Journal of Hazardous Materials 431 (2022) 128643

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Investigation of the adsorption behavior of Pb(II) onto natural-aged microplastics as affected by salt ions

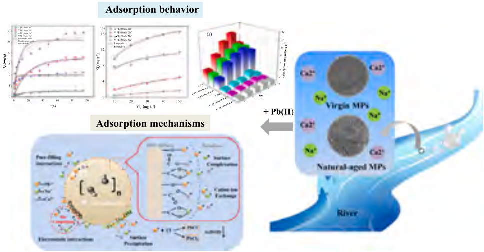
Si Liu [a,b], JinHui Huang [a,b,*], Wei Zhang [a,b], LiXiu Shi [c], KaiXin Yi [a,b], ChenYu Zhang [a,b], HaoLiang Pang [a,b], JiaoNi Li [a,b], SuZhou Li [a,b]

[a] *College of Environmental Science and Engineering, Hunan University, Changsha 410082, PR China*
[b] *Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, PR China*
[c] *College of Hydraulic and Environmental Engineering, Changsha University of Science and Technology, Changsha 410114, PR China*

HIGHLIGHTS

- The adsorption capacity of Pb(II) on natural-aged microplastics was higher than virgin ones.
- Divalent Ca^{2+} markedly inhibited the sorption of Pb(II) compared to monovalent Na^{+}.
- The pH and fulvic acid affected the Pb (II) sorption in different electrolyte solutions.
- Competition and complexation interactions played great roles in the adsorption process.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Natural-aged microplastics
Lead(II)
Sorption
Coexisting electrolyte ions
Interaction mechanisms

ABSTRACT

In this study, the adsorption behavior of Pb(II) on natural-aged and virgin microplastics in different electrolyte solutions was investigated. The results demonstrated that natural-aged microplastics exhibited higher adsorption capacity for Pb(II) compared to virgin ones, and the addition of $CaCl_2$ strongly inhibited the adsorption amount of Pb(II). The adsorption kinetics of Pb(II) adsorption were better fitted by the pseudo-second order model and Elovich equation, and were slowed down greatly at higher ionic strength. The rate-limiting steps of adsorption process were dominated by intra-particle diffusion. The adsorption isotherm of Pb(II) onto microplastics affected by salt ions can be well described by Freundlich model, the greater adsorption efficiency of natural-aged microplastics proved that adsorption process was multilayer and heterogeneous. In addition, pH significantly influenced the adsorption of Pb(II) due to the changes electrostatic interactions. The effect of fulvic acid in the electrolyte solutions was also revealed and attributed to the complexation with Na^{+} and Ca^{2+}. Furthermore, the higher pH and ionic strength in different environmental water dramatically decreased adsorption capacity onto microplastics. Finally, it's confirmed that the adsorption mechanisms affected by salt ions mainly involve electrostatic interaction, surface complexation, and ionic exchange. These findings indicate that salt ions exert an important influence on the adsorption of heavy metals for MPs, which should be further concerned.

---

[*] Corresponding author at: College of Environmental Science and Engineering, Hunan University, Changsha 410082, PR China.
E-mail address: huangjinhui_59@163.com (J. Huang).

https://doi.org/10.1016/j.jhazmat.2022.128643
Received 24 January 2022; Received in revised form 26 February 2022; Accepted 5 March 2022
Available online 9 March 2022

1. Introduction

The application of plastic products has penetrated into all aspects of people's lives over the past two decades, and the consequent plastic pollution has become a problem that needs to be solved urgently (Wagner et al., 2018). Among the concerns about plastic pollution, microplastics (MPs), an emerging pollutant with a size range from 1 μm to 5 mm, are used as an additive in personal care products such as facial cleansers and toothpaste, or are degraded from large pieces of plastic by natural weathering processes (Thompson et al., 2004; Ouyang et al., 2020). Recently, it has been widely reported that microplastics have been found in the ocean, lake, river, reservoir, and even the polar regions (Di and Wang, 2018; Eerkes-Medrano et al., 2015). The widespread distribution of microplastics could pose a potential threat to aquatic biota and human health.

The unique properties of MPs surface, such as large specific surface areas, high hydrophobicity, and diffusivity, influence the adsorption of chemicals (Zhang et al., 2020a). It seems to be exploit MPs as an low-cost adsorbents for contaminant removal from aqueous environments. However, adsorption onto MPs can promote long-term transportation of various pollutants in the aquatic environment, extending their spatial distribution and providing more possibilities for food chain transfer and bioaccumulation (Liu et al., 2022; Mittal et al., 2021). Until now, many reports have demonstrated that MPs are highly effective adsorbent for heavy metals. For instance, Lin et al. (2021) compared the adsorption performance of lead (Pb) onto three kinds of MPs (polyethylene PE, polyvinylchloride PVC, polystyrene PS) and found that the adsorption of Pb(II) by MPs was followed the order of PVC < PE < PS. Gao et al. (2019) measured the accumulation of heavy metals of virgin polypropylene (PP) and PVC in the seawater within 6 months and detected different heavy metals such as chromium (Cr), cadmium (Cd), manganese (Mn), copper (Cu), zinc (Zn), arsenic (As) and Pb were adsorbed by MPs. However, MPs will inevitably undergo weathering or aging process under the influence of various factors once entering the environment, which will change the particle size, surface morphology, and microstructure of MPs (Lambert and Wagner, 2016; Holmes et al., 2012). The availability of adsorption sites of MPs increased as a result of these changes, leading to higher adsorption capacity for heavy metals (Khalid et al., 2021). Brennecke et al. (2016) found that aged PVC have the stronger affinity for Cu and Zn. The possible explanation is that the higher surface area, polarity, and aging degree of PVC, providing more adsorption sites for metal ions. Mao et al. (2020) investigated the adsorption of heavy metals on PS by exposing to UV radiations; the result revealed that the adsorption capacity of PS to different metal ions was enhanced with increasing degree of aging, which may be attributed to the increased surface roughness and the oxygen-containing functional groups. Similarly, higher concentrations of trace metals were accumulated with the aging/ biofouling of MPs (Holmes et al., 2014). Therefore, the significant changes of MPs surface properties under environmental stresses affected the interaction with heavy metals. Noteworthy, previous studies have focused mainly on exploring the short-term aging properties of MPs, and the aging process only involves a single external force, which cannot truly simulate the aging of MPs in the actual environment affected by various environmental conditions. Therefore, comparing the adsorption behavior and interaction mechanism of aging MPs to heavy metals, it is more representative to choose natural-aged MPs. Fu et al. (2021) revealed that natural-aged MPs exhibited a stronger adsorption capacity for Pb(II), and the main mechanism might be dominated by electrostatic force between the oxygen-containing functional groups of MPs and Pb(II).

Heavy metal pollution in freshwater environments is eliciting increasing attention, among which Pb(II) is one of the most common and toxic metals (Huang et al., 2018). Pb(II) can induce serious toxic effects on aquatic ecosystem because of its persistence and bioaccumulation (Huang et al., 2016a,b; Huang et al., 2019). Several field investigations have proved that the interaction between MPs and Pb(II) is universal in the freshwater system (Purwiyanto et al., 2020; Guan et al., 2020; Ta and Babel, 2020). For example, Purwiyanto et al. (2020) found that a high concentration of Pb(II) on MPs in the Musi River, and deduced that possible mechanism is physical sorption with weak bonds. Notably, the adsorption of Pb(II) onto MPs is generally affected by environmental factors such as pH, salinity, and dissolved organic matter (DOM). Most studies have regarded salinity as an influencing factor to explore the interaction mechanism between MPs and Pb(II). Tang et al. (2020) evaluated the effect of salinity on Pb(II) adsorption onto aged nylon MPs, and they found higher NaCl concentration inhibited the adsorption of Pb(II), which may be related to the increased competition for binding sites of nylon MPs and reduced charged ions activity. Lin et al. (2021) demonstrated that the adsorption of Pb(II) on MPs was observed to decrease at high ionic strength, the possible reason is that electrostatic screening by salt ions between negative MPs surface and positive Pb(II). A similar adsorption inhibition of Pb(II) on MPs was also reported by Zou et al. (2020). Different concentrations and types of salt play an important role in freshwater systems. The adsorption of heavy metals onto MPs affected by salts may have significant implications for their fate and transport in the natural environment. However, the influence of salts on the interaction between Pb(II) and MPs in the real freshwater environment has not been clearly clarified. It has been reported that the mechanism of antibiotics onto MPs in real freshwater systems may be different from that of pure water systems (Zhang et al., 2021; Li et al., 2018). Thus, the adsorption of Pb(II) onto MPs in the different electrolyte solutions should be carefully considered.

In this study, we aim to explore the influence of salt ions on the adsorption behavior of Pb(II) on natural-aged PE MPs (A-PE) and virgin PE MPs (V-PE). The influence of ionic species and strength were analyzed by applying various kinetics and isotherm models to the experimental data. Besides, we revealed the effects of solution pH and fulvic acid (FA) affected by salt ions on the adsorption of Pb(II), and further explored the adsorption capacity of Pb(II) in different environmental water. Finally, the possible mechanism between MPs and Pb(II) was discussed. The results provide thorough information for understanding the influence of solution chemistry on the adsorption of heavy metals onto MPs.

