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Assessment and prediction of the effect of ageing on the adsorption of nonylphenol in black carbon-sediment systems

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ABSTRACT

Black carbon (BC) is a promising sediment amendment, as proven by its considerable adsorption capacity for hydrophobic organic pollutants and accessibility, but its reliability when used for the removal of pollutants in natural sediments still needs to be evaluated. For example, the ageing process, resulting in changing of surface physicochemical properties of BC, will decrease the adsorption capacity and performance of BC when applied to sediment pollution control. In this study, how the ageing process and BC proportion affect the adsorption capacity of BC-sediment systems was modelled and quantitatively investigated to predict their adsorption capacity under different ageing times and BC additions. The results showed that the ageing process decreased the adsorption capacity of both BC-sediment systems, due to the blockage of the non-linear adsorption sites of BC. The adsorption capacity of rice straw black carbon (RC)-sediment systems was higher than that of fly ash black carbon (FC)-sediment systems, indicating that RC is more efficient than FC for nonylphenol (NP) pollution control in sediment. The newly established model for the prediction of adsorption capacity fits the experimental data appropriately and yields acceptable predictions, especially when based on parameters from the Freundlich model. However, to fully reflect the influence of the ageing process on BC-sediment systems and make more precise predictions, it is recommended that future work considering more factors and conditions, such as modelling of the correlation between the adsorption capacity and the pore volume or specific surface area of BC, be applied to build an accurate and sound model.

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Introduction

Black carbon (BC), a highly aromatic organic carbon widely existing in sediments of natural water bodies, is mainly derived from incomplete combustion of biomass and fossil fuels (Cornelissen et al., 2005; Cousins et al., 1998). It is proposed to play a potentially important role in the sorption and desorption of contaminants, especially hydrophobic organic contaminants (HOCs), in sediments due to its intrinsically high surface activity (Huang et al., 2018a; Kim et al., 2016; Lee et al., 2018; Li et al., 2017; Suo et al., 2019; Wang et al., 2019; Zhao et al., 2018). The adsorption capacity of BC is several orders of magnitude higher than that of other natural organic matter in sediments. For example, Xiao et al. (2004) found that organic carbon-normalised single-point K_{OC} values were comparable to those reported in the literature for purified kerogen and BC, which means that hard carbon contributes most of the adsorption capacity of sediments (Yu et al., 2006). In addition, the adsorption process for HOCs on BC is partially irreversible, which will reduce the risk of re-release after applying BC to adsorb contaminants (Wang et al., 2017). Moreover, as BC is a part of natural sediment, some of its features, such as widespread sources, eco-friendly production processes (Wang et al., 2019), low cost and no secondary pollution, also promote its reliability. Hence, BC is considered to be a promising adsorbent material when applied to control sediment organic pollution.

However, the adsorption capacity of BC for organic compounds in sediments can vary with respect to the contact time between BC and sediment. This is because after BC is introduced into sediment, it might interact with other substances, such as humic acids, minerals, and native organic matter (Yang et al., 2020; Huang et al., 2018b; Zhang and He, 2013). The main effects of these interactions on BC include surface coverage, pore blockage and surface oxidation, which might affect its sorptive properties. This procedure is called ageing (Jing et al., 2018), and few studies have been conducted on the influence of BC ageing in sediment on its adsorption capacity. Lou et al. (2012) found that the adsorption capacity of BC from both agricultural and industrial sources for pentachlorophenol decreased with ageing time. In addition, the adsorption capacity of aged BC in sediment was also related to the BC mass proportion (Bao-Son et al., 2017; Hagner et al., 2015). Previous studies have shown the effects of BC addition or ageing time on the adsorption capacity of BC-sediment systems, but the combined effect of the two variables on adsorption capacity and their contributions to the adsorption or distribution effects of BC-sediment systems have not been quantitatively studied.

Therefore, in this paper, nonylphenol (NP), a kind of environmental oestrogen (Lepretti et al., 2015; Shirdel and Kalbassi, 2016), was selected as a representative hydrophobic contaminant. NP is ubiquitous in environmental matrices worldwide, for example, it was found in bivalves and sediments on the northeast coast of China (Wang et al., 2010). NP can interfere with the growth of organisms and might cause cancer or malformation at a low concentration, and Yuan et al. (2019) found that 4-NP can lead to potential DNA damage to Chinese hamster ovary cells. In addition, NP has only limited affinity in the sediment phase, which means that the improvements obtained by adding BC will be prominent. Rice straw and fly ash were selected to produce BC, representing BC from agricultural and industrial sources, respectively. Then, how the adsorption capacity for NP of BC-sediment systems is affected by different ageing times and BC amendment rates was investigated to verify the feasibility of BC-based remediation by utilizing the Freundlich and dual-mode models for fitting and analysing changes in parameters. For example, Acharya et al. (2020) applied the Freundlich model to fit the removal process of benoxacor and furilazole by granular activated carbon. Furthermore, the relations between adsorption capacity and BC mass proportion and between adsorption capacity and ageing time were modelled so that the adsorption capacity can be predicted from the known BC mass proportion and ageing time, avoiding tedious experimental processes. In conclusion, it is expected that this study will enable evaluation of the feasibility of the application of BC for NP pollution control and provide a theoretical foundation for the remediation of organic pollution in sediments.

