



Adsorption-desorption behavior of florpyrauxifen-benzyl on three microplastics in aqueous environment as well as its mechanism and various influencing factors

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ABSTRACT

Microplastics (MPs) and pesticides are two categories contaminants with proposed negative impacts to aqueous ecosystems, and adsorption of pesticides on MPs may result in their long-range transport and compound combination effects. Florpyrauxifen-benzyl, a novel pyridine-2-carboxylate auxin herbicide has been widely used to control weeds in paddy field, but the insights of which are extremely limited. Therefore, adsorption and desorption behaviors of florpyrauxifen-benzyl on polyvinyl chloride (PVC), polyethylene (PE) and disposable face masks (DFMs) in five water environment were investigated. The impacts of various environmental factors on adsorption capacity were evaluated, as well as adsorption mechanisms. The results revealed significant variations in adsorption capacity of florpyrauxifen-benzyl on three MPs, with approximately order of DFMs > PE > PVC. The discrepancy can be attributed to differences in structural and physicochemical properties, as evidenced by various characterization analysis. The kinetics and isotherm of florpyrauxifen-benzyl on three MPs were suitable for different models, wherein physical force predominantly governed adsorption process. Thermodynamic analysis revealed that both high and low temperatures weakened PE and DFMs adsorption, whereas temperature exhibited negligible impact on PVC adsorption. The adsorption capacity was significantly influenced by most environmental factors, particularly pH, cations and coexisting herbicide. This study provides valuable insights into the fate of florpyrauxifen-benzyl in presence of MPs, suggesting that PVC, PE and DFMs can serve as carriers of florpyrauxifen-benzyl in aquatic environment.

1. Introduction

Pesticides have made significant contributions in effectively managing crop diseases and pests, promoting grain yield and elevating quality of human life. Among these chemicals, herbicides have emerged as predominant weed control strategy in contemporary agricultural production (Samsidar et al., 2018; Song et al., 2022). However, the post-application average utilization rate of pesticides is merely around 20–40 %, with a significant portion being released into the natural environment, resulting in multi-media pollution and significant threat to structure and function of ecosystem (Kim et al., 2017; Liu et al., 2021). As a result, global water pollution caused by pesticides is a pervasive

issue (Challis et al., 2018; Stone et al., 2014).

The environmental impact of pesticides has gradually garnered significant attention years, and the development of new pesticides has primarily focused on adhering to principles of environmental friendliness, resulting in compounds with heightened activity and selectivity. Florpyrauxifen-benzyl, a novel pyridine-2-carboxylate auxin herbicide, exhibits a unique mechanism of action that not only addresses known resistance issues but also demonstrates remarkable efficacy against *Echinochloa crusgalli*, a challenging weed to control in paddy field (Lim et al., 2021). However, limited research has been conducted on its environmental migration, transformation and toxicology; previous studies have predominantly focused on synthesis and development

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strategies, weed control and analytical methods (Herrera et al., 2021; Lim et al., 2021). While our previous work investigated residue analysis method, dissipation dynamics, hydrolytic behavior and mechanism of florpyrauxifen-benzyl (Zhou et al., 2022; Zhou et al., 2023), no reports exist regarding its adsorption behavior. Therefore, conducting a comprehensive investigation on its environmental fate is crucial for better assessing environmental risks.

The utilization of plastic film mulching is widely adopted as an effective approach agricultural yield. PVC or PE are primary constituents of plastic film materials. However, most of plastic film exhibits impermeability to water and air and show resistance towards microbial degradation. Consequently, the lack of efficient recycling methods has resulted in a significant accumulation of waste plastic film residues in farmlands worldwide (Jin et al., 2020). These residues have potential to impede plant root growth, alter soil microbial community structure, and ultimately diminish soil productivity (Liu et al., 2014). Meanwhile, the COVID-19 pandemic has led production and utilization of DFMs on a large scale. According to the WHO's estimation, approximately 89 million DFMs were being used worldwide each month to control the widespread transmission of COVID-19 (WHO, 2020). However, inadequate handling and management practices have led to challenges associated with non-biodegradability of these plastic-based DFMs, thereby impacting natural environments (Tesfaldet and Ndeh, 2022).

Furthermore, rapid weathering breakdown of abandoned plastic film residues and DFMs can result in a substantial generation of MPs within a short timeframe. MPs has posed a significant threat to both the ecosystem, causing serious pollution in soil, water and atmosphere, as well as human health (Aragaw, 2020; Leslie et al., 2022; Lin et al., 2022). Moreover, due to their small particle size, large specific surface area, strong hydrophobicity and stable chemical structure, MPs persistently in soil and water for extended periods. This persistence enables them to accumulate organic pollutants (Li et al., 2021), heavy metals (Imran et al., 2019) and antibiotics (Lin et al., 2022), while facilitating their synergistic migration. They can serve as vectors to affect the secondary release of contaminants and their biological toxicity, thereby altering original environmental behaviors and harmfulness of contaminants and facilitating their persistence and transmission (Kinigopoulou et al., 2022; Lin et al., 2022). For example, the degradation, residue, adsorption, desorption and migration of agricultural chemicals can be affected to varying degrees with presence of MPs in natural environment (Fang et al., 2019; Wang et al., 2020a; Wu et al., 2020). Furthermore, the pesticides can be readily absorbed on the surface of MPs when both coexist in water environment. Consequently, plankton ingests pesticides bound to MPs which then enters food chain. The accumulation of pesticides bound to MPs in higher trophic organisms chains and webs has the potential to result in compound pollution, which can cause imperceptible harm not only to individual organisms but also to entire ecosystems (Zou et al., 2017).

Since the pathways and mechanisms of interaction between MPs and pesticides are the prerequisite for evaluating their environmental risks and toxicological mechanisms, the objectives of this study were to (1) investigate adsorption and desorption kinetics, isotherms and thermodynamics of florpyrauxifen-benzyl on three MPs (PVC, PE and DFMs) in five distinct water environment (ultrapure water, tap water, lake water, paddy water and seawater) using a batch equilibrium method; (2) elucidate the effects of various environmental factors (pH, coexisting MPs, fertilizers, cations, anions, surfactants, coexisting herbicide, humic acid, biochar) on adsorption process; (3) reveal interaction mechanisms between three MPs and florpyrauxifen-benzyl. These findings presented herein will contribute to assessment of the safe and rational utilization of florpyrauxifen-benzyl, as well as its potential risk on water environments. Additionally, they will aid in estimating the environmental behaviors of PVC, PE and DFMs, while providing an analytical framework for comprehending their role in facilitating the transport of other pollutants.

2. Materials and methods

2.1. Instruments and reagents[×]

2.2. Adsorption-desorption kinetics tests

Adsorption-desorption kinetics of PVC and PE were as follows (Li et al., 2021; Lin et al., 2022; Wang et al., 2020b). For adsorption kinetics, 0.5 g of PVC or PE were accurately weighted and placed into 50 mL brown glass reagent bottles. Subsequently, 7 mg/L florpyrauxifen-benzyl aqueous solutions with a volume of 20 mL were added, respectively. The samples were continuously shaken at 298 K and 150 rpm using a constant temperature air oscillator under dark condition. The solutions were collected at given time intervals (0.5, 1, 2, 4, 6, 8, 10, 12, 24, 36, 48 and 60 h) for further extraction and purification prior to analysis. To determine the desorption equilibrium time point after reaching adsorption equilibrium (48 h), desorption tests were immediately conducted by removing the remaining aqueous solutions from adsorption tests followed by adding corresponding blank aqueous solutions with a volume of 20 mL into their respective bottles. The samples were then subjected to continuous shaking under identical conditions as mentioned above (298 K and 150 rpm). The solutions were collected at given time intervals (0.5, 1, 2, 4, 6, 8, 10, 12, 24, 36, 48 and 60 h). The other steps were the same as the adsorption tests. The desorption tests of PE was not conducted since PE in this test was not easily separated from aqueous solution.

Adsorption-desorption kinetics of DFMs were as follows (Li et al., 2021; Lin et al., 2022; Wang et al., 2020b). The DFMs (2.8126–2.9325 g) were placed into 250 mL brown glass reagent bottles. Subsequently, 7 mg/L florpyrauxifen-benzyl aqueous solutions with a volume of 200 mL were added, respectively. The remaining steps were the same as above.

2.3. Isothermal adsorption-desorption tests

Isothermal adsorption-desorption of PVC and PE were as follows (Li et al., 2021; Lin et al., 2022; Wang et al., 2020b). 0.5 g of PVC or PE were accurately weighted and placed into 50 mL brown glass reagent bottles. Subsequently, florpyrauxifen-benzyl aqueous solutions with different mass concentrations (2.5, 5, 7, 25, 50 mg/L) in a volume of 20 mL were added, respectively. The samples were continuously shaken at 298 K and 150 rpm for 48 h based on foregoing kinetic experiments. The adsorption amounts were calculated by determining reduction in concentration before and after adsorption. Isothermal desorption tests followed immediately after completion of isothermal adsorption tests. The remanent florpyrauxifen-benzyl solutions from isothermal adsorption tests were removed and replaced with corresponding blank aqueous solutions in a volume of 20 mL. The samples were shaken continuously at 298 K and 150 rpm for 48 h to measure desorption amounts by analyzing concentrations after desorption.

