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Tetracycline uptake by photoaged polypropylene and high-density polyethylene microplastics in water

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ABSTRACT

Microplastics coexist with various contaminants in aquatic environments and may serve as critical sorbents for those contaminants, including tetracycline (TC) antibiotics. In this study, the adsorption behavior of TC on pristine and photoaged polypropylene (PP) and high-density polyethylene (HDPE) microplastics was systematically investigated in aqueous systems. Fourier-transform infrared spectroscopy and scanning electron microscopy were used to observe the structural and surface changes induced by photoaging process. Batch adsorption experiments were conducted to study the effects of contact time, solution pH, and initial TC concentration. Photoaging significantly enhanced TC uptake on both type of polymers, with PP exhibiting a higher adsorption capacity than HDPE. Adsorption on both polymers followed pseudo-second-order kinetics, with photoaged materials exhibited lower rate constants but higher equilibrium capacities. The Langmuir model provided the best fit to equilibrium data, indicating the monolayer coverage of TC on polymer surfaces. The estimated maximum adsorption capacity (Q_{max}) of PP increased by more than twofold following photoaging, rising from 4.15 mg g^{-1} to 9.22 mg g^{-1}, whereas HDPE exhibited consistently low capacities (1.59–1.60 mg g^{-1}) regardless of aging. These results demonstrate that polymer type and environmental aging critically govern the role of microplastics as antibiotic carriers in aquatic environments.

Keywords: adsorption, high-density polyethylene, microplastics, polypropylene, photoaging, tetracycline.

INTRODUCTION

Due to their versatility, plastics are widely used in numerous aspects of modern life. The continuing increase of plastic production results in the introduction of nearly 8 million tons of plastic waste into the worldwide ocean every year (Jambeck et al., 2015). Major fractions of plastic waste will undergo fragmentation, generating microplastics that are now ubiquitous in aquatic systems. These tiny particles possess a large surface area and high hydrophobicity that enable the adsorption of various types of micropollutants such as antibiotics, heavy metals, pharmaceuticals, and pesticides from the environment (Atugoda et al., 2021; Medyńska-Juraszek and Jadhav, 2022; Tursi et al., 2022). Such interactions raise concerns regarding the role of microplastics as the vectors that facilitate contaminant transport in the environment (Tanaka et al., 2013; Liu et al., 2022; Avio et al., 2015).

Various microplastic polymers such as polyesters, polyvinyl chloride, polyethylene, polypropylene (PP), polyethylene terephtalate, polystyrene, and others are found in aquatic ecosystem (Castro-Castellon et al., 2022; Ashrafy et al., 2023). Due to their extensive use in a variety of consumer goods, polypropylene (PP) and high-density polyethylene (HDPE) are the most prevalent forms of microplastic polymers in the environment (Ruangpanupan et al., 2022; Rübsam et al., 2017; Newrick et al., 2025). Despite their similar hydrocarbon-based structures, PP and HDPE have differences in their chain architecture, crystallinity, and susceptibility to environmental degradation. These factors may influence their interactions with dissolved pollutants.

In parallel with microplastics, antibiotic pollution has become a growing global concern due to its persistence and ability to accelerate the development of antimicrobial resistance. Among antibiotics, tetracycline (TC) is commonly found in surface waters and wastewater effluents (Amangelsin et al., 2023; Danner et al., 2019; Javid et al., 2016) due to its widespread use in human and veterinary medicine, as well as in animal growth promotion and preventive healthcare (Semenova et al., 2025; Jeong et al., 2010).

Recent researches have underscored the ability of microplastics to serve as carriers for various pollutants in the environment, including TC antibiotics (Likpalimor et al., 2025). The ability of microplastics to adsorb such pollutants depends on various factors, including the weathering processes of microplastics, such as photoaging. Exposure to sunlight can induce photochemical oxidation, which further alters the surface morphology and functional groups of microplastic polymers, which eventually may alter their ability in uptaking other pollutants (Xu et al., 2024).