1. Materials and methods

1.1. *Chemicals and materials*

NP (purity > 99%) was purchased from Aladdin (China) and prepared to generate a concentrated stock solution with acetonitrile.

Rice straw black carbon (RC) was prepared from air-dried rice straw collected from the Hua-Jia-Chi farm of Zhejiang University in China. The rice straw was burnt on a stainless steel plate in an open field under controlled conditions. Fly ash black carbon (FC) was collected from the electrostatic precipitation stage of a thermoelectric plant in Hangzhou, Zhejiang Province, China. To obtain purified RC and FC, first, the BC samples were treated with 6 mol/L HCl once. Then, they were treated with 6 mol/L HCl and 1 mol/L:1 mol/L HCl:HF solutions 5 times each (throughout the process, each gram of sample required 20 mL solution for every step). After that, the samples were washed with distilled water and oven-dried overnight at 105 °C. Finally, the RC and FC were finely ground (< 250 µm particle size). The surface and chemical properties of the RC and FC were characterised, such as the elemental composition, surface area and pore volume, surface acidity and basicity, and surface functional groups. RC exhibited a higher H/C ratio (0.032), surface area (72.1 m^2/g) and porosity (0.133 mg/L) than FC (Appendix A Table S1). RC and FC had 2.995 and 0.984 mmol/g acidic groups, respectively, and no basic groups due to the acid treatments (Appendix A Table S2). From the Fourier transform infrared spectroscopy (FT-IR) spectra of RC, the peak of C–H stretching vibration of aliphatic C was present at 2920 cm^{-1}, which contributed importantly to the high adsorption affinity of BC for HOCs (Chefetz and Xing, 2009). The other main peaks of two BCs were assigned as follows: C = C stretching vibration of aromatic C (1620 cm^{-1}), O–H stretching vibration of carboxylic C (3435 cm^{-1}) and C–O stretching vibration of carboxylic C (1090 cm^{-1}) (Zhang et al., 2013) (Appendix A Fig. S1).

1.2. *Collection of sediment samples*

A clam sampler was used to collect surface sediment from the Qian-tang River, Hangzhou, Zhejiang Province, China. Then, debris, decayed leaves, and other impurities were removed. Sediment samples were stored at −4 °C in the dark after freeze-drying, ground and screened. The contents of organic matter and BC in the sediment, determined according to GB 9834-88 and the method of Lim and Cachier (1996), were 0.96% and 0.37%, respectively.

1.3. *Ageing treatment of BC*

To achieve BC proportions (dry weight basis) of 0% (control treatment, in order to eliminate effect of the indigenous BC, ensuring reliability and validity of the results), 0.5%, 2.0% and 5.0% (W/W), sediment was mixed in a 50 mL glass tube with specific quantities of RC or FC. Then, the sediments and amendments were thoroughly mixed, and 0.02 Appendix A NaN_3 solution was added to restore the initial moisture content (Chen et al., 2004). After that, the sediments were placed in the dark and aged for 7, 21, 63 and 120 days at 25 °C. Each treatment was replicated three times.

1.4. *Adsorption isotherm of NP in aged BC-sediment systems*

After ageing for a period of time, the sediments were treated with 30 mL electrolyte solution (200 mg/L NaCl and 200 mg/L NaN_3). To reach NP concentrations ranging from 0.2 to 4.0 mg/L, every tube was supplemented with a specific volume of 1000 mg/L NP solution. After that, the tubes were shaken at 180 r/min on a shaker at 25 ± 1 °C for 16 hr in the dark. Then, the sediment and water phases were separated by centrifugation at 3000 r/min for 20 min. Finally, 1 mL liquid supernatant in each tube was collected for analysis by high-performance liquid chromatography (HPLC) (Agilent 1100, Agilent, USA). Each treatment was replicated three times.

1.5. *Data analysis*

The Freundlich and dual-mode models were used to fit the adsorption isotherm data.

Freundlich model:

$$Q_e = K_f C_e{}^n \quad (1)$$

where C_e (mg/L) is the equilibrium concentration of the adsorbate; Q_e (mg/kg) is the adsorption amount; K_f ((mg/kg)/(mg/L)^n) is the adsorption capacity coefficient,; and n is the heterogeneity factor of point energy, which reflects the magnitude and change of energy in a particular adsorption process and is often used to indicate the degree of nonlinearity.

