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Effects of dissolved organic matter from sewage sludge on sorption of tetrabromobisphenol A by soils

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Abstract

Sorption of tetrabromobisphenol A (TBBPA) by soil influences its fate and transport in the environment. The presence of dissolved organic matter (DOM) may complicate the sorption process in soil. The effects of DOM from sewage sludge on TBBPA sorption by three soils were investigated using batch equilibration experiments in the study. DOM was observed to be sorbed on the soils and the isotherms could be fitted by the Langmuir model. The effects of DOM on TBBPA sorption were dependent on the characteristics of soils and the concentrations of DOM present. TBBPA sorption by Henan (HN) soil (sandy loam) and Liaoning (LN) soil (loamy clay) was promoted in the presence of DOM at low concentration (\leq 90 mg organic carbon (OC)/L), and the sorption was promoted by HN soil and inhibited by LN soil at DOM added concentration of 180 mg OC/L. TBBPA sorption by Guangxi (GX) soil (silt loam) was always inhibited in the presence of DOM. It was also found that the amount of TBBPA sorbed decreased with the increase in the solution pH value in the absence of DOM. The influencing mechanisms of DOM on the sorption of TBBPA by soils were also discussed.

Key words: tetrabromobisphenol A (TBBPA); sorption; dissolved organic matter; pH; soil; sewage sludge

Introduction

The amount of sewage sludge produced worldwide is ever increasing, and a growing proportion is being applied to agricultural land. Sewage sludge applied to soil has been considered as an effective way to improve the soil fertility and increase the organic matter content (Wang, 1997; Düring and Gäth, 2002). However, sewage sludge often contains high concentrations of potentially toxic organic contaminants and heavy metals (Wang and Jones, 1994; Walter *et al.*, 2002). Moreover, sewage sludge applied to soil can also produce dissolved organic matter (DOM) (Ashworth and Alloway, 2004). The influx of a large amount of DOM into soils can significantly affect the transport and fate of organic compounds in the soils (Nelson *et al.*, 1998; Williams *et al.*, 2000; Huang and Lee, 2001).

According to batch and column studies, it has been clearly demonstrated that DOM can form complexation with relatively polar pesticides or uptake hydrophobic organic contaminants, which increases the apparent aqueous solubility and potentially decreases the sorption (Chiou *et al.*, 1986; Nelson *et al.*, 1998; Williams *et al.*, 2000; Huang and Lee, 2001). In addition, the presence of DOM in solution may also compete with organic contaminants for adsorptive sites on the soil surface (Kögel-Knabner

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et al., 2000). Nevertheless, DOM can be sorbed to the soil surface, which may elevate the content of soil organic matter (Kaiser and Guggenberger, 2000). The sorbed DOM can make the soil more capable of sorbing organic compounds, resulting in an overall enhancement in sorption and retardation (Totsche et al., 1997). The source of DOM and the amount mixed with soil also influence the magnitude of sorption (Celis et al., 1998; Ling et al., 2005a; Gao et al., 2007). Celis et al. (1998) reported that DOM extracted from limed sewage sludge reduced atrazine sorption to soil, but DOM extracted from composted sludge enhanced atrazine sorption to soil owing to the increasing sorption of atrazine by DOM associated with the soil. Ling et al. (2005b) found that there was critical concentration of DOM derived from sewage sludge between promotion and inhibition of atrazine sorption by soil; the presence of DOM with concentration lower than the critical concentration promoted atrazine sorption, whereas the presence of DOM with concentration higher than the critical concentration tended to inhibit atrazine sorption. These studies clearly indicated that DOM can affect the sorption of hydrophobic compounds on soil, and the impact will be dependent on the intrinsic nature of solute, soil, and DOM, and the competition among solutesoil, solute-DOM, and DOM-soil interactions (Seol and Lee, 2000).