Despite the increasing interest in microplastics and antibiotic interactions, most studies have concentrated on pristine microplastics, while the effects of environmental aging of microplastics, such as photoaging, remain underexplored, particularly in the case of non-aromatic polymers like PP and HDPE. This study investigates the adsorption of TC onto pristine and photoaged PP and HDPE microplastics in aqueous systems. The study aims to investigate the physicochemical changes in the microplastics' surface induced by UV-photoaging and to evaluate the adsorption of TC on microplastics before and after photoaging, in order to elucidate the underlying adsorption mechanisms.

METHODS

Materials

PP and HDPE nurdles were purchased from the local shop in Yogyakarta, Indonesia. Analytical-grade chemicals (NaCl, HCl, tetracycline) were purchased from Sigma-Aldrich (Germany) and used as received. Distilled water was utilized throughout every step of this investigation.

Microplastics preparation and photoaging

PP and HDPE nurdles were ground and sieved using a 30-mesh sieve (pore diameter: 0.6 mm). The uniform microplastics were rinsed with distilled water to remove contaminants and then air-dried. Photoaging of microplastics was conducted in a closed UV reactor (20 W UV lamp, 365 nm). Microplastic particles were irradiated for 7 days (equivalent to photoaging for 5 days under strong tropical sunlight). Pristine and photoaged microplastics were characterized using Scanning Electron Microscope (Thermo Fisher Scientific, Phenom ProX) and Fourier Transform Infrared Spectrophotometer (FTIR, Shimadzu IRTracer-100).

Batch adsorption studies

Adsorption experiments were performed in closed reactors containing 50 mg microplastics and 25 mL TC solution. After 24 h of agitation at room temperature, suspensions were filtered, and residual TC concentration was determined spectrophotometrically at 357 nm (Shimadzu UV-1700 Pharmaspec). Equation 1 and Equation 2 were used to calculate the adsorption rate (%) and the quantity of TC adsorbed per unit mass of microplastics (mg g^{-1}), respectively.

$$Adsorption\ rate\ (\%) = \frac{(C_0 - C_e)}{C_0} \times 100\% \quad (1)$$

$$Adsorption\ capacity\ (Qe) = \frac{(C_0 - C_e)}{m} \times v \quad (2)$$

where: m denotes the mass of microplastics (g), v stands for the volume of the solution (L), and Co and Ce denote the concentration of TC before and after the adsorption process, respectively (mg L^{-1}).

The influence of solution pH (3–9), initial TC concentration (5–50 mg L^{-1}), and contact time (10–1440 min) was systematically investigated. All experiments were conducted in duplicate, and mean values were reported. Control experiments without microplastics were performed to check the potential TC losses unrelated to adsorption. Kinetic data were fitted to pseudo-first-order and pseudo-second-order models, while equilibrium data were analyzed using Langmuir and Freundlich isotherms.

RESULTS AND DISCUSSION

Surface characterization

FTIR spectra of pristine and photoaged PP and HDPE microplastics before and after TC adsorption are presented in Figure 1a and 1b, respectively. The FTIR spectra of the pristine microplastics showed the presence of characteristic functional groups for each plymer type. The HDPE microplastics exhibit peaks at 1470 cm^{-1} and 720 cm^{-1}, corresponding to C-H bending of CH$_2$ bonds and CH$_3$ rocking in HDPE (Benítez et al., 2013). The distinctive peaks of PP seen at 1460 cm^{-1} and 1380 cm^{-1} correspond to the symmetric bending of CH$_3$ bonds, whereas the peak at 972 cm^{-1} is attributed to CH$_3$ rocking bonds (Andreassen, 1999). Distinctive peaks at 2900 cm^{-1} corresponded to the C-H bond. No new absorption bands are observed in the FTIR spectra of PP and HDPE after photoaging. This indicates that the photooxidation of microplastics in this study is likely still in its early stages. FTIR analysis confirmed that the chemical structures of PP and HDPE remained unchanged after photoaging, as no new characteristic absorption bands were observed. However, the photoaging process caused the physical or morphological changes of the microplastics. This is shown by the yellowing of microplastics and the increasing of surface roughness, confirmed by the SEM analysis in Figure 2 (Xu et al., 2024; Burrows et al., 2024; Abaroa-Pérez et al., 2022). After TC adsorption, a decrease in FTIR transmittance was observed. Such decrease was more pronounced for photoaged microplastics. This suggests a greater adsorption of TC molecules onto the photoaged surface, likely due to the increased surface area and higher surface energy of the photoaged polymer. These findings are consistent with previous studies (Bhagat et al., 2022).