Isothermal adsorption-desorption of DFMs were as follows (Li et al., 2021; Lin et al., 2022; Wang et al., 2020b). The DFMs (2.8126–2.9325 g) were placed into 250 mL brown glass reagent bottles. Subsequently, florpyrauxifen-benzyl aqueous solutions of different mass concentrations (2.5, 5, 7, 25, 50 mg/L) with a volume of 200 mL were added, respectively. The remaining steps remained unchanged.

All adsorption and desorption tests were performed in triplicate, while simultaneous blank tests without MPs were conducted.

2.4. Adsorption-desorption thermodynamics tests

The adsorption-desorption thermodynamics were investigated by isothermal adsorption and desorption tests at five temperatures (278, 288, 298, 308, 318 K).

2.5. Effects of environmental factors on adsorption

2.5.1. Initial pH

The effects of initial pH of florpyrauxifen-benzyl aqueous solutions (7 mg/L) on its adsorption on three MPs were investigated by isothermal adsorption tests. The pH of the florpyrauxifen-benzyl aqueous solutions were adjusted to 3, 4, 5, 6, 7, 8, 9 and 10 using 0.010 mol/L of HCl or NaOH before test, respectively.

2.5.2. Coexisting MPs

The effects of adding PE or DFM (0.5, 1, 2, 5 %) to florpyrauxifen-benzyl aqueous solutions (7 mg/L) on its adsorption on PVC were investigated by isothermal adsorption tests. Here, DFM were cut into tiny pieces. Similarly, PVC or DFM were added to florpyrauxifen-benzyl aqueous solutions (7 mg/L) in isothermal adsorption tests of PE, and PVC or PE were added to florpyrauxifen-benzyl aqueous solutions (7 mg/L) in isothermal adsorption tests of DFM.

2.5.3. Other environmental factors

The effects of adding fertilizers [calcium magnesium phosphate fertilizer (CMPF), urea, organic fertilizer, potash fertilizer, compound fertilizer, organic and inorganic compound fertilizer (OICF)] with varying contents (0.01, 0.1, 1 %), cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Fe^{3+} , Cu^{2+} , Mn^{2+} , Zn^{2+} , Al^{3+}) with different ionic concentrations (0.01, 0.05, 0.1 mol/L), NO_3^- (0.1, 1, 10 mg/L) and NO_2^- (0.01, 0.1, 1 mg/L), surfactants including Tween80 (1, 2, 5 CMC), cetyltrimethyl ammonium bromide (CTAB) (0.1, 0.5, 1 CMC) and sodium dodecylbenzene sulfonate (SDBS) (0.1, 0.5, 1 CMC), coexisting herbicide propyrisulfuron (1, 10, 100 mg/L), humic acid and biochar with different contents (0.01, 0.1, 1 %) to florpyrauxifen-benzyl aqueous solutions (7 mg/L) on its adsorption on three MPs were investigated by isothermal adsorption tests.

2.6. Analytical method for florpyrauxifen-benzyl^{*}

2.7. Mathematical models and data analyses

2.7.1. Adsorption and desorption capacities of MPs. The equilibrium and non-equilibrium adsorption amounts of florpyrauxifen-benzyl on three MPs were calculated using Eq. (1) and Eq. (2) (Lin et al., 2022):

$$Q_{e(t)}^{\text{ads}} = \frac{(C_0 - C_{e(t)}^{\text{ads}}) \times V}{m} \quad (1)$$

$$Q_e^{\text{des}} = \frac{(C_0 - C_e^{\text{des}} - C_{e(t)}^{\text{des}}) \times V}{m} \quad (2)$$

in which Q_e^{ads} (μg/g) and Q_t^{ads} (μg/g) are adsorption amounts of MPs at adsorption equilibrium and at any time t (h), respectively; C_0 (μg/L) represents the initial mass concentration of florpyrauxifen-benzyl; C_e^{ads} (μg/L) and C_t^{ads} (μg/L) are the mass concentrations of florpyrauxifen-benzyl at adsorption equilibrium and at any time t (h), respectively; V (L) denotes the volume of florpyrauxifen-benzyl aqueous solution; m (g) is the mass of MPs; Q_e^{des} (μg/g) and Q_t^{des} (μg/g) are the adsorption amounts of MPs at desorption equilibrium and at any time t (h), respectively; C_e^{des} (μg/L) and C_t^{des} (μg/L) are the mass concentrations of florpyrauxifen-benzyl at desorption equilibrium and at any time t (h), respectively.

2.7.2. Adsorption kinetics models^{*}.

2.7.3. Adsorption isotherm models^{*}.

2.7.4. Adsorption thermodynamics^{*}.

2.8. Characterization^{*}

The detailed information of sections marked by “^{*}” were shown in Supplementary Material (SM).

3. Results and discussion

3.1. Characterization of PVC, PE and DFM

The SEM images, EDX spectra, XRD spectra, contact angles and specific surface area of PVC, PE, outer layer, middle layer, inner layer and ear band of the DFM were presented in Fig. S1-S5. From the SEM images, it is evident that both PVC and PE surface exhibit irregular structures such as folds, protrusions and fractures, and four part of the DFM consist of numerous cylindrical fibers, which may provide sufficient adsorption sites for florpyrauxifen-benzyl. The EDX spectra reveal the presence of C and Cl elements in PVC while only C element appears in PE as well as outer layer, middle layer and inner layer. Moreover, ear band exhibits C, O and N elements. However, H elements can not be displayed in EDX spectra. Furthermore, it may be confirmed that outer layer, middle layer and inner layer are composed of polypropylene (PP) whereas polyurethane (PU) and polyester (PET) are used to fabricate ear band through the XRD and FTIR spectra (Fig. 1) (Lin et al., 2022). The contact angles of PVC, PE, outer layer, middle layer, inner layer and ear band of the DFM are 88.568°, 112.521°, 126.643°, 133.197°, 111.919° and 110.707°, respectively, indicating a high hydrophobicity on their surfaces except PVC (Lin et al., 2022). Consequently, florpyrauxifen-benzyl may be adsorbed on these surfaces through hydrophobic interaction in water environment. Meanwhile, FTIR was employed to analyze the changes of the surface functional groups of the PVC, PE, four parts of the DFM before and after the florpyrauxifen-benzyl adsorption. As shown in Fig. 1, the peaks at 2969 — 2814 cm⁻¹ correspond to the C-H stretching vibrations of aliphatic structures (-CH, -CH₂ and -CH₃). The peaks at 1000 — 720 cm⁻¹ can be attributed to the C-H vibration out of plane. The peaks at 1506 — 1340 cm⁻¹ are assigned to C-C stretching. For PVC, the peak at roughly 610 cm⁻¹ is associated C-Cl. For ear band, the peaks around approximately 1711 and 1240 cm⁻¹ are ascribable to C=O and C-O stretching present of -COO- and -NHCOO-. After florpyrauxifen-benzyl adsorption, none of new peaks appeared or disappeared, indicating that no new covalent bonds were formed during the adsorption process which was primarily dominated by physical force (Lin et al., 2022). Furthermore, as demonstrated by the SEM-EDX mapping (Fig. 2), some elements that only appear in the florpyrauxifen-benzyl in contrast to three MPs, such as F, Cl, O and N elements, were evenly adsorbed on three MPs. SEM-EDX and FTIR results verified that florpyrauxifen-benzyl was adsorbed on the MPs; however, it is noteworthy that this adsorption process was mainly controlled by intermolecular Van Der Waals Force and the microporous filling mechanism and so on. Moreover, the specific surface area of PVC, PE, outer layer, middle layer, inner layer and ear band of the DFM are 1.1490, 0.0750, 0.3600, 1.8318, 0.3261, 0.3132 m²/g, respectively.