Dual-mode model:

$$Q_T = Q_P + Q_A \quad (2)$$

$$Q_T = K_{om} C_e + Q_{max,D} \quad (3)$$

where Q_T (mg/kg) represents the total adsorption amount of HOCs on the adsorbent. Q_P (mg/kg) and Q_A (mg/kg) are the distribution amount and the adsorption amount, respectively. K_{om} (L/kg) represents the distribution system, and $Q_{max,D}$ (mg/kg) is the saturated adsorption capacity estimated from the high-solute-concentration data.

2. Results and discussion

2.1. *Adsorption of NP in aged BC-sediment systems*

Figs. 1 and 2 show the adsorption isotherms of NP in RC-sediment and FC-sediment systems, respectively. It can be seen from Figs. 1 and 2 that regardless of the ageing time, the adsorption capacities of the two BC-sediment systems for NP and nonlinearity of the adsorption isotherms both increased with increasing BC proportion, and the adsorption capacity of the aged RC-sediment system for NP was significantly higher than that of the aged FC-sediment system, which is consistent with our previous research results (Lou et al., 2014). According to the characteristics of RC and FC (Appendix A Tables S1 and S2, Fig. S1), the presence of aliphatic carbon (Chefetz and Xing, 2009), the higher carbon sorption sites, specific surface area (SSA), and pore volume of RC increased the adsorption capacity to NP. Therefore, the better adsorption performance of RC is attributed to its aliphatic carbon, higher C content, pore volume and SSA, which has been discussed in detail in our previous research (Cheng et al., 2017).

To obtain a deeper understanding of the NP adsorption performance of the two aged BC-sediment systems treated for different ageing times, the adsorption isotherms were fitted by the FM (Table 1), and the fitting parameters of the Freundlich model that changed with ageing time were comparatively analysed (Fig. 3).

As shown in Table 1, in both BC-sediment systems, the adsorption isotherm was well fitted by Freundlich model, with R^2 ranging from 0.968 to 0.9997. Both values of K_f increased with the BC proportion, while both values of n decreased, indicating that BC can improve the adsorption capacity and nonlinearity of NP in BC-sediment systems. For example, after the systems were aged for 120 days, in the RC-sediment system, the value of K_f increased from 73.21 ± 0.79 (pure sediment) to 853.28 ± 8.76 (mg/kg)/(mg/L)^n as the mass percentage of BC content changed from 0% to 5.0%. For the FC-sediment system, the K_f value shifted from 73.21 ± 0.79 (pure sediment) to 148.97 ± 1.22 (mg/kg)/(mg/L)^n. The values of K_f in both aged BC-sediment systems decreased with increasing ageing time, but the values of n showed an opposite trend. For example, when the BC-sediment systems containing 5% BC mass were aged from 7 to 120 days, in the RC-sediment system, the value of K_f decreased from 969.41 ± 14.29 to 853.28 ± 8.76 (mg/kg)/(mg/L)^n, while that in the FC-sediment system decreased from 190.73 ± 2.79 to 148.97 ± 1.22 (mg/kg)/(mg/L)^n.

The changes in the adsorption parameters fitted by the Freundlich model in the two aged BC-sediment systems with ageing time are shown in Fig. 3, and a linear fit was made to analyse the relationship between adsorption parameters and ageing time. The results showed that with increasing ageing

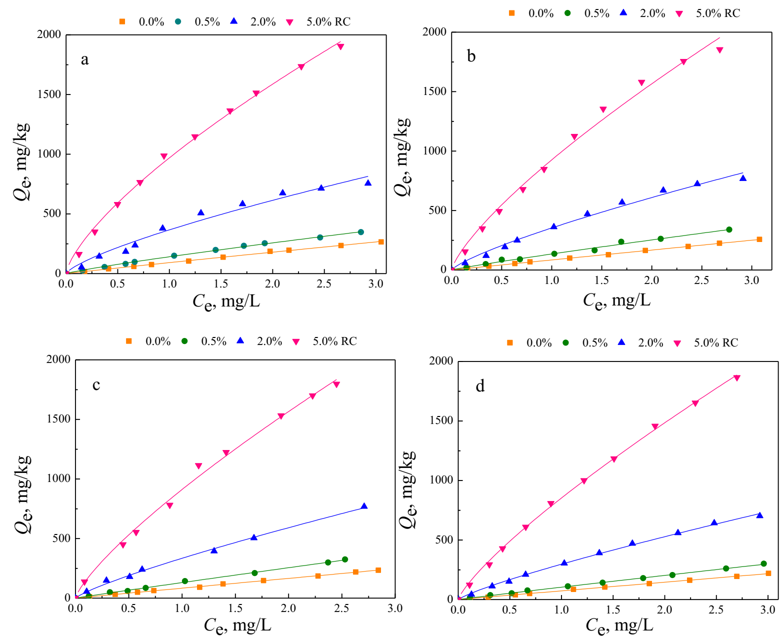

Fig. 1 – Sorption isotherms of NP on aged RC-sediment system. (a):7-day ageing; (b): 21-day ageing; (c): 63-day ageing; (d): 120-day ageing.