The surface morphologies of pristine and photoaged PP and HDPE were examined by SEM. The pristine PP (Figure 2a) and HDPE (Figure 2c) surfaces were relatively smooth and compact, indicating their intact polymeric structure before the UV light exposure. In contrast, the photoaged particles (Figure 2b and 2d) exhibited rougher and more irregular surfaces with visible cracks, pits, and flake-like structures. Although the photoaging process used in this study is not strong enough to induce the chemical changes on microplastics, the photoaging can still induce the morphological changes, such as the formation of new cracks and porosity on the surface of microplastics. Such increase on surface roughness and microdefects are known to enhance surface area and potential adsorption sites for other pollutants (Gao et al., 2024). Similar surface deterioration has been reported in other photoaged polymers (Wang et al., 2024). The photoaging effects are dependent on the polymer types, with PP being more susceptible to surface modification (Ducoli et al., 2025).

Effect of pH on tetracycline uptake

Solution pH is a crucial variable influencing the adsorption process. In the adsorption of TC onto microplastic particles, pH affects the surface potential of microplastics and the speciation of TC in solution (Nguyen et al., 2021). Consequently,

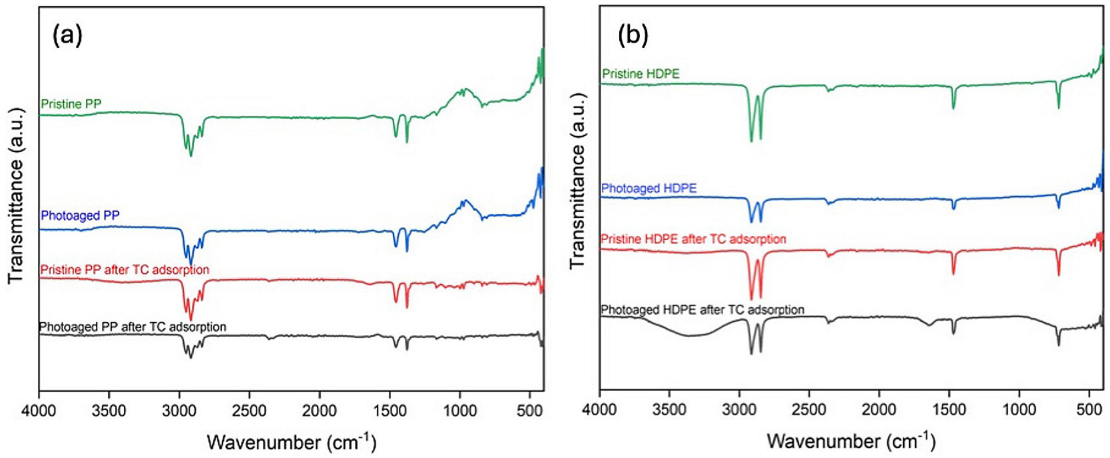

Figure 1. FTIR spectra of pristine and photoaged PP (a) and HDPE (b) microplastics before and after adsorption of TC antibiotics