2. Materials and methods

2.1. Materials and chemicals

Natural-aged polyethylene plastics were collected from Changsha section of the Xiangjiang river ($28^{\circ}8^{\prime}$N, $112^{\circ}56^{\prime}$E). In the laboratory, the plastic surface was carefully rinsed with ultrapure water to eliminate the attached sand. After air-drying, cut them into micro-sized plastics less than 5 mm in diameter. Virgin polyethylene microplastics were prepared using purchased PE plastics. Both types of micro-sized plastics were passed through 0.84–0.6 mm sieves for the batch experiments. Lead nitrate ($Pb(NO_3)_2$), sodium chloride (NaCl), potassium hydroxide (KOH), calcium chloride ($CaCl_2$), nitric acid (HNO_3) all are 99.0% pure and purchased from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China). Lead standard solution of 1000 mg L^{-1} and fulvic acid (FA) were of analytic grade and obtained from Shanghai Aladdin Bio-Chem Technology Co., Ltd.

2.2. Characterization of microplastics

The surface morphology of MPs was observed by scanning electron microscopy (SEM, TESCAN MIRA4, Czech). Energy dispersive spectroscopy (EDS, Xplore, UK) was used to analyze the elements distribution on the MPs. Fourier transform spectroscopy (FTIR, Thermo Scientific Nicolet iS20, USA) with a wavelength of 400–4000 cm^{-1} was used to determine the functional groups of MPs before and after Pb(II) adsorption. X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, USA) was applied to characterize the adsorbents. The specific surface area of MPs were measured by Brunauer-Emmett-Teller (BET, Micromeritics TriStar II 3020, USA). Additionally, the zeta potentials (Malvern Zetasizer Nano ZS90, UK) was performed to determine the surface charge.

2.3. *Adsorption experiments*

Prior to the adsorption experiments, we have evaluated the metal content such as Cd, Pb, Mn, and Zn adsorbed on the MPs according to the previous studies (Li et al., 2019), the detailed results are shown in Table S1 in Supporting Information (SI). In adsorption kinetics experiments, 50 mg of MPs were separately added to 150 mL glass conical flasks with 100 mL of 20 mg L^{-1} Pb(II) solution. Two commonly found salts in the aquatic environment, sodium chloride (NaCl) and calcium chloride ($CaCl_2$), are used as background solutions. Generally, the concentration of monovalent Na^+ is higher than that of divalent Ca^{2+} (He et al., 2018). To simulate the actual freshwater environment, two concentration gradients of the salts in NaCl (10 mM and 50 mM) and $CaCl_2$ (1 mM and 5 mM) were selected (Dong et al., 2019). The initial pH of the solutions was adjusted using 0.1 M HNO_3 or 0.1 M KOH solution. All the samples were shaken in a dark and constant temperature shaker (25 ± 1 °C, 150 rpm). Samples were collected at twelve time intervals within 96 h and passed through a 0.45 μm filter to analyze aqueous concentrations of Pb(II) measured by flame atomic absorption spectrophotometer (FLAAS, PerkinElmer (Suzhou), PinAAcle900F, China). The control group was carried out using the same conditions but without MPs. Each test was performed in triplicate. All glass vials in the experiments were soaked in 10% HNO_3 for at least 24 h and rinsed with Milli-Q water before use. The adsorption isotherm experiments were carried out with a series of concentrations of Pb(II) (10, 20, 30, 40, 50 mg L^{-1}). The rest conditions were the same as aforementioned adsorption kinetics experiment. All the adsorption models and parameters were shown in Table S2.

The effect of solution pH and FA on the adsorption of Pb(II) by MPs in the 10 mM NaCl or 1 mM $CaCl_2$ background electrolyte solutions. The initial solution pH was adjusted to a range of 3–7 using 0.1 M HNO_3 or 0.1 M KOH solution. The concentrations of FA (0, 3, 5, 10, 15 mg L^{-1}) referred to actual freshwater systems were used (Zhou et al., 2020a). Furthermore, the uptake of Pb(II) by MPs in complex environmental water conditions was investigated. Five different types of water including Milli-Q water, Xiangjiang river water, Taozi lake water, stormwater, and tap-water were selected, and the aqueous medium was filtered through 0.22 μm polycarbonate membranes (Millipore). Table S3 provides different parameters of the water matrices. The same experimental conditions as in the adsorption kinetics were performed.

3. Results and discussions

3.1. *Characterization of MPs*

According to the external surface color (Fig. S1), it could be obviously observed that the darker color was visualized in natural-aged MPs compared to the virgin MPs, which may be related to the oxidation of MPs surface and the attachment of organic material (Chen et al., 2021). The SEM images further revealed that the difference in surface morphology of MPs as shown in Fig. 1a, b. The surface of V-PE was relatively smooth and flat, whereas A-PE presented a rough and cracked surface. The SEM-EDS elemental mapping (Fig. S2) also verified that the

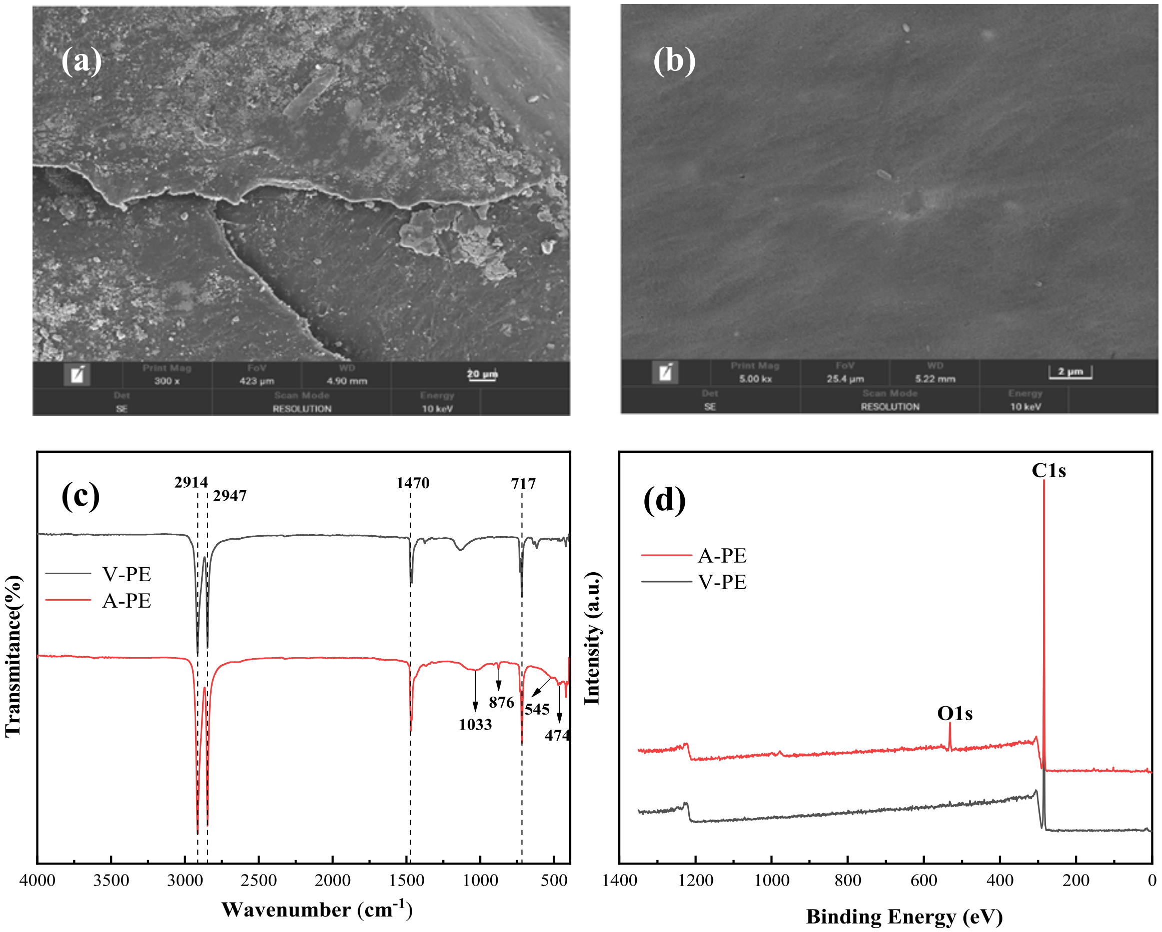


Fig. 1. SEM images of natural-aged (a) and virgin (b) microplastics; FTIR spectra of A-PE and V-PE (c); XPS spectra of survey scan of A-PE and V-PE (d).

element of Si (4.67%), Al (4.51%) associated with clay material was observed on the surface of A-PE, which may enhance the adsorption capacity for metal ions. The specific surface area of A-PE displayed 0.521 $m^2 g^{-1}$, which was larger than that of V-PE (0.144 $m^2 g^{-1}$). In addition, the differences of metal contents between natural-aged MPs and virgin MPs measured by ICP-MS were shown in Table S1. The concentration of metals such as Mg, Al, Mn, Fe was higher in the A-PE than in the V-PE, indicating that clay minerals could precipitate onto the A-PE in the natural aquatic environment. The difference of the concentration of Na, K and Ca could attributed to the leaching of additives and the sorption from surrounding environment of A-PE. However, there was no significant difference of Pb content on MPs, which can be ignored in subsequent adsorption comparisons.