In the last decades, brominated flame retardants (BFRs)

have been widely used in industrial practice to improve the flame resistance of polymeric materials. Tetrabromobisphenol A (TBBPA) is a kind of BFR that was found in the wider industrial applications, which accounted for more than half of BFRs usage. The worldwide market demand for TBBPA was estimated at 1.2×10^8 kg in 2001 (Birnbaum and Staskal, 2004). TBBPA has been reported to cause adverse impacts to mammal and human health. Prolonged exposure of rats to TBBPA disturbed the liver heme metabolism, as well as the neural system activities (Syzmanska et al., 2000; Mariussen and Fonnum, 2003). Vitro assay results indicated that TBBPA manifested up to 10 times more potent than thyroxine (T4, thyroid hormone) to bind to human transthyretin (thyroid hormone transport protein) (Meerts et al., 2000). TBBPA is a relatively persistent compound, and is widely found in the environment, such as stream sediments, municipal wastewater, sewage sludge, soils, and biota (Sellström and Jansson, 1995; WHO/IPCS, 1995; Öberg et al., 2002; Morris et al., 2004). At a contaminated site in Israel, the concentration of TBBPA in the upper 20 cm of the soil layer was more than 50 mg/kg of soil, which can transfer to the deeper soil profile and may contaminate groundwater (Arnon et al., 2006).

Sorption is an important process for organic contaminants in soil influencing transport, degradation, and bioavailability of these compounds in the environment. However, to our knowledge, no studies were conducted to investigate TBBPA sorption by soil and the effects of DOM on the sorption to date. The objective of this study was to investigate the impacts of DOM from sewage sludge of a municipal wastewater treatment plant on TBBPA sorption by several soils. The results will provide a better evaluation of sorption and transport of TBBPA in the soils where sewage sludge is applied for land use.

1 Materials and methods

1.1 Materials

TBBPA (4,4'-isopropylidenebis(2,6-dibromophenol)) was obtained from Aldrich chemicals (USA) with a purity > 97%. The chemical structure of TBBPA is shown in Fig.1. The aqueous solubility of TBBPA is 4.16 mg/L at 25°C, and $\log K_{ow}$ is 4.50 (WHO/IPCS, 1995). Methanol (HPLC grade) was obtained from Tedia Company (USA). All other chemicals were of analytical grade.

Three soil samples were collected from surface layers (0–20 cm) in Fengqiu County of Henan (HN) Provice, Shenyang City of Liaoning (LN) Province, and Hechi City of Guangxi (GX) Province, China, respectively. The soil

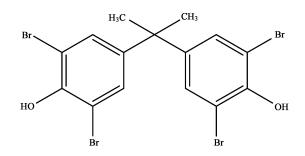


Fig. 1 Chemical structure of Tetrabromobisphenol A (TBBPA).

samples were air-dried and passed through a 0.25-mm sieve. The selected soil characteristics are listed in Table 1. Soil organic carbon (OC) was determined by the method of dichromate oxidation, and soil cation exchange capacity (CEC) was determined by the method of NH_4Ac exchange (Lu, 1999).

1.2 Dissolved organic matter extraction

Liquid sewage sludge was collected from Suojincun Municipal Wastewater Treatment Plant, Nanjing, China. The sample was collected in a 20-L plastic bottle and transported to the laboratory for DOM extraction. DOM was separated from the liquid sewage sludge using centrifugation and filtration methods. The liquid sewage sludge was centrifuged (10,000 r/min for 15 min) and then the supernatant was filtered through a 0.45-µm cellulose filter. The filtrate, i.e., DOM solution, was analyzed for pH (7.99) and conductivity (1,708 µs/cm). After filtration, NaN₃ was added to the filtrate at a concentration of 100 mg/L to inhibit microbial activities. The DOM content presented as OC concentration was measured using a total organic carbon analyzer (TOC-5000A, Shimadzu, Japan). The content of DOM in the solution was 277.7 mg OC/L. The filtrate was stored in refrigerator at 4°C for sorption experiments.

1.3 Dissolved organic matter sorption on soils

DOM sorption on the three soils was performed by adding 10 ml DOM solutions with a series of initial concentrations in each 15 ml glass tube containing 1.00 g soil. All the DOM solutions contained $0.01 \text{ mol/L CaCl}_2$ and 100 mg/L NaN_3 , and the pH of the solutions were adjusted to 8.0 (about the pH of the initial extracted DOM solution) with 0.1 mol/L HCl or 0.1 mol/L NaOH. The tubes were shaken at 150 r/min for 48 h at 25°C. After centrifugation at 4,000 r/min for 10 min, the DOM concentrations in solutions (presented as OC) were measured using a total organic carbon analyzer (TOC-5000A, Shimadzu, Japan).

Table 1 Selected physical and chemical properties of soils

Soil	pH ^a	OC (g/kg)	CEC (cmol(+)/kg)	Sand (wt.%)	Silt (wt.%)	Clay (wt.%)	Texture
HN soil	8.16	8.47	10.8	62.4	24.2	13.4	Sandy loam
LN soil	7.81	13.7	22.6	51.3	21.8	26.9	Loamy clay
GX soil	7.11	21.3	9.50	41.3	45.9	12.8	Silt loam

^a Soil:water = 1 g:2.5 ml. HN: Henan; LN: Liaoning; GX: Guangxi; OC: organic carbon; CEC: cation exchange capacity.