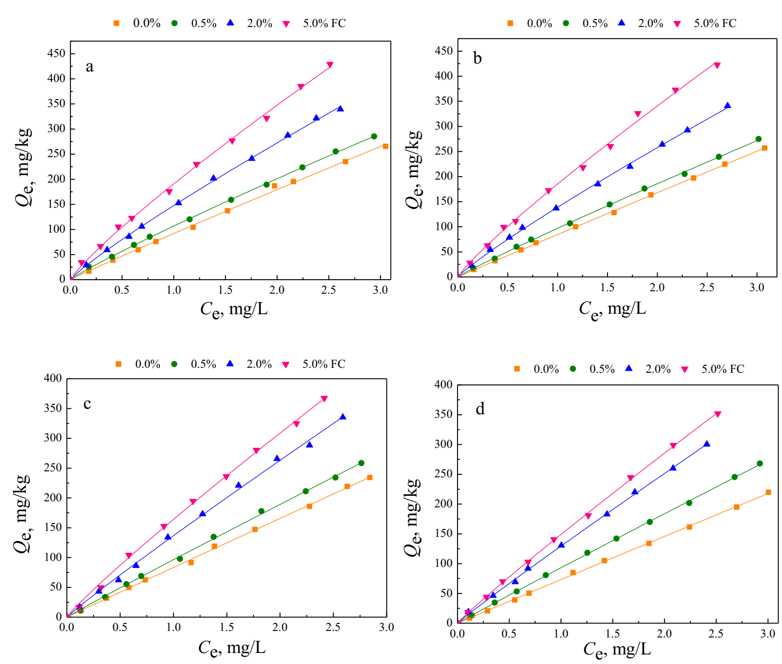

Fig. 2 – Sorption isotherms of NP on aged FC-sediment system. (a):7-day ageing; (b): 21-day ageing; (c): 63-day ageing; (d): 120-day ageing.

time, the value of K_f decreased faster in the aged RC-sediment system than in the FC-sediment system when the BC content was the same. However, even though aged RC-sediment systems lost their adsorption capacity faster than FC-sediment systems, they still maintained a high adsorption performance for NP after 120 days of ageing. This result indicated that RC is a promising remediation material for controlling organic contamination of sediments.

2.1.1. *Mechanism by which the ageing process affects the adsorption performance of BC*

Huang and Weber (1998) proposed that organic matter in sediment can be divided into two categories: one is a highly amorphous rubbery soft carbon component, which has a linear distribution of HOCs, no competitive adsorption, and a reversible desorption process; the other is a compact, ordered, highly aromatic glassy hard carbon component that exhibits nonlin-

Table 1 – Parameters of sorption of NP on aged sediment-BC systems fitted by Freundlich model.

Adsorbent	Parameters	7 days	21 days	63 days	120 days
S	K_f ((mg/kg)/(mg/L)^n)	91.97±1.84	84.48±0.73	83.35±1.22	73.21±0.79
	n	0.96±0.023	0.99±0.0096	0.99±0.017	0.99±0.012
	R^2	0.998	0.9996	0.999	0.999
0.5%	K_f ((mg/kg)/(mg/L)^n)	141.13±2.24	134.53±5.40	130.51±2.89	103.92±1.67
RC	n	0.87±0.020	0.91±0.051	0.97±0.028	0.97±0.019
	R^2	0.998	0.987	0.998	0.999
2.0%	K_f ((mg/kg)/(mg/L)^n)	366.33±18.80	353.71±12.43	334.55±8.91	297.19±5.05
RC	n	0.75±0.063	0.78±0.043	0.82±0.033	0.83±0.020
	R^2	0.968	0.987	0.995	0.997
5.0%	K_f ((mg/kg)/(mg/L)^n)	969.41±14.29	925.81±25.10	911.56±20.68	853.28±8.76
RC	n	0.71±0.020	0.76±0.036	0.78±0.032	0.80±0.014
	R^2	0.996	0.989	0.993	0.9998
0.5%	K_f ((mg/kg)/(mg/L)^n)	106.75±0.92	96.93±0.95	96.39±1.30	93.08±0.74
FC	n	0.91±0.010	0.94±0.011	0.97±0.016	0.98±0.0092
	R^2	0.999	0.999	0.999	0.9997
2.0%	K_f ((mg/kg)/(mg/L)^n)	147.69±1.49	138.88±1.45	136.49±2.44	129.47±1.32
FC	n	0.88±0.013	0.89±0.013	0.95±0.024	0.96±0.015
	R^2	0.999	0.999	0.998	0.999
5.0%	K_f ((mg/kg)/(mg/L)^n)	190.73±2.79	185.22±3.21	163.95±2.31	148.97±1.22
FC	n	0.87±0.020	0.88±0.024	0.91±0.020	0.94±0.011
	R^2	0.998	0.997	0.998	0.999

S: sediment; RC: rice straw black carbon; FC: fly ash black carbon.