The surface functional groups of A-PE and V-PE were characterized by FTIR and shown in Fig. 1c. The PE MPs displayed several similar FTIR absorption bands. The obvious peaks at 2915, 2848, 1463, 715 cm^{-1} corresponded to the $–CH_2$- stretching vibration, and the band intensity was higher in natural-aged PE than in virgin ones. Compared to the V-PE, a broadened peak located at 1033 cm^{-1} was observed on the A-PE, which may be attributed to the C-O-C stretching of ester (Ye et al., 2020). The new peak appearing at 535 cm^{-1} represents C═O bending vibration of aliphatic ketone. Moreover, the peaks at 876 and 474 cm^{-1} could be assigned to the Si-O band and the wobble of CO_2^- (Fu et al., 2021; Aghilinasrollahabadi et al., 2021), confirming the presence of organic matter accumulated on the surface of A-PE. According to the XPS spectrum (Fig. 1d), the virgin PE MPs only contain element C appeared at 284.8 eV, while a new peak of O1s was detected on the aged PE MPs. Additionally, the high resolution scan of C1s of A-PE (Fig. S3) contains a new peak at 286.4 eV, which could be assigned to C-O or C-O-C band. These results indicate that the oxygen-containing functional groups were generated on the natural-aged MPs, which may affect the adsorption capacity of heavy metals onto MPs.

3.2. Adsorption kinetics

The adsorption kinetics data of Pb(II) onto aged and virgin MPs in different background solutions are presented in Fig. 2. The values of kinetic models parameters are listed in Table S4. According to the R^2 values, the pseudo-second-order model fits better than the pseudo-first-order model in the different electrolyte solutions, which indicated that the adsorption process of Pb(II) onto MPs may be dominated by chemical adsorption (Liang et al., 2021; Yu et al., 2022). The adsorption of Pb (II) on A-PE and V-PE occurs quickly initially within the first 12 h in the Milli-Q water, and the adsorption equilibrium was sufficiently reached in 96 h. Considering the dynamic adsorption process of metal ions onto MPs and the higher adsorption capacity of Pb(II), thus a longer adsorption time was required. However, due to the competitive behavior of different salt ions for the active sites of MPs surface, the adsorption rate was decreased by adding Na^+ and Ca^{2+}. When the initial concentration of Na^+ and Ca^{2+} increased to 50 mM and 5 mM, the adsorption capacity was greatly decreased for A-PE and V-PE, which was consistent with the results of the previous study (Tang et al., 2020). Additionally, the inhibitory effects of Ca^{2+} on Pb(II) adsorption were much greater than that of Na^+. The possible reason is that Ca^{2+} and Pb(II) have the same valence state, thus strongly hindering the binding of Pb(II) to the limited active sites of MPs (Ho and Leung, 2021). Besides, Elovich equation was used to describe the adsorption process that occurred on the heterogeneous surface, and the higher correlation coefficient (R^2 =0.89–0.99, see Table S3) indicated that the adsorption of Pb(II) on MPs involves valence forces through electron exchange or sharing (Wu et al., 2018).

Intra-particle diffusion model was conducted to reveal the mass transfer steps of Pb(II) sorption. As shown in Fig. 2b, d, f, the adsorption process can be divided into three linear segments, which suggested different procedures of Pb(II) sorption onto MPs (Wang and Wang, 2018). The initial linear segments represented Pb(II) diffuses rapidly in the solid surface through film diffusion, and the latter stage was related to the intra-particle diffusion process. The fitting curves do not cross the origin, indicating that the rate-limiting step was simultaneously controlled by film and intra-article diffusion (Hu et al., 2020). In addition, the relatively high R^2 values (Table S5) showed the intra-particle diffusion was dominant in the adsorption of Pb(II) onto V-PE in comparison to A-PE in the different water systems. The k_t values decreased continuously throughout the adsorption process until reached sorption quasi-equilibrium in the third stage, and diffusion rate was much lower with the addition of Na^+ and Ca^{2+}, which may be related to the decrease of effective active sites on the adsorbent or the decrease of Pb(II) concentration (Guan et al., 2020). The S values referring to the boundary layer effect of A-PE were larger, verifying the surface sorption played a key role in the adsorption of Pb(II) (Qi et al., 2021).

3.3. Adsorption isotherms

The adsorption isothermal curves of Pb(II) onto MPs at different initial concentrations (10–50 mg L^{-1}), salt types and concentrations were shown in Fig. 3. The results indicated that the adsorption amount of Pb(II) increased with the equilibrium concentrations. The adsorption rate was fast between 10 and 20 mg L^{-1}, which may be attributed to more adsorption sites were occupied by Pb(II) at low concentrations. When the concentration of Pb(II) continue to increase, the adsorption tendency slowed down because of the adsorption mainly occurred on the external surface of the MPs and reached a certain adsorption amount affecting subsequent process (Shen et al., 2021).

The adsorption data were fitted by the Langmuir and Freundlich models and the related parameters were listed in Table S6. The adsorption isothermal curves can be better fitted by Freundlich model based on the R^2 values (0.852–0.998), which revealed that Pb(II) sorption onto MPs was a heterogeneous and multilayer adsorption process (Chen et al., 2017; Ahmad et al., 2022). Furthermore, the higher K_F values suggested that the high affinity of A-PE for Pb(II), the porous structure and the formation of oxygen-containing groups may be the explanation for the MP adsorption capacity. The comparison of Pb(II) adsorption over recently reported MPs was listed in Table S7. As can be seen, in pure water system, Q_m was 18742.9, 10283.3 μg g^{-1} for A-PE and V-PE, respectively. The higher adsorption capacity of A-PE for Pb(II) may be due to its large specific surface area and attached organic matter. A-PE in this study exhibited the highest Q_m values compared to the other materials, indicating that Pb(II) adsorption process on natural-aged microplastics was more favorable. Noteworthy, we found that the Langmuir model was slightly better fit for the data of A-PE in 10 mM NaCl solution, but the Q_m value (17.730 mg g^{-1}) obtained from Langmuir was much higher than the experimental data, which may not be suitable for estimate the actual adsorption. Besides, compared to the pure water system, the types of salt obviously affected the adsorption of Pb(II) onto MPs. Analysis of variance demonstrated that the adsorption capacity of MPs for Pb(II) exists significant difference under adding Na^+ and Ca^{2+} conditions ($p < 0.001$). With the increase of electrolytes concentrations, the adsorption amount of Pb(II) onto MPs was decreased, which may be related to the charged sites of the adsorbent were shielded by the salt ions (Tang et al., 2021). Holmes et al. (2014) also observed a similar phenomenon in the estuarine conditions. However, divalent Ca^{2+} influenced the adsorption process of different Pb(II) concentrations more prominently than monovalent Na^+. This is because of the stronger charge neutralization and higher electrical double layer compression of Ca^{2+} (Campinas and Rosa, 2006; Shams et al., 2020). Moreover, the metal ion species could be changed by the increase of Cl^- concentration. The presence of salt ions will increase the complexity of metal sorption onto MPs, thus the possible adsorption mechanisms in the different water systems would be further analyzed in the next Section.

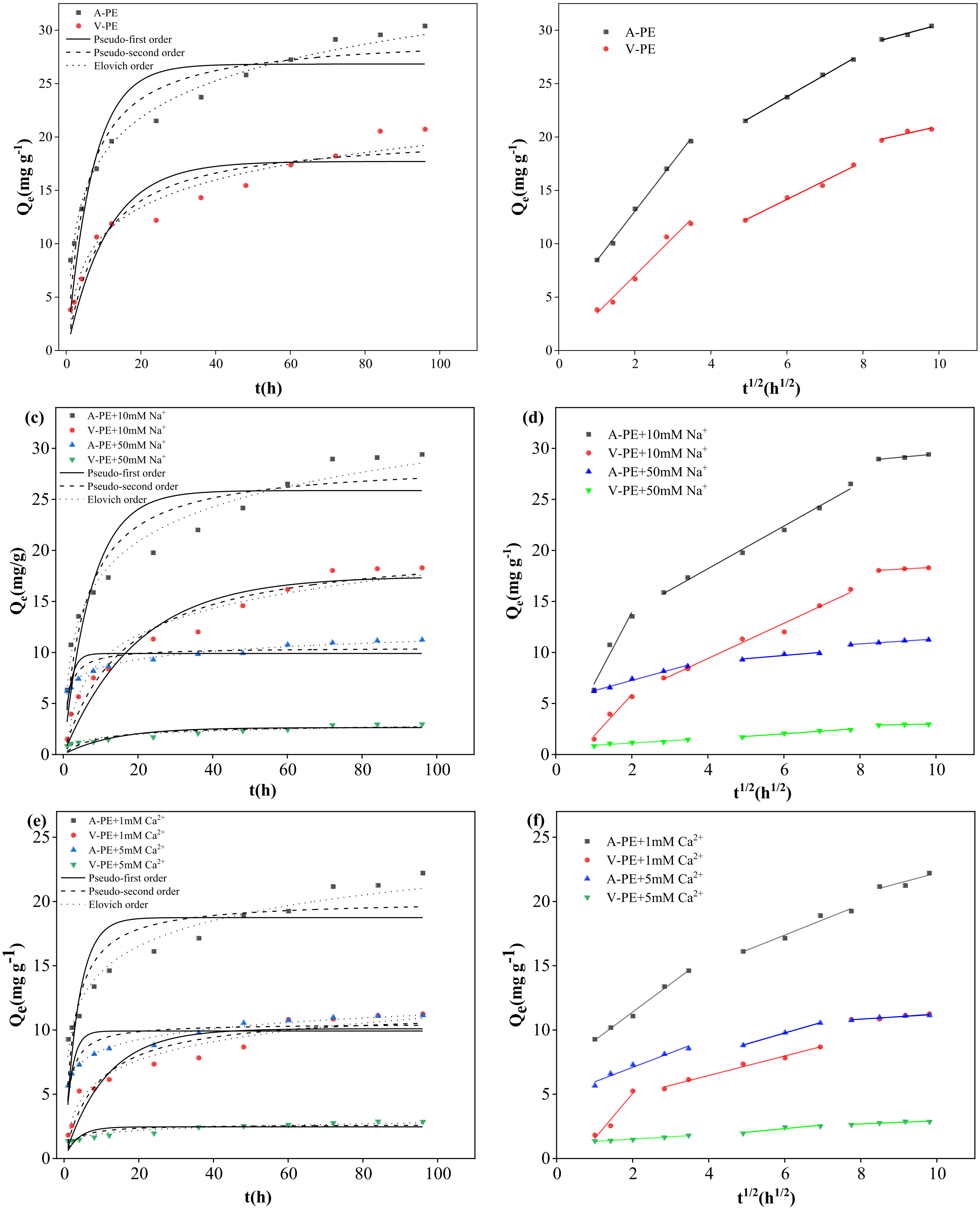


Fig. 2. Adsorption kinetics of Pb(II) on A-PE and V-PE in the presence of pure water (a,b), NaCl (c,d), $CaCl_2$ (e,f). Experimental conditions: $[Pb(II)]_0 = 20$ mg L^{-1}; T = 25 ℃ ; pH = 6.0 ± 0.1; [adsorbents] = 0.5 g/L.