Sorbed organic carbon was calculated from the difference between the OC content of the DOM solution, which was initially added, and from that found in equilibrium solution with the soil, of which the amount of native DOM released from the air-dried soil samples was subtracted. All sorption samples were conducted in duplicate and the means are shown in Fig.2.

1.4 TBBPA sorption by soils

DOM solutions (10 ml) with different concentrations were added to the soils in 15 ml glass tubes with PTFElined screw caps. The solid-to-solution ratios were adjusted to attain 20%-80% of the initially added TBBPA uptake by the soils. All the DOM solutions contained 0.01 mol/L CaCl₂ to maintain a constant ionic strength and 100 mg/L NaN₃ to inhibit potential microbial activities, and the pH values of the solutions were adjusted to 8.0 with 0.1 mol/L HCl or 0.1 mol/L NaOH. TBBPA was mixed at high concentration in methanol before being added to the DOM solutions. Methanol concentrations were always less than 0.1% of the total solution volume to avoid the cosolvent effect. The tubes were shaken at 150 r/min for 48 h at 25°C. Preliminary studies showed that sorption equilibrium was approached within this time period. After mixing, the tubes were centrifuged at 4,000 r/min for 10 min, and 1.0 ml of the supernatant was removed into a sampling vial for analysis. All sorption samples were conducted in duplicate. The sorption experiments were conducted at different pH values in the absence of DOM by addition of HCl and NaOH as required to solutions containing 0.01 mol/L CaCl₂ and 100 mg/L NaN₃. The investigated pH value ranged from 5.5 to 9.0 since TBBPA mainly exists in molecular form when the pH is below 6.0 and in anionic form when the pH is above 8.5. The initially added concentration of TBBPA was 0.4 mg/L. After shaking and centrifugation, the pH values of the supernatants were measured using a 320-S pH meter (Mettler-Toledo Instruments Co., Ltd., China).

The samples were analyzed by high performance liquid chromatography (HPLC) equipped with a UV detector with a wavelength at 210 nm (1100, Agilent, USA). The column was a 4.6×250 mm model (ZORBAX SB-C18, Agilent, USA). The mobile phase was a mixture of 85:15 (*V*/*V*) methanol and 0.2% acetic acid aqueous solution at a flow rate of 1 ml/min. The loss of the TBBPA from photochemical decomposition, volatilization, and sorption to the tube was found to be negligible.

TBBPA sorption was calculated from the difference between the total amount of TBBPA initially added to the solution and the amount remaining in the solution at equilibrium. The distribution coefficient (K_d) was calculated using the linear sorption model (Eq.(1)):

$$q_{\rm e} = K_{\rm d} C_{\rm e} \tag{1}$$

where, q_e (mg/kg) and C_e (mg/L) denote the soil sorbed concentration and the concentration in solution, respectively.

2 Results and discussion

2.1 Sorption of DOM on soils

Sorption isotherms of DOM on the three soils are shown in Fig.2. The nonlinear isotherms were observed, which can be fitted by the Langmuir model (Eq.(2)) well ($R^2 > 0.985$):

$$q_{\rm DOM} = q_0 b C_{\rm DOM} / (1 + b C_{\rm DOM}) \tag{2}$$

where, q_{DOM} (mg OC/kg) is the amount of DOM adsorbed per unit weight of soil, *b* is the adsorption constant, q_0 (mg OC/kg) is the maximum amount of DOM adsorbed, and C_{DOM} (mg OC/L) is the equilibrium concentration of DOM in solution. The calculated maximum amounts of DOM sorbed on the soils were 394.6, 278.5, and 451.2 mg OC/kg for HN soil, LN soil, and GX soil, respectively. The distribution coefficient (K_{DOM}) between soil and DOM was calculated using Eq.(3):

$$K_{\rm DOM} = q_{\rm DOM} / C_{\rm DOM} \tag{3}$$

Within the investigated concentration, the K_{DOM} values were 1.24 ± 0.22, 2.04 ± 0.51, and 3.92 ± 0.84 L/kg OC for HN soil, LN soil, and GX soil, respectively. The sorption of DOM on GX soil was higher than that on HN soil and LN soil, which may be caused by the lower pH of GX soil. As the pH decreases, the charges on the DOM molecules and the mineral surface of soil become less negative, which decreases the repulsion between the surface and the additional adsorbed DOM molecules, and promotes the ligand exchange interaction between DOM and hydrous oxide (Shen, 1999).

