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Influence of the coexisting contaminants on bisphenol A sorption and desorption in soil

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Abstract

The effects of different heavy metals (Cd, Pb), surfactants (cetyltrimethylammonium bromide (CTAB), cetylpyridine chloride (CPC)) and the ionic strength (Ca^{2+}, NH_4^+) on breakthrough curves (BTCs) for sorption and desorption of bisphenol A (BPA) were studied using soil column experiment. Results showed that the presence of heavy metals and cationic surfactants caused a significant increase on the BPA sorption. In addition, the volume required when effluent concentration reached half of the influent concentration ($V_{C_{1/2}}$) increased due to the introduction of heavy metals and surfactants. It was also found that the larger amount of BPA was absorbed with higher ionic strength. The cationic surfactants enhanced the desorption ability of BPA from the soil. The results provided a better understanding of BPA behavior in environment and facilitated more accurate assessment of its ecological risk and identification of appropriate management strategies.
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Keywords: BPA; Heavy metals; Surfactants; Sorption; Desorption

1. Introduction

BPA, an alkylphenolic compound, is a principle monomer in the production of polycarbonates, epoxy resins, and other plastics [1]. The epoxies are used as food-contact surface coatings for cans, automobile parts, and adhesives and as a coating for PVC water pipe walls. BPA is suspected to enter rivers, streams and surface waters from effluents of facilities that manufacture epoxy and polycarbonate plastics and elution from the products containing BPA [2,3]. It is antioxidant, non biodegradable and highly resistant to chemical degradation. It is present in surface waters, as well as industrial wastewaters at concentrations that are of environmental concern and present a risk to humans and animals [4,5]. BPA has been reported to interact with the humans and wildlife and show adverse effect. These effects include formation of additional female organs, enlarged accessory sex glands, and a stimulation of egg and egg mass production resulting in increased female mortality [6]. One recent paper has demonstrated that laboratory mice exposure of about 20 mg/l of BPA have developed chromosomal abnormalities in eggs, raising earlier concerns about safe level of BPA exposure [7]. Therefore, BPA has been identified as the endocrine disrupting chemicals (EDCs) by the U.S Environmental Protection Agency (EPA), World Wide Fund for Nature (WWF) [8,9] and is becoming a social issue of increasing interest in the public [10–12].

Sorption and desorption are key factors in evaluating the transport, transformation, degradation and fate of BPA in environment. There have been a number of studies focused on the adsorption/partition behavior of BPA and they showed that the effects of the presence of organic pollutants on the sorption and other related fate and transport processes of BPA and other EDCs should not be ignored [10,11]. Heavy metals and surfactants are commonly coexisting with BPA in contaminated surface water and groundwater systems. Cadmium and lead as important EDCs [13] are well-detected in the natural water body, particularly in heavily contaminated wastewaters and in hazardous wastes areas [14,15]. BPA can also synergize with heavy metal cadmium and ultraviolet radiation, leading to increasing harm to the organisms [16,17]. Surfactants were also widely found in environment due to their applied strength in the fields of oil exploitation, synthetic detergents, printing and dyeing. They were found to affect

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the fate, transformation and transportation of pollutants in soils and sediments [18]. However, none of the previous studies have examined the effects of the presence of heavy metals and surfactants on the sorption and desorption of BPA in soils using soil column experiment.

Taking into account the above, the overall objective of this study was to investigate the influence of heavy metals and surfactants on the sorption and desorption of BPA. This evaluation will be carried out by studying its sorption isotherm by batch experiment. The effects of heavy metals, surfactants and ionic strength on breakthrough curves (BTCs) of BPA will be investigated using soil column experiment. The extent of surfactants desorption BPA from soil will also be measured.

2. Materials and methods

2.1. Materials

The soil sample was taken from the experimental farm, Shanghai Jiaotong University, Shanghai, China. The sample was crushed to pass through a 0.2 mm mesh sieve after air-dried. The basic characteristics of samples were analyzed using the methods described by He et al. [19] and are given in Table 1. BPA, calcium chloride, ammonium chloride, lead chloride, cadmium chloride, CTAB and CPC are all of analytical grade obtained from Shanghai Chemical Reagent Co. in China.