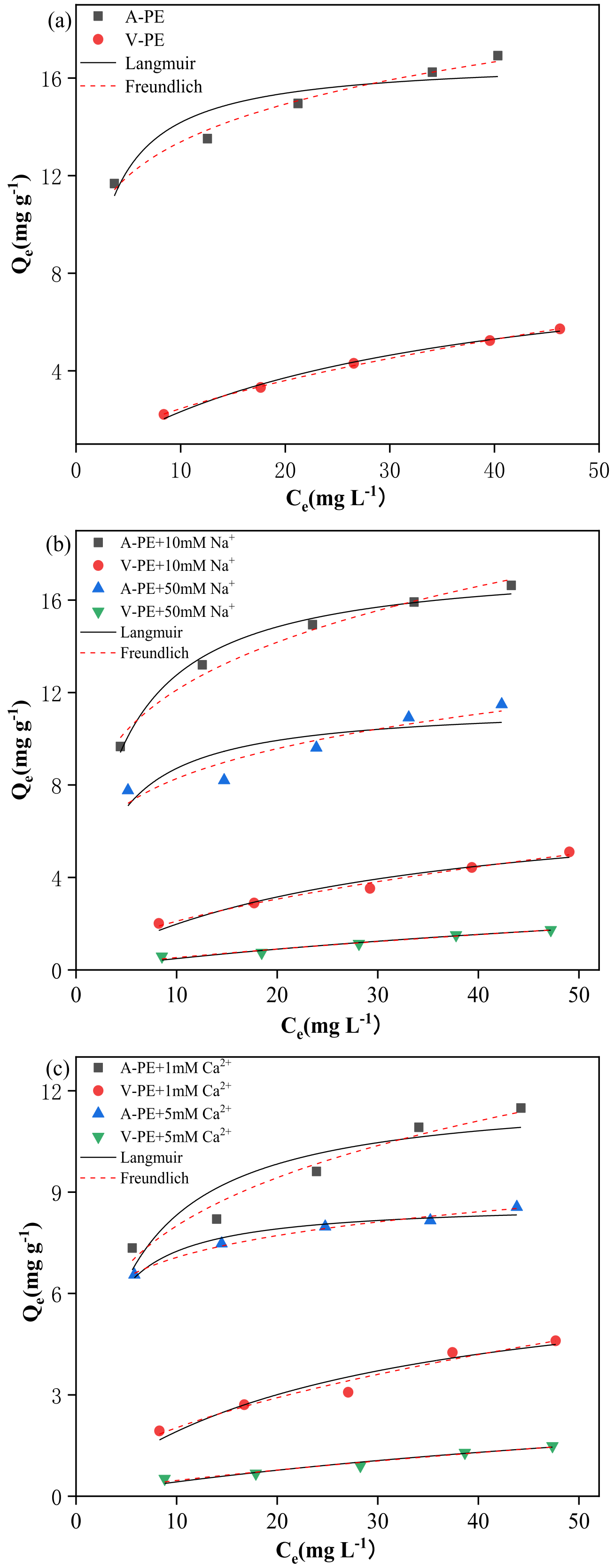


Fig. 3. Adsorption isotherms of Pb(II) on A-PE and V-PE in the pure water (a), NaCl (b), $CaCl_2$ (c) systems. Experimental conditions: T = 25 ℃ ; pH = 6.0 ± 0.1; t = 96 h; [adsorbents] = 0.5 g/L.

3.4. Effect of pH on adsorption

The solution pH may affect the surface charge of the adsorbents and species of metal ions (Huang et al., 2017), thus influencing the adsorption of Pb(II) onto MPs in different aqueous systems. The pH dependence of Pb(II) adsorption on MPs ranging from 3.0 to 7.0 at different background solutions was shown in Fig. 4a. In the pure water system, the amount of Pb(II) adsorbed gradually increases with the increase of pH values from 3 to 6. The main reason is the H^+ competes with Pb(II) for the same adsorption sites at a lower pH value, while as the pH increased, the changes of surface charge and the deprotonation of surface functional groups promote the adsorption ability of metal ions (Tang et al., 2021; Shi et al., 2019). The highest adsorption capacity of Pb(II) were observed at pH 6.0. A similar phenomenon was also reported by Wang et al. (2022). In addition, the surface of A-PE and V-PE showed electro-negativity of pH ranging from 3 to 7 and decreased gradually with the increase of pH value (Fig. S4), which was conductive to the electrostatic attraction of the MPs to Pb(II). However, when pH further increased to 7, Pb(II) adsorption decreased due to Pb(II) species exist in the form of Pb^{2+}, $Pb(OH)^+$, $Pb(OH)_2$ (Aghilinasrollahabadi et al., 2021).

Similar phenomenon occurred in the different electrolyte solutions. Additionally, the results suggested Pb(II) uptake decreased at different pH for the given salt concentration (i.e. 10 mM NaCl and 1 mM $CaCl_2$), which could be ascribed to the synergistic competitive effect of H^+ and Na^+ or Ca^{2+}. Because of the organic matter and minerals attached to the MPs surface in the natural environment, the A-PE accumulated more negative charges than V-PE (Kim and Sansalone, 2008), resulting in higher adsorption amount of Pb(II) onto MPs. Furthermore, Zhang et al. (2020b) found that three MPs possessed negative charge in different aquatic systems (i.e. Milli-Q water, Na^+ and Ca^{2+} solutions). The electro-negativity of MPs in Ca^{2+} solution was weakened due to the screening effect of MPs surface charges, which could further account for the lower adsorption of Pb(II) in the $CaCl_2$ solution. Overall, these findings indicated that electrostatic interaction between metal ions and MPs strongly affects the adsorption behavior.

3.5. Effect of FA on adsorption

Generally, organic matter such as fulvic acid (FA) exist in the form of dissolved molecules in the soil, sediments, and water. The adsorption of heavy metals can be affected by the presence of FA via complex interactions. Huang et al. (2020) have reported that the FA-metal complexes formed affected the adsorption of metal ions onto MPs. In this study, the effect of FA on the adsorption capacity of MPs in different aquatic systems was investigated. As shown in Fig. 4b, the adsorption of Pb(II) was inhibited with the increase of FA concentration in the pure water systems, particularly for the A-PE. On the one hand, a number of negatively charged functional groups (i.e. –OH and -COOH) were owned on the FA surface (Tan et al., 2021), which may lead to the combination with Pb(II) through complexation interaction. Tang et al. (2021) also reported that the formation of bidentate complexes (FA2Cu) significantly decreased the amount of Cu onto nylon MPs. On the other hand, the FA molecules were difficult to be adsorbed on the negatively charged MPs surface due to electrostatic repulsion, resulting in the increase of competitive adsorption between FA and MPs with Pb(II) (Sun et al., 2019). In addition, the effect of FA on Pb(II) adsorption was also affected by the presence of salt ions. For a given electrolyte solution, the amount of Pb(II) decreased with the increase of FA concentration, but the reduction rate was lower than that of pure water system. The most likely explanation is that the aggregation of FA occurred when the salt ions (especially Ca^{2+}) were present. Correspondingly, the competitive effect was reduced because of the formation of large aggregates would be less adsorbable (Kilduff et al., 1996). In contrast, we observed a slight increase of Pb(II) sorption onto MPs in the presence of 15 mg L^{-1} FA, which may be attributed to the relative stability of Ca^{2+}-FA complexes (Iskrenova-Tchoukova et al., 2010). Moreover, the conformation of FA

