Investigation on the adsorption-desorption behavior of antibiotics by polybutylene succinate and polypropylene aged in different water conditions

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Research Article

Keywords: Biodegradable MPs, DOM, Salinity, Ageing, Adsorption, Desorption.

Posted Date: October 11th, 2022

DOI: https://doi.org/10.21203/rs.3.rs-2085043/v1

Abstract

Microplastics (MPs) are widely present in the aqueous environment and aged by natural components, such as salinity (SI) and dissolved organic matter (DOM). The effects of multi-conditioned ageing on the physicochemical properties and environmental behavior of MPs are not completely investigated. In this study, the degradable MPs polybutylene succinate (PBS) was used to investigate the environmental behavior of sulfamethoxazole (SMZ), and compared it with polypropylene (PP). The results showed that the single conditions of DOM and SI, particularly DOM, promoted the ageing process of MPs more significantly, especially for PBS. The degree of MPs ageing under multiple conditions is lower than that of under single condition. Compared to PP, PBS has a greater specific surface area, crystallinity and hydrophilicity, so it has a stronger adsorption capacity for SMZ. The adsorption behavior of MPs fitted well with the pseudo-second-order kinetic and Freundlich isotherm models, indicating a multilayer adsorption. The desorption experiments showed that the desorption amount of SMZ on MPs in simulated intestinal fluid was greater than that in Milli-Q water. Both the original PBS and the aged PBS had a stronger desorption capacity than the PP. The desorption quantity of PBS was 1.23–1.84 times greater than PP, while the desorption rates were not significantly different. This experiment would provide a theoretical basis for assessing the ecological risks of degradable MPs in complex water conditions.

1. Introduction

Due to the low price, easy processing, stable performance, and other characteristics, plastics are widely used in agriculture, commerce, industry, daily necessities and many other fields (Jia et al., 2020; YooeunChae. & Youn-JooAn, 2018). Plastic products exposed to the environment are further broken down by heat, salt, and dissolved organic matter (DOM) into many different particle sizes (Lingchen Liu, 2022), and out of those which are smaller than 5 mm are called microplastics(MPs) (Andrady, 2017a).

Due to their light weight and small dimensions, MPs can easily enter the water environment through atmospheric deposition, and effluent discharges (Kp & Sm, 2021). Studies demonstrated that about 1.5–4.5% of the total plastics produced globally are released directly into the ocean (Nizzetto et al., 2016).Therefore, marine plastic pollution has become a research hotspot for many researchers (Chen et al., 2021; Li, 2019; Nakanoa & Arakawab, 2022). In the actual marine environment, plastics undergo a series of ageing processes, including physical wear and tear, ultraviolet radiation, biodegradation, chemical oxidation, etc. (Andrady, 2017b; Jahnke et al., 2017). The additives and intermediates released from the aging process of MPs, further increases the ecological risk of MPs (Liu et al., 2021; Luo et al., 2019). In addition, aged MPs have a large specific surface area, and high hydrophobicity and mobility, so they act as carriers of other pollutants in the environment by adsorbing them (Hongwei et al., 2022b). At the same time, it poses a threat to the health of marine life because it is small, not readily degradable and easily ingested by marine life (Bhatt et al., 2021; Golaa et al., 2021).

However, most of the current articles on MPs are limited to aging in pure water, ignoring the natural components of water (Fan et al., 2021a; Meng et al., 2021). The natural components in the water have an

influence on the ageing effect of MPs, such as changing their microstructure, surface morphology, microstructure and environmental behavior etc. (Liu et al., 2022). Salinity (SI) and DOM (Ling et al., 2021a; Schmidt et al., 2017) are the main components of seawater. Studies have shown that the functional groups (e.g. -OH, C - H) of conventional plastics aged in seawater change significantly, with an increase in oxygen-containing functional groups, resulting in the high hydrophobicity and fluidity of the aged MPs (Ling et al., 2021b). However, the current research on MPs ignores the influence co-existence SI and DOM during the aging process (Gao et al., 2021).

In order to reduce the pollution caused by the traditional plastics, degradable plastics are widely used in our daily lives (Sato et al., 2017). However, there are few investigations on degradable MPs (Ling et al., 2022). Petroleum-based degradable plastics can be completely degraded by the action of microorganisms in water (Zhu & Wang, 2020). Polybutadiene-styrene (PBS) is a petroleum-based degradable plastic that is widely used in food packaging films, plant protection films, bone tissue engineering and other applications. During the aging process, PBS can release millions of plastic fragments to the environment (Xin-Feng et al., 2022). Meanwhile, aged degradable plastics fragments have a larger specific surface area and can absorb more pollutants than aged conventional plastic plastics (Fan et al., 2021b; Yu et al., 2019). Although there are some researches on degradable MPs, but there is a lack of research on degradable MPs that aged in seawater.

Antibiotics are a class of antibacterial drugs that have been classified as a new type of pollutants due to their overuse (Roy et al., 2020). Sewage discharges and mariculture have resulted in large amounts of antibiotics in seawater (Du et al., 2019). The antibiotic content of seawater has been estimated to be as high as 181 ng/L (Shuang et al., 2020). Due to their small dimensions, marine organisms confuse MPs with food, which leads to accidental ingestion, causing MPs to accumulate in the food chain (Borrelle et al., 2017). Antibiotic-loaded MPs may migrate through the food chain, and adversely affecting individuals and different communities of organisms (Capolupo et al., 2019; Philipp et al., 2019). Therefore, there is a need to understand the mechanisms of interaction between MPs and antibiotics, which can help to determine the potential ecological risks of degradable MPs on pollutants.