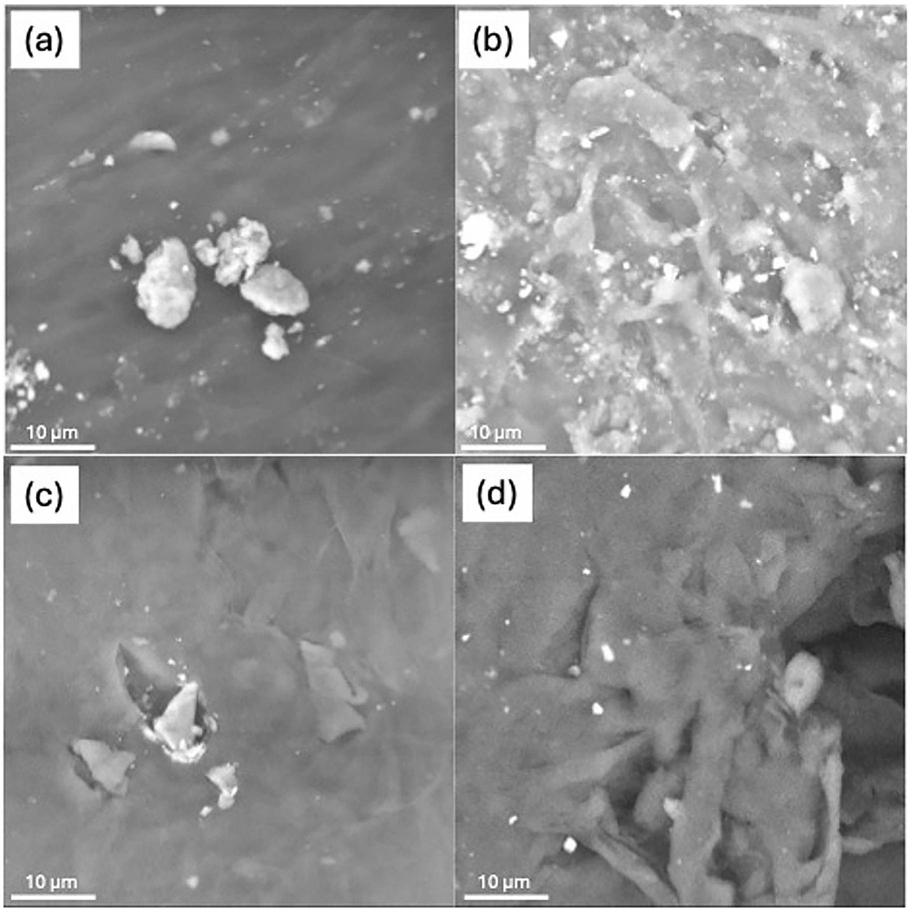

Figure 2. Surface morphologies of pristine PP (a); photoaged PP (b); pristine HDPE (c); and photoaged HDPE (d)

variations in pH dictate the electrostatic interactions between the microplastic surface and the TC antibiotic, thereby directly influencing adsorption efficiency. The effect of pH on TC adsorption onto PP and HDPE microplastics is given in Figure 3. The adsorption of TC on the pristine PP microplastics was optimum at pH 3, while the adsorption on photoaged PP was optimum at pH 5. This observed shift in the optimal pH suggests a modification in the surface charge characteristics of the microplastic surface following photodegradation, thereby changing the dominant adsorption mechanisms (Liu et al., 2024). In addition, for pristine and photoaged HDPE, the optimal pH for adsorption is 9. This variation in optimal pH across different microplastic types suggests a complex interaction of surface chemistry, degree of oxidation, and the corresponding ionic forms of TC in the aqueous environment (Anbarani et al., 2023). It is also notable that the adsorption rate of TC increases after the photoaging process of both types of microplastic polymers. This enhancement in adsorption rate is consistent with prior research indicating that environmental aging processes, such as photooxidation, induce surface modifications, which can provide more active sites for pollutant (Ho et al., 2023). In addition, such modifications often increase surface area and roughness, thereby physically entrap more TC molecules and improve overall adsorption efficacy (Moura et al., 2023). The ability of pristine and photoaged PP microplastics to adsorb TC is higher than that of the HDPE microplastics. The different adsorption behavior between PP and HDPE microplastics is due to differences in their properties, such as hydrophobicity, crystallinity, and intrinsic structure. The presence of a more labile tertiary carbon atom in the backbone of PP makes it more susceptible to photo-oxidative chain scission and subsequent oxidation during UV exposure than HDPE, which consists of a linear polyethylene backbone (Moura et al., 2023).