Table 2 – Parameters of sorption of NP on sediment-BC systems fitted by Dual-mode model.

Absorbent	Parameters	7 days	21 days	63 days	120 days
S	K_{om} (L/kg)	85.97±3.94	83.05±0.97	83.11±2.44	72.18±1.38
	$Q_{max,D}$ (mg/kg)	7.44±8.61	1.49±2.02	−0.96±5.13	1.05±3.19
	R^2	0.990	0.999	0.996	0.999
0.5%	K_{om} (L/kg)	106.84±3.42	118.79±12.33	123.87±4.07	99.70±1.98
RC	$Q_{max,D}$ (mg/kg)	43.41±6.86	13.81±23.42	7.27±7.35	2.46±4.12
	R^2	0.995	0.958	0.996	0.998
2.0%	K_{om} (L/kg)	216.87±21.88	217.62±22.27	255.78±6.59	235.62±4.96
RC	$Q_{max,D}$ (mg/kg)	201.60±39.14	174.04±45.25	73.38±11.52	62.95±8.32
	R^2	0.961	0.950	0.998	0.998
5.0%	K_{om} (L/kg)	571.68±14.27	567.90±57.96	646.24±40.31	635.04±18.27
RC	$Q_{max,D}$ (mg/kg)	446.91±23.48	420.39±107.96	256.59±75.32	221.52±27.81
	R^2	0.998	0.950	0.985	0.996
0.5%	K_{om} (L/kg)	93.42±1.37	87.79±1.55	87.13±1.49	89.01±1.22
FC	$Q_{max,D}$ (mg/kg)	12.98±2.94	9.24±3.36	8.28±3.43	5.05±2.25
	R^2	0.999	0.998	0.999	0.999
2.0%	K_{om} (L/kg)	120.38±3.71	119.29±1.57	116.18±3.96	120.79±0.55
FC	$Q_{max,D}$ (mg/kg)	30.85±7.26	17.63±3.07	15.38±6.91	8.75±1.00
	R^2	0.995	0.999	0.994	0.999
5.0%	K_{om} (L/kg)	156.05±6.33	153.56±7.84	140.72±2.73	135.51±3.31
FC	$Q_{max,D}$ (mg/kg)	34.54±12.28	32.33±14.15	26.26±4.74	13.95±5.91
	R^2	0.993	0.987	0.998	0.998

S: sediment; RC: rice straw black carbon; FC: fly ash black carbon.

ear adsorption of HOCs and hysteretic desorption (Huang and Weber, 1998; LeBoeuf and Weber, 1997). Based on these properties of organic matter, the dual-mode model proposed by Xing and Pignatello (1997) can well explain the two adsorption mechanisms (linear distribution and nonlinear adsorption) of HOCs adsorption behaviour in BC-sediment systems. In this study, the dual-mode model was used to analyse and investigate the changes in the adsorption performance of BC during ageing (Table 2).

Fig. 4 compares the changes in K_{om} and $Q_{max,D}$ in the two aged BC-sediment systems with ageing time. The value of K_{om} changed slightly, but the value of $Q_{max,D}$ decreased significantly with ageing time, indicating that other coexisting substances in the sediment affected the nonlinear adsorption sites of BC but had less of an influence on linearly distributed sites when BC was added to the sediment. Our previous research showed that blocking of the pores of BC is the dominant mechanism of nonlinear adsorption for NP in BC-sediment