In this study, the PBS and PP (Polypropylene) were used as target MPs, and Sulfamethoxazole (SMZ) was used as the target pollutant. The main objectives of this research were to; (1) investigate the effect of ageing on the physical properties of MPs under different conditions (i.e. DOM, SI, DOM/SI); (2) study on the behavior of PBS and PP in adsorption of SMZ after ageing; (3) explore the difference of MPs desorption behavior for SMZ in Milli -Q water and simulated intestinal fluid. This study broadens the scope of research on MPs and contributes to a more comprehensive assessment of the potential ecological risks of MPs.

2. Materials And Methods

2.1. Materials

PBS and PP used in this experiment were purchased from the Shanghai Guanbu Electromechanical Technology Co. (Shanghai, China). The average particle size of these two MPs was 40 μm. Chemicals that simulated seawater, such as NaCl, $MgCl_2$, Na_2SO_4, $CaCl_2$, FA (fulvic acid) were purchased from Aladdin Industrial Co. (USA), with purity ≥ 98%. The drugs used in the experiment sodium taurocholate (ST) and bovine serum albumin (BSA) with purity ≥ 98%, were purchased from Aladdin Industrial Corporation (USA).

2.2. MPs aging experiment

To simulate the ageing behavior of MPs in water, three different solutions were prepared; a 4% SI solution, a 6mg/L DOM solution, and a mixture of the two solutions. 4% SI solution was prepared with an inorganic salt's concentration of 28 g/L NaCl, 4.67g/L Na_2SO_4, 6.05g/L $MgCl_2$, and 1.32g g/L $CaCl_2$. PBS and PP were aged under UV irradiations for 48 h with different water conditions. In order to better simulate the ageing process of MPs in nature, 30w UV shortwave radiation was used. The aged samples were first washed, filtered, dried, and then were stored in a dark area and dried finally.

2.3. Characterization

The surface morphology of the MPs was characterized by scanning electron microscopy (SEM, Hitachi S-4800). The specific surface area of the samples was measured by an ASAP 2020 instrument (Micromeritics, USA). Characterization of the functional groups of original and aged MPs was done by X-ray photoelectron spectroscopy (XPS, ULVAC-PHI Inc., Japan) and Fourier transform infrared spectrometer (FTIR, Tensor 27., Bruker). Variation in the crystallinity of the reaction sample was performed by X-ray diffraction (XRD, X'Pert PRO MPD., Netherlands). By measuring the contact angle (Dataphysics OCA20, German) of the samples, the changes in hydrophilicity were investigated.

2.4. Adsorption experiments

In the adsorption kinetics test, the time intervals were set from 30-2880 min, and experiments were conducted at 25℃ by mixing the 50mg MPs samples with 50 mL of SMZ solution (50 mg/L) in the centrifuge tubes. Centrifuge tubes were shaken at 150 rpm in a dark air bath thermostatic shaker. Remove the centrifuge tubes at the set time intervals and filtered the sample solution through a 0.45μm membrane filter. The concentration of SMZ in the filtrate was determined by high performance liquid chromatography (HPLC).

Adsorption isothermal experiments were carried out in 100 mL centrifuge tubes, where SMZ concentrations were ranged from 1–20 mg/L. 50 mg samples were placed in the centrifuge tubes containing 50 mL of solution for the experiment. The experiments were carried out at 25℃ with a shaking speed of 150 rpm. The equilibrium time was set at 48 h. After the oscillation, the sample solution was filtered through a 0.45μm membrane filter, then loaded into a brown injection bottle for testing.

2.5. Desorption experiments

In the desorption experiment, 500 mg of sample was mixed with 500 mL SMZ solution with a concentration of 15 mg/L. After adsorption saturation of the MPs, the samples were filtered, then placed in a dark place, and later dried at low temperature. The desorption experiments were carried out in Milli-Q water, and simulated intestinal fluid. Centrifuge tubes containing 50mL of pure water and simulated intestinal fluid were taken and then the glass centrifuge tubes were placed in the air bath constant temperature oscillator shaker (150 rpm) in the dark at 25℃. The samples were shaken for 1-48h, and after that they were filtered through a 45μm membrane filter, and then measured by HPLC.

2.6. Date analysis

For the adsorption kinetics studies the pseudo-first-order kinetic model, and pseudo-second-order kinetic model were selected, and their equations are as follows (Pin et al., 2022):

$$ln\left(Q_e - Q_t\right) = lnQ_e - k_1 t$$

1

$$t/Q_t = \left(1/k_2 Q_e^2\right) + \left(1/Q_e\right) \times t$$

2

where Q_t(mg/g) is the SMZ adsorption capacity of the MPs at time t(min). k_1 (min^{-1}) and k_2(g/mg·min) are the rate constants of the pseudo-first-order, and pseudo-second-order, respectively.

To understand the rate at which SMZ was adsorbed by the three different types of MPs, an intraparticle diffusion model was used.

$$Q_t = k_p t^{0.5} + C$$

3

where, k_p (mg/ (g min$^{0.5}$)) is the internal diffusion rate and C is the mean boundary-layer thickness.

Adsorption isotherms can be fitted with the following models.