2.2. Sorption and desorption experiments

2.2.1. Sorption isotherm

A total of 1.5 g of soil was filled in 25 ml glass bottles with 20 ml of BPA solutions, which already contained $CaCl_2$ and NaN_3 at the concentrations of 5 mmol/l and 100 mg/l, respectively. $CaCl_2$ was used to give the appropriate ionic strength for the background groundwater and also could reduce the dispersion of particle. NaN_3 was used to inhibit the microbial activity. The glass bottles were capped with a well-fitted stopper, sealed with parafilm and then shaken in a DZK-2 thermostatted waterbath (Shanghai Jinghong Instrument Factory, China) at $25 \pm 1\,^{\circ}C$ for 24 h. Then soil suspensions were centrifuged at 10,000 rpm for 20 min using a high-speed refrigerated centrifuge (Sartorius 3–18 K, Sigma, Germany). A blank sorption experiment with no soil was performed using the same procedure to determine the loss of the BPA that might be absorbed on the glassware.

2.2.2. Sorption column experiment

The soil column experiment was carried out in a PVC column with an inside diameter of 5 cm and a length of 10 cm. One hundred grams of soil was added into the column with tapping to achieve a uniform porous medium. The upper part of the column
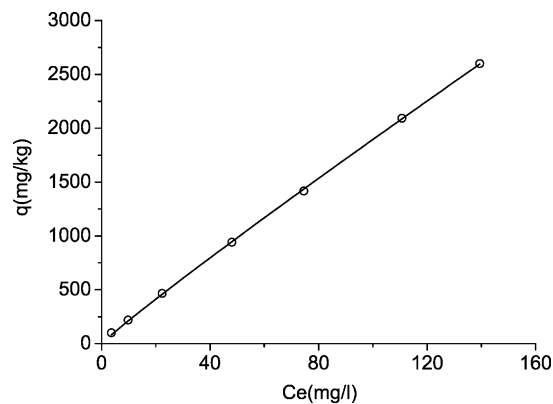

Fig. 1. The sorption isotherm of BPA in soil.

was uniformly covered with glass beads and nylon mesh over the whole of the surface of the column to facilitate the distribution of the solution, as well as minimize surface distribution. The bottom of the column was also uniformly covered with glass beads and nylon mesh to prevent the loss of the soil. The deionized water was used to wash the soil in an upflow rate, then in a downflow rate in order to reach a constant flow rate [20]. Immediately after this, the BPA solution at the concentration of 80 mg/l containing heavy metals, surfactants and ionic strength, respectively, was continuously fed to the top of the column at a constant flow rate controlled by a BT00–300M peristaltic Pump (Baoding Longer Precision Pump Co., China) until the solute effluent concentration, C_e (mg/l), approached the influent concentration, C_0 (mg/l), i.e., $C_e/C_0 = 1$ [21]. In all column tests, the effluent samples were intermittently collected in glass tubes by a BSZ-160 automatic fraction collector (shanghai Qinpu, Co., China) and measured by the UV 2102-PC spectrophotometer (Shanghai Unico Co., China) at the wavelength of 276 nm.

2.2.3. Desorption column experiment

For these studies, the solution containing 20 mg/l CTAB and CPC, respectively, was pumped into the top of the column, in which the initial soil was contaminated with 80 mg/l BPA solution. The solution flowed through the contaminated column until the $C_e/C_0 = 0$ was approached. All the experiments were conducted three times at $25\,^{\circ}C$ and the average values are shown in all Figs.