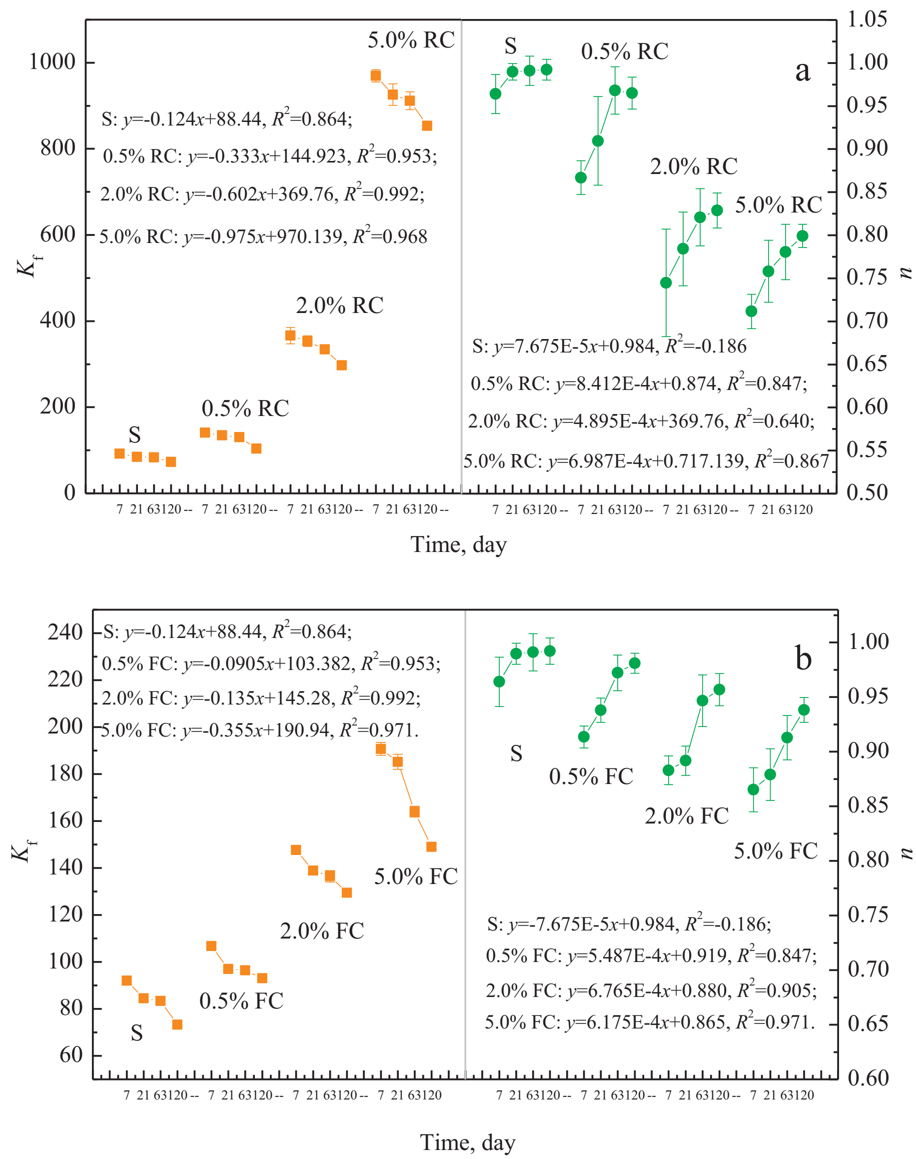

Fig. 3 – Chang of sorption parameters fitted by Freundlich model of NP on aged BC-sediment system with ageing time. (a): RC-sediment; (b): FC-sediment system.

systems (Lou et al., 2014). Therefore, the ageing process of the two BC-sediment systems reduces the value of $Q_{max, D}$, which indicates that the occupation and blockage of micropores are the main reasons for BC ageing in sediments. Moreover, the decline rate of $Q_{max, D}$ in the RC-sediment system was steeper than that in the FC-sediment system. As the proportion of BC increased, the decline rate of $Q_{max, D}$ in both aged BC-sediment systems decreased.

For example, the value of $Q_{max, D}$ in the RC-sediment system (5% mass proportion) decreased by 50.43% when the ageing time increased from 7 to 120 days, but the values decreased by 94.33% and 68.77% in systems that contained 0.5% and 2.0% mass proportions of RC, respectively. This result occurred because most of the micropores in the system exist within the BC, and although more micropores will likely be exposed to the sediment when the BC content in the sediment increases, only a small number of surface pore sites can be occupied; these conditions also indicate that 120 days are not long enough to allow coexisting substances (dissolved organic matter, background pollutants, minerals, etc.) in the sediment to diffuse into the internal pores of BC, which indicates that the coexisting substances can only block the entrances of micropores.

2.1.2. *Establishment of a model to describe the changes in adsorption parameters with BC addition and ageing time*

As mentioned above, linear regression was used to fit the four parameters of Freundlich and dual-mode models with respect to ageing time and BC content. According to the fitting results, the parameters fitted by the Freundlich and dual-mode models in both BC-sediment systems showed a good linear trend with the ageing time. Hence, further analysis was performed to discover the relationship among the fitting parameters, ageing time and BC content.