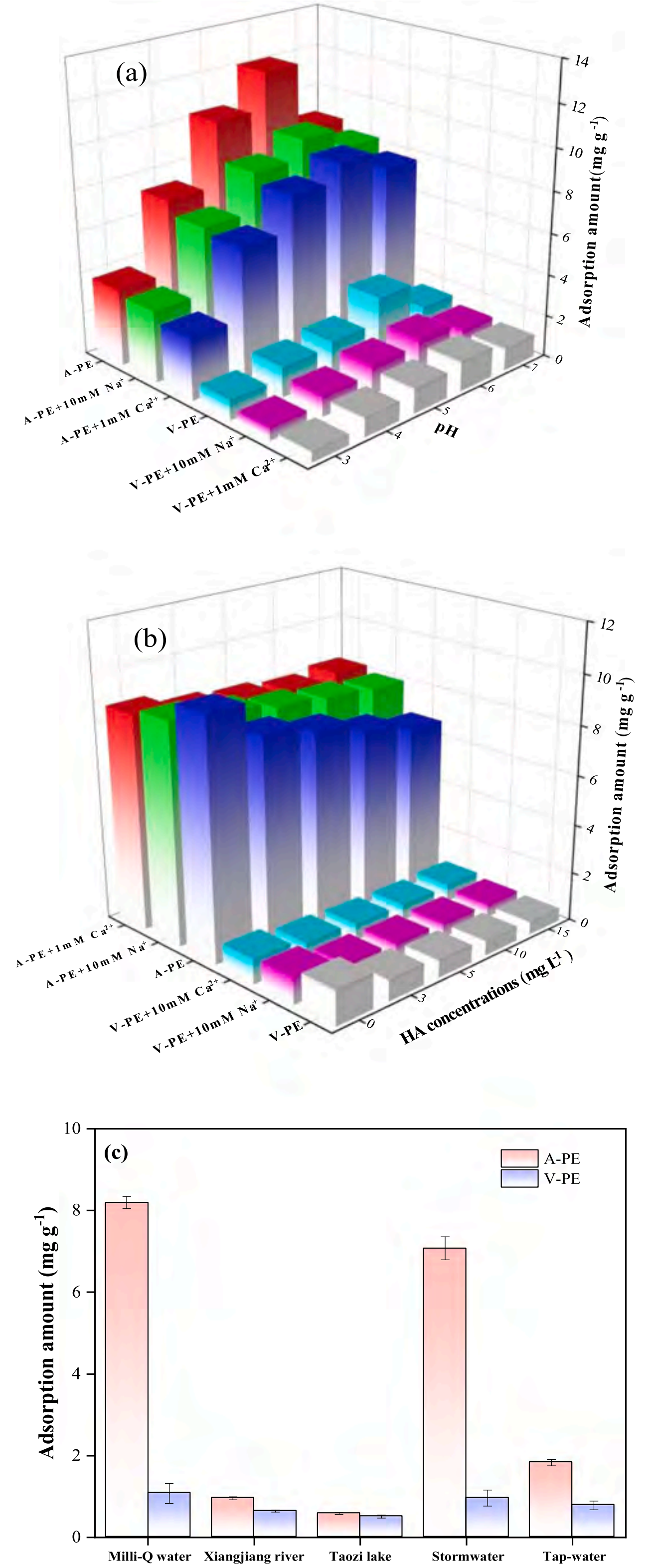


Fig. 4. Effect of the pH (a), FA (b), and environmental water conditions (c) on the adsorption of Pb(II) on A-PE and V-PE. Experimental conditions for a, b: [Pb (II)]$_0$ = 20 mg L^{-1}; T = 25 ℃ ; pH = 6.0 ± 0.1; t = 96 h; [adsorbents] = 0.5 g/L, [salinity] = 10 mM NaCl and 1 mM CaCl$_2$.

was changed by cross-linking with Ca^{2+} in the higher concentration of natural organic matter (Pivokonsky et al., 2015), thus affecting the adsorption process of metal ions. Similar results were also observed for the removal of arsenic by ferric chloride (Kong et al., 2017).

3.6. Effect of environmental water conditions on adsorption

Different types of environmental water presented different parameters and ion concentrations (Table S3). The speciation distributions of Pb (II) in aqueous medium at different pH values were shown in Fig. S5. The major species present at pH 7–8 were Pb^{2+}, Pb(OH)$^{+}$. As Fig. 4c demonstrated, the adsorption amount of Pb(II) in Milli-Q water (pH=5.46) and stormwater was relatively higher, which was consistent with the results of the pH experiment. However, a significantly lower Pb (II) sorption by both A-PE and V-PE in the Xiangjiang river and TaoZi lake water, which may be related to higher pH values, organic matter content and ionic strength. In contrast, the inhibition effect of Pb(II) sorption onto MPs was slightly reduced in tap-water. The main reason for the above results is that the higher concentrations of Na^{+}, Mg^{2+}, Ca^{2+}, K^{+} will compete for electrostatic binding to the MPs surface, thus decreasing the activity of Pb ions. Ahamed et al. (2020) also confirmed that the presence of cations such as K^{+}, Na^{+}, Ca^{2+} in synthetic drinking water at pH 7.8 inhibited the adsorption of Pb(II). Remarkably, the adsorption of Pb(II) in the stormwater mainly depends on the pH variation due to the relatively weak ionic strength. Therefore, MPs could serve as vectors of heavy metals in the stormwater and transport from terrestrial to aquatic environments, which may further increase the potential risks of MPs to ecological environment.

4. Adsorption mechanism

As reported, the adsorption of heavy metals onto MPs was mainly dominated by the intermolecular interactions between plastic polymers and adsorbates (Li et al., 2015; Zhou et al., 2020b). Due to specific characteristics, microplastic adsorbents may exhibit selective adsorption behavior. Table S8 summarized the different metal ions adsorbed by MPs and possible mechanisms. However, the potential interaction will be affected by the aquatic chemistry such as pH and salinity. Tang et al. (2021) demonstrated that divalent metal ions (Cu, Zn, Ni) adsorption onto MPs was salinity-dependent significantly, and the electrostatic interaction would be inhibited with the increasing of NaCl concentrations. Besides, Zhao et al. (2011) considered that the adsorption of Cd(II) on bentonite was related to the ion exchange of Cd(II) with the coexisting electrolyte cations. To further explore the interaction mechanisms between Pb(II) and MPs, the SEM, SEM-EDS, and FTIR were used to analyze the surface chemistry of MPs after the adsorption of Pb(II) in the different electrolyte solutions.

As shown in Fig. S6, compared to the smooth surface of V-PE, some tiny components were incorporated into the crakes of A-PE after the adsorption of Pb(II), which implicated that the pore filling effect was involved in the adsorption process (Zhang et al., 2018). Fig. 5 showed the main elemental composition on the surface of A-PE and V-PE after adsorption. We observed that Pb(II) was located in similar regions with the element of Fe and Mn on the A-PE surface, inferring that the attached clay minerals promoted the accumulation of Pb(II). In previous studies, Fe and Mn oxides were considered as a principal factor affecting the adsorption of heavy metals onto MPs due to its ability to form metal-organic complexes or ion exchange reactions (Ashton et al., 2010). In addition, after the adsorption of Pb(II) in the electrolyte solutions, a higher proportion of element Ca was detected on the A-PE surface compared with element Na. The result revealed that the coexisting Ca^{2+} might be strongly accumulated onto MPs, resulting in the decrease of available adsorption sites for Pb(II).

Fig. 6 shows the FTIR spectra of A-PE and V-PE before and after adsorption. There are no obvious changes of V-PE before and after adsorption in the different aqueous systems, suggesting that physical adsorption may play a dominant role in the adsorption process (Guan et al., 2020). However, after the adsorption of Pb(II), the C-H group on the surface of A-PE was shifted from 717 cm^{-1} to 730 cm^{-1}, which verified that C-H group participated in the adsorption and involved the electrostatic force (Wang et al., 2022; Yu et al., 2021). The appearance

