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Adsorption of 17 α-ethyl estradiol with the competition of bisphenol A on the marine sediment of Hong Kong

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Abstract:

The present experimental study was to characterize the adsorption behavior of 17 α-ethyl estradiol (EE2) onto marine sediment in both the single and binary solute systems. Stepwise spiking was innovatively performed to better understand the competition effects. Adsorption of EE2 on the marine sediment can be well fitted by the Freundlich model with an affinity coefficient (K_F) varying from 15.8 to 39.8 L/kg. It was significantly influenced by SOM and the particle properties. Co-presence of BPA brought about a significant competition effect on the adsorption of EE2, leading to a reduced EE2 adsorption. The competitive effect imposed by EE2 to BPA, however, was even more serious owing probably to the large molecular structure and high hydrophobicity of EE2. The sediment sample with the highest SOM and SSA presented a mild competition effect, while the sediment with the lowest SOM and largest particle size exhibited the most serious competition effect.

Key words: 17 α-ethyl estradiol (EE2); bisphenol A (BPA); Hong Kong marine sediment; adsorption; competitive adsorption

1. Introduction

Endocrine disrupting chemicals (EDCs) have attracted growing attentions from the academic community and general public in recent years, for their chronic harm to the reproduction of organisms in the ecosystem (Colborn et al., 1993; Collins, 2008). 17 α-ethyl estradiol (EE2) is one of common EDCs found in the aquatic environment. EE2 is generally used in our daily life as contraceptive pills. Owing to its long half-life in living organisms such as fish, the toxicity of EE2 was reported to be significantly greater than many other similar hormones, e.g. estrone and β-estradiol (Li et al., 2014). Bisphenol A (BPA) is another typical EDC that had been widely employed previously as a plasticizer in industrial production. Due to releases from various sources and insufficient removal from wastewater, EE2, BPA and other EDCs have entered rivers, lakes, soils and marine waters (Li et al., 2013b). Occurrence of EE2 in high abundance has been found in surface water (Peng et al., 2008), agricultural soil (Azzouz and Ballesteros, 2012), river and coastal sediment (Azzouz and Ballesteros, 2012; Zhang et al., 2009).

For hydrophobic compounds such as EE2, strong adsorption onto soil or sediment is expected to greatly affect their fate and transport in the environment (Sun et al., 2012; Voice and Weber, 1983). However, compared to the numerous studies on the occurrences and toxicities of EE2 and other EDCs, limited research has been conducted on their adsorption behavior, especially in the marine water-sediment system. Recently, adsorption of EE2 has been reported on engineered materials such as biochars (Sun et al., 2011) and carbon nanotubes (Pan et al., 2008), as well as on natural solids including minerals (Shareef et al., 2006) and soil samples (Li et al., 2013a; Sun et al., 2012). According to the experimental studies, the properties of the absorbents, including the particle size, surface feature and the organic matter content, all affect the adsorption of EE2. Most of these investigations, however, were focused on the adsorption of EE2 only under the single-solute condition, which is rather different from the multi-solute system in the natural environment. Compared to single-solute EE2 adsorption, multi-solute adsorption with the competition of other EDCs remains to be investigated. Pan and Xing (2010) reported the competitive adsorption of BPA and EE2 on carbon nanomaterials. Similar tests on the co-adsorption of BPA and EE2 were conducted by Li et al. (2013b) on soils and Yu and Huang (2005) and Li et al. (2014) on river sediments. In general, research on the competitive adsorption of EDCs on natural materials is largely limited. There is no report of experimental studies on the adsorption of EE2, either individually or competitively, in marine sediment.

In the present study, the adsorption behavior of EE2 on marine sediment was investigated in both single and binary solute systems. Natural marine sediment samples with varied particle properties and organic matter contents were collected for the experimental studies. To further illustrate the competitive effects, the tests were conducted not only in the way of simultaneous spiking of the two solutes (EE2 and BPA) but also by the approach of stepwise spiking of the competing agent and the target compound. The experimental findings are important to the better understanding of (1) the influences of the marine sediment properties on the adsorption of EE2 and (2) the competitive effect imposed by BPA to EE2 or EE2 to BPA during the sediment adsorption in the binary-solute system.

2. Materials and methods

2.1. Chemicals

Pure EE2 and BPA used in the present study were of analytical grade provided by Sigma-Aldrich. The main physicochemical properties of EE2 and BPA are given in Table 1. EE2 are more hydrophobic than BPA, with a lower water solubility (S_w) and higher octanol-water partition coefficient (K_{ow}). Other reagents and solvents, including hydrochloric acid (HCl), hydrogen peroxide (H_2O_2), calcium chloride ($CaCl_2$), sodium azide (NaN_3), and acetonitrile, used in the experimental study were of analytical grade purchased from Sigma-Aldrich. The stock solution of either EE2 or BPA was firstly prepared in methanol, which was then diluted to working concentrations before use. The organic solvent in the working solutions after purged by pure N_2 was less than 0.1% to avoid co-solvent effects (Plaza et al., 2009).