Langmuir isotherm: $C_e/Q_e = 1/\left(K_L Q_{max}\right) + C_e/Q_{max}$ (4)

Freundlich isotherm: $lnQ_e = lnK_F + \left(1/n_F\right) lnC_e$ (5)

where Q_{max}(mg/g) is the maximum value of the SMZ adsorption capacity of the MPs. k_L(L/mg) and k_F [(mg/g) (L/mg)] are the Langmuir and Freundlich distribution coefficients, respectively. $1/n_F$ is the Freundlich model parameter that reflects the adsorption intensity and heterogeneity of the adsorbent.

3. Results And Discussion

3. 1. Characteristics

3. 1. 1. Degree of crystallinity, morphology and surface properties

Table 1 compared the changes in crystallinity of PBS and PP before and after ageing. The crystallinity of PBS increased from 24.82–26.31% (Pure water-aged), 29.24% (DOM-aged), 28.56% (SI-aged) and 26.68% (DOM/SI-aged), respectively. The crystallinity of PP increased from 14.14% to 19.48–25.98%. The crystallinity of the MPs increased during the ageing. The findings were consistent with previous studies (Qinke et al., 2022). The single conditions (i.e. DOM, SI) promoted the ageing of MPs. The increase of crystallinity is due to the destruction of the non-crystalline structures of the MPs by reactive oxygen species (ROS) during the ageing process, resulting in localized areas of secondary crystallization (Peipei et al., 2021). In the condition of severe photodegradation, the crystallinity of the MPs increased significantly (Olga et al., 2022). Therefore, an in-depth study of PBS was carried out. The fragmentation of the PBS surface was observed by SEM.

Table 1
The Degree of crystallinity and $S_{BET}(m^2/g)$ of the original and aged PBS, PP.

Microplastics	Aging environment	Degree of crystallinity	$S_{BET(}m^2/g)$
PBS	Original	24.82%	0.35
	Pure water-aged	26.31%	0.43
	Dom-aged	29.24%	0.51
	SI-aged	28.56%	0.47
	DOM/SI-aged	26.68%	0.45
PP	Original	14.15%	0.37
	Pure water-aged	19.48%	0.45
	Dom-aged	25.98%	0.82
	SI-aged	23.17%	0.67
	DOM/SI-aged	21.91%	0.45

Fig. 1 showing the SEM images of original PBS and PBS aged in different water conditions (i.e. pure water, DOM, SI, DOM/SI). The surface of the original PBS is smooth (Fig. 1a). The PBS aged in pure water for 48h shows small cracks on the surface (Fig. 1b). According to Figs.1(c-e), the PBS aged in DOM, SI, or

DOM/SI component exhibits visible holes and cracks on the surface. It can be clearly seen from the images that some of the plastic fragments have come off the whole. This is because MPs ageing generally has two modes of degradation i.e. cracking and flaking (Gao et al., 2021), and PBS is degraded mainly by cracking (Si et al., 2022). Fig. 1 shows the complexed water environments accelerates the formation of pores and cracks on the PBS surface, which increases the adsorption sites for pollutants, thus it increases the ability to adsorb them.

The specific surface area (S_{BET}) figures for PBS and PP are illustrated in Table 1. The S_{BET} of the original PBS and PP MPs were 0.35 m^2/g and 0.37 m^2/g, respectively, and increased to 0.43 m^2/g and 0.45 m^2/g after aging in pure water. As it can be seen in Table 1, natural components in the water column promote the ageing process of MPs. For example, the S_{BET} of PBS increased to 0.51 m^2/g, 0.47 m^2/g, 0.45 m^2/g in the DOM, SI, and DOM/SI respectively. The S_{BET} of PP after ageing in three components are 0.82 m^2/g, 0.67 m^2/g, and 0.45 m^2/g, under the conditions of DOM, SI, and DOM/SI, respectively. The S_{BET} of the MPs surface increases due to photodegradation. Under single conditions, DOM and SI accelerated the ageing process of MPs, and after ageing, the S_{BET} of the MPs increased, which was similar to the results of previous studies (Yinghua et al., 2022a). Under the coexistence of SI and DOM, SI will reduce the solubility of DOM and forms solidification or precipitation (Robert et al., 2018), which slows down the MPs photodegradation. This suggests that single conditions in water promote the ageing process of MPs. The results for the specific surface area are in agreement with the XRD and SEM results.

3.1.2. Surface functional groups and contact angles

Figure 2 reveals the changes in the functional groups of MPs before and after ageing. For PBS (Fig. 2a), the peak near 3441 cm^{-1} is the absorption peak caused by the hydroxyl group(-OH), and the peak at 2960 cm^{-1} is the absorption peak caused by the anti-symmetric stretching vibration of the methyl group(-CH_3). The absorption peak of the carbonyl group(C = O) is located near 1725 cm^{-1} and the absorption peak of -CH groups is located near 1152 cm^{-1} (Meng et al., 2022). According to Fig. 1a, the intensity of the carbonyl peak of PBS increased after ageing due to the oxidation of C-H. For PP, the main characteristic peaks are around 3426 cm^{-1}, 2964 cm^{-1}, 1720 cm^{-1}, corresponding to the functional groups -OH, -CH_3 and C = O, respectively. The peak near 738 cm^{-1} is produced by the -CH_2 bending vibration (Jiang et al., 2022). The high oxygen content of original PP may be caused by plasticizers added during the production process (Yinghua et al., 2022b). The weakening of the absorption peak of -CH_3 in aged PP is due to oxidation process during ageing.