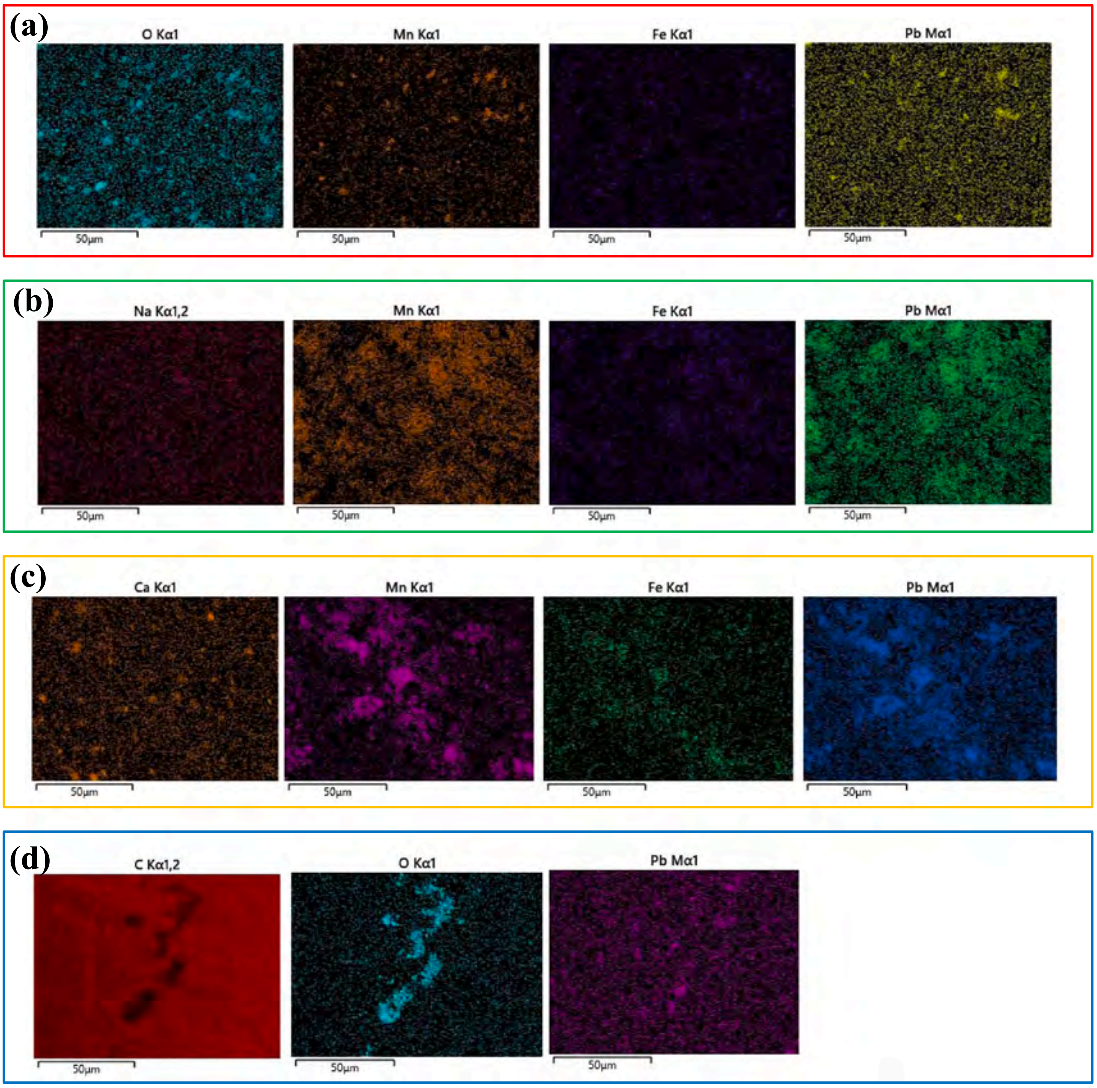


Fig. 5. SEM-EDS elemental maps of A-PE in the pure water (a), NaCl (b), $CaCl_2$ (c) systems and V-PE in the pure water system (d) after Pb(II) adsorption.

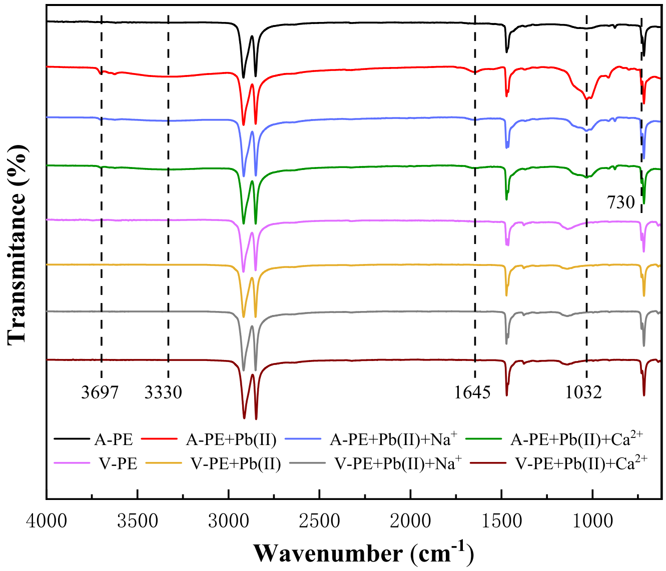


Fig. 6. FTIR spectra of A-PE and V-PE before and after adsorption of Pb(II) in the different aqueous systems.

of a broadened peak at 3330–3697 cm^{-1} was attributed to the free –OH group on the A-PE surface. The presence of peak at 1645 cm^{-1} was responsible for the stretching of –COOH. The above findings indicated that the significant role of the oxygen-containing functional groups involved in the Pb(II) adsorption process (Zhou et al., 2020b). In particular, the intensity of the peaks at 1645 and 1032 cm^{-1} was slightly weak in the electrolytes solution; the possible reason is that Na^+ and Ca^{2+} are complex with the carboxylate anion and C-O-C groups (Clement et al., 1998). Therefore, the result of SEM-EDS and FTIR spectra proved that the adsorption of Pb(II) onto A-PE was a complex physical and chemical process.

In this study, the presence of NaCl and $CaCl_2$ affects both adsorption kinetics and isotherms, also being associated with solution parameters such as pH and DOM, which can change the adsorption process. Firstly, the addition of electrolyte ions not only led to a reduction in zeta potential, but also screen the charged sites of the adsorbent and compress the electrical double layers of MPs (Hu et al., 2013). Thus, the reduced Pb(II) adsorption amount was attributed to the suppression of electrostatic interactions. Moreover, the compression degree of electrical double layers depends on the concentration and valence of electrolyte cations (Zhang et al., 2019). Obviously, Ca^{2+} and higher ionic strength inhibited the adsorption of Pb(II) onto MPs. Secondly, Pb(II) was easy to form outer-sphere complexes with the adsorption sites on the MPs surface. Generally, compared to the inner-sphere complexes, the electrolyte ions exist on the same plane as the outer-sphere complexes, leading to the outer-sphere complexes are more susceptible to the variation of ionic strength (Sheng et al., 2010). Therefore, Na^+ and Ca^{2+} compete effectively with Pb(II) for outer-sphere adsorption sites, and Ca^{2+} could compete with Pb(II) for inner-sphere sites better than Na^+. Thirdly, the adsorption process was sensitive to the changes of the background electrolyte because of the solid phase concentration was closely related to the activity of ions in the solution (Zhang et al., 2019). With the addition of NaCl and $CaCl_2$, the amounts of Pb^{2+} decreased while more $PbCl^+$ and $PbCl_2$ ion pairs are generated, thus decreasing the activity coefficient of Pb(II) ions (Mattigod et al., 1979). Meanwhile, $CaCl_2$ forms ion pairs of greater stability with metal ions than NaCl which forms weaker ion pairs, which limits the transfer of Pb(II) ions to the MPs surface.

According to the above analysis, we conclude that the adsorption mechanisms of Pb(II) onto A-PE in the different solutions mainly involve physical adsorption (including pore-filling and surface precipitation) and chemical adsorption (including electrostatic interaction, surface complexation, and ionic exchange). Also, the surface O-containing groups of natural-aged MPs play an important role in the adsorption process. The detailed interaction mechanisms of Pb(II) adsorption onto MPs were illustrated in Fig. 7.

5. Conclusion

In this study, the adsorption behavior and interaction mechanisms of Pb(II) onto natural-aged MPs and virgin MPs affected by salt ions were studied. This study provided new insights on how salt ions play an important role in the fate and transport of heavy metals adsorbed by MPs. However, the current research on the adsorption of heavy metals onto MPs is mostly focused on the pure water systems. Future research should consider the effect of ionic species and strength between laboratory simulation and actual aqueous environments, thus increasing the environmental relevance. The major conclusions obtained from the results are as follows:

(1) It was evident that A-PE exhibited higher adsorption capacity than V-PE in different aqueous systems, which may be attributed to the attachment of organic matter and the formation of oxygen-containing functional groups.

(2) The pseudo-second-order model and Elovich equation fitted better for adsorption kinetics data, evidencing Pb(II) adsorption onto MPs were dominated by chemisorption. Diffusion mechanisms suggested that the adsorption of Pb(II) on V-PE was mainly controlled by intra-particle diffusion while the binding diffusion for A-PE.

(3) Compared to the V-PE, A-PE showed the largest Pb(II) adsorption amount based on the Freundlich isotherm model, implying that heterogeneous and multilayer adsorption occurred.

(4) Owing to the electrostatic interaction, the highest adsorption efficiency of Pb(II) was reached at pH 6. However, the increasing of FA concentrations exhibited different inhibition effects in the presence of Na^+ and Ca^{2+}, indicating that aggregation and surface complexation cannot be ignored. More importantly, the decrease of Pb(II) adsorption in different environmental water conditions further demonstrated that pH and ionic strength play a crucial role on the adsorption capacity of MPs.

(5) The results of experiment data, FTIR and SEM-EDS demonstrated that the adsorption mechanisms of Pb(II) onto natural-aged MPs in electrolyte solutions involves complex interaction, including pore filling, surface precipitate, electrostatic interaction, surface complexation, and ion exchange.

CRediT authorship contribution statement

Si Liu: Methodology, Validation, Investigation, Data curation, Formal analysis, Writing – original draft. **Jinhui Huang:** Conceptualization, Methodology, Writing – review & editing, Supervision, Funding acquisition. **Wei Zhang:** Conceptualization, Validation, Formal analysis, Investigation. **Lixiu Shi:** Resources, Data curation, Formal analysis. **Kaixin Yi:** Investigation, Visualization, Data curation. **Chenyu Zhang**: Software, Writing – review & editing. **Haoliang Pang**: Investigation, Methodology. **Jiaoni Li**: Formal analysis. **Suzhou Li**: Writing – review & editing.