3.2. Adsorption-desorption kinetics

The temporal profiles of adsorption amounts of florpyrauxifen-benzyl on PVC, PE and DFM in five water were shown in Fig. 3(A-C). As shown, all adsorption processes initially occurred relatively rapidly within the first 12 h and then proceeded gradually until reaching equilibrium at approximately 48 h. Therefore, 48 h was selected as the adsorption equilibrium time for subsequent experiments. The three MPs exhibited notable adsorption capacities for florpyrauxifen-benzyl; however, adsorption capacities at equilibrium were not identical and followed a general trend of DFM > PE > PVC. Specifically, the adsorption capacities of DFM and PVC in five water exhibited significant variation, ranging from 174.247 to 429.913 μg/g and from 100.258 to 211.744 μg/g respectively. In contrast, the adsorption capacity of PE in five water showed minimal variation with values ranging from 231.865 to 242.465 μg/g. The highest adsorption capacity of DFM was observed in ultrapure water while it exhibited the lowest adsorption capacity in paddy water. In contrast, PVC demonstrated its strongest

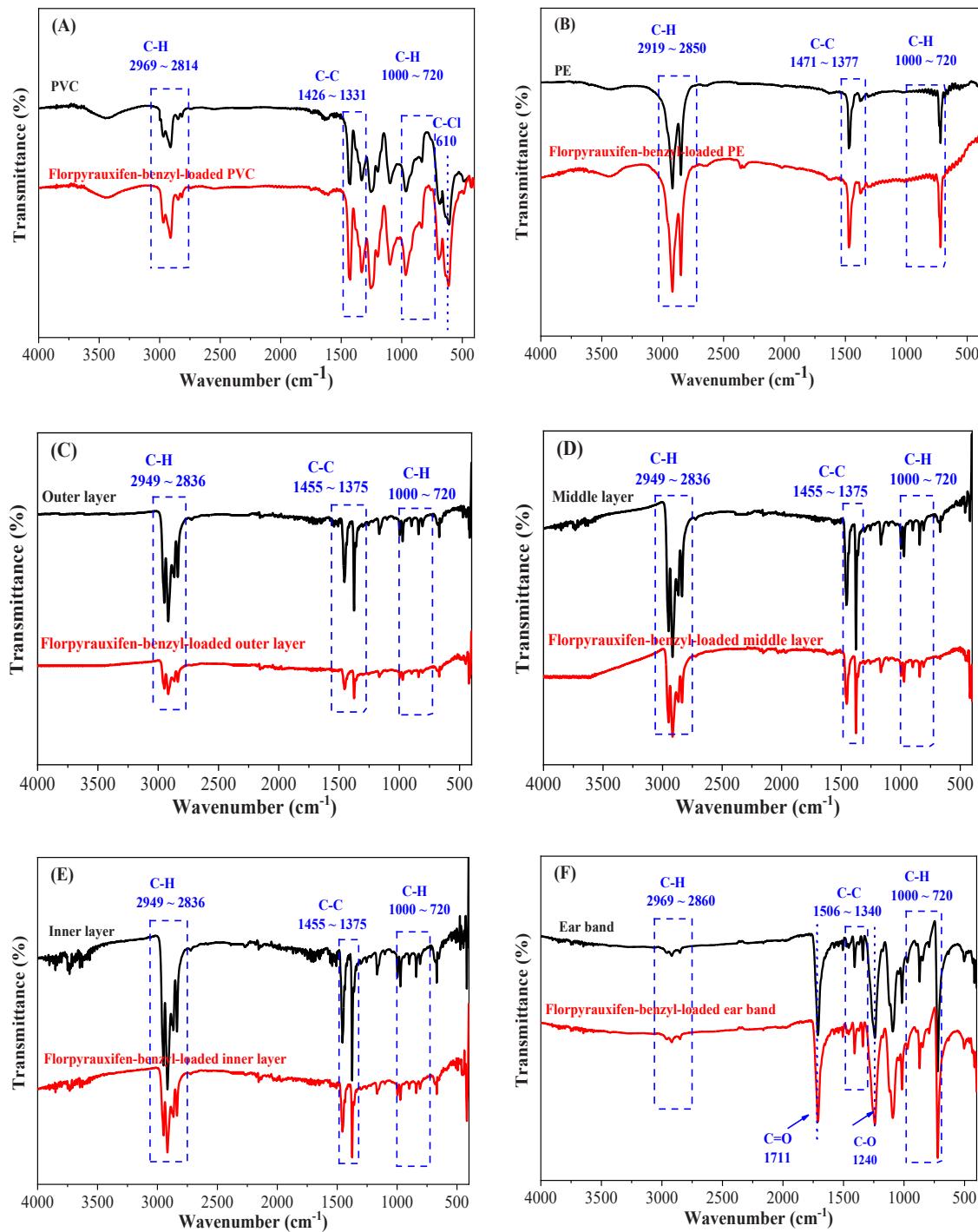


Fig. 1. FTIR spectra of (A) PVC, (B) PE, (C) outer layer, (D) middle layer, (E) inner layer and (F) ear band of the DFMs with and without florpyrauxifen-benzyl loaded.

adsorption capacity in seawater but displayed its weakest adsorption capacity in tap water. These findings suggest that both molecular structure characteristics and physicochemical properties of MPs and properties of different water can affect their respective adsorption behaviors. Additionally, the superior adsorption capacity of DFMs (429.913 µg/g) may be attributed to its larger specific surface area and contact angle in comparison to PVC and PE. However, the relatively lower adsorption capacity of PVC (100.258 µg/g) may primarily result from its weak hydrophobicity with a contact angle < 90°. After achieving adsorption equilibrium, the concentration of florpyrauxifen-benzyl in blank tests (MPs free) was analyzed. The results indicate that effects of photolysis, hydrolysis and microbial decomposition on

adsorption process of florpyrauxifen-benzyl on three MPs are limited. Therefore, the reduction in solution concentration primarily result from the adsorption of florpyrauxifen-benzyl on three MPs.

The adsorption kinetic model can establish correlation between adsorption capacity and time, enabling analysis of pesticide adsorption mechanisms on MPs (Li et al., 2021). Hence, pseudo-first-order kinetics model (PFOM), pseudo-second-order kinetics model (PSOM), Elovich model (EM) and intraparticle diffusion model (IDM) were applied to describe adsorption kinetics data (Bian et al., 2022). The corresponding adsorption kinetic parameters were listed in Table 1. The coefficient of determination (R^2) indicates that EM ($0.8706 \leq R^2 \leq 1.0000$) provides a better fit to the experimental data for PVC adsorption compared to other

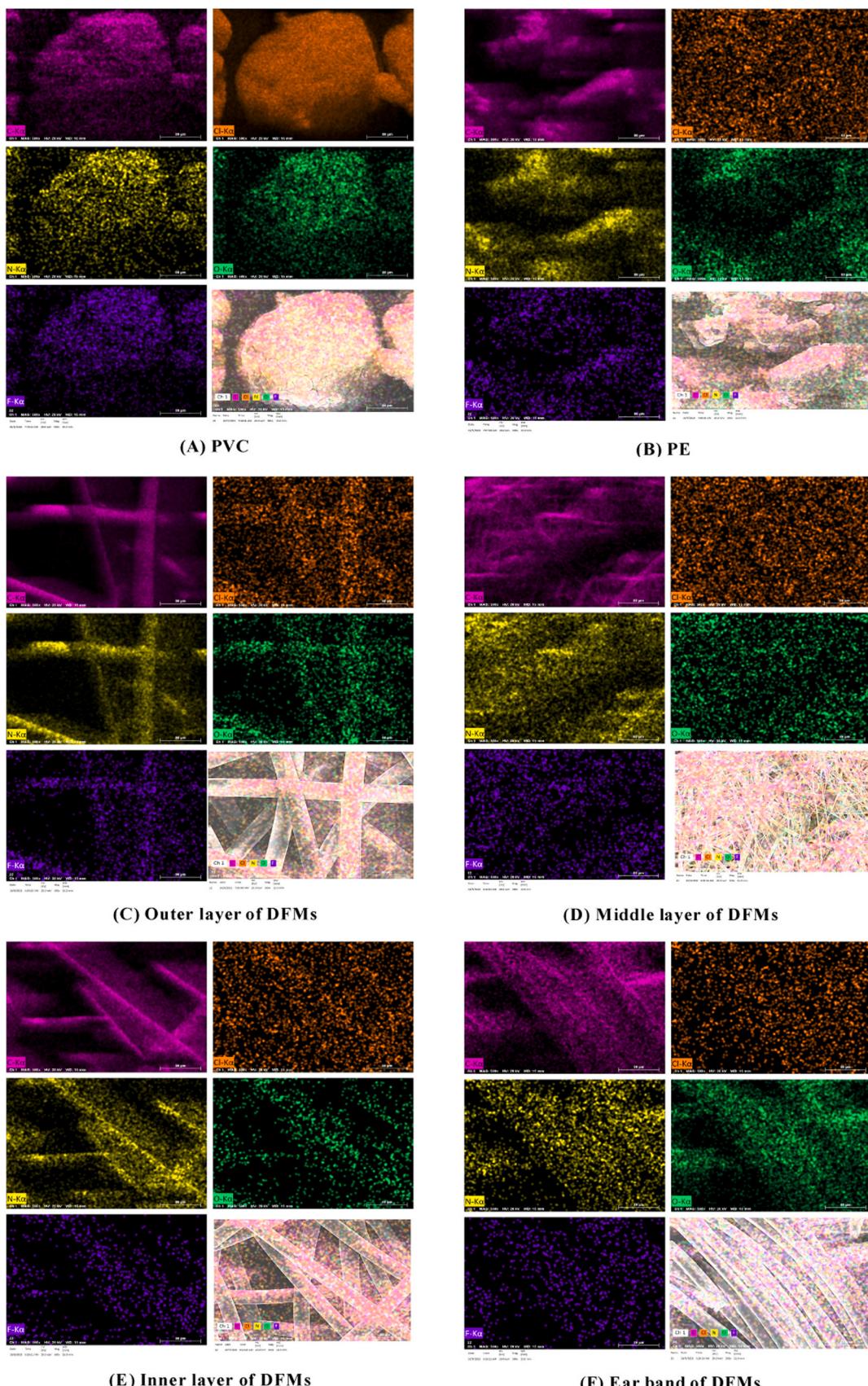


Fig. 2. SEM-EDX mapping images of (A) PVC, (B) PE, (C) outer layer, (D) middle layer, (E) inner layer and (F) ear band of the DFMs after adsorption of florpyrauxifen-benzyl in ultrapure water.