3. Result and discussion

3.1. Single sorption isotherm

Fig. 1 shows the sorption isotherm of BPA, which can be fitted by the Freundlich model,

$$q = K_f C_e{}^N$$

Table 1
The properties of soil

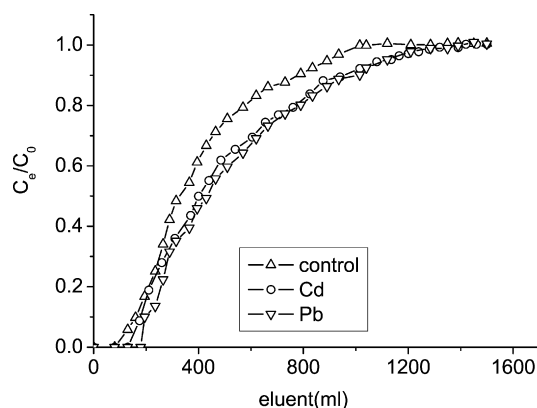| Sand (%)	Silt (%)	Clay (%)	Cation exchange capacity (c mol/kg)	Organic carbon (%)	pH (s:w = 1:2.5)
80	15	5	12.4	1.27	7.34

Fig. 2. Sorption breakthrough curves for BPA through soil in the presence of different heavy metals (0.12 mmol/l Cd and Pb, respectively).

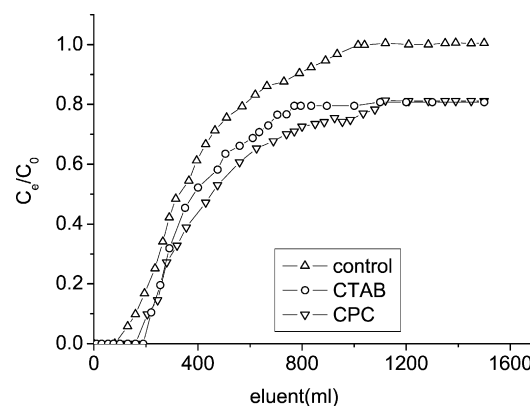
Fig. 3. Sorption breakthrough curves for BPA through soil in the presence of different surfactants (20 mg/l CTAB and CPC, respectively).

where K_f is the Freundlich unit-capacity coefficient $(mg/kg)(mg/l)^{-N}$, which is also called affinity constant and N is Freundlich exponent which shows the extent of isotherm nonlinearity. The sorption isotherm of BPA was nonlinear with N value 0.946. This suggests that partition was not the unique mechanism for the sorption of BPA in soil and the adsorption played an important role.

3.2. The effect of heavy metals on BPA sorption

The results obtained from the soil column experiments have been represented in the form of BTCs, in which the ratio (C_e/C_0) is plotted as a function of the cumulative volume of eluent, V (ml). The effect of heavy metals (0.12 mmol/l Cd and Pb, respectively) on BPA sorption is presented in Fig. 2. The parameters $V_{C_{1/2}}$ obtained from BTC are given in Table 2. The $V_{C_{1/2}}$ is the volume of eluent when the effluent concentration reaches half of the influent concentration in the sorption process [22]. Evidently, the presence of Cd and Pb significantly increased the sorption of BPA and resulted in a latter breakthrough. Also, as can be seen from Table 2, the $V_{C_{1/2}}$ decreases with the addition of Cd and Pb, with the value of 407 ml and 442 ml, respectively, compared to 332 ml in the absence of Cd and Pb. The results observed could be well explained by the polymer-based dual-domain concept, which considers natural organic matter (NOM) to contain a mixture of "glassy and rubbery" components. It is commonly

Table 2
The parameters for BPA obtained from the BTCs

Parameter	Added	Value
$V_{C_{1/2}}$ (ml)	Control	328–339(332)[a]
	Cd	391–415(407)
	Pb	372–386(442)
	CTAB	389–401(392)
	CPC	437–459(444)
	Ca^{2+}	534–542(536)
	NH_4^+	485–506(494)
$V_{D_{1/2}}$ (ml)	Control	214–231(224)
	CTAB	176–195(188)
	CPC	186–204(193)

[a] Figures in the parentheses indicate mean values.

accepted that sorption of organic compounds to soils/sediments consists of adsorption in glassy domain and partition in rubbery domain. Adsorption is characteristic of nonlinearity while partition is linear [23–25]. BPA is a weak acid with a pK_a of 10.23 and exists in molecular form in the solution with pH of 7.34 used in this study. Freundlich exponent of BPA isotherm in the absence of heavy metals is 0.946, which indicated that BPA was adsorbed in the glassy matrix by the adsorption, as well as partition in the rubbery matrix. It was likely that the addition of Cd and Pb could result in the aggregation and flocculation of humus in soil and increase of the adsorption percentage. The increasing glassiness would be more favorable to the BPA adsorption onto soil, leading to the enhancement of adsorption. Tao [26] reported the similar observations that the presence of heavy metals increased the adsorption of organic pollutants to soil, and the adsorption became a more significant contributor to the overall sorption behavior of atrazine when Cu is present.