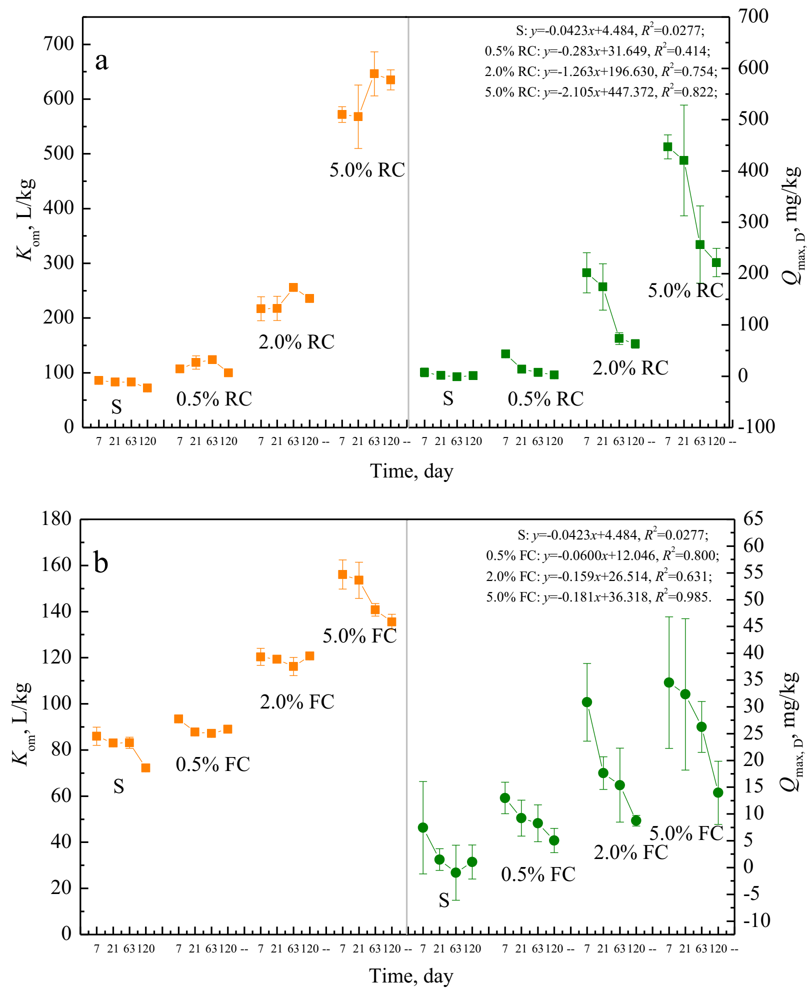

Fig. 4 – Chang of sorption parameters fitted by dual-mode model of NP on aged BC-sediment system with ageing time. (a): RC-sediment; (b): FC-sediment system.

Linear fitting of parameters with respect to ageing time:

$$P = A \times T + B \quad (4)$$

P represents the fitting parameters of Freundlich and dual-mode models. P= K_f, n, K_{om} or $Q_{max,\,D}$. A is the slope of the fitting parameters with respect to ageing time, and B is the intercept. T is the ageing time.

As shown in Tables 1 and 2, Figs. 3 and 4, A and B will change with BC content (C: RC or FC). In addition, when P=K_{om}, A = 0 because there is no significant difference between K_{om} values at different ageing times. Based on this relationship, we can obtain:

$$A = M_A \times C + N_A \quad (5)$$

$$B = M_B \times C + N_B \quad (6)$$

M is the slope of A with respect to BC content, and B is the intercept.

Bringing Eqs. (5) and (6) into Eq. (4) yields the following:

$$P = (M_a \times C + N_a) \times t + (M_b \times C + N_b) \quad (7)$$

For the FM, P= K_f and n.

$$K_f = (M_{A1} \times C + A_{A1}) \times T + (M_{B1} \times C + N_{B1})$$

$$n = (M_{A2} \times C + N_{A2}) \times T + (M_{M2} \times C + N_{B2})$$

K_f and n can be substituted into Eq. (1). Therefore, Freundlich model can be expressed as:

$$Q_e = [(M_{A1} \times C + N_{A1}) \times T + (M_{B1} \times C + N_{B1})] \times {C_e}^{[(MA2\times C+NA2)\times T+(MB2\times C+NB2)]} \quad (8)$$

For dual-mode model, P = K_{om} and $Q_{max,D}$.

$$K_{om} = (M_{B3} \times C + N_{B3})$$

$$Q_{max,D} = (M_{A4} \times C + N_{A4}) \times T + (M_{B4} \times C + N_{B4})$$

K_{om} and $Q_{max,\,D}$ can be substituted into Eq. (3). Thus, dual-mode model can be expressed as:

$$Q_T = (M_{B3} \times C + N_{B3}) \times C_e + [(M_{A4} \times C + N_{A4}) \times T + (M_{B4} \times C + N_{B4})] \quad (9)$$

Eqs. (8) and (9) show that the only variables are BC content (C) and ageing time (T). Therefore, in theory, the adsorption

Table 3 – The expressions of parameters fitted by FM and the DM.