After ageing, the oxygen-containing functional groups such as hydroxyl and carbonyl groups on the surface of the MPs increased, which is similar to the results of previous studies (Jianxin et al., 2022). The C = O stretching vibration is more pronounced in aged MPs. This is because the ageing conditions contain Cl^{-1} (Mao et al., 2020), which is more susceptible to substitution reactions and therefore has a higher degree of oxidation. Qiu et al. (2022b) points out that DOM can release ROS and promotes the

ageing process of MPs. The interaction of PP with the aromatic structure in FA results in a more pronounced change in the characteristic peak under the ageing condition of DOM (Hongwei et al., 2022a). However, the co-existence of DOM and SI conditions did not show a higher degree of oxidation, probably because the DOM and the SI are mutually constraining, thus reducing the ageing process of the MPs. The increase in the number of oxygen-containing functional groups increases the hydrophilicity of the aged MPs.

Figure S1 shows the changes in contact angles of the MPs before and after ageing. The contact angle of the original PBS was 81.11°. After ageing, the contact angle of the PBS is reduced to 55°-64°. The contact angle of the original PP was 125.83°, while all the contact angles were less than 80° after ageing. This phenomenon indicates that PP changes from hydrophobic to hydrophilic, and this is similar to the results of previous studies (You et al., 2021). These results suggested that ageing increases the oxygen-containing functional groups and therefore increased the hydrophilicity of the MPs. This is in agreement with the FTIR results. This study shows that UV radiations and water environment factors are conducive to enhance the hydrophilicity, and accelerates the ageing process of MPs.

3.1.3. The O/C ratio and CI index

The O/C ratio and carbonyl index (CI) are used to indicate the degree of ageing of MPs (Fan et al., 2021c). As can be seen from Table 2, the O/C ratio of PBS increased from 0.412 to 0.416 (pure water), 0.432 (DOM), 0.420 (SI) and 0.417 (DOM/SI), respectively. The O/C ratio of PP increased from 0.281 to 0.348–0.383. Under the same conditions, the O/C ratio of PBS was higher than the PP, which means that PBS is more susceptible to ageing than PP.

The CI index is being used as a metric to indicate the degree of ageing of MPs. The CI index of PBS increased from the original 0.077 to 0.082 (pure water), 0.220 (DOM), 0.190 (SI), 0.162 (DOM/SI), respectively (Table 2). PP increased from 0.129 to 0.171–0.254. The pattern of change in the CI index is consistent with the O/C ratio. The MPs aged under DOM conditions have a higher degree of ageing. This may be attributed to the fact that ROS produced by DOM under UV irradiations promote the ageing process of MPs (Qiu et al., 2022a). The CI index of MPs aged SI conditions is lower than aged in DOM. This is probably due to the presence of Cl^-could impeded the photo aging process of MPs (Xiaowei et al., 2021).

Table 2
Changes of O/C ratio and CI index in PBS and PP after UV radiation ageing.

Samples	Wide-Scan elemental content			CI index
	C (%)	O (%)	O/C	
Original PBS	70.70	29.19	0.412	0.077
Pure water-aged PBS	70.51	29.40	0.416	0.082
Dom-aged PBS	70.53	30.51	0.432	0.229
SI-aged PBS	70.52	29.67	0.420	0.190
DOM/SI-aged PBS	70.50	29.40	0.417	0.162
Original PP	77.95	21.93	0.281	0.129
Pure water-aged PP	75.57	26.28	0.348	0.171
Dom-aged PP	72.11	27.62	0.383	0.254
SI-aged PP	72.62	26.69	0.368	0.231
DOM/SI-aged PP	74.11	27.62	0.372	0.183

3.2. Adsorption kinetics

In order to better analyze the adsorption process and elucidate the adsorption mechanism, kinetic equations were used to fit the experimental data, and the resulting parameters are summarized in Table S1. The pseudo-second-order model ($R^2 > 0.99$) was better fitted to the SMZ adsorption data for PBS and PP. This suggested that the adsorption processes of MPs on SMZ are mainly chemisorption (Bao et al., 2020).

The k_2 values of aged MPs decreased during the ageing process. The low values of k_2 indicated that the adsorption rate decreased with time, and absorption rates are proportional to the number of unoccupied sites (Gupta et al., 2010). As shown in Fig. S2, the adsorption quantity of MPs increases gradually during the first 1500 min, and after 1500 min, the adsorption quantity hardly varies with time, so saturation is reached at this point.

According to Fig. S2, the adsorption quantity of SMZ by the aged MPs increased with an increase in time. MPs aged under DOM condition shows a strong carrier capacity for pollutants, especially for PBS (Fig. S2). For example, the adsorption quantity of PBS increased from 4.56 mg/g to 5.74 mg/g, and PP increased from 2.80 mg/g to 3.41 mg/g. The adsorption capacity of MPs is related to their physicochemical properties. Compared to PP, FTIR results showed that PBS had more oxygen-containing functional groups on its surface, which facilitates the adsorption of SMZ on PBS. At the same time, PBS has larger specific surface area (Fig. 1, Table 1), thus increasing the adsorption sites on the surface of the

MPs, and enhancing its ability to adsorb pollutants. Previous studies have also confirmed these results (Huimin et al., 2021). In addition, the crystallinity of the MPs increased during ageing, which facilitated the enhanced adsorption to the SMZ (Shi et al., 2021a). Thus, compared to PP, PBS can adsorb more antibiotics, has a greater carrier capacity, and poses a greater ecological risk to water environment.