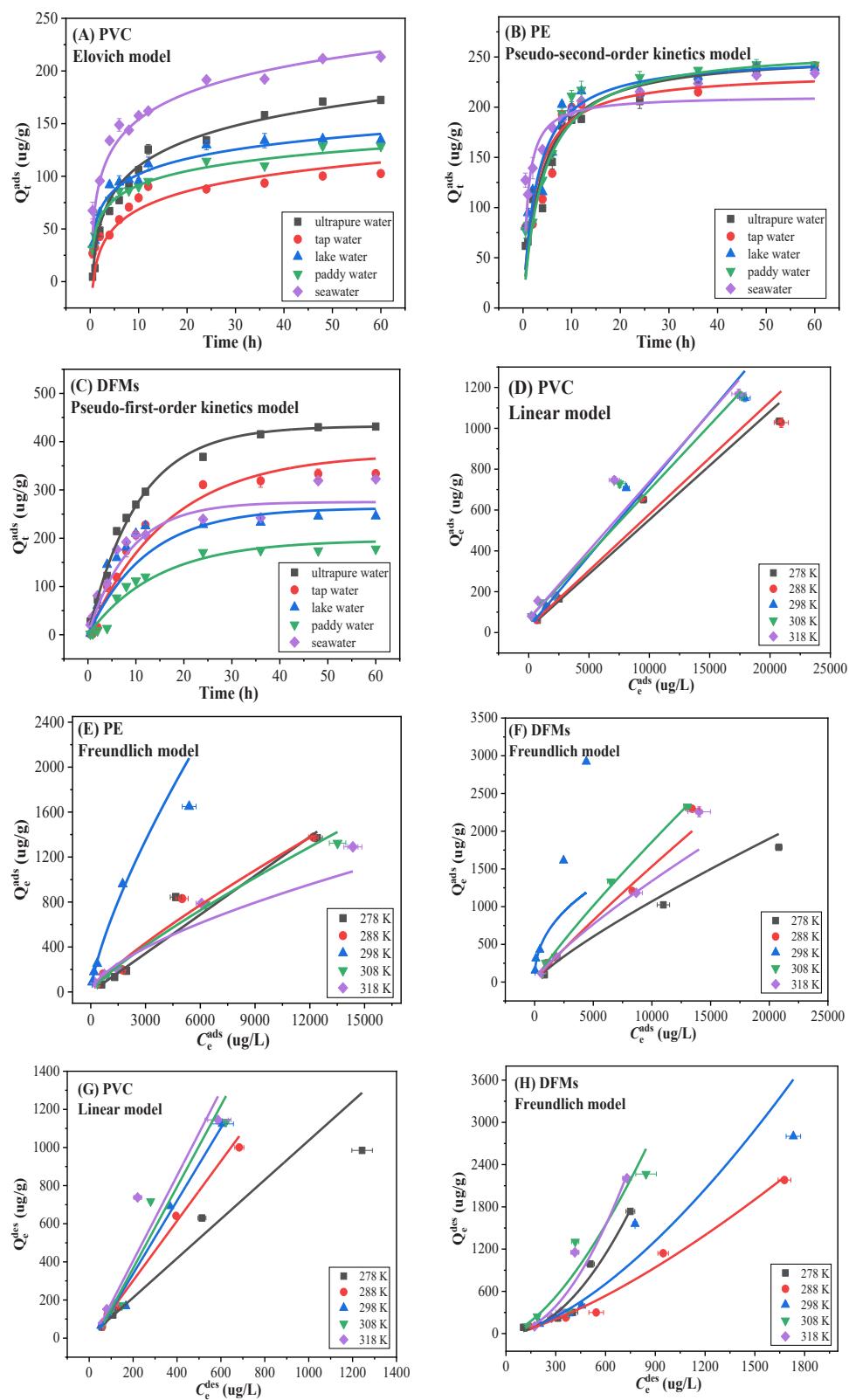


Fig. 3. Adsorption kinetics of floryrauxifen-benzyl on (A) PVC, (B) PE and (C) DFM in five water ($n = 3$); adsorption and desorption isotherms of floryrauxifen-benzyl on (D, G) PVC, (E) PE and (F, H) DFM at different temperatures ($n = 3$).

models, the simulated adsorption amounts obtained by EM show good agreement with experimental values. For PE adsorption, PSOM ($0.9067 \leq R^2 \leq 0.9841$) is found to be more suitable. This finding aligns with Wang et al.'s study where they investigated the adsorption of five

pesticides on PE and observed a better fit with PSOM (Wang et al., 2020b). These results probably suggest that in addition to surface site adsorption, the adsorption process of PE involves mass transfer and intraparticle diffusion (Wang et al., 2020b). In contrast, PFOM

Table 1

Adsorption kinetic parameters fitted by using the pseudo-first-order kinetics model (PFOM), pseudo-second-order kinetics model (PSOM), Elovich model (EM) and intraparticle diffusion model (IDM).

MPs	Water	PFOM			PSOM			EM			IDM		
		K_1 / 1/h	Q_e , cal / $\mu\text{g/g}$	R^2	K_2 / g/ ($\mu\text{g}\cdot\text{h}$)	Q_e , cal / $\mu\text{g/g}$	R^2	K_3 / g/ μg	Q_e , cal / $\mu\text{g/g}$	R^2	K_4 / $\mu\text{g}/ (\text{g}\cdot\text{h}^{0.5})$	Q_e , cal / $\mu\text{g/g}$	R^2
PVC	ultrapure water	0.0707 ± 0.0071	174.9446 ± 1.0796	0.9997	0.0003 ± 5.41E-5	219.6460 ± 7.5559	0.9996	0.0285 ± 0.0001	164.5825 ± 0	0.9998	23.7840 ± 0.2320	/	0.9990
	tap water	121.2571 ± 0	52.1017 ± 5.7882	/	0.0012 ± 0.0002	121.4438 ± 7.5062	0.9316	0.0402 ± 0.0047	107.5531 ± 0	0.8706	12.1282 ± 2.0996	22.2672 ± 5.8652	0.7464
	lake water	603.8517 ± 0	97.1822 ± 0.7594	/	0.0016 ± 3.59E-5	146.9874 ± 0.8054	0.9976	0.0466 ± 8.82E-5	135.4824 ± 0	1.0000	9.3895 ± 0.1561	70.4755 ± 0.4459	0.9970
	paddy water	39.9705 ± 7.20E9	46.6585 ± 14.6961	/	0.0033 ± 0.0010	116.3708 ± 5.8819	0.7925	0.0512 ± 0.0038	122.0610 ± 0	0.9437	11.4154 ± 1.3091	54.2289 ± 4.4636	0.8722
	seawater	97.9922 ± 0	108.0422 ± 10.4214	/	0.0019 ± 0.0001	214.5162 ± 5.2062	0.9804	0.0277 ± 0.0007	210.4725 ± 0	0.9929	20.9750 ± 1.4914	67.1763 ± 3.6974	0.9471
	PE	467.9348 ± 0	240.1017 ± 0.5589	/	0.0011 ± 0.0001	254.4166 ± 1.2314	0.9841	0.0278 ± 0.0016	232.1062 ± 0	0.9648	16.1992 ± 2.0807	114.6559 ± 16.1139	0.8442
7	ultrapure water	0.2804 ± 0.0607	212.8761 ± 2.1555	0.8901	0.0016 ± 0.0004	235.3271 ± 5.9938	0.9259	0.0252 ± 0.0018	239.8104 ± 0	0.9448	29.2856 ± 3.0577	68.2523 ± 14.8564	0.8919
	tap water	49.2660 ± 2.13E11	110.2446 ± 36.2600	/	0.0014 ± 0.0003	251.7327 ± 6.3259	0.9067	0.0302 ± 0.0036	235.6358 ± 0	0.8651	14.9605 ± 2.8230	127.4732 ± 17.9038	0.7112
	paddy water	258.7245 ± 0	140.8627 ± 12.4337	/	0.0009 ± 7.95E-5	261.0561 ± 6.7443	0.9758	0.0224 ± 0.0011	239.2375 ± 0	0.9727	22.7346 ± 1.8277	81.7730 ± 5.6527	0.9332
	seawater	0.8898 ± 0.0684	191.5637 ± 3.7632	0.9367	0.0053 ± 0.0005	211.5992 ± 3.3096	0.9772	0.0293 ± 0.0010	246.6870 ± 0	0.9880	27.4373 ± 3.9065	97.9954 ± 10.4454	0.8146
	DFMs	0.0963 ± 0.0013	432.4103 ± 2.4074	0.9967	0.0002 ± 8.18E-6	495.1087 ± 4.1054	0.9964	0.0109 ± 0.0004	421.5328 ± 0	0.9856	35.5913 ± 2.8345	170.4156 ± 12.8410	0.9344
	tap water	0.0599 ± 0.0169	375.3462 ± 56.6406	0.9216	8.20E-5 ± 5.45E-5	531.0237 ± 125.5281	0.9026	0.0128 ± 0.0012	316.1514 ± 0	0.9165	63.9658 ± 6.3864	/	0.9003
7	lake water	0.0822 ± 0.0240	262.7465 ± 29.5006	0.9354	0.0002 ± 0.0001	355.0364 ± 66.9649	0.9171	0.0175 ± 0.0006	263.8054 ± 0	0.9850	45.2225 ± 4.6029	/	0.8967
	paddy water	0.0685 ± 0.0167	196.1642 ± 23.6676	0.9375	0.0002 ± 0.0001	274.8290 ± 52.2126	0.9232	0.0228 ± 0.0020	173.9191 ± 0	0.9232	35.3156 ± 3.3851	/	0.9074
	seawater	0.1162 ± 0.0174	275.0346 ± 18.9256	0.9714	0.0004 ± 9.44E-5	328.6227 ± 19.4317	0.9858	0.0162 ± 0.0006	292.5380 ± 0	0.9837	54.6382 ± 6.1287	17.3594 ± 21.2164	0.8771