Environmental Implication

Current researches on the adsorption of heavy metals by microplastics in real freshwater system and the mechanistic interactions are limited. Also, the adsorption capacity of natural-aged microplastics should be carefully considered. In this work, the adsorption of Pb(II) onto natural-aged and virgin microplastics were investigated in different aqueous systems, and reveal the effects of environmental factors on the uptake of Pb(II). Finally, the possible mechanisms were deeply explored

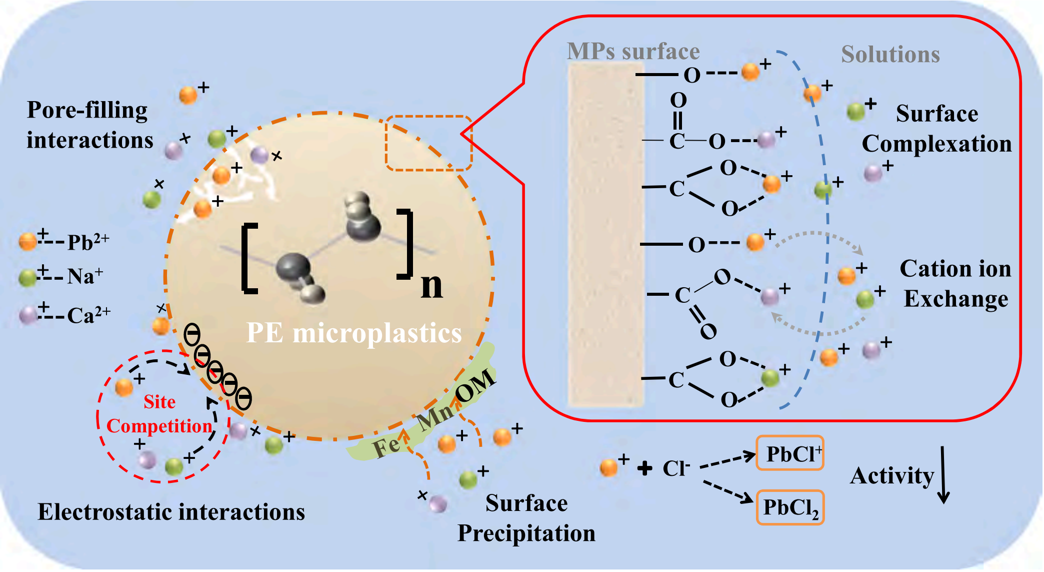


Fig. 7. Schematic illustration of adsorption mechanisms of Pb(II) on MPs in the presence of NaCl and $CaCl_2$.

to provide fundamental understandings of the MPs- Pb(II) complicated pollution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This study was supported by the Innovative Province Construction Special Fund of Hunan Province (2020SK2016) and the Water Conservancy Science and Technology Project of Hunan Province (XSKJ2021000-47).

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.128643.

References

Aghilinasrollahabadi, K., Salehi, M., Fujiwara, T., 2021. Investigate the influence of microplastics weathering on their heavy metals uptake in stormwater. J. Hazard. Mater. 408, 124439.

Ahamed, T., Brown, S.P., Salehi, M., 2020. Investigate the role of biofilm and water chemistry on lead deposition onto and release from polyethylene: an implication for potable water pipes. J. Hazard. Mater. 400, 123253.

Ahmad, R., Ansari, K., Ejaz, M.O., 2022. Enhanced sequestration of heavy metals from aqueous solution on polyacrylamide grafted with cell@Fe_3O_4 nanocomposite. Emergent Mater.

Ashton, K., Holmes, L., Turner, A., 2010. Association of metals with plastic production pellets in the marine environment. Mar. Pollut. Bull. 60, 2050–2055.

Brennecke, D., Duarte, B., Paiva, F., Caçador, I., Canning-Clode, J., 2016. Microplastics as vector for heavy metal contamination from the marine environment. Estuar., Coast. Shelf Sci. 178, 189–195.

Campinas, M., Rosa, M.J., 2006. The ionic strength effect on microcystin and natural organic matter surrogate adsorption onto PAC. J. Colloid Interface Sci. 299, 520–529.

Chen, C.C., Zhu, X., Xu, H., Chen, F., Ma, J., Pan, K., 2021. Copper adsorption to microplastics and natural particles in seawater: a comparison of kinetics. Isotherms, Bioavailab., Environ. Sci. Technol. 55, 13923–13931.

Chen, Z., Wang, J., Pu, Z., Zhao, Y., Jia, D., Chen, H., Wen, T., Hu, B., Alsaedi, A., Hayat, T., Wang, X., 2017. Synthesis of magnetic Fe_3O_4/CFA composites for the efficient removal of U(VI) from wastewater. Chem. Eng. J. 320, 448–457.

Clement, O., Rapko, B.M., Hay, B.P., 1998. Structural aspects of metal–amide complexes. Coord. Chem. Rev. 170, 203–243.

Di, M., Wang, J., 2018. Microplastics in surface waters and sediments of the Three Gorges Reservoir, China. Sci. Total Environ., 616- 617, 1620–1627.

Dong, Z., Zhang, W., Qiu, Y., Yang, Z., Wang, J., Zhang, Y., 2019. Cotransport of nanoplastics (NPs) with fullerene (C60) in saturated sand: effect of NPs/C60 ratio and seawater salinity. Water Res. 148, 469–478.

Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75, 63–82.

Fu, Q., Tan, X., Ye, S., Ma, L., Gu, Y., Zhang, P., Chen, Q., Yang, Y., Tang, Y., 2021. Mechanism analysis of heavy metal lead captured by natural-aged microplastics. Chemosphere 270, 128624.

Gao, F., Li, J., Sun, C., Zhang, L., Jiang, F., Cao, W., Zheng, L., 2019. Study on the capability and characteristics of heavy metals enriched on microplastics in marine environment. Mar. Pollut. Bull. 144, 61–67.

Guan, J., Qi, K., Wang, J., Wang, W., Wang, Z., Lu, N., Qu, J., 2020. Microplastics as an emerging anthropogenic vector of trace metals in freshwater: significance of biofilms and comparison with natural substrates. Water Res. 184, 116205.

He, L., Wu, D., Rong, H., Li, M., Tong, M., Kim, H., 2018. Influence of nano- and microplastic particles on the transport and deposition behaviors of bacteria in Quartz Sand. Environ. Sci. Technol. 52, 11555–11563.

Ho, W.K., Leung, K.S., 2021. The crucial role of heavy metals on the interaction of engineered nanoparticles with polystyrene microplastics. Water Res. 201, 117317.

Holmes, L.A., Turner, A., Thompson, R.C., 2012. Adsorption of trace metals to plastic resin pellets in the marine environment. Environ. Pollut. 160, 42–48.

Holmes, L.A., Turner, A., Thompson, R.C., 2014. Interactions between trace metals and plastic production pellets under estuarine conditions. Mar. Chem. 167, 25–32.

Hu, B., Li, Y., Jiang, L., Chen, X., Wang, L., An, S., Zhang, F., 2020. Influence of microplastics occurrence on the adsorption of 17β-estradiol in soil. J. Hazard. Mater. 400, 123325.

Hu, Y., Guo, T., Ye, X., Li, Q., Guo, M., Liu, H., Wu, Z., 2013. Dye adsorption by resins: effect of ionic strength on hydrophobic and electrostatic interactions. Chem. Eng. J. 228, 392–397.

Huang, J., Yuan, F., Zeng, G., Li, X., Gu, Y., Shi, L., Liu, W., Shi, Y., 2017. Influence of pH on heavy metal speciation and removal from wastewater using micellar-enhanced ultrafiltration. Chemosphere 173, 199–206.

Huang, J., Guo, S., Zeng, G.-M., Li, F., Gu, Y., Shi, Y., Shi, L., Liu, W., Peng, S., 2018. A new exploration of health risk assessment quantification from sources of soil heavy metals under different land use. Environ. Pollut. 243, 49–58.

Huang, J., Li, F., Zeng, G., Liu, W., Huang, X., Xiao, Z., Wu, H., Gu, Y., Li, X., He, X., He, Y., 2016a. Integrating hierarchical bioavailability and population distribution into potential eco-risk assessment of heavy metals in road dust: a case study in Xiandao District, Changsha city, China. Sci. Total Environ. 541, 969–976.

Huang, J.H., Liu, W.C., Zeng, G.M., Li, F., Huang, X.L., Gu, Y.L., Shi, L.X., Shi, Y.H., Wan, J., 2016b. An exploration of spatial human health risk assessment of soil toxic metals under different land uses using sequential indicator simulation. Ecotoxicol. Environ. Saf. 129, 199–209.

Huang, J., Peng, S., Mao, X., Li, F., Guo, S., Shi, L., Shi, Y., Yu, H., Zeng, G.-M., 2019. Source apportionment and spatial and quantitative ecological risk assessment of heavy metals in soils from a typical Chinese agricultural county. Process Saf. Environ. Prot. 126, 339–347.

Huang, X., Zemlyanov, D.Y., Diaz-Amaya, S., Salehi, M., Stanciu, L., Whelton, A.J., 2020. Competitive heavy metal adsorption onto new and aged polyethylene under various drinking water conditions. J. Hazard. Mater. 385, 121585.

Iskrenova-Tchoukova, E., Kalinichev, A.G., Kirkpatrick, R.J., 2010. Metal cation complexation with natural organic matter in aqueous solutions: molecular dynamics simulations and potentials of mean force. Langmuir 26, 15909–15919.

Khalid, N., Aqeel, M., Noman, A., Khan, S.M., Akhter, N., 2021. Interactions and effects of microplastics with heavy metals in aquatic and terrestrial environments. Environ. Pollut. 290, 118104.

Kilduff, J.E., Karanfil, T., Weber, W.J., 1996. Competitive interactions among components of humic acids in granular activated carbon adsorption systems: effects of solution chemistry. Environ. Sci. Technol. 30, 1344–1351.

Kim, J.-Y., Sansalone, J.J., 2008. Zeta potential of clay-size particles in urban rainfall–runoff during hydrologic transport. J. Hydrol. 356, 163–173.

Kong, Y., Kang, J., Shen, J., Chen, Z., Fan, L., 2017. Influence of humic acid on the removal of arsenate and arsenic by ferric chloride: effects of pH, As/Fe ratio, initial As concentration, and co-existing solutes. Environ. Sci. Pollut. Res. 24, 2381–2393.

Lambert, S., Wagner, M., 2016. Characterisation of nanoplastics during the degradation of polystyrene. Chemosphere 145, 265–268.