2.2. Marine sediment

Natural marine sediment samples were collected from 5 sites in Victoria Harbour, Hong Kong, marked as #1-#5. The latitudes/longitudes of sites #1-#5 are 22°14.800/114°16.000, 22°17.500/114°13.500, 22°17.470/114°11.180, 22°18.400/114°06.500, and 22°18.142/114°03.051, respectively (Fig. 1). The sediment samples were collected from the top 30 cm surface of the sediment by Peterson grab. After shells and gravels were removed, the sediment was stored below 4°C in refrigerator and transferred to laboratory as soon as possible before the experimental use. the sediments were air-dried, homogenized and then ground to pass through 60-mesh sieve (Xu et al., 2008). The sediment samples from different sites were expected to have different organic contents and particle sizes distribution, as site #1 is closer to the mouth of Pearl River on its left, #5 is closer to the open ocean to its right, and site #4 is at the middle of Victoria Harbour that receives stormwater runoffs from the urban areas of both sides of the Harbour (Fig. 1).

The fraction of organic matter in the sediment (f_{OM}), or sediment organic matter (SOM), was measured by the means of ignition at 550°C (Beaudoin, 2003; Fei et al., 2011). To determine the content of black carbon in sediment (f_{BC}), the raw sediment was treated first by HCl and then by H_2O_2 to remove the carbonates and labile organic matter, respectively. The organic residual in the treated sediment was then measured and its fraction was recorded as f_{BC} (Xu et al., 2008; Zhao et al., 2001). The particle size distributions of the sediment samples were measured by a particle size analyzer (Coulter LS 13320, Beckman, USA). The specific surface area (SSA) was determined for the sediments by a surface analyzer (Coulter SA 3100, Beckman, USA) using the multi-point Brunauer–Emmett–Teller (BET) nitrogen adsorption method. The physicochemical characteristics of the marine sediment samples are summarized in Table 2.

2.3. Adsorption of EE2 on the marine sediments

Tests on the adsorption of EE2 by the 5 sediments were conducted following the batch equilibration approach (OECD, 2000). The laboratory tests on the adsorption isotherms were performed using 12-ml amber glass screw-cap vials (minimal headspace) with Teflon-lined septa (Xing and Pan, 2010; Xing and Pignatello, 1997). Briefly, for each sediment sample, the sediment was placed in a series of vials, each with 0.4 g dry sediment. The vials were filled with 10 mL of the background solution containing 0.01M $CaCl_2$ for the basic ionic strength and 200 mg/L NaN_3 for microbial inhibition (Xing and Pan, 2010). EE2 solution was then added into the vials to different concentrations from 0.5 to 8.0 mg/L. If not stated otherwise, the pH of the solutions was 7.2 for the sediment adsorption tests.

The sealed vials were placed in a temperature-controlled shaking incubator (Polyscience, USA) for adsorption at 25^{o}C. According to the preliminary tests, the equilibrium of EE2 adsorption by the sediment could be reached within 24 hrs. Upon completion of the adsorption tests, the sediment mixtures from the test vials were centrifuged to remove the solids. The EE2 concentrations in the aqueous phase were measured using a high-performance liquid chromatography (HPLC). Concentration loss caused by adsorption of EE2 to the glass walls or by other reasons was found to be less than 3%. Thus, from the difference between the initial and final EE2 concentrations in the aqueous phase, the amount of EE2 adsorbed by the sediment in each test vial could be calculated. The results from a series of initial concentrations were then used to determine the adsorption isotherm of EE2 for each sediment sample.

2.4. Adsorption of EE2 by the sediment with the competition of BPA

The EE2 adsorption experiments also were conducted in the presence of BPA as a competitor chemical. The batch isotherm tests followed the similar procedure as described above, except for the extension of the equilibration time to 2 days. As a competing compound, BPA was dosed at 40 mg/L (about 10% of its solubility in water). The competitive adsorption tests were carried out in 3 sub-groups: (1) adsorption of EE2 only as the reference, (2) synchronous competitive adsorption of EE2 and BPA when both chemicals were simultaneously spiked initially, and (3) lag-competitive adsorption which had EE2 spiked 1 day after BPA to have the competing agent adsorbed onto the sediment before the competition. Upon the completion of the tests, both EE2 and BPA were analyzed for their equilibrium concentrations in the aqueous phase.

In addition, the effect of the EE2 presence on the adsorption of BPA also was investigated. For the tests, EE2 as a competitor had a fixed concentration at 3 mg/L (about 10% of its solubility in water), while the BPA concentration varied from 5 to 50 mg/L for adsorption by the sediment. Similarly, the competitive adsorption tests were conducted in 3 sub-groups: (1) adsorption of BPA alone as the reference, (2) synchronous competitive adsorption of BPA and EE2, and (3) lag-competitive adsorption which had BPA spiked 1 day after EE2. The equilibrium concentrations of BPA and EE2 in the aqueous phase were measured after the adsorption tests.

2.5. Chemical analysis

The EE2 and BPA concentrations in water were measured by an HPLC (Waters 2695) with a C18 column (5 μm, 2.1×150 mm) for separation and a photodiode array detector (Waters 2996) for chemical detection and quantification. The mobile phase was a mixture of Milli-Q water and acetonitrile (50:50, v/v). The flow rate was set at 0.5 ml/min, and the sample injection volume was 50 μL. Under this chromatographic condition, EE2 and BPA could be separated and detected at around 6 and 8 min, respectively. The areas of peaks at the wavelength of 225 nm were used for quantification, and the limits of BPA and EE2 quantifications were 0.02 mg/L and 0.1 mg/L, respectively.