3.3. Effect of surfactants on BPA sorption

Fig. 3 describes the BTCs of BPA without surfactant and in the presence of CTAB and CPC with the concentration of 20 mg/l, respectively. It can be seen from Fig. 3, the BTCs in the presence of CTAB and CPC is significantly displaced to the right of the BTCs of BPA alone. As can also be seen from Table 2, the $V_{C_{1/2}}$ values in the presence of CTAB and CPC are 392 ml and 444 ml, respectively. The result clearly indicated that the sorption of BPA to soil was greatly enhanced in the presence of CTAB and CPC. In addition, as evidently shown in Fig. 3, a clear difference between the BTCs is the "tailing" in the presence of CTAB and CPC (slow approach to $C_e/C_0 = 1$), which is characteristics of nonequilibrium sorption during BPA transport. This may be due to a slower sorption rate in the presence of CTAB and CPC [21,27].

The influence of cationic surfactants on BPA sorption to soil might be explained by considering the following two impacts. When a surfactant is added to the soil-water system, two new phases are created with which the organic contaminants may become associated [28–30]. On the one hand, the surfactant in monomer form may be adsorbed onto the soil and may increase
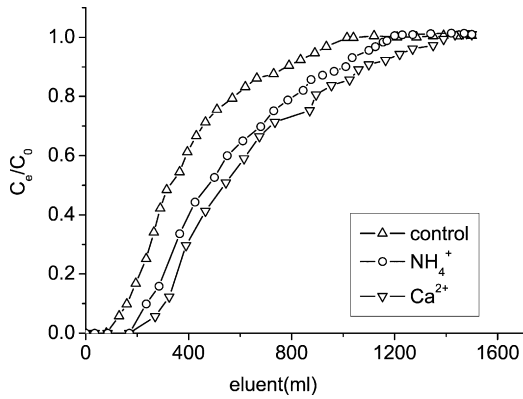
Fig. 4. Sorption breakthrough curves for BPA through soil in the presence of different salts as intermediate ionic strength (0.5 mol/l Ca^{2+} and NH$_4^+$, respectively).

Fig. 5. Desorption breakthrough curves for BPA through soil in the presence of different surfactants (20 mg/l CTAB and CPC, respectively).

the affinity of the BPA for the soil. On the other hand, due to its special surface properties, the surfactants would increase the water solubility of some organic chemicals [31], that is, the surfactants may increase the solubility of BPA, hence increase affinity of the BPA for the solution and decrease the sorption of BPA to soil. Since both actions give rise to opposite influences on the organic contaminants sorption, the net influence of the surfactant on the BPA sorption will depend on the balance of both of them; that is, whether the increase in sorption or the increase in solubilization will be more important [30–32]. The cationic surfactants are favorably sorbed by the soil phase because the soil surface is negatively charged and hence the amount of cationic surfactants sorbed by the soil is highly greater than that of remaining in solution. Therefore, the cationic surfactants enhanced the BPA sorption significantly.