Where “/” represents the value is negative and values represent means ± standard errors.

($0.9216 \leq R^2 \leq 0.9967$) is considered more appropriate for adsorption of florpyrauxifen-benzyl on DFMs, indicating that physical adsorption rather than chemisorption predominantly governs the process. Therefore, rate constants K_3 , K_2 and K_1 are considered to be more applicable for directly describing the adsorption kinetic process of PVC, PE and DFMs, respectively (Li et al., 2019).

The desorption kinetics of florpyrauxifen-benzyl from PVC and DFMs in five water were illustrated in Fig. S5(G–H). Compared to the adsorption process, the desorption process of PVC exhibited a similar trend, with rapid release occurring within the initial 12 h followed by a more gradual rate until reaching equilibrium at approximately 48 h. The desorption amounts from PVC in ultrapure water, tap water, lake water,

paddy water and seawater at 48 h were 27.096, 8.993, 17.721, 17.386 and 38.372 µg/g, respectively. Similarly for DFMs, the desorption process occurred relatively rapidly during the first 4 h and then reached dynamic equilibrium within 48 h. The desorption amounts from DFMs in ultrapure water, tap water, lake water, paddy water and seawater at 48 h were 15.950, 10.486, 10.549, 6.696 and 29.300 µg/g, respectively. The difference in desorption amounts may be attributed to different polymeric properties and the number of effective active adsorption sites of PVC and DFMs as well as different conditions (pH, dissolved organic matter, salinity) of water environments, which leads to differences in their adsorption capacities of florpyrauxifen-benzyl, and ultimately obtains different desorption amounts (Chen et al., 2021).

Table 2

Parameters of the isotherm models for the adsorption and desorption of the florpyrauxifen-benzyl on three MPs in ultrapure water.

Adsorption									
MPs	Temperature / K	Langmuir model			Freundlich model			linear model	
		K_L / L/µg	$Q_{e, \text{cal}}$ / µg/g	R2	K_F / (µg/g)/(µg/L) ^{1/n}	n	R2	K_I / L/g	R2
PVC	278	3.25E-5 ± 4.91E-6	2588.8286 ± 363.9834	0.9910	0.2068 ± 0.0318	1.1612 ± 0	0.9838	0.0531 ± 0.0050	0.9654
	288	2.67E-5 ± 1.07E-5	2931.3887 ± 1037.9794	0.9750	0.2350 ± 0.0797	1.1797 ± 0	0.9787	0.0551 ± 0.0054	0.9622
	298	2.50E-5 ± 1.20E-5	3848.6806 ± 1539.4237	0.9788	0.2261 ± 0.1078	1.1387 ± 0	0.9724	0.0700 ± 0.0066	0.9659
	308	7.01E-5 ± 6.68E-5	2019.2689 ± 1352.2694	0.6856	1.6247 ± 1.1382	1.5094 ± 0	0.8654	0.0632 ± 0.0056	0.9697
	318	0.0018 ± 0.0012	278.7723 ± 126.7992	0.4246	5.4285 ± 2.3432	2.0044 ± 0	0.7241	0.0672 ± 0.0110	0.9016
PE	278	3.01E-6 ± 1.76E-5	3.89E4 ± 2.23E5	0.9451	0.1109 ± 0.0658	0.9961 ± 0	0.9446	0.1163 ± 0.0139	0.9452
	288	5.70E-5 ± 3.00E-5	3354.0388 ± 1326.6356	0.9489	0.5626 ± 0.2798	1.2041 ± 0	0.9545	0.1141 ± 0.0137	0.9447
	298	0.0006 ± 0.0002	1901.4954 ± 470.6744	0.9590	3.5912 ± 0.7121	1.3506 ± 0	0.9737	0.4983 ± 0.0947	0.8696
	308	3.54E45 ± 3.31E47	141.8493 ± 59.0436	/	0.5437 ± 0.0982	1.2086 ± 0	0.9925	0.1140 ± 0.0072	0.9842
	318	0.0008 ± 0.0006	492.9382 ± 250.4682	0.6411	2.2894 ± 0.8888	1.5564 ± 0	0.9152	0.1076 ± 0.0089	0.9735
DFMs	278	/	167.9028 ± 70.7851	/	0.5206 ± 0.4824	1.2073 ± 0	0.7737	0.1028 ± 0.0334	0.6792
	288	6.84E46 ± 0	150.9131 ± 63.9366	/	0.3707 ± 0.2227	1.1061 ± 0	0.9078	0.1592 ± 0.0280	0.8870
	298	0.0113 ± 0.0091	527.9304 ± 115.6731	0.6202	34.2359 ± 28.3732	2.3663 ± 0	0.7307	0.5743 ± 0.1092	0.8695
	308	4.83E46 ± 0	286.0298 ± 175.6771	/	0.5701 ± 0.2553	1.1386 ± 0	0.9733	0.1785 ± 0.0157	0.9698
	318	/	285.4422 ± 159.9886	/	0.7981 ± 0.2256	1.2404 ± 0	0.9921	0.1298 ± 0.0079	0.9854
Desorption									
MPs	Langmuir model			Freundlich model			linear model		R2
	Temperature / K	K_L / L/µg	$Q_{e, \text{cal}}$ / µg/g	R2	K_F / (µg/g)/(µg/L) ^{1/n}	n	R2	K_I / L/g	
PVC	278	0.0002 ± 0.0002	4857.5829 ± 3860.4146	0.9565	1.4618 ± 0.5497	1.0609 ± 0	0.9383	1.0303 ± 0.1435	0.9267
	288	1.36E-6 ± 0.0025	1.10E6 ± 1.98E9	/	1.2033 ± 0.3766	0.9623 ± 0	0.9787	1.5677 ± 0.1119	0.9799
	298	3.74E-7 ± 0.0040	4.40E6 ± 4.72E10	/	0.3684 ± 0.2199	0.7870 ± 0	0.9768	1.9128 ± 0.1936	0.9603
	308	5.23E-7 ± 0.0025	3.48E6 ± 1.66E10	/	0.8607 ± 0.8048	0.8839 ± 0	0.9009	2.1359 ± 0.3006	0.9252
	318	2.16E-6 ± 0.0086	8.20E5 ± 3.27E9	/	0.7301 ± 0.8675	0.8377 ± 0	0.6650	2.2338 ± 0.7568	0.6584
DFMs	278	1.29E-7 ± 0.0029	1.32E7 ± 2.98E11	/	0.0036 ± 0.0080	0.5048 ± 0	0.9487	2.1898 ± 0.4790	0.8326
	288	9.11E-8 ± 0.0011	1.25E7 ± 1.52E11	/	0.0783 ± 0.0681	0.7242 ± 0	0.9824	1.3362 ± 0.1397	0.9577
	298	6.73E45 ± 1.07E47	396.5541 ± 1193.8743	/	0.0372 ± 0.0693	0.6494 ± 0	0.7580	2.4257 ± 0.4356	0.8824
	308	1.21E-7 ± 0.0028	1.52E7 ± 3.55E11	/	0.0736 ± 0.1638	0.6428 ± 0	0.7657	3.0738 ± 0.7631	0.7919
	318	/	507.4834 ± 547.2370	/	0.0049 ± 0.0118	0.5052 ± 0	0.9099	3.7819 ± 0.6640	0.8871

Where “/” represents the value is negative and values represent means ± standard errors.