In order to clarify the adsorption mechanism of SMZ on original and aged MPs, an intraparticle diffusion model was used to fit the adsorption kinetic data. The results are summarized in Fig. 3, Table S2. It is clear from Fig. 3 that the internal particle diffusion model is a linear relationship, where $k_{p1}>k_{p2}>k_{p3}$. The slope determines the rate of adsorption, and the fitting parameter(C) reflects the causes that influence the rate of adsorption (Inyang et al., 2016). Therefore, the adsorption process of SMZ on MPs is divided into three main stages (Li et al., 2021). Stage : external mass transfer; Stage : Interfacial diffusion; Stage : Intraparticle diffusion.

At the Stage , the adsorption of MPs has reached an equilibrium. The k_{pi} values of the aged MPs are higher than that of the original MPs. This may be due to the increased number of MPs adsorption sites after ageing, which facilitates faster access of SMZ to the adsorption sites. Meanwhile, the k_{pi} values for aged PBS were higher than that of aged PP, indicating that the antibiotics diffused more rapidly in aged PBS. PBS has developed large cracks and higher crystallinity during the ageing process, which makes it easier for antibiotics to be adsorbed (Pin et al., 2022; Shi et al., 2021b).

3.3. Adsorption isotherms

The results of the Langmuir and Freundlich isothermal sorption models fitted to the sorption isotherm data are shown in Table 3. By comparing the R^2 of the two models, it was found that the Freundlich isotherm model ($R^2 > 0.93$) better describes the adsorption behavior of SMZ on MPs than Langmuir ($R^2 < 0.89$). This indicated that the adsorption of SMZ on PBS and PP was multi-layer adsorption. In Freundlich isotherm, k_F (mg/g(L/mg)$^{1/n}$) and $1/n_F$ are the parameters reflecting adsorption capacity of the adsorbents and the parameter reflecting adsorption strength, respectively.

Table 3
Isotherm model parameters of SMZ adsorption by Original and aged PBS and PP.

Adsorbent	Langmuir			Freundlich		
	Q_{max}(mg/g)	k_L	R^2	$1/n_F$	k_F	R^2
Original PBS	9.3806	0.0650	0.6972	0.4285	0.5390	0.9924
Pure water aged-PBS	8.1633	0.0928	0.6476	0.7509	0.6834	0.9918
Dom-aged PBS	14.26534	0.1438	0.8470	0.7125	1.7220	0.9889
SI-aged PBS	5.9988	0.3527	0.7859	0.4507	1.5952	0.9960
DOM/SI-aged PBS	20.9205	0.0289	0.5769	0.5035	0.5995	0.9853
Original PP	5.6883	0.0083	0.1830	0.9899	0.1959	0.9866
Pure water aged-PP	4.6067	0.0102	0.1234	0.9975	0.7900	0.9716
Dom-aged PP	11.1732	0.1127	0.4743	0.8094	0.9108	0.9368
SI-aged PP	9.7561	0.0201	0.1628	0.9751	0.4078	0.9303
DOM/SI-aged PP	5.5096	0.0251	0.2647	0.9608	0.3901	0.9415

For PBS, the k_F and $1/n_F$ increased after ageing. Of the changes, the most pronounced effect was seen in PBS aged in DOM, where k_F increased from 0.5390 mg/g(L/mg)$^{1/n}$ to 1.722 mg/g(L/mg)$^{1/n}$, and $1/n_F$ increased from 0.4285 mg/g(L/mg)$^{1/n}$ to 0.7125 mg/g(L/mg)$^{1/n}$. This suggests that MPs aged in DOM have a strong adsorption capacity. This is because DOM accelerates the ageing process of the MPs, creating cracks and pits on his surface and increasing his adsorption sites. Compared to PBS, PP has a weaker adsorption capacity. The k_F values increased from 0.1959 mg/g(L/mg)$^{1/n}$ to 0.3910–0.9108 mg/g(L/mg)$^{1/n}$. However, the values of $1/n_F$ do not differ much, which may be due to the short ageing time.

PBS has a stronger adsorption capacity for SMZ than PP, which may be due to the different physicochemical properties of PBS and PP. The reasons for the different physicochemical properties are following; (1) According to Fig. 2, aged MPs contain more oxygen functional groups. The oxygen-containing functional groups increased the hydrophilicity of MPs as they make hydrogen bonds with water (Fig. S1), thus enhancing the adsorption of antibiotics (Ke et al., 2022);(2) Aged MPs have a larger specific surface area and more adsorption sites, which will increase the ability of degradable MPs to adsorb antibiotics (Yanji et al., 2022), and as a result, PBS is more likely to carry higher levels of contaminants such as antibiotics in water environments.

3.4. Desorption kinetics

The data for the desorption of MPs in pure water and intestinal fluid are shown in Fig. 4, Fig. S3. It can be seen that the amount of desorption may be related to several factors; (1) the environment of desorption (Wai-Kit & Kelvin Sze-Yin, 2019); (2) types of MPs; and (3) the ageing conditions of the MPs. As can be seen in Fig. 6, the amount of desorption in the intestinal fluid is significantly greater than that in the water.