2.6. Data analysis

The isotherm data of sediment adsorption of EE2 were arranged to fit for typical adsorption models, including the linear partition model, Freundlich model, and Langmuir model (Eq. 1-3) (Voice and Weber, 1983; Xing et al., 2011), i.e.,

$$q_e = K_d C_e \tag{1}$$

$$q_e = K_F C_e^{1/n} \tag{2}$$

$$q_e = \frac{k q_{\max} C_e}{1 + k C_e} \tag{3}$$

where C_e and q_e are the equilibrium concentrations of the chemical in the aqueous phase and sediment, respectively; K_d is the partition coefficient in the linear model; K_F and $1/n$ are the affinity coefficient and curvature index, respectively, in the Freundlich equation; and k and q_{max} are the equilibration parameter and the adsorption capacity, respectively, for the Langmuir model.

Furthermore, for some isotherms, K_d was normalized by the SOM content (f_{OM}) to obtain the organic normalized adsorption coefficient K_{OM} (Eq. 4), i.e.,

$$K_{OM} = \frac{K_d}{f_{OM}} \tag{4}$$

Microsoft Excel 2010 and SPSS 13.0 were employed for data analysis and Origin 8 was used for graph plotting.

3. Results and discussion

3.1. Adsorption of EE2 on marine sediments

The adsorption isotherms for EE2 on the marine sediments are presented in Fig. 2, and the parameters of model fittings are summarized in Table 3. According to the comparison between the R^2 values, the isotherms of EE2 adsorption in the concentration range tested on the marine sediments can be better fitted by the Freundlich model and the linear model than the Langmuir model. The partition coefficient, K_d, from the linear model varied between 9.2 to 23.9 L/kg, and the affinity coefficient, K_F, from the Freundlich model ranged between 15.8 to 39.8 L/kg for EE2 adsorption. The Freundlich adsorption isotherms had a curvature index ($1/n$) that varied in a range of 0.5-0.8 (<1). This may suggest that the adsorption would become saturated when the aqueous EE2 concentration increased (Xing et al., 2011).

Despite of the variations, the adsorption capacity of the marine sediment in the present study is generally lower compared to that reported in the literature. EE2 is considered to be a moderately to strongly adsorbed chemical compound by soil and sediment, with a K_d value ranging from 2.33 to 339 L/kg (Chen et al., 2012; Pan et al., 2009; Sun et al., 2012). Extremely high adsorptive capacity usually occurred for the sediment or soil samples with a very high organic carbon content (f_{oc}) or SSA (Sun et al., 2012). The adsorption performance of the marine sediment in the present study is close to that of the soil sample collected from Indiana, USA, with similar K_d (2.33-23.4 L/kg), f_{oc} (0.22-2.91%), and SSA (6.4-7.3 m^2/g) values (Lee et al., 2003).

Among the 5 sediment samples, sediment #1 exhibited the highest EE2 adsorption and #5 sediment the lowest (Table 3). This may be attributed to the fact that sediment #1 was of smallest particle sizes with a high silt fraction according to the particle size distribution, while #5 was of larger particles with a high fraction of sand (Table 2). More importantly, sediment #1 had the second most f_{OM} and highest SSA, while #5 had the least f_{OM} and the lowest SSA. As shown in Fig. 1, site #1 is close to the mouth of Pearl River, with fine particles and a high organic content in the sediment, in comparison to site #5 next to the open ocean, with coarse sand and a lower organic fraction in the sediment. The adsorption capacity (e.g., K_F) of the marine sediments tested in the present study can be well correlated with the

f_{OM} or SSA values (Fig. 3A), which is in general agreement with previous studies on the adsorption of organic pollutants by soil and sediment (Chiou et al., 1979; Karickhoff et al., 1979; Sun et al., 2012). Along with a 80% f_{OM} increase in the sediment, K_F increased by a factor of more than 2.5, demonstrating the important role of SOM in EE2 adsorption.

The K_{OM} values, i.e. the normalized K_F by f_{OM}, for EE2 adsorption by the 5 sediment samples varied from 212 to 347 L/kg-SOM (Table 3). There is no direct correlation found between K_{OM} and f_{OM}, or between K_{OM} and f_{BC}. Sediment from site #1 had a much higher K_{OM} than other sediment samples which apparently is related to the highest SSA found in sediment #1. Besides the SOM factor, the surface properties of the sediment would also affect its adsorption capacity (Sun et al., 2012; Xing et al., 2011). As shown in Fig. 3B, when the SSA increased by 155% from sediment #5 to #1, the K_F value increased by 152%, suggesting a correlation between K_F and SSA for EE2 adsorption.

3.2. Influence of BPA on EE2 adsorption

The adsorption isotherms and model-fitting results for the binary-solute adsorption by the marine sediments are summarized in Fig. 4 and Table 4. Adsorption of EE2 in the concentration range tested can be fitted by the linear model and the Freundlich model, either with or without BPA. As a phenomenon of competitive adsorption (Li et al., 2013b), the presence of BPA in the binary-solute system affected the adsorption of EE2. With the simultaneous existence of BPA, K_d of EE2 in the linear partition model decreased by 7-18%, and K_F of the Freundlich model decreased by 13-23%.