2.2 Effects of DOM on sorption of TBBPA by soils

The sorption isotherms of TBBPA by the three soils in the absence and presence of DOM are shown in Fig.3. The sorption isotherms were fitted by the linear sorption model well ($R^2 > 0.975$), and the parameters of the linear sorption model are given in Table 2. In the absence of DOM, K_d in the tested soils ranged from 18.5 to 394.1 L/kg, which suggested that TBBPA has high affinity with the soils.

In the presence of DOM, the sorption behaviors of TBBPA by the soils were different. The impact of DOM on

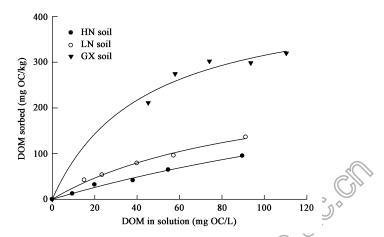


Fig. 2 Sorption isotherms of dissolved organic matter (DOM) on soils.

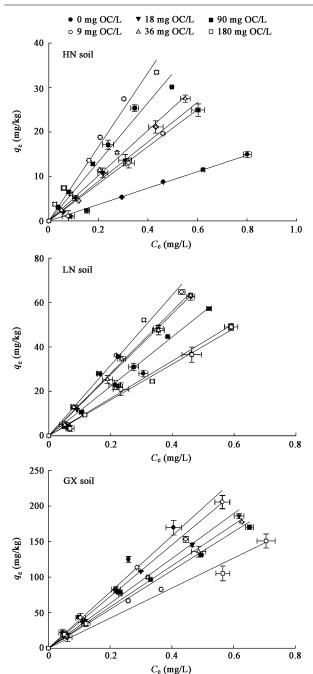


Fig. 3 Sorption isotherms of TBBPA by soils in the absence and presence of DOM.

TBBPA sorption was quantified by comparing the apparent distribution coefficient of TBBPA in the presence of DOM (K_d^*) and the absence of DOM (K_d) . The sorption was promoted when the ratio of K_d^*/K_d was more than 1, and was inhibited when the ratio of K_d^*/K_d was less than 1. The curves of K_d^*/K_d as a function of DOM added concentrations are displayed in Fig.4. Compared with the sorption of TBBPA in the absence of DOM, TBBPA sorption by HN soil was promoted in the presence of DOM, and the sorbed amount increased with increasing concentration of DOM. TBBPA sorption by LN soil was also enhanced in the presence of DOM at low concentration (\leq 90 mg OC/L), while the increment of sorption decreased with increasing concentration of DOM. In the presence of 180 mg OC/L

 Table 2
 Linear sorption model parameters for isotherms in the absence and presence of DOM, and the pH values of the supernatants at equilibrium

	-		
DOM (mg OC/L)	$K_{\rm d}~({\rm L/kg})$	R^2	pH
HN soil			
0	18.5±0.3	0.998	7.64 ± 0.02
9	41.7±1.3	0.988	7.69 ± 0.01
18	44.4±1.5	0.984	7.80 ± 0.03
36	50.9±1.3	0.993	7.92 ± 0.01
90	66.4±2.5	0.983	7.95 ± 0.02
180	83.2±3.3	0.984	8.01 ± 0.01
LN soil			
0	84.2±2.5	0.987	7.41±0.03
9	158.5 ± 4.6	0.987	7.42 ± 0.02
18	140.5 ± 4.9	0.982	7.51±0.01
36	137.4±2.4	0.995	7.61±0.01
90	111.3±2.0	0.995	7.72 ± 0.02
180	80.6±2.0	0.991	7.90 ± 0.02
GX soil			
0	394.1±15.7	0.977	7.17±0.01
9	363.2±7.0	0.994	7.34±0.02
18	315.4±10.2	0.984	7.46±0.02
36	289.7±6.6	0.994	7.58 ± 0.04
90	273.0±9.8	0.989	7.74 ± 0.04
180	211.2±9.3	0.978	7.97 ± 0.01

 K_d : distribution coefficient; R^2 : correlation coefficient.