Other than pH, the chemistry of water such as the presence of dissolved organic matter, salts, and other coexisting contaminants may influence the adsorption of TC onto microplastics.

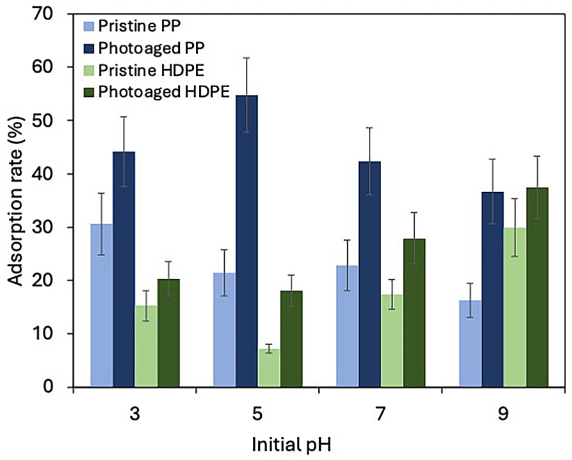

Figure 3. Effect of pH on TC adsorption onto pristine and photoaged PP and HDPE microplastics

Dissolved organic matter can compete for adsorption sites or form complexes with tetracycline, while increased ionic strength from salts may change the electrostatic interactions and affect adsorption behavior of TC on microplastics (Shang et al., 2023).

Adsorption isotherms

The adsorption results were analyzed using both the Langmuir and the Freundlich isotherm models (Figure 4a and 4b) to better clarify the adsorption mechanism of TC onto PP and HDPE microplastics. The adsorption process conformed closely to the Langmuir isotherm model, as shown in Table 1. The robust alignment with the Langmuir isotherm indicates a monolayer adsorption process, in which TC molecules occupy distinct, homogeneous active sites on the microplastic surface, rather than forming multiple layers. Langmuir's maximum adsorption capacity (Q_{max}) shows the highest amount of TC that might be adsorbed, given the finite amount of available adsorption sites on microplastic surfaces. The Q_{max} values for pristine and photoaged PP are 4.15 and 9.22 mg g^{-1}, respectively, while for pristine and photoaged HDPE, the values are 1.59 and 1.60 mg g^{-1}, respectively. The increase in Q_{max} values of photoaged PP suggests the significant enhancement in adsorption capacity resulting from the surface modification caused by photoaging (Hu et al., 2024). For PP microplastics, the pristine microplastics show a higher K_L value that indicates a stronger affinity between TC and the pristine PP surface at the initial stages of adsorption, despite the photoaged PP eventually exhibits a higher overall capacity. In contrast, the photoaged HDPE shows a higher KL value than that of the pristine HDPE. This indicates the distinct surface and interaction mechanisms that develop during photoaging in these two types of polymers. The surface modifications induced by photoaging in HDPE may create sites with a more immediate and effective binding capability for TC, even if the overall capacity increase is less pronounced than in PP. However, it is important to note that a higher KL value for pristine PP does not indicate a superior overall adsorption, but rather a more favorable initial binding affinity, as demonstrated by the higher Q_{max} of photoaged PP. The higher KL for photoaged HDPE suggests that photoaging enhances HDPE's adsorption affinity. The good fit of the Langmuir model indicates a homogeneous adsorption process in which each binding site has equal affinity for the adsorbate (Anbarani et al., 2023)