A similar competitive effect was observed for EE2 adsorption when BPA was spiked before EE2, especially for sediments #1 and #3 (Table 4). For sediment #5, the competition caused by the prior presence of BPA was somewhat stronger, as suggested by the further decrease of K_d and K_F compared to the co-existence tests. The different observations among the three sediment samples might be mainly attributed to their different properties, such as the pore structure, active sites on surface, as well as the SOM composition, i.e. the rubbery domain and glassy domain contents. Based on the dual model for organic adsorption (Xing and Pignatello, 1997; Xing et al., 2011), sorption by SOM usually includes unspecific partitions in the amorphous rubbery domain and site specific adsorption in the glassy domain. As Pan and Xing (2010) proclaimed that, owing to its unique butterfly structure and small molecular size, BPA has the advantage in wedging into small pores and high-energy sites in the adsorbent, that is, in the glassy domain represented by the BC content (Weber et al., 2002). Thus, when the sediment had fine pore structures with abundant small pores that EE2 cannot utilize, or with a high f_{BC} which provides specific sites for BPA, the adsorption would be less affected by the competition. In the present study, sediment #5 was composed of large sediment particles with the lowest f_{OM}, and its f_{BC} was only 53% and 60% of that of #1 and #3, respectively (Table 2). Thus, the adsorption by the glassy domain of sediment #5 was rather limited compared to the other two sediment samples. Moreover, whilst the ‘buffering’ by small pores was inconspicuous, sediment #5 contained much less fine clay particles with the lowest SSA. Thus, all of these led to a more notable competition effect of BPA on EE2 adsorption of by sediment #5.

3.3. *Influence of EE2 on BPA adsorption*

Similar to EE2, the isotherms of BPA adsorption by the sediments are fitted well with the Freundlich model (Table 4). The non-linear isotherms, as shown in Fig. 5, had the curvature index values that are all significantly lower than 1. With the simultaneous presence of EE2, the adsorption of BPA was hindered by 9-14% according to the decrease of K_d, or by 2-29% based on the decrease of K_F. The change of $1/n$ in the Freundlich model suggests that the linearity of BPA adsorption isotherms slightly increased with the EE2 competition.

As for the tests with the prior EE2 spiking into the sediment before the BPA addition and adsorption, the competition effect appeared to be more significant (Fig. 5). The decreases of K_d and K_F for BPA were 11-28% and 15-30%, respectively, which are considerably higher than the changes obtained for the simultaneous competition tests. Moreover, the competition effect of EE2 on BPA adsorption was more than that BPA imposed to EE2. As far as the binding forces are concerned, hydrophobicity played a crucial role in the adsorption of non-ionic organic pollutants (Voice and Weber, 1983; Xing et al., 2011). Stronger competitions have been found when the competitors were more hydrophobic than the solutes of interest (Yu and Huang, 2005). Additionally, a larger molecular size would also facilitate the competition, as the molecules would take up more surface sites and/or block small pores. Thus, with a larger size and higher hydrophobicity, EE2 brought about a more significant competition effect on BPA adsorption by the sediment than BPA did on EE2. Nonetheless, the competition effect caused by EE2 to BPA could still vary between different sediment samples. For sediment #1, which had the highest f_{OM}, especially f_{BC}, and the highest SSA compared to the other sediments, the adsorption of BPA appeared to be much less affected by the EE2 competition.

4. Conclusions

- Single-solute adsorption of EE2 on the marine sediment can be well described by the Freundlich model, and the adsorption behavior of EE2 was affected by both the SOM content and the surface properties of the sediment.
- In the binary-solute system of EE2 and BPA, a competition effect was observed on the adsorption for either EE2 or BPA, as sediment adsorption of either chemical would be suppressed by the co-existence of the other one. Prior spiking of the competitor chemical would bring about a more serious competition as compared to the synchronous spiking.

- The competition between BPA and EE2 for adsorption could be reduced for the sediment with a high black carbon content and fine-pore structure. The sediment sample with the highest f_{BC} and SSA displayed the mildest competition effect during the binary-solute adsorption, while the sediment with the lowest f_{BC} and larger particle sizes exhibited the most serious competition effect.

Acknowledgments

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Table captions:

Table 1. Chemical properties of the selected EDCs: BPA and EE2.

Table 2. Physicochemical characteristics of the marine sediment samples.

Table 3. Coefficients fitted for the different isotherm models for the adsorption of EE2 by the marine sediment samples.

Table 4. Coefficients fitted for the isotherm models for the competitive adsorption of EE2 and BPA on the marine sediment samples.

Figure captions:

Fig. 1. Locations of the sediment sampling sites in Victoria Harbour, Hong Kong.

Fig. 2. Single-solute adsorption isotherms of EE2 on the marine sediment.

Fig. 3. Correlations between the adsorption coefficient of EE2 and the sediment properties.

Fig. 4. The competition effect of BPA on the adsorption of EE2 onto the marine sediment.

Fig. 5. The competition effect of EE2 on the adsorption of BPA onto the marine sediment.

Table 1. Chemical properties of the selected EDCs: BPA and EE2 (Sun et al., 2010).

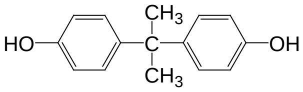
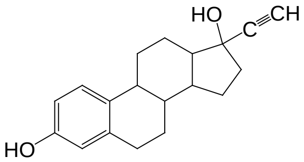
	BPA	EE2
Molecular structure		
Molecular diameter (Å)	4.3	6.0
Water solubility, S_W (mg/L)	380	7.6
Octanol-water distribution coefficient, $logK_{ow}$	2.2	4.2

Table 2. Physicochemical characteristics of the marine sediment samples.