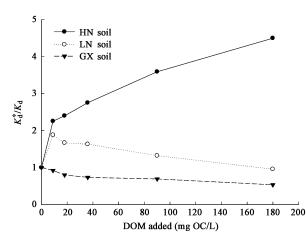


Fig. 4 Ratio of distribution coefficients of TBBPA in the presence of DOM and the absence of DOM (K_d^*/K_d) as a function of DOM added concentrations for soils.

DOM, the amount of TBBPA sorbed by LN soil was 4.3% lower than that in the absence of DOM. However, TBBPA sorption by GX soil was always inhibited in the presence of DOM, and the amount of TBBPA sorbed decreased with increasing concentration of DOM. For example, in the presence of 9 mg OC/L DOM, the K_d^* values for TBBPA sorption by HN soil and LN soil were 125.4% and 88.2% larger than their corresponding K_d values, whereas the K_d^* value for TBBPA sorption by GX soil was 7.9% lower than the K_d value. In the presence of 90 mg OC/L DOM, the K_{d}^{*} values for TBBPA sorption by HN soil and LN soil were 258.9% and 32.2% greater than their corresponding $K_{\rm d}$ values, whereas the $K_{\rm d}^*$ value for TBBPA sorption by GX soil was 32.3% lower than the K_d value. The results suggested that the effects of DOM on TBBPA sorption are dependent on the soil characteristics, as well as the presence of DOM concentrations.

No. 9

2.3 Effects of pH on sorption of TBBPA by soils

TBBPA is a kind of hydrophobic ionizable organic compound. The pK_{a1} and pK_{a2} of TBBPA are 7.5 and 8.5, respectively (WHO/IPCS, 1995). Thus, the aqueous solubility, sorption, and bioavailability of TBBPA are pH dependent. The effects of pH on the TBBPA sorption by soils are shown in Fig.5. Nearly all initially added TBBPA was sorbed by soils as the solution pH was below 6.0 (data not shown). When the pH was higher than 6.0, it was clear that the amount of TBBPA sorbed decreased as the solution pH increased in each soil in the absence of DOM. TBBPA exists in both molecular and anionic forms owing to the ionization in the pH range examined in the study. As the solution pH increases, the anionic form becomes dominant. Therefore, the electrostatic repulsion between the TBBPA anionic form and the negatively charged surface of soil becomes more pronounced and the amount of TBBPA partitioned to soil decreases. In addition, an increasing pH may enhance the release of native organic matter from the soils into solution, therefore result in the decrease of TBB-PA sorption. Similar results have been previously reported that increasing pH values decrease the sorption capacity of ionizable organic compounds to soils/sediments (Sheng et al., 2005; Zeng et al., 2006; Li et al., 2007). At the same pH, the amount of TBBPA sorbed by HN soil was the lowest, followed by that of LN soil, and the amount of sorbed by GX soil was the highest, which was probably owing to the different organic matter content of the soils.

2.4 Mechanisms of TBBPA sorption in the presence of DOM

As seen in Table 2, the pH values of the supernatants increased with increasing concentration of DOM at equilibrium, which was attributed to the good buffer capacity of DOM solutions. As mentioned above, pH plays an important role in TBBPA sorption by soils. Therefore, in the presence of DOM, TBBPA sorption by soil was controlled by the competition among soil-TBBPA, soil-DOM, and TBBPA-DOM interactions, as well as the solution pH. As displayed in Fig.5, the K_d values decreased with increasing solution pH, and according to the data, the curve of K_d as a function of pH (pH ranged from 7.0 to 8.2)

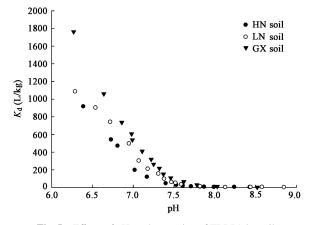


Fig. 5 Effects of pH on the sorption of TBBPA by soils.

in the absence of DOM could be fitted by Eq.(4):

$$\log K_{\rm d} = a \times \rm pH + b \tag{4}$$

where, a and b are empirical constants, respectively. The parameters of the equation are shown in Table 3.

Table 3 Parameters of the equation of $\log K_d$ as a function of pH

Soil	а	b	R^2	п
HN soil	-1.7734	14.772	0.9977	9
LN soil	-2.0474	17.062	0.9910	9
GX soil	-1.7385	15.021	0.9962	9

a and *b* are empirical constants, and *n* is sample number.