According to Fig. 4a, the desorption of the original PBS in the intestinal fluid was 1.14 mg/g, which increased after aging to 1.75 mg/g (pure water-aged), 3.15 mg/g (DOM-aged), 2.94 mg/g (SI-aged), and 2.46 mg/g (DOM/SI-aged). From Fig. S3a, it can be seen that the desorption rate increased from 29.00–36.44%, 54.83%, 53.90%, 46.51%, respectively (Fig. S3a). The amount of original PP desorbed in the simulated intestinal fluid increased from 1.10 mg/g to 1.71mg/g, and the desorption increased from 39.36–50.59%. The quantity and rate of desorption of antibiotics in Milli-Q water is much smaller than the amount in the intestinal fluid (Fig. S3b).

This indicates the presence of certain substances in the intestinal fluid that facilitate the release of antibiotics from the MPs. The active agent on the intestinal surface can increase the desorption of the polymer by increasing the diffusion rate of the pores within the particles. In addition, hydrophobic organic pollutants can be easily desorbed from the MPs after adsorption onto the surface of the MPs (Hartmann et al., 2017). According to Fig. 4 and Fig. S3, PBS has a stronger desorption capacity for SMZ than PP. The high desorption efficiency of PBS may be due to the large surface area, and hydrophobic interaction between SMZ and PBS, resulted in the release of more SMZ from the PBS surface (Yali et al., 2022).

4. Conclusions

The changes in the physicochemical properties and environmental behavior of PBS aged under different water environments were systematically investigated. The PBS and PP MPs were used in this research, the key findings of the study are as follows;

(1) The pattern of variation in crystallinity and O/C ratio indicated that PBS was more susceptible to ageing and degradation than PP. The hydrophilicity and specific surface area of MPs increased during the ageing process. The ageing process of MPs was encouraged under single conditions (i.e. DOM, SI). The coexistence of multiple aqueous environmental factors (i.e. DOM and SI) did not have a synergistic action on the ageing process of MPs, which may be mutually constrained and the degree of ageing was lower than under single conditions.

(2) The adsorption kinetics tests showed that the adsorption of SMZ by MPs occurs mainly through surface adsorption and intraparticle diffusion. The results of the adsorption isotherms indicated that the adsorption of SMZ by MPs was multilayer adsorption on non-uniform surfaces. Compared to PP aged under the same conditions, PBS shows better adsorption capacity. The ageing process increases the adsorption capacity and strength of MPs, and also increases the ecotoxicity of MPs.

(3) The desorption experiments showed that the quantity of MPs desorbed on the simulated intestinal fluid is approximately 10 times higher than that in Milli-Q water. Ageing enhances the desorption ability of SMZ by MPs. Compared to PP, PBS showed a higher desorption capacity. This research would provide a theoretical basis for assessing the ecological risks of degradable plastics in the complex water conditions.

Declarations

Acknowledgements

We are grateful for the grants for the National Natural Science Foundation of China (NO. 52000153), the Natural Science Foundation of the Jiangsu Higher Education Institutions of China (NO. 22KJA610005), the Jiangsu Association for Science and Technology Young Scientific and Technological Talents Project (No. TJ-2021-061), the Water Conservancy Technology Project of Jiangsu Province (NO. 2021077), the Xuzhou Key Research and Development Plan Project (social development) (NO. KC20163). The authors would like to thank Shiyanjia Lab (www.shiyanjia.com) for the HPLC analysis.

Author Contributions

Xiulei Fan*: Conceptualization, Resources, Writing - Review & Editing, Supervision, Funding acquisition. Weiyi Li: Data curation, Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Writing-Review & Editing. Easar Alam: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Data curation, Visualization, Writing-Review & Editing, Supervision, Project administration, Funding acquisition. Binwen Cao: Writing - Review & editing. Shenwen Qian: Writing - Review & editing. Shang Shi: Writing - Review & editing. Yangyang Yang: Writing - Review & editing.

Funding

This work was supported by the National Natural Science Foundation of China [grant numbers 52000153], the Natural Science Foundation of the Jiangsu Higher Education Institutions of China [grant number 22KJA610005], and the Jiangsu Association for Science and Technology Young Scientific and Technological Talents Project [grant number TJ-2021-06], the Water Conservancy Technology Project of Jiangsu Province [grant number 2021077], the Xuzhou Key Research and Development Plan Project (social development) [grant number KC20163]. Meanwhile, they had no involvement in study design, in the collection, analysis and interpretation of data, in the writing of the report, and in the decision to submit the article for publication.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Competing interests

The authors declare no competing interests.

Data availability

Not applicable.