#	SOM (%)			SSA	Particle size distribution (%)		
	f_{OM}	f_{BC}	$\frac{f_{BC}}{f_{OM}}$	(m^2/g)	Sand (> 60 µm)	Silt (2-60 µm)	Clay (< 2 µm)
1	6.9±0.7	4.9±0.4	71	21.2	0.3	93.5	6.2
2	6.7±1.1	4.6±0.6	69	12.4	22.5	77.3	0.2
3	6.4±0.3	4.3±0.4	67	9.2	0.2	98.1	1.7
4	7.7±0.9	5.3±0.5	69	15.2	29.4	70.4	0.2
5	4.2±0.4	2.6±0.3	62	8.3	44.2	55.7	0.1

Table 3. Coefficients fitted for the different isotherm models for the adsorption of EE2 by the marine sediment samples.

#	Linear			Freundlich			Langmuir		
	K_d (L/kg)	R^2	K_{OM} (L/kg-SOM)	K_F (L/kg)	$1/n$	R^2	q_{max} (mg/kg)	k (L/mg)	R^2
1	23.9±1.1[a]	0.97	346.9	39.8±1.0[a]	0.6±0.08[a]	0.97	157.0±18.1[a]	0.3±0.002[a]	0.92
2	14.2±0.7[b]	0.97	212.1	25.1±1.0[b]	0.5±0.04[b]	0.97	98.9±10.4[b]	0.3±0.004[b]	0.92
3	15.5±0.8[c]	0.96	243.0	25.1±1.1[b]	0.7±0.06[c]	0.96	127.4±19.3[c]	0.2±0.005[c]	0.84
4	23.1±0.9[a]	0.94	300.1	31.6±1.1[c]	0.8±0.05[d]	0.97	255.8±59.7[d]	0.1±0.003[d]	0.67
5	9.2±0.7[d]	0.97	217.4	15.8±1.1[d]	0.6±0.04[a]	0.92	66.7±1.8[e]	0.3±0.02[e]	0.70

[*] Different letters in the same column indicate a significant difference in statistics at level of p = 95%.

Table 4. Coefficients fitted for the isotherm models for the competitive adsorption of EE2 and BPA on the marine sediment samples.

#	Adsorbate	Linear model		Freundlich model		
		K_d (L/kg)	R^2	K_F (L/kg)	$1/n$	R^2
1	EE2 (single)	20.6±1.1^{a}	0.99	25.1±0.6^{a}	0.7±0.03^{a}	1.00
	EE2 (with BPA)	17.4±0.8^{b}	0.99	21.9±0.5^{b}	0.7±0.04^{a}	0.99
	EE2 (after BPA)	20.0±0.4^{a}	0.99	21.4±0.5^{c}	0.8±0.03^{b}	0.99
	BPA (single)	4.21±0.1^{a}	0.98	10.9±0.5^{a}	0.7±0.01^{a}	1.00
	BPA (with EE2)	3.82±0.1^{b}	0.99	10.7±0.4^{a}	0.7±0.01^{b}	1.00
	BPA (after EE2)	3.05±0.2^{c}	0.96	9.3±1.0^{b}	0.6±0.03^{c}	0.98
3	EE2 (single)	25.3±1.0^{a}	0.97	30.2±0.6^{a}	0.8±0.02^{a}	0.99
	EE2 (with BPA)	20.6±0.5^{b}	0.99	23.4±0.4^{b}	0.8±0.02^{b}	1.00
	EE2 (after BPA)	22.5±0.5^{c}	0.99	20.9±1.4^{c}	0.8±0.05^{ab}	0.97
	BPA (single)	4.81±0.2^{a}	0.99	11.8±0.6^{a}	0.7±0.02^{a}	1.00
	BPA (with EE2)	4.15±0.1^{a}	0.99	9.6±0.4^{b}	0.8±0.01^{b}	1.00
	BPA (after EE2)	4.29±0.2^{a}	0.99	8.3±0.5^{c}	0.8±0.02^{b}	1.00
5	EE2 (single)	17.9±0.4^{a}	0.99	20.8±0.5^{a}	0.8±0.03^{a}	0.99
	EE2 (with BPA)	16.6±0.5^{b}	0.97	17.0±0.7^{b}	1.0±0.05^{b}	0.99
	EE2 (after BPA)	14.3±0.2^{c}	1.00	14.6±0.3^{c}	1.0±0.03^{b}	1.00

BPA (single)	1.97±0.1^{a}	0.94	6.8±1.0^{a}	0.7±0.04^{a}	0.97
BPA (with EE2)	2.20±0.06^{b}	0.99	4.9±0.3^{b}	0.8±0.02^{b}	1.00
BPA (after EE2)	1.72±0.1^{c}	0.96	4.9±0.7^{b}	0.7±0.04^{a}	0.98

[*] Different letters in the same column in each section indicate a significant difference in statistics at level of p = 95%.