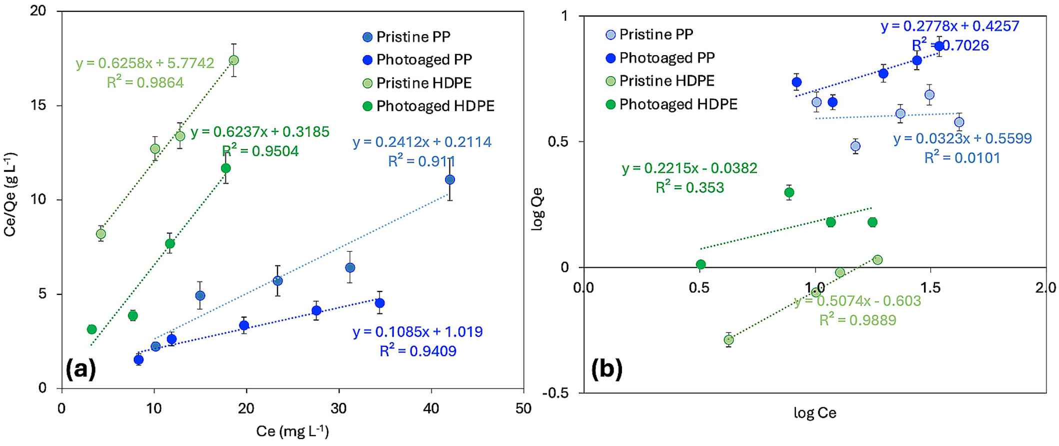

Figure 4. Ce versus Ce/Qe plot (a) and log Ce versus log Qe plot of tetracycline adsorption onto microplastics

Table 1. Langmuir isotherm parameters of tetracycline adsorption onto microplastics

Microplastics	Q_{max} (mg g^{-1})	KL	R^2
Pristine PP	4.15	1.141	0.911
Photoaged PP	9.22	0.098	0.941
Pristine HDPE	1.59	0.108	0.986
Photoaged HDPE	1.60	1.96	0.950

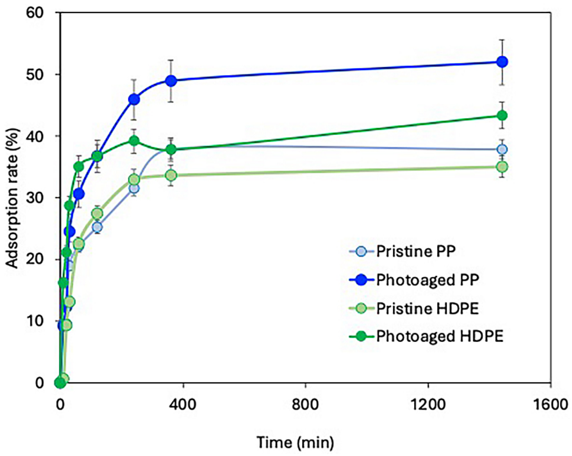

Figure 5. Effect of interaction time on adsorption rate (%) of TC on pristine and photoaged microplastics

After equilibrium, the adsorbed TC may desorb from the microplastics when the solution conditions such as pH, ionic strength, or the presence of competing pollutants. The desorption is also strongly influenced by the polymer type, as differences in surface chemistry, crystallinity, hydrophobicity, and functional groups among polymers can alter adsorption strength and desorption behavior (Fan et al., 2021).

Adsorption kinetics

Figure 5 shows the effect of interaction time on the adsorption rate of TC onto PP and HDPE microplastics. The equilibrium was reached within 24 h. The adsorption rates of photoaged microplastics are higher than those of the pristine ones. This enhancement is likely due to the surface modifications induced by photoaging, including increased surface roughness, formation of cracks and pits, and higher surface polarity. These changes cause the stronger interactions between TC molecules and microplastic surfaces. Using the values compiled in Table 2, the pseudo-first-order and pseudo-second-order kinetic models (Figure 6a and 6b) were used to study the adsorption kinetics. The adsorption of TC on both polymers, before and after photoaging, followed the pseudo-second-order kinetic model. The pristine microplastics show higher rate constants (k_2) values that suggests that the adsorption of TC on pristine microplastics is initially faster, but the aged microplastics exhibit higher adsorption capabilities. The enhanced adsorption on aged microplastics can also be attributed to the development of a more intricate pore structure, which increases the total accessible surface area for TC molecules (Ma et al., 2024). The results of this study are similar to those of previous studies (Anbarani et al., 2023). According to the pseudo-second-order kinetic model, chemisorption – which involves valence forces arising from the microplastics and TC antibiotics exchanging or sharing electrons – is the rate-determining step in the adsorption process.

