



Insights into tetrabromobisphenol A adsorption onto soils: Effects of soil components and environmental factors



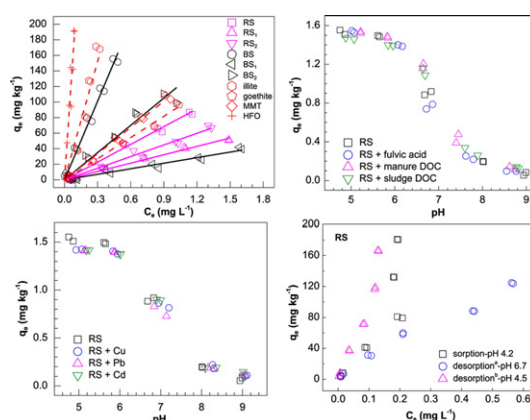
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HIGHLIGHTS

- Adsorption of TBBPA by soils decreases with increasing pH.
- SOM, iron oxides and phyllosilicate minerals are active surfaces for TBBPA sorption.
- DOC exhibits limited effect on TBBPA adsorption only at neutral condition.
- Heavy metals (Cu, Pb, Cd) show no direct interference with TBBPA sorption in pH 5–9.
- The alkaline river water significantly enhances TBBPA desorption from red soil.

GRAPHICAL ABSTRACT



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ABSTRACT

Concerns regarding tetrabromobisphenol A (TBBPA), the most widely utilized brominated flame retardant in the world, are growing because of the wide application and endocrine-disrupting potential of this compound. To properly assess its environmental impacts, it is important to understand the mobility and fate of TBBPA in soil environments. In this study, the effects of soil components, dissolved organic carbon (DOC) and heavy metal cations on TBBPA adsorption onto two Chinese soils (red soil and black soil) were investigated using batch sorption experiments. The desorption behavior of TBBPA when the two soils are irrigated with eutrophicated river water was also investigated. The results showed that pH greatly affects the adsorptive behavior of TBBPA in soils. Iron oxide minerals and phyllosilicate minerals are both active surfaces for TBBPA sorption, in addition to soil organic matter (SOM). DOC (50 mg OC L⁻¹) exhibited a limited effect on TBBPA sorption only under neutral conditions. TBBPA sorption was only minimally affected by the heavy metals (Cu²⁺, Pb²⁺ and Cd²⁺) in the studied pH range. Eutrophicated river water significantly enhanced the desorption of TBBPA from red soil due to the change in soil solution pH. These findings indicate that mobility of TBBPA in soils is mainly associated with soil pH, organic matter and clay fractions: it will be retained by soils or sediments with high organic matter and clay fractions under acidic conditions but becomes mobile under alkaline conditions.

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1. Introduction

Tetrabromobisphenol A (TBBPA) is the most widely utilized brominated flame retardant (BFR) in the world and has been widely detected in various environmental matrices with concentration levels ranging from ng g^{-1} to $\mu\text{g g}^{-1}$ (Covaci et al., 2009). Concerns regarding its use are growing because it is a suspected endocrine disrupter (Covaci et al., 2009). To date, most studies regarding TBBPA have concentrated on its occurrence, distribution, toxicity and degradation in the environment (Birnbaum and Staskal, 2003; Covaci et al., 2009). It can enter soil environments through disposal of waste electric and electronic equipment (WEEE) without thermal processing or in effluents of municipal wastewater treatment plants (Morf et al., 2005; Öberg et al., 2002; Wager et al., 2012; Zhou et al., 2013). In soil environments, TBBPA can be retained by soils (Jin et al., 2006; Yu and Hu, 2007) or it can undergo degradation or transformation by soil microbes (George and Haggblom, 2008; Shi et al., 2009). Among these environmental processes, the adsorption of TBBPA by soils has rarely been documented (Han et al., 2013; Sun et al., 2008) although it is critical to understanding the fate of TBBPA in soils or sediments.

TBBPA is a hydrophobic organic compound with low water solubility (0.171 mg L^{-1} , pH 3.05) and relatively high $\log K_{ow}$ (6.53, pH 3.05) (Kuramochi et al., 2008), thus it tends to partition into the organic matter of soils or sediments under acidic conditions. Sun et al. (2008) found that soil organic matter (SOM) contributes to over 90% of TBBPA sorption. However, the TBBPA molecule contains two phenolic hydroxyl groups which can undergo deprotonation with $\text{p}K_{a1}$ and $\text{p}K_{a2}$ of 7.5 and 8.5 (WHO/IPCS, 1995), which means that TBBPA mainly exists in its ionic form