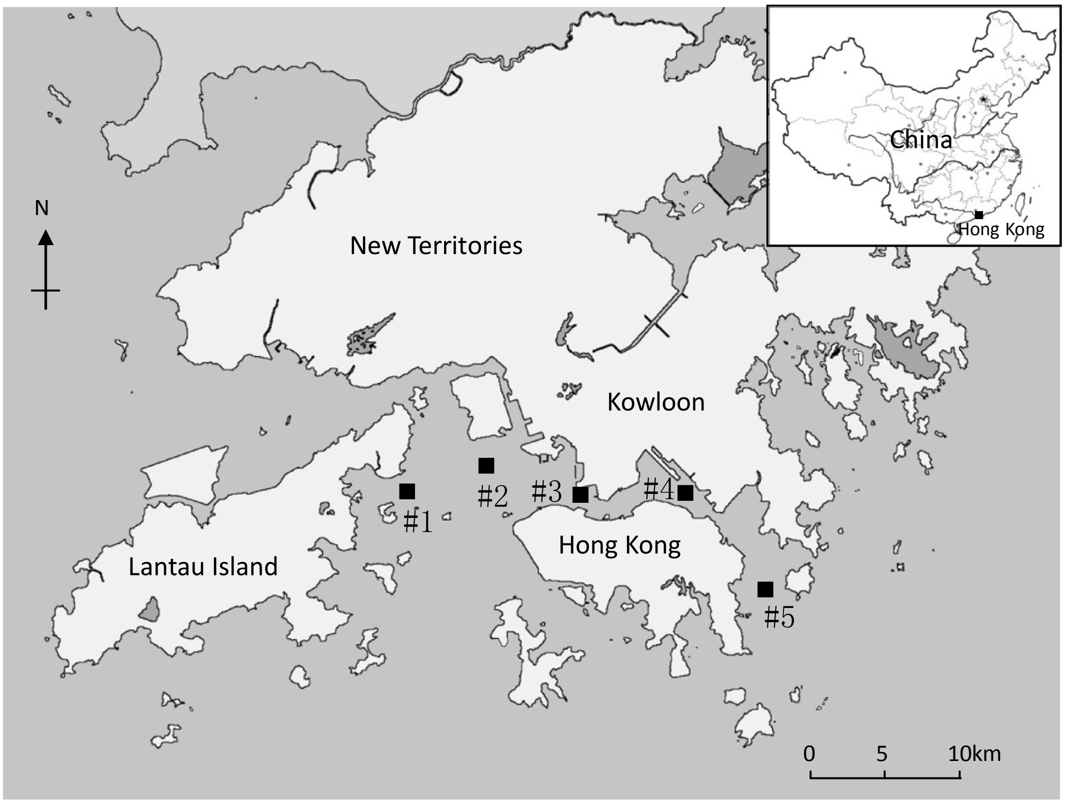


Fig. 1. Locations of the sediment sampling sites in Victoria Harbour, Hong Kong.