 K_d at a given pH can be estimated by Eq.(4) (pH ranging from 7.0 to 8.2). Assume that the increment of the sorption (ΔK_d) can be presented by the difference between K_d^* and calculated K_d at the same pH, i.e., $\Delta K_d = K_d^* K_{d}$. ΔK_{d} represents the net impacts of DOM on TBBPA sorption without the effect of pH. The curves of ΔK_d as a function of DOM added concentrations are displayed in Fig.6. The ΔK_d values in the presence of DOM were greater than that in the absence of DOM in each soil, which suggested that DOM plays a main role in the increase of TBBPA sorption. The influence of DOM on organic contaminant sorption may be attributed to their interaction in solution, competition for sorption sites, co-sorption, and cumulative sorption (Totsche et al., 1997; Celis et al., 1998; Spark and Swift, 2002; Ling et al., 2006; Gao et al., 2007). The competition of DOM with relative polar organic contaminant for sorption sites tended to reduce the organic contaminant sorption by soils (Spark and Swift, 2002), and the clay mineral surface generally contributed to the sorption of DOM (Satterberg et al., 2003). The presence of DOM promoted TBBPA sorption, and thus the competition of TBBPA with DOM for sorption sites did not seem to be an important mechanism of the influence of DOM on TBBPA sorption. The association of organic compound with DOM may reduce the sorption of organic compound (Williams et al., 2000), or the complexation of organic contaminant and DOM, and its sorption by soil (co-sorption) may promote the sorption of organic contaminant (Celis et al., 1998). However, the co-sorption

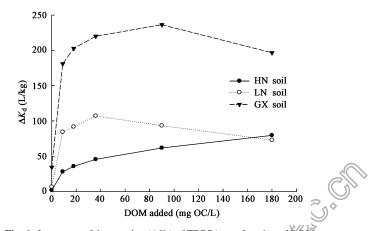


Fig. 6 Increment of the sorption (ΔK_d) of TBBPA as a function of DOM added concentrations for soils.

did not seem to be a major mechanism controlling TBBPA sorption because the sorption in the presence of DOM could be described by the linear model well (Ling *et al.*, 2006; Gao *et al.*, 2007). The present DOM was sorbed on the soil, which increased the organic matter content of soil and provided more sorption sites for TBBPA. Such cumulative sorption will increase TBBPA sorption and may be a major mechanism to the enhancement of TBBPA sorption by the tested soils.

However, the increase in solution pH had adverse effect on the promotion of TBBPA sorption that was caused by the addition of DOM solutions. Seol and Lee (2000) investigated the effect of DOM in treated effluent on transport of triazines, and found that the observed decrease in sorption was considerably more than the predicted values by model, which only consider the impact of DOM. Another study suggested that high pH and buffer capacity of the effluent induced increase in soil-solution pH, and prometryn, with a pK_a 4.5, showed highest sensitivity to pH changes and thereby the large pH-related effect on the enhanced movement in soils (Seol and Lee, 2001). Therefore, other factors should be considered to evaluate the effects of DOM on the sorption and transport of hydrophobic ionizable organic compounds in soil in addition to DOM.

The ΔK_d value was the greatest at DOM added concentration of 36 mg OC/L for LN soil and 90 mg OC/L for GX soil (Fig.6). When the DOM added concentrations were higher (\geq 90 mg OC/L for LN soil and 180 mg OC/L for GX soil), the ΔK_d value decreased, which was owing to the enhancement of TBBPA associated with DOM in solution. Gao *et al.* (2007) found that the sorption of phenanthrene by soils in the presence of exotic DOM increased at low concentration (\leq 28 mg OC/L) and decreased at higher concentration (\geq 52 mg OC/L). At the same concentration of DOM added (\leq 90 mg OC/L), the greatest ΔK_d value was observed in GX soil, then in LN soil, and the lowest ΔK_d value in HN soil, which was owing to the different amounts of DOM sorbed on the soils within the investigated concentrations.

3 Conclusions

TBBPA has high affinity with the tested soils, which suggests that soil may be one of the major pools for TBBPA in the environment. The effects of DOM on TBBPA sorption were dependent on the characteristics of soils and the concentrations of DOM present. TBBPA sorption by soils decreased with increasing solution pH in the absence of DOM. DOM was sorbed on the soils, which increased the organic matter content of soil and provided more sorption sites for TBBPA, while increasing pH caused adverse effect on the promotion of sorption resulting from good buffer capacity of DOM solution. The results of the study will be useful for better understanding of the behavior of TBBPA in soil and the ecological risk from land application of sewage sludge.

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