3.4. Effect of ionic strength on BPA sorption

The effect of ionic strength (0.5 mol/l Ca^{2+}and NH$_4^+$, respectively) on sorption of BPA was shown in Fig. 4. It can be seen from Fig. 4 that the BTCs in the presence of Ca^{2+} and NH$_4^+$displaced to the right and clearly approach C_e/C_0 more slowly. This is also in accordance with the $V_{C_{1/2}}$ values obtained from BTC which are more for Ca^{2+} 536 ml, NH$_4^+$ 494 ml. The result indicated that the sorption capacity of BPA increased with the addition of Ca^{2+}, NH$_4^+$. This pronounced increase of the BPA adsorption can be attributed to the following several reasons. The first reason is that the Ca^{2+} and NH$_4^+$ could react with negative charges of the soil surface[33,34], thus decreasing the repulsion force between BPA and negatively charged surface of the soil and leading to an increasing sorption of BPA to soil. In addition, the Ca^{2+} and NH$_4^+$ could reduce the water solubility of BPA and hence enhancing BPA sorption to soil. According to the observations of researchers [35], the sorption amount increased as the solubility of compounds decreased, in good agreement with our result. Finally, at much higher ionic strength, the Ca^{2+} and NH$_4^+$ could inhibit the release of organic matter from the soil and diminished the pH values of suspension, which also increase the sorption of capacity.
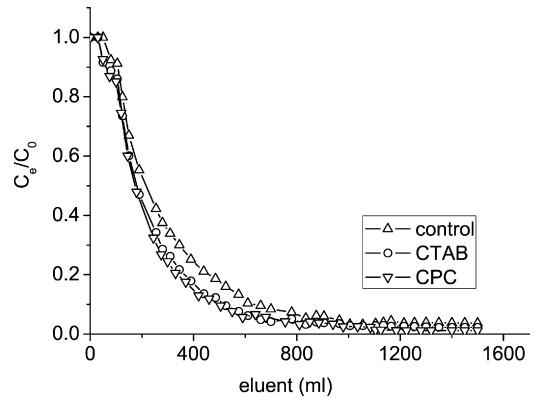
3.5. Effect of surfactants on BPA desorption

The effect of surfactants on BPA desorption was studied to evaluate the extent of BPA desorption from soil. The results are presented in Fig. 5 and parameters $V_{D_{1/2}}$ obtained from BTC are shown in Table 2. The $V_{D_{1/2}}$ is the volume of eluent when the effluent concentration reaches half of the initial concentration in desorption process.

As shown in Fig. 5, the BPA remaining in the soil column was rapidly reduced during the first desorpiton, after which the removal rate decreased significantly. For example, desorption with 400 ml CTAB solution removed 80% of the BPA during the first desorption, while only another 18% was removed for additional 800 ml. These results agree with those obtained previously [36,37]. For example, the results of Rodríguez-Cruz [37] indicated that there was initial rapid removal of the atrazine and metalaxyl, followed by a slower removal rate after the first desorption.

In addition, as can be seen from Fig. 5, at first desorption stage, the effect of surfactant was insignificant. As can also be seen from Table 2, the $V_{D_{1/2}}$ values corresponding to CPC and CTAB are almost equal to deionized water, resulting in an insignificant decrease in $V_{D_{1/2}}$ values from 224 ml for deionized water, to 188 ml for CPC and 193 ml for CTAB, respectively. At the first stage, the surfactant did not adsorb to the soil surface completely, therefore insignificant effect was observed. However, as desorption continued, the surfactants became increasingly more effective in desorption BPA from soil. The effective desorption of BPA from the soil could be explained by the increased solubility of BPA and the change of surface tension in the presence of surfactants. Usually, even at very low concentration, surfactants can reduce the surface tension of soil water to a large extent, which would result in a higher wettability of soil particles [38]. A higher wettability of soil particles would reduce the amount of other pollutants adsorbed by soil and hence enhancing the solubility of BPA. Therefore, this lead to the substantial effect in enhancing desorption of BPA from soil.

4. Conclusions

As mixed contaminants are often found in contaminated surface water and groundwater systems, the study of complex

pollution on the sorption, other related fate and transport processes of individual chemicals has become an issue of increasing interest in the environmental science. Results from this study indicate that the addition of heavy metal and cationic surfactants increase the sorption of BPA in soil. The surfactants enhance the desorption ability of BPA and facilitate the transport of BPA. The results obtained in this paper could be useful in order to complete the knowledge about the fate of BPA, and other phenolic endocrine disrupting chemicals.

Acknowledgments

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