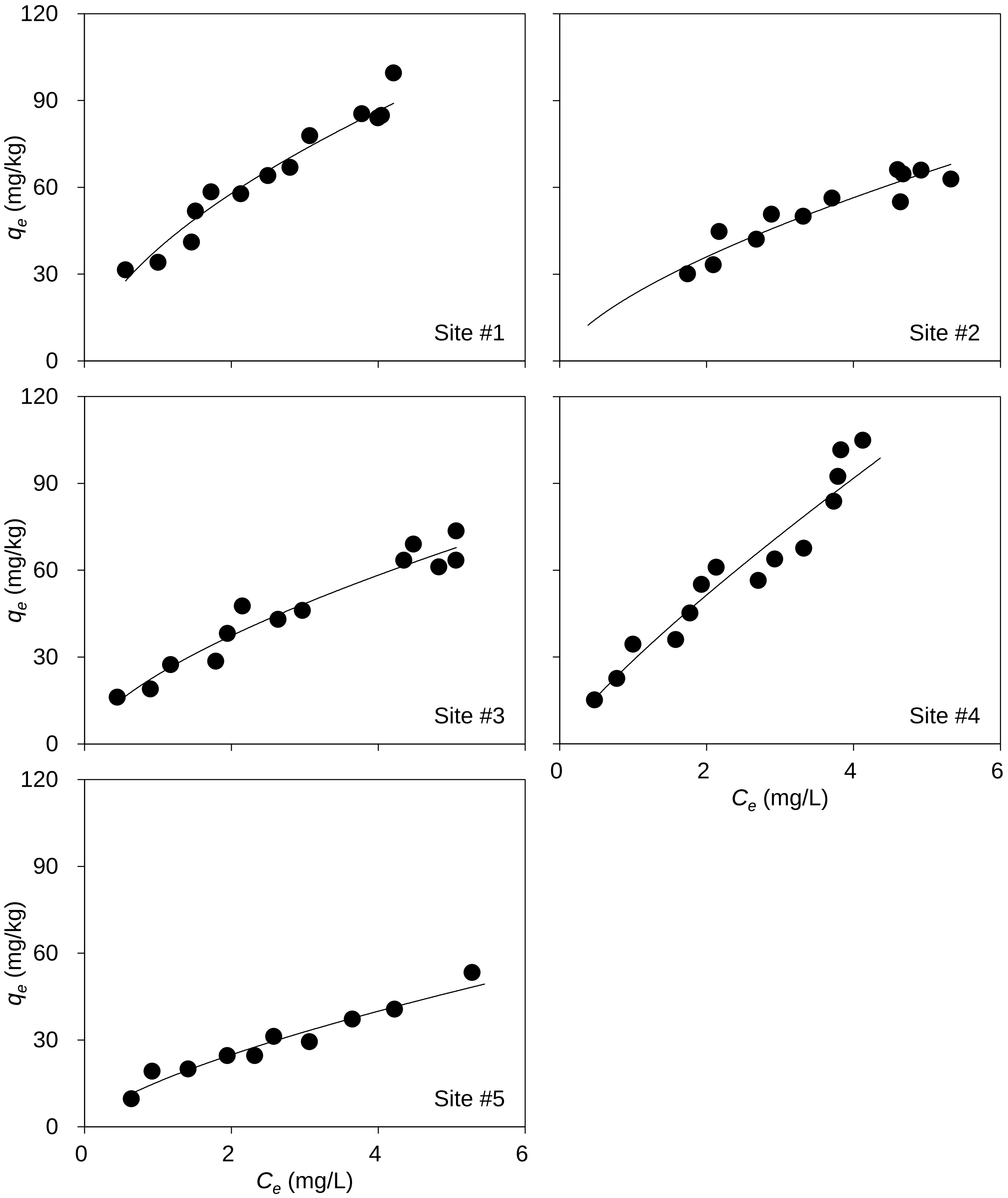


Fig. 2. Single-solute adsorption isotherms of EE2 on the marine sediment.

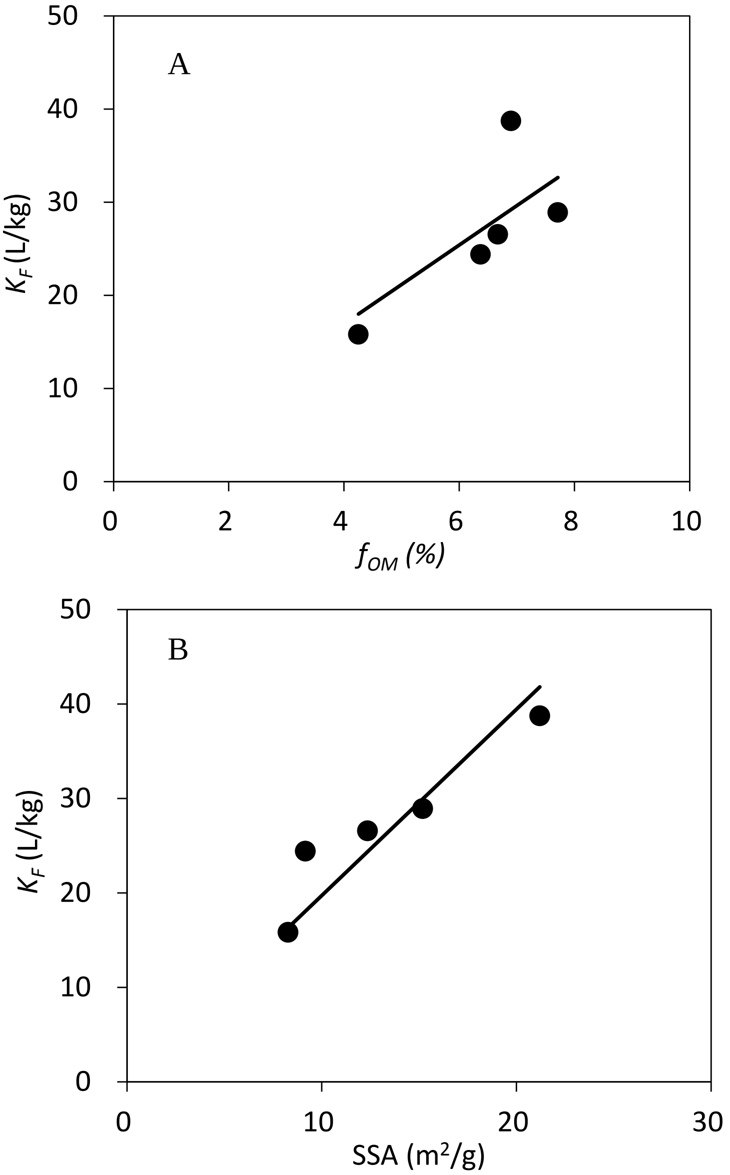


Fig. 3. Correlations between the adsorption coefficient of EE2 and the sediment properties.

