). It was estimated that 693 species in 2015 had been affected by marine plastic pollution, while 267 species were recorded to have marine plastic debris in their bodies (Gall and Thompson, 2015). For example, plastic fragments or MPs were identified in the guts, stomachs, and tissues of marine organisms, such as seabirds (Blight and Burger, 1997), fishes (Collard et al., 2015; Miranda and de Carvalho-Souza, 2016), turtles (Bjorndal et al., 1994), whales (Fossi et al., 2012; Tarpley and Marwitz, 1993), crabs (Watts et al., 2016), and even the alimentary tract of gudgeons (*Gobio gobio*) (Sanchez et al., 2014). Table S9 shows the spatial distribution of MPs in several aquatic organisms from different areas. The amount of MPs in bivalves was reported to range from 4.3 to 57.2 items/individual of a Chinese fishery market (Li et al., 2015). In addition, the number of plastic fibers found was up to 178 items/individual in *Mytilus edulis* from Canada (Mathalon and Hill, 2014), while the levels of MPs (0.03 ± 0.18 items/individual) in demersal fish from the North Sea and Baltic Sea were approximately four orders of magnitude lower than that in *Mytilus edulis* (Rummel et al., 2016). Thus, the plastic debris in the ocean is not only a kind of global environmental pollution problem, but also brings potential harms to marine organisms.

Plastic debris is related to the injury of organism (Derraik, 2002; Gregory, 2009), and may even lead to sub-vital and mortality effects on individuals (Browne et al., 2015; Cole and Galloway, 2015). The harm of plastic fragments to marine life is mainly manifested in physical, chemical and biological effects.

With regards to physical aspects, plastic debris has a direct mechanical effect on marine creatures through entanglement and ingestion. For example, large plastic debris or fragments block the passage of food and affect appetite, and can even cause intestinal perforation and reproductive impairment (Derraik, 2002; Gregory, 2009; Moore, 2008; Wilcox et al., 2015). To be specific, plastic debris (particularly synthetic fibers) swallowed by animals may cause a false feeling of satiation or lead to internal blockage or damage to the digestive system. All these effects would then reduce food intake and affect digestion, and finally cause starvation and death (Duis and Coors, 2016; Foekema et al., 2013; Hjelmeland et al., 1988; Jackson et al., 2000; Ryana, 1988; Wright et al., 2013). According to von Moos et al. (2012), ingested plastic particles may stay in the alimentary canal and cause internal wear and inflammation. Predation further transfers MPs into the food chains. For example, pelagic zooplankton, are more susceptible to microplastic uptake, which indirectly causes serious harm to the food chain due to bioaccumulation and biomagnification of microplastics and chemicals (Cole and Galloway, 2015; Mohamed Nor and Obbard, 2014; Moore, 2008). Another example is the blue mussel (*Mytilus edulis*). They can ingest and transport microplastic particles to the gut, which then accumulate in the digestive cavity and tubules (Browne et al., 2008). Overall, the physical harm of MPs are to cause injury after ingesting plastic fragments, affect the digestive system, and even cause starvation (Donohue et al., 2001). But one important factor is that the MP concentrations in these lab experiments were orders of magnitude higher than those which can be found in the environment. Such physical injuries of MP should only occur with MP heavily-polluted areas.

With regards to chemical aspects, MPs indirectly affect the organism because they can sorb and concentrate harmful chemicals from the surroundings and transport these chemicals into organisms (Mato et al., 2001; Teuten et al., 2007, 2009). As mentioned earlier, MPs or plastic fragments could be an importance transport route or vector for chemicals into the marine food web (Bakir et al., 2012). Chemicals sorbed on the surface of MPs and plasticizers or additives released from plastics can enter the organism with the particles, and could be released through desorption processes to cause effects, such as lesions to the organism and ultimately to humans (Bakir et al., 2014; Cole et al., 2011; Farrell and Nelson, 2013; Neves et al., 2015; Watts et al., 2014). According to Ryana (1988), the amount of ingested plastic particles had a positive correlation with the concentrations of PCBs in the adipose tissues of great shearwaters (*Puffinus gravis*). In addition, MPs have a larger surface area than plastic debris, which make them more accessible to sorb organic chemicals and plasticizers from surrounding seawater (Lee et al., 2014). Microplastics are also small enough to enter the living body, while large plastic fragments can be blocked without a hitch. Thus, MPs may potentially cause greater damage than large plastic fragments to creatures in marine environment (Mohamed Nor and Obbard, 2014). Chemicals absorbed to MPs, such as PCBs, and those released from MPs, such as BPA, have carcinogenic, teratogenic and mutagenic effects on organisms (Zhang et al., 2016). However, some researchers are still controversial about the vector function of MPs. According to Gouin et al. (2011) and Beckingham and Ghosh (2017), the transfer of organic contaminants into biological organisms by dietary MPs may be a small or limited contribution compared to other natural routes of exposure in most cases. Therefore, MPs can introduce these harmful chemicals into organisms only when fugacity of a chemical in microplastics is greater than that in organism, which may result in an exposure risk.

With regards to biological aspects, MPs may become gathering places for some microorganisms. This will introduce new or non-native species into other areas through transport of plastic debris by ocean currents and wind patterns, such as bryozoans (Barnes, 2002; Derraik, 2002).

Last but not least, MPs indirectly have a harmful effect on human health. Bivalves such as mussels (*M. edulis*) and oysters (*Crassostrea gigas*) are popular seafood, in which MPs have been discovered in both farmed and wild bivalves (Mathalon and Hill, 2014; Van Cauwenberghe and Janssen, 2014). Therefore, humans at the top of the food chain may be subject to potential health risks after the consumption of seafood polluted by MPs and toxic chemicals (Seltenrich, 2015).

In summary, the MPs in the ocean not only cause marine environmental pollution issues, affecting marine biodiversity, but also have a potential of causing harmful effects on marine organisms and humans (Gall and Thompson, 2015).

6. Conclusions

This review analyzes the distribution characteristics of chemicals on MPs, and discusses the sorption behavior between MPs and chemicals. Chemicals adhered to MPs sampled from larger cities or highly industrialized areas are higher than that in smaller cities or remote areas. In addition, chemicals sorbed on black or aged MPs are at a greater concentration than that on colored or white MPs. Furthermore, chemical levels on MPs will also be reduced due to the prohibition on use of some organic chemicals and to the degradation of chemicals. These results are related to the sorption mechanisms between MPs and chemicals, which are mainly controlled by the physical-chemical properties of MPs, nature of chemicals, temperature, and solution chemistry. Finally, MPs may enter the body of organisms through ingestion, enter the food chain via predation, and eventually reach the human bodies. They can cause hazardous effects on ecological and human health. This review article calls for public attention on microplastic pollution from a scientific viewpoint.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.watres.2018.04.003.

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