3.3. Adsorption-desorption isotherms

Variation in temperature may influence adsorption and desorption processes, therefore adsorption and desorption isotherms between florpyrauxifen-benzyl and three MPs in ultrapure water at 278, 288, 298, 308 and 318 K were depicted in Fig. 3(D–H). The adsorption and desorption isotherms are commonly applied to illustrate the distribution of pollutants between solid and liquid phases at equilibrium (Wang et al., 2020b). Langmuir, Freundlich and linear models were used to fit the adsorption and desorption isotherms of florpyrauxifen-benzyl on three MPs for analyzing adsorption mechanism, the fitting parameters of three models were presented in Table 2.

The linear model ($0.9016 \leq R^2 \leq 0.9697$) was found to be more suitable for PVC adsorption. Several studies have consistently reported that adsorption of hydrophobic compounds on MPs followed a linear model (Li et al., 2019; Razanajatovo et al., 2018; Wang et al., 2015). The equilibrium adsorption amounts of PVC increased linearly with increasing initial mass concentrations of florpyrauxifen-benzyl. Since the adsorption isotherms did not reach a plateau, a linear model was employed to fit the isotherm data. Whereas, the K_L values from Langmuir model were extremely close to 0 and the n values from Freundlich model were approximately equal to 1, indicating that Freundlich and Langmuir equations tend to be linear equation. Therefore, the adsorption process of florpyrauxifen-benzyl on PVC was best described by linear model, suggesting that partitioning mechanism dominates this process (Lin et al., 2022). The constants in the linear model (K_l) represent the adsorption affinity between florpyrauxifen-benzyl and PVC. At 278, 288, 298, 308 and 318 K, K_l were 0.0531, 0.0551, 0.0700, 0.0632 and 0.0672 L/g, respectively. These results indicated that temperature had no obvious effect on florpyrauxifen-benzyl adsorption on PVC and no correlations was observed between K_l and temperature (Li et al., 2019).

The Langmuir model exhibited a poor linear fit, indicating that the adsorption processes of florpyrauxifen-benzyl on PE and DFMs were not molecular adsorption on uniform surface. Conversely, the Freundlich model fitting results agreed well with the experimental data for both PE ($0.9152 \leq R^2 \leq 0.9925$) and DFMs ($0.7307 \leq R^2 \leq 0.9921$) adsorption, which was consistent with the previous studies (Fang et al., 2019; Wang et al., 2020b). These results imply that interactions between florpyrauxifen-benzyl and PE or DFMs involve multilayer adsorption on heterogeneous surfaces affected by both chemical and physical adsorption processes. Furthermore, the parameter n in the Freundlich model has been utilized to determine the type of adsorption process. When $n > 1$ indicates a predominance of physical process, whereas when $n < 1$ suggests a prevalence of chemical process (Li et al., 2021). The n ranged from 0.9961 to 2.3663, indicating that the adsorption process between florpyrauxifen-benzyl and PE or DFMs was mainly physical process. Meanwhile, the n values were close to 1, which indicated that the adsorption process was non-linear and easily occurred on heterogeneous surfaces of the MPs (Fang et al., 2019). Besides, the results suggested that the adsorption processes were favorable, implying that increasing concentrations of florpyrauxifen-benzyl would enhance the adsorption (Li et al., 2021). The calculated K_F values at 298 K follow the order of DFMs > PE, in agreement with the adsorption kinetics, which indicated that adsorption capacity of DFMs to florpyrauxifen-benzyl was the strongest (Fang et al., 2019).

Similarly, PVC desorption followed a linear model ($0.6584 \leq R^2 \leq 0.9799$) and DFMs desorption was better described by Freundlich model ($0.7580 \leq R^2 \leq 0.9824$), respectively.

3.4. Adsorption-desorption thermodynamics

To further elucidate the adsorption mechanism of florpyrauxifen-benzyl on three MPs, ΔG was calculated using equilibrium adsorption data, as shown in Table S1. The absolute values of ΔG were less than 40 kJ/mol, indicating the adsorption was a physical process (Bian et al.,

2022). Both high and low temperatures weaken adsorption of florpyrauxifen-benzyl on PE or DFMs. Low temperature may reduce the activity of adsorption sites and thus the ability to adsorb florpyrauxifen-benzyl. Moreover, a higher temperature may increase solubility of florpyrauxifen-benzyl in water, thus increasing its desorption capacity from MPs and subsequently reducing their adsorption capacity. This is in accord with a previous study that both high and low temperatures were not conducive to adsorption of pydiflumetofen in eight different types of soils (Bian et al., 2022).

3.5. Effects of environmental factors on adsorption

A multitude of studies have demonstrated that adsorption of organic contaminants on MPs is influenced by various factors, including type of MPs, solution pH, ionic strength and presence of coexisting dissolved organic matter, etc (Li et al., 2019).

3.5.1. Initial pH

As depicted in Fig. 4 (A), it is evident that pH has a significant impact on adsorption. The adsorption amounts of three MPs exhibit a similar trend with variations in solution pH. The adsorption amounts are greater under acidic condition compared to alkaline condition in the range of 3 – 10, as florpyrauxifen-benzyl are stable under acidic conditions (Zhou et al., 2023), which is beneficial for the adsorption of florpyrauxifen-benzyl on MPs. When pH was 3, the adsorption amounts of florpyrauxifen-benzyl on PVC and DFMs were significantly higher than that of others. The adsorption amount of florpyrauxifen-benzyl on PE was significantly higher at solution pH 4 compared to other pH. Therefore, the pH is a critical parameter that can significantly influence the adsorption performance of florpyrauxifen-benzyl. Wang et al. suggested that high alkalinity could induce surface erosion of MPs, leading to an increased surface area and subsequently enhancing their adsorption capacity for pesticides (Wang et al., 2018). However, the adsorption of florpyrauxifen-benzyl on three MPs under alkaline conditions contradicted Wang et al.' findings; it is plausible that the properties of florpyrauxifen-benzyl play a more significant influence on its adsorption. Consequently, a lower pH facilitates the adsorption of florpyrauxifen-benzyl on three MPs. The zeta potentials of them in a series of pH conditions are presented in Fig. S6. The zeta potentials of PVC decreased with increasing pH and the zeta potential is negative at the pH values were 2 to 10. The point of zero charge (pH_{PZC}) of PE and DFMs was 4. The different surface charges of them at various pHs possibly affected the adsorption behaviors by electrostatic effects (Xu et al., 2018).

3.5.2. Coexisting MPs

In general, MPs in the natural environment do not exist in isolation. The effects of coexisting MPs on adsorption of florpyrauxifen-benzyl on three MPs were investigated, as depicted in Fig. 4(B–D). The adsorption amounts of florpyrauxifen-benzyl on PVC, PE and DFMs increased with increasing coexisting MPs, with PVC being particularly influenced by coexisting DFMs exhibiting 1.170 – 1.437 times higher adsorption amounts. These findings demonstrate a promoting effect of coexisting MPs on adsorption of florpyrauxifen-benzyl (Chen et al., 2021).