		Expression
RC	K_f	[(−16.012±2.269) × C-(0.208±0.0614)] × T+[(17862.125±1052.147) × C+(58.401±28.452)]
	n	[(−0.00317±0.00124) × C+(8.570E-4 ± 1.026E-5)] × T+[(−4.672±1.656) × C+(0.925±0.0448)]
	K_{om}	(8599.076±613.448) × C+(76.864±3.112)
	$Q_{max,D}$	[(−41.268±6.058) × C-(0.150±0.164)] × T+[(9025.128±395.142) × C+(0.813±10.685)]
FC	K_f	[(−5.065±1.253) × C-(0.0812±0.0339)] × Tt+[(2020.558±233.924) × C+(94.125±6.326)]
	n	[(−0.00197±0.00) × C+(7.158E-4 ± 0.00)] × T+[(−1.946±0.946) × C+(0.948±0.0256)]
	K_{om}	(1633.553±154.289) × C+(83.458±2.592)
	$Q_{max,D}$	[(−2.790±0.944) × C-(0.0583±0.0255)] × T+[(604.690±137.334) × C+(8.503±3.714)]

C: carbon; P: parameters; RC: rice straw black carbon; FC: fly ash black carbon.

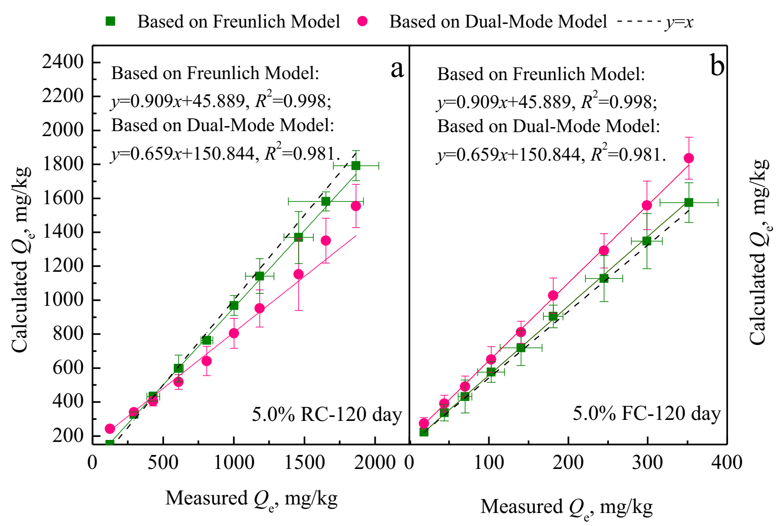

Fig. 5 – Correlations between observed and calculated Q_e in 5% BC-sediment system after 120-day ageing. (a): RC- sediment system; (b): FC- sediment system.

capacity for NP can be predicted by the mass proportion of BC and the ageing time of the BC-sediment system. The values of A and B can be directly obtained by linear fitting of the experimental data, as shown in Figs. 3 and 4. All parameter values are shown in Table 3.

We then verified the above model by setting the ageing time to 120 days (t = 120 days) and BC mass proportion to 5.0% (RC or FC) and then compared the correlation between the measured and model-calculated adsorption capacities, as shown in Fig. 5. The results showed that there was a significant linear correlation between the measured and model-calculated Q_e values in the two aged BC-sediment systems. Compared with the results obtained with the dual-mode model, the Q_e linear curve calculated by Eq. (8) for the two systems is closer to the actual measured linear curve $y = x$. This result shows that in aged BC-sediment systems with different BC contents, the newly established model based on the fitting parameters of Freundlich model can predict the adsorption capacity for NP more accurately. Hence, the adsorption capacity of NP in BC-sediment systems can be predicted from the known BC mass proportion and ageing time when BC is applied for organic pollution control, which can avoid tedious experimental processes. However, the models established in this study only work for RC and FC, not for other BCs. Therefore, further studies are needed, such as analysis of the correlations between adsorption capacity and ageing time and between adsorption capacity and the pore volume or specific surface area of BC, and the results are expected to be applicable to more forms of BC.

3. Conclusion

The ageing process will decrease the adsorption capacity and the performance of BC when applied for sediment pollution control. How the ageing process and BC content affect the adsorption capacity of two BC-sediment systems was modelled and quantitatively investigated in this study. The results showed that the ageing process decreased the adsorption capacity of BC-sediment systems due to the blockage of non-linear adsorption sites of BC by coexisting materials in the sediment during ageing. Furthermore, the RC-sediment system was more efficient for NP pollution control than the FC-sediment system. The established model appropriately fits the experimental data and yields an acceptable prediction, especially for the model based on parameters from Freundlich model. However, this model is limited to only the two kinds of BC in this study. To fully reflect the influences of the ageing process on BC-sediment systems and make more precise predictions, it is recommended that future work considering more factors and conditions, such as modelling of the correlation between adsorption capacity and the pore volume or specific surface area of BC, be applied to build an accurate and sound model.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.09.008.

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