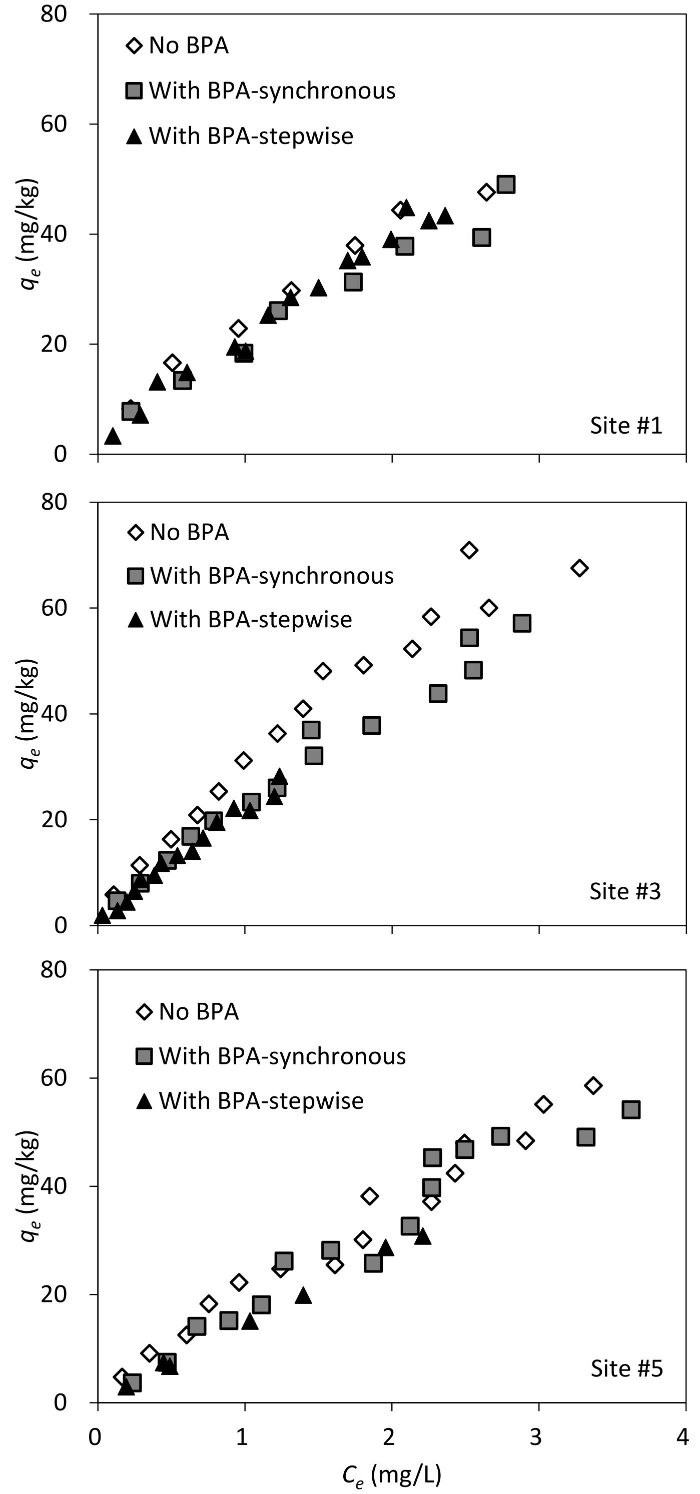


Fig. 4. The competition effect of BPA on the adsorption of EE2 onto the marine sediment.

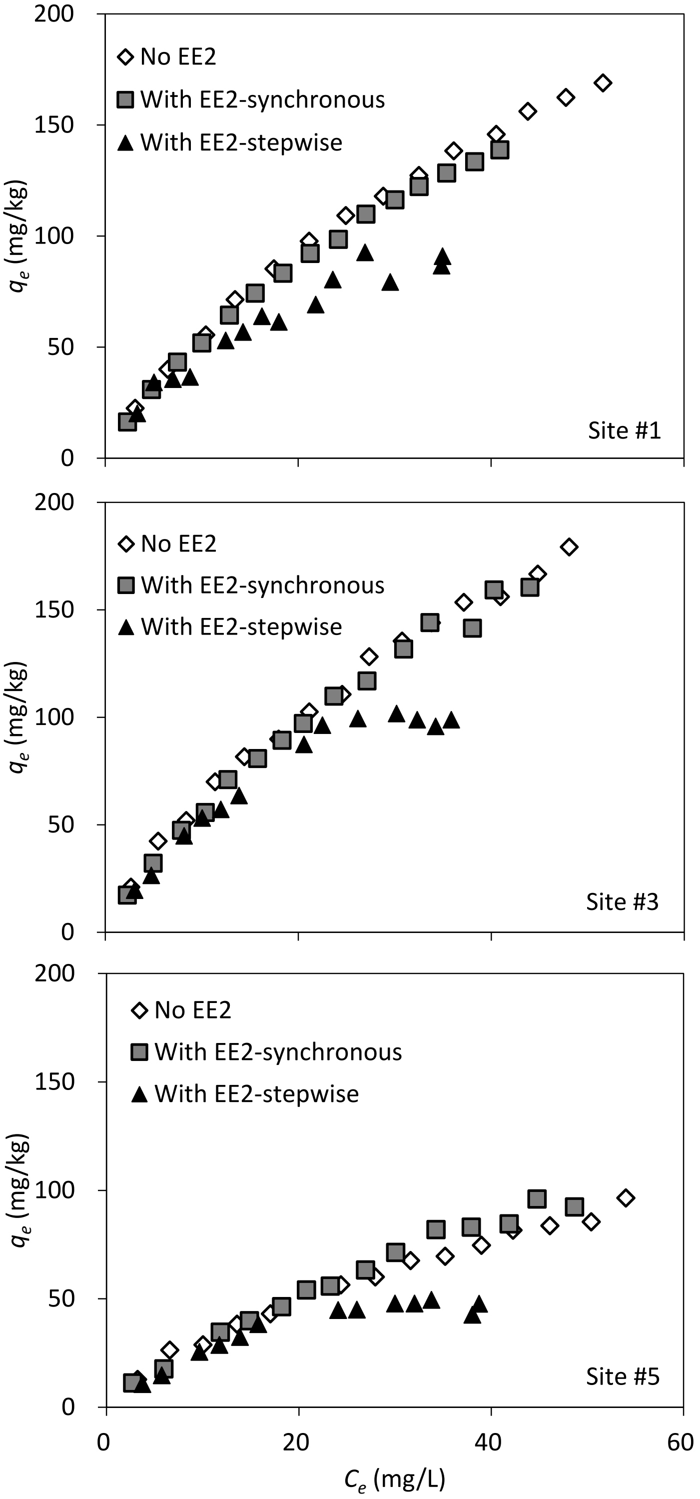


Fig. 5. The competition effect of EE2 on the adsorption of BPA onto the marine sediment.