Li, F., Huang, J., Zeng, G., Huang, X., Liu, W., Wu, H., Yuan, Y., He, X., Lai, M., 2015. Spatial distribution and health risk assessment of toxic metals associated with receptor population density in street dust: a case study of Xiandao District, Changsha, Middle China. Environ. Sci. Pollut. Res. 22, 6732–6742.

Li, J., Zhang, K., Zhang, H., 2018. Adsorption of antibiotics on microplastics. Environ. Pollut. 237, 460–467.

Li, X., Mei, Q., Chen, L., Zhang, H., Dong, B., Dai, X., He, C., Zhou, J., 2019. Enhancement in adsorption potential of microplastics in sewage sludge for metal pollutants after the wastewater treatment process. Water Res. 157, 228–237.

Liang, L., Xi, F., Tan, W., Meng, X., Hu, B., Wang, X., 2021. Review of organic and inorganic pollutants removal by biochar and biochar-based composites. Biochar 3, 255–281.

Lin, Z., Hu, Y., Yuan, Y., Hu, B., Wang, B., 2021. Comparative analysis of kinetics and mechanisms for Pb(II) sorption onto three kinds of microplastics. Ecotoxicol. Environ. Saf. 208, 111451.

Liu, S., Huang, J., Zhang, W., Shi, L., Yi, K., Yu, H., Zhang, C., Li, S., Li, J., 2022. Microplastics as a vehicle of heavy metals in aquatic environments: a review of adsorption factors, mechanisms, and biological effects. J. Environ. Manag. 302, 113995.

Mao, R., Lang, M., Yu, X., Wu, R., Yang, X., Guo, X., 2020. Aging mechanism of microplastics with UV irradiation and its effects on the adsorption of heavy metals. J. Hazard. Mater. 393, 122515.

Mattigod, S.V., Gibali, A.S., Page, A.L., 1979. Effect of ionic strength and ion pair formation on the adsorption of nickel by kaolinite. Clays Clay Miner. 27, 411–416.

Mittal, J., Ahmad, R., Mariyam, A., Gupta, V.K., Mittal, A., 2021. Expeditious and enhanced sequestration of heavy metal ions from aqueous environment by papaya peel carbon: a green and low cost adsorbent. Desalin. Water Treat. 210, 365–376.

Ouyang, W., Zhang, Y., Wang, L., Barcelo, D., Wang, Y., Lin, C., 2020. Seasonal relevance of agricultural diffuse pollutant with microplastic in the bay. J. Hazard. Mater. 396, 122602.

Pivokonsky, M., Naceradska, J., Brabenec, T., Novotna, K., Baresova, M., Janda, V., 2015. The impact of interactions between algal organic matter and humic substances on coagulation. Water Res. 84, 278–285.

Purwiyanto, A.I.S., Suteja Trisno, Y., Ningrum, P.S., Putri, W.A.E., Rozirwan, F. Agustriani, Fauziyah, M.R.Cordova, Koropitan, A.F., 2020. Concentration and adsorption of Pb and Cu in microplastics: case study in aquatic environment. Mar. Pollut. Bull. 158, 111380.

Qi, K., Lu, N., Zhang, S., Wang, W., Wang, Z., Guan, J., 2021. Uptake of Pb(II) onto microplastic-associated biofilms in freshwater: adsorption and combined toxicity in comparison to natural solid substrates. J. Hazard. Mater. 411, 125115.

Shams, M., Alam, I., Chowdhury, I., 2020. Aggregation and stability of nanoscale plastics in aquatic environment. Water Res. 171, 115401.

Shen, M., Song, B., Zeng, G., Zhang, Y., Teng, F., Zhou, C., 2021. Surfactant changes lead adsorption behaviors and mechanisms on microplastics. Chem. Eng. J. 405.

Sheng, G., Li, J., Shao, D., Hu, J., Chen, C., Chen, Y., Wang, X., 2010. Adsorption of copper(II) on multiwalled carbon nanotubes in the absence and presence of humic or fulvic acids. J. Hazard. Mater. 178, 333–340.

Shi, L., Huang, J., Zhu, L., Shi, Y., Yi, K., Li, X., 2019. Role of concentration polarization in cross flow micellar enhanced ultrafiltration of cadmium with low surfactant concentration. Chemosphere 237, 124859.

Sun, B., Hu, Y., Cheng, H., Tao, S., 2019. Releases of brominated flame retardants (BFRs) from microplastics in aqueous medium: kinetics and molecular-size dependence of diffusion. Water Res. 151, 215–225.

Ta, A.T., Babel, S., 2020. Microplastics pollution with heavy metals in the aquaculture zone of the Chao Phraya River Estuary, Thailand. Mar. Pollut. Bull. 161, 111747.

Tan, M., Liu, L., Zhang, M., Liu, Y., Li, C., 2021. Effects of solution chemistry and humic acid on the transport of polystyrene microplastics in manganese oxides coated sand. J. Hazard. Mater. 413, 125410.

Tang, S., Lin, L., Wang, X., Feng, A., Yu, A., 2020. Pb(II) uptake onto nylon microplastics: interaction mechanism and adsorption performance. J. Hazard. Mater. 386, 121960.

Tang, S., Lin, L., Wang, X., Yu, A., Sun, X., 2021. Interfacial interactions between collected nylon microplastics and three divalent metal ions (Cu(II), Ni(II), Zn(II)) in aqueous solutions. J. Hazard. Mater. 403, 123548.

Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304, 838.

Wagner, S., Huffer, T., Klockner, P., Wehrhahn, M., Hofmann, T., Reemtsma, T., 2018. Tire wear particles in the aquatic environment - A review on generation, analysis, occurrence, fate and effects. Water Res. 139, 83–100.

Wang, Q., Zhang, Y., Zhang, Y., Liu, Z., Wang, J., Chen, H., 2022. Effects of biofilm on metal adsorption behavior and microbial community of microplastics. J. Hazard. Mater. 424, 127340.

Wang, W., Wang, J., 2018. Comparative evaluation of sorption kinetics and isotherms of pyrene onto microplastics. Chemosphere 193, 567–573.

Wu, W., Sheng, H., Gu, C., Song, Y., Willbold, S., Qiao, Y., Liu, G., Zhao, W., Wang, Y., Jiang, X., Wang, F., 2018. Extraneous dissolved organic matter enhanced adsorption of dibutyl phthalate in soils: Insights from kinetics and isotherms. Sci. Total Environ., 631- 632, 1495–1503.

Ye, S., Cheng, M., Zeng, G., Tan, X., Wu, H., Liang, J., Shen, M., Song, B., Liu, J., Yang, H., Zhang, Y., 2020. Insights into catalytic removal and separation of attached metals from natural-aged microplastics by magnetic biochar activating oxidation process. Water Res. 179, 115876.

Yu, S., Pang, H., Huang, S., Tang, H., Wang, S., Qiu, M., Chen, Z., Yang, H., Song, G., Fu, D., Hu, B., Wang, X., 2021. Recent advances in metal-organic framework membranes for water treatment: a review. Sci. Total Environ. 800, 149662.

Yu, S., Tang, H., Zhang, D., Wang, S., Qiu, M., Song, G., Fu, D., Hu, B., Wang, X., 2022. MXenes as emerging nanomaterials in water purification and environmental remediation. Sci. Total Environ. 811, 152280.

Zhang, H., Pap, S., Taggart, M.A., Boyd, K.G., James, N.A., Gibb, S.W., 2020a. A review of the potential utilisation of plastic waste as adsorbent for removal of hazardous priority contaminants from aqueous environments. Environ. Pollut. 258, 113698.

Zhang, H., Wang, J., Zhou, B., Zhou, Y., Dai, Z., Zhou, Q., Chriestie, P., Luo, Y., 2018. Enhanced adsorption of oxytetracycline to weathered microplastic polystyrene: kinetics, isotherms and influencing factors. Environ. Pollut. 243, 1550–1557.

Zhang, Y., Zhu, C., Liu, F., Yuan, Y., Wu, H., Li, A., 2019. Effects of ionic strength on removal of toxic pollutants from aqueous media with multifarious adsorbents: a review. Sci. Total Environ. 646, 265–279.

Zhang, P., Huang, P., Sun, H., Ma, J., Li, B., 2020b. The structure of agricultural microplastics (PT, PU and UF) and their sorption capacities for PAHs and PHE derivates under various salinity and oxidation treatments. Environ. Pollut. 257, 113525.

Zhang, Y., Ni, F., He, J., Shen, F., Deng, S., Tian, D., Zhang, Y., Liu, Y., Chen, C., Zou, J., 2021. Mechanistic insight into different adsorption of norfloxacin on microplastics in simulated natural water and real surface water. Environ. Pollut. 284, 117537.

Zhao, D., Chen, S., Yang, S., Yang, X., Yang, S., 2011. Investigation of the sorption behavior of Cd(II) on GMZ bentonite as affected by solution chemistry. Chem. Eng. J. 166, 1010–1016.

Zhou, N., Liu, Y., Cao, S., Guo, R., Ma, Y., Chen, J., 2020a. Biodegradation of bisphenol compounds in the surface water of Taihu Lake and the effect of humic acids. Sci. Total Environ. 723, 138164.

Zhou, Y., Yang, Y., Liu, G., He, G., Liu, W., 2020b. Adsorption mechanism of cadmium on microplastics and their desorption behavior in sediment and gut environments: the roles of water pH, lead ions, natural organic matter and phenanthrene. Water Res. 184, 116209.

Zou, J., Liu, X., Zhang, D., Yuan, X., 2020. Adsorption of three bivalent metals by four chemical distinct microplastics. Chemosphere 248, 126064.