3.5.3. Fertilizers

Fertilizers are extensively utilized in agricultural production, consequently leading to their present in water environment. To investigate their impacts on adsorption behavior, six common fertilizers were introduced into the florpyrauxifen-benzyl-MPs adsorption system, as depicted in Fig. 4(E–G). Overall, these fertilizers had minimal influence on the adsorption capacity of three MPs. Lower content of fertilizers promoted adsorption while higher content inhibited it. Notably, 0.1 % compound fertilizer exhibited the most significant promoting effect on PVC; conversely, 1 % OICF demonstrated the greatest inhibiting effect on PVC. The largest promoting effect on PE was 0.01 % organic

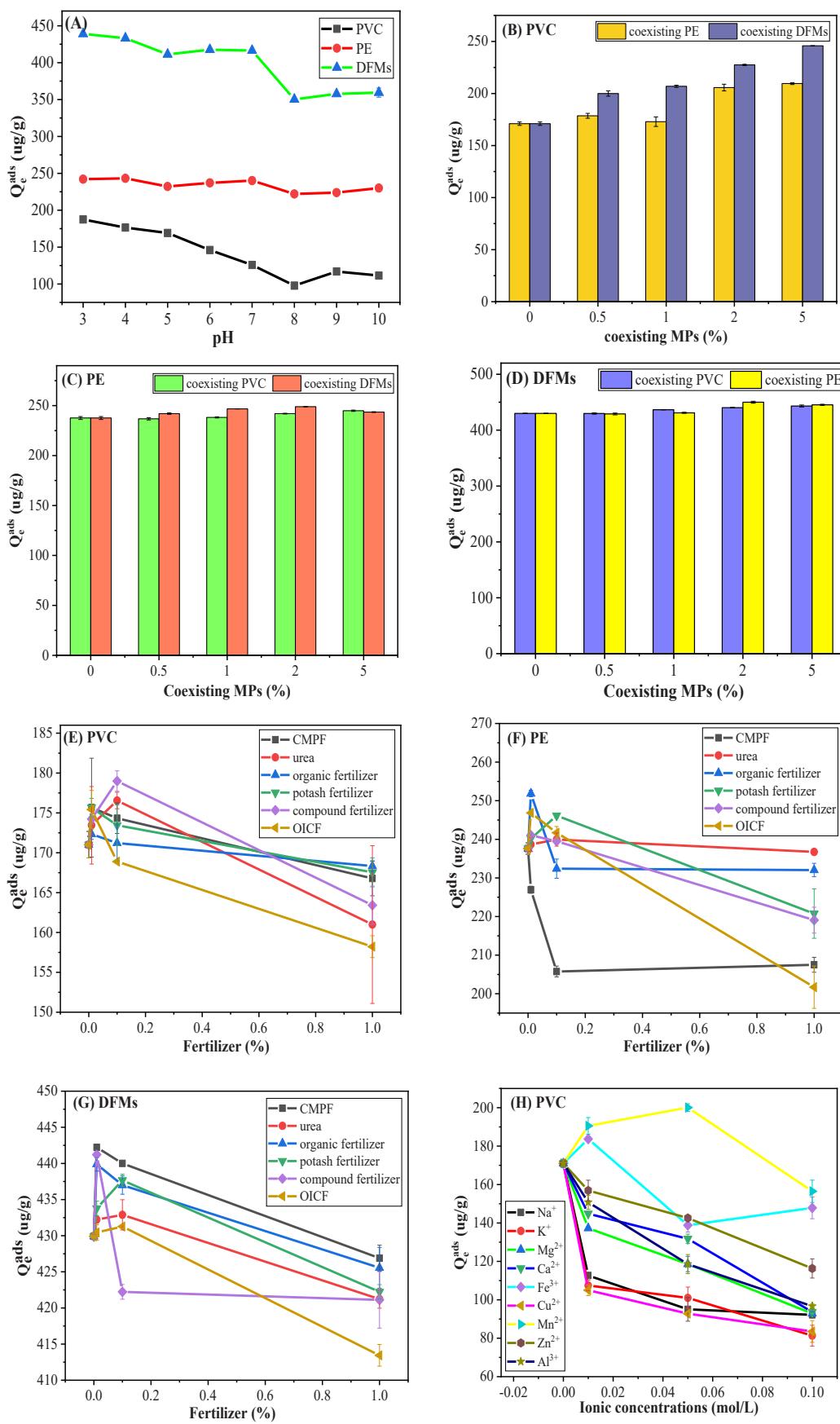


Fig. 4. Effects of pH (A), coexisting MPs (B-D), fertilizers (E-G), cations (H-J), anions (K-L), surfactants (M-O), coexisting herbicide (P), humic acid and biochar (Q-S) on adsorption of florpyrauxifen-benzyl on three MPs in ultrapure water ($n = 3$).

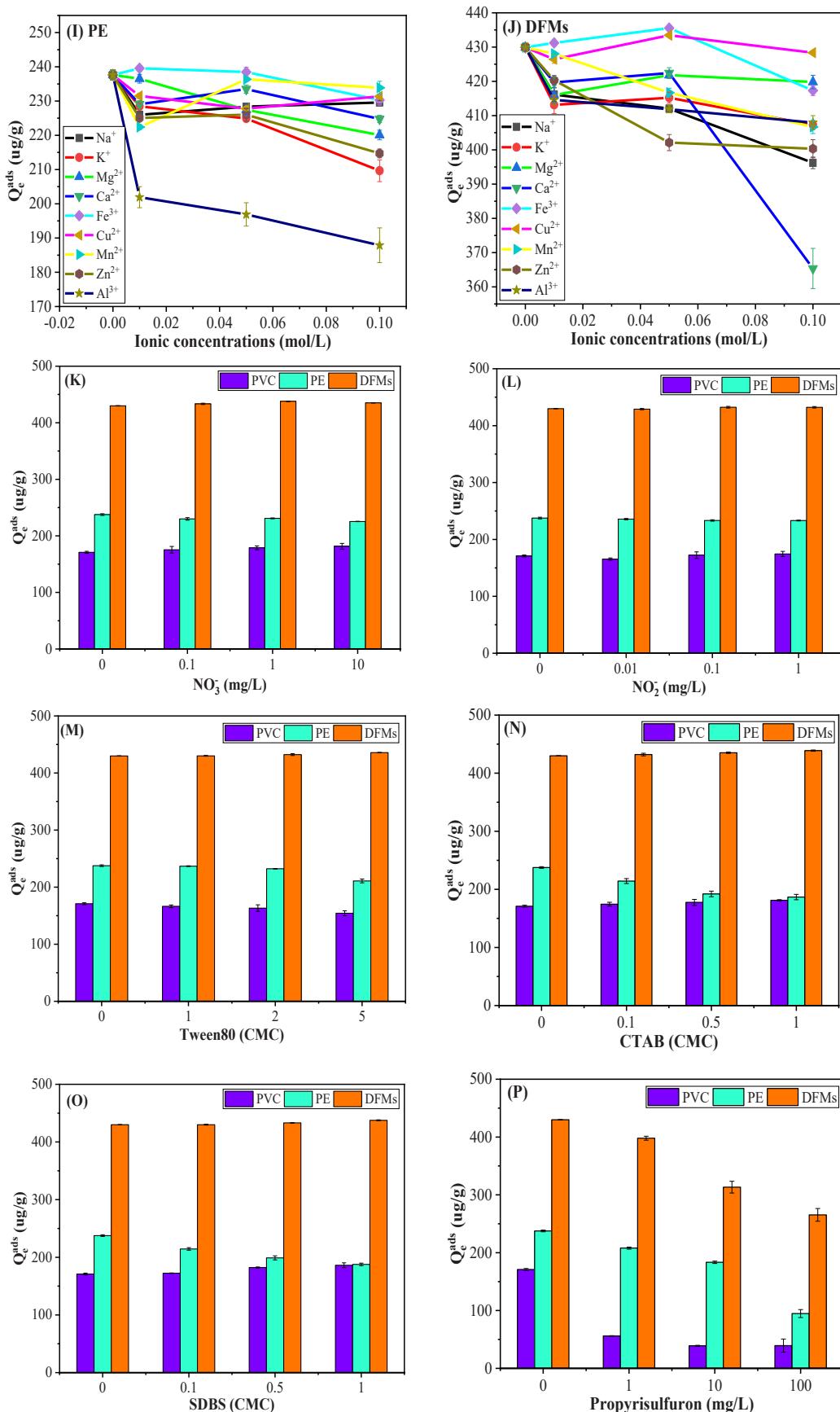


Fig. 4. (continued).