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Figures

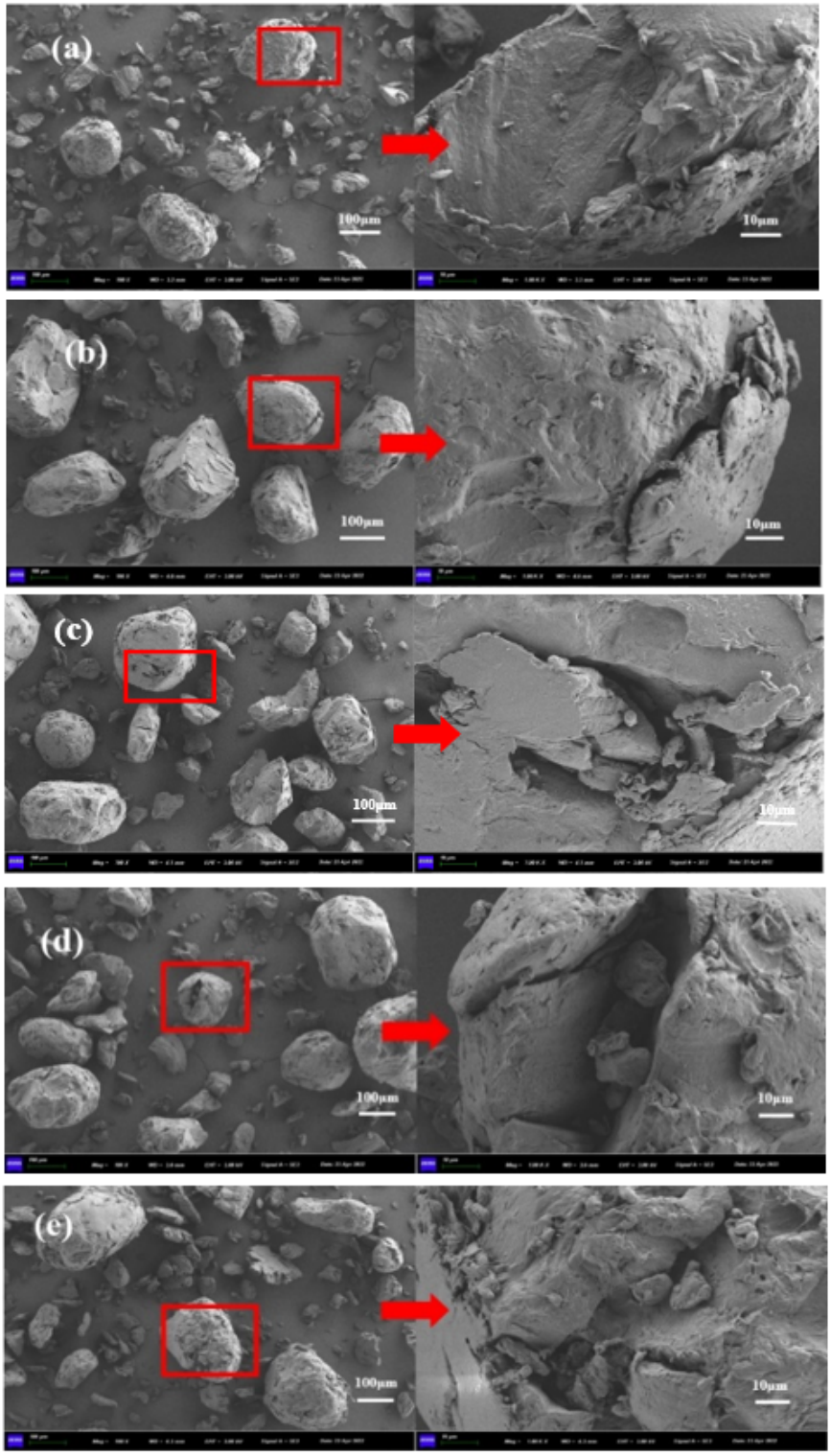

Figure 1

SEM images of original PBS (a), pure water-aged (b), DOM-aged(c), SI-aged(d), DOM/SI-aged(e)