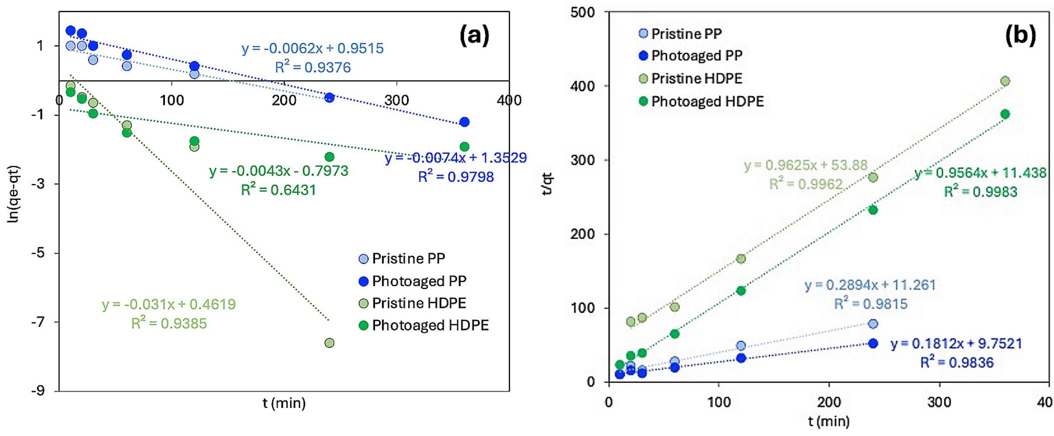

Figure 6. Plot of t versus ln(qe-qt) of pseudo-first-order (a) and plot of t versus t/qt for pseudo-second-order kinetic model (b)

Table 2. Kinetic features related to TC adsorption on pristine and photoaged microplastics

Adsorption kinetic parameters	Pristine PP	Photoaged PP	Pristine HDPE	Photoaged HDPE
Pseudo-first order				
Qe (mg g^{-1})	2.590	4.033	1.587	0.451
k_1 (min^{-1})	0.006	0.007	0.031	0.004
R^2	0.938	0.980	0.938	0.643
Pseudo-second order				
Qe (m g^{-1})	3.455	5.519	1.039	1.046
k_2 (g $mg^{-1}min^{-1}$)	0.007	0.003	0.017	0.08
R^2	0.982	0.984	0.996	0.998

Similar observations have been reported in previous studies showing that microplastic polymers can adsorb TC and that various aging processes often increase adsorption capacity by modifying surface morphology and the available sorption sites of microplastics (Gao et al., 2022; Cheng et al., 2024).

CONCLUSIONS

This study demonstrates that photoaging significantly influences the adsorption of tetracycline (TC) onto microplastics in aqueous solution. The photoaging process doubled the adsorption capacity of polypropylene (PP) microplastics, and for high-density polyethylene (HDPE), the adsorption capacity was relatively unchanged. Photoaging induced physical and morphological changes, such as increased surface roughness and the formation of cracks. The formation of new oxygen-containing functional groups is not readily apparent because the photoaging process is likely still at an early stage. The pseudo-second-order model provided the best description of the adsorption kinetics for all adsorption processes. Pseudo-second-order model indicated a surface-controlled adsorption process. However, the decrease in the pseudo-second-order rate constant (k_2) after photoaging suggests that TC adsorption on aged microplastics occurs more slowly, likely due to diffusion limitations and increased surface complexity. Langmuir isotherm further confirmed a monolayer adsorption mechanism. Overall, these findings highlight that photoaged microplastics can act as an adsorbent for TC antibiotics, emphasizing their potential role as long-term contaminant carriers in aqueous systems.

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