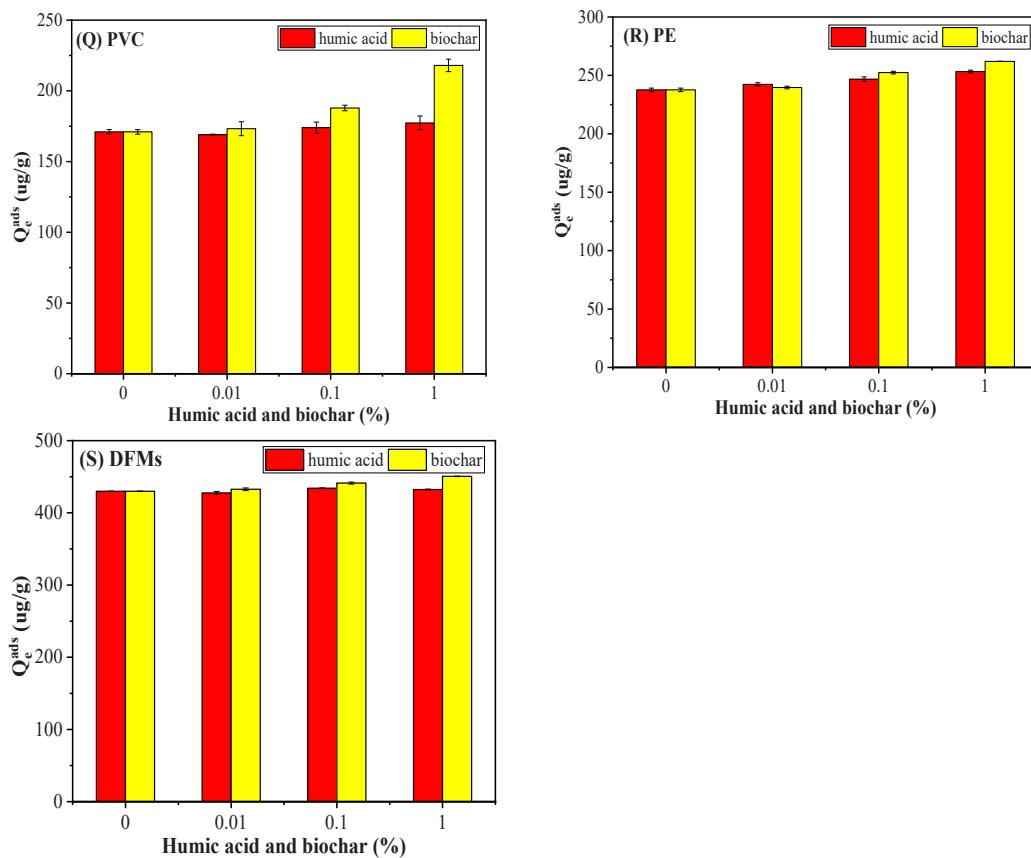


Fig. 4. (continued).

fertilizer, whereas 1 % OICF exerted the strongest inhibiting effect. Similarly, 0.01 % CMPF displayed the largest promoting effect on DFM, and inhibiting effect of 1 % OICF was significant. This could be attributed to occupation adsorption sites of MPs by free fertilizers at high content, thereby impeding the movement of florporauxifen-benzyl molecules within solution and consequently weakening capacity of MPs to adsorb florporauxifen-benzyl (Sun et al., 2023).

3.5.4. Cations

The effects of nine cations with varying ionic concentrations on adsorption process were also assessed, as shown in Fig. 4(H–J). The low concentration of Fe^{3+} and Mn^{2+} slightly promoted PVC adsorption, low concentration of Fe^{3+} slightly increased PE adsorption, and low concentration of Fe^{3+} and Cu^{2+} slightly enhanced DFM adsorption. Conversely, all other cations had a certain inhibiting effect on the adsorption of three MPs, with PVC being particularly affected.

The strong acid and base salts, such as NaCl , KCl , and CaCl_2 , may exhibit a salting-out effect on the water solubility of florporauxifen-benzyl. The competition between Na^+ , K^+ and Ca^{2+} and florporauxifen-benzyl molecules for solvent molecules in water affects the adsorption process (Yang et al., 2006). Additionally, MgCl_2 , $\text{Fe}_2(\text{SO}_4)_3 \cdot \text{xH}_2\text{O}$, CuSO_4 , $\text{MnCl}_2 \cdot 4 \text{H}_2\text{O}$, $\text{ZnSO}_4 \cdot 7 \text{H}_2\text{O}$ and $\text{Al}(\text{NO}_3)_3 \cdot 9 \text{H}_2\text{O}$ are strong acid and weak base salts that can precipitate $\text{Mg}(\text{OH})_2$, $\text{Fe}(\text{OH})_3$, $\text{Cu}(\text{OH})_2$, $\text{Mn}(\text{OH})_2$, $\text{Zn}(\text{OH})_2$ and $\text{Al}(\text{OH})_3$, respectively. These resulting precipitates may potentially affect the adsorption process. Meanwhile, dissolved cations in aqueous solution may increase cations concentration near surface of MPs, thereby leading to competition for adsorption sites and consequently excluding florporauxifen-benzyl attachment (Li et al., 2021). Moreover, when the ionic strength reaches a certain level, it may cause decomposition of the florporauxifen-benzyl, resulting in a reduced concentration of florporauxifen-benzyl in the aqueous solution, and consequently diminishing its adsorption on MPs (Wang et al.,

2020b). This result indicates that cation exchange and electrostatic interaction play significant roles in interaction mechanisms between florporauxifen-benzyl and three MPs (Lin et al., 2022).

3.5.5. Anions

The impacts of NO_3^- and NO_2^- on adsorption process were also investigated, as depicted in Fig. 4(K–L). Except for 0.01 mg/L NO_2^- , which slightly inhibited adsorption of PVC and DFM, other concentrations of NO_3^- and NO_2^- had a certain promoting effect on PVC and DFM, with NO_3^- having a stronger promoting effect than NO_2^- . Both NO_3^- and NO_2^- displayed inhibiting effect on PE, with adsorption capacity of adding NO_3^- being lower than that of NO_2^- .

3.5.6. Surfactants

The surfactants induced variations in adsorption capacity, as shown in Fig. 4(M–O). To PVC adsorption, adding Tween80 inhibited adsorption, while SDBS and CTAB both promoted it, and promoting effect was SDBS > CTAB. The addition of surfactants inhibited PE adsorption, and inhibiting effect was CTAB > SDBS > Tween80. The presence of three surfactants promoted DFM adsorption, and the promoting effect was CTAB > SDBS > Tween80.

On the one hand, as an organic matter, surfactants may be adsorbed by MPs, thereby competing with florporauxifen-benzyl for adsorption sites of MPs and resulting in a reduction in adsorption capacity. On the other hand, they can act as effective adsorbents to enhance the overall adsorption capacity. Furthermore, monomer molecules of surfactant in water may influence solubility of florporauxifen-benzyl (Xia et al., 2020). Therefore, there are multiple factors contributing to impact of surfactants on adsorption of florporauxifen-benzyl on MPs, and adsorption amount is determined by a complex interplay of various competing effects.

3.5.7. Coexisting herbicide

The adsorption amount of florporauxifen-benzyl by MPs significantly decreased with increasing concentration of herbicide propyrisulfuron, as shown in Fig. 4(P). The extent of decrease followed the order: PVC > PE > DFMs. It indicates that propyrisulfuron competes strongly with florporauxifen-benzyl for adsorption sites of MPs (Yu et al., 2019).

3.5.8. Humic acid and biochar

Humic acid is a naturally occurring, organic polymeric substance which contains both hydrophilic and hydrophobic molecules (Dong et al., 2021). Biochar is widely used in ecological restoration, agriculture and environmental protection because of its excellent pore structure, large specific surface area and abundant adsorption sites (Dai et al., 2019). The adsorption of florporauxifen-benzyl on MPs was positively affected by increase in humic acid and biochar content, as depicted in Fig. 4(Q-S). This can be attributed to abundant presence of functional groups within humic acid, such as carboxylic and alcohol hydroxyl groups, which facilitate pesticide adsorption through hydrogen bonding. Meanwhile, dissolved humic acid can enhance pesticides solubility, thereby increasing the overall adsorption capacity (Yu et al., 2019). Specifically, biochar significantly increased adsorption capacity, with a tendency towards saturation of adsorption amounts observed at addition of 1 % biochar for both PE and DFMs. The remarkable improvement in adsorption capacity by biochar may be attributed to its porosity and large specific surface area (Bian et al., 2022).

4. Conclusion

The adsorption and desorption behaviors of florporauxifen-benzyl on PVC, PE and DFMs were comprehensively investigated as a function of temperature, pH, coexisting MPs, fertilizers, cations, anions, surfactants, coexisting herbicide, humic acid and biochar. The adsorption capacity was found to be influenced by molecular structure characteristics and physicochemical properties of MPs, properties of different water and various environmental factors. The adsorption of florporauxifen-benzyl on three MPs exhibited a pronounced pH dependence. Furthermore, the influence of ionic concentrations on adsorption processes were not negligible, suggesting that cation exchange and electrostatic interaction play significant roles in the interaction mechanisms. The adsorption process was predominantly governed by physical force, as evidenced by the characterization of SEM-EDX and FTIR, along with utilization of adsorption kinetic models, adsorption isotherms models and thermodynamics analysis. This study provides novel insights into potential combined environmental pollution risks posed by pesticides and MPs in agricultural fields. To our knowledge, this is the first study to evaluate interactions between florporauxifen-benzyl, PVC, PE, DFMs and environmental factors, providing a valuable baseline for further investigations.

CRediT authorship contribution statement

Dong Zemin: Writing – review & editing. **Li Baotong:** Supervision, Funding acquisition. **Zhou Rendan:** Writing – original draft, Data curation. **Li Yuqi:** Validation. **Xing Lei:** Validation. **Wu Tianqi:** Investigation. **Lin Wei:** Investigation. **Chang Hailong:** Investigation. **Li Zhuo:** Methodology. **Zhou Wenwen:** Validation.

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.

Data Availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2024.116066.

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