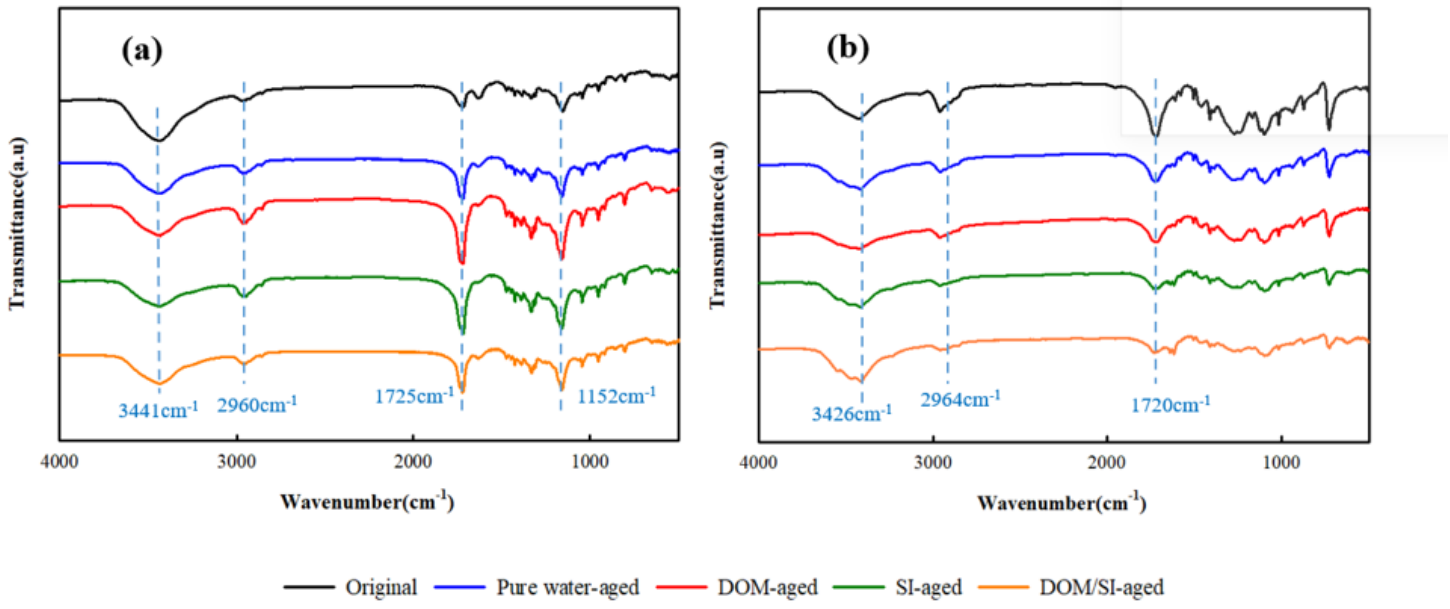

Figure 2

FTIR spectra of PBS(a) and PP(b) aged in different water conditions

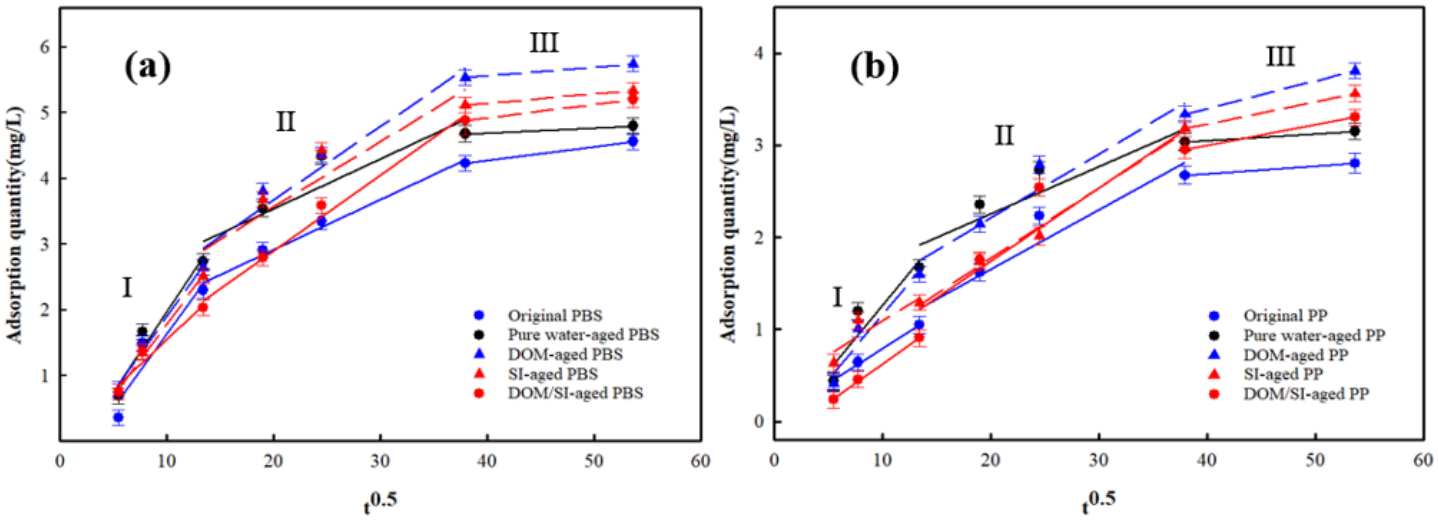

Figure 3

Intra-particle diffusion model plots for SMZ adsorption by original, aged PBS(a) and PP(b)

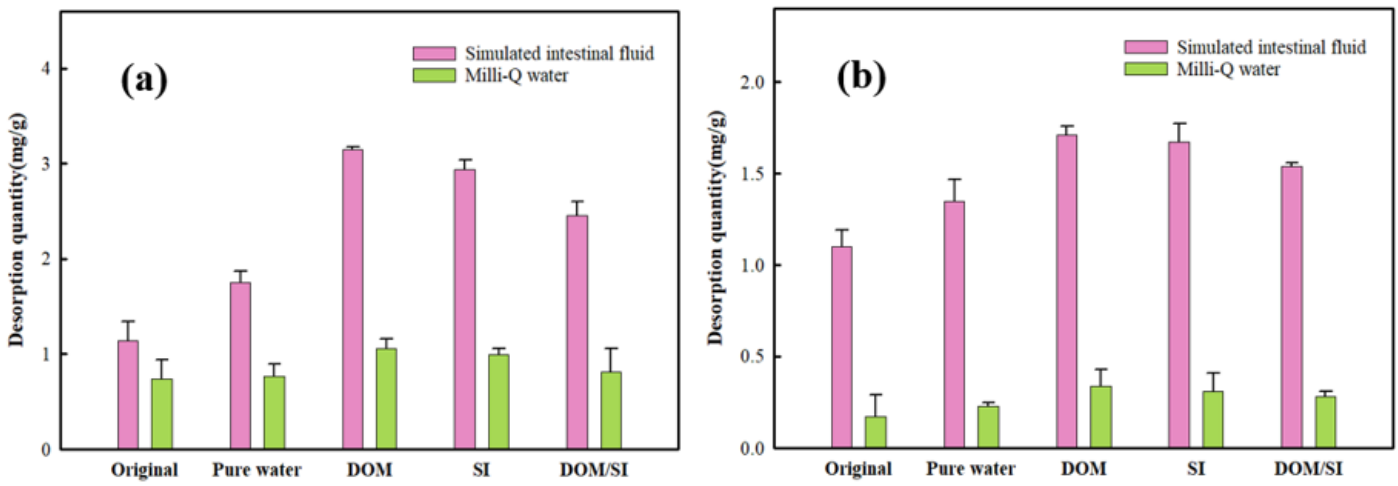

Figure 4

Desorption quantity of SMZ in Milli-Q water and simulated intestinal fluid by PBS(a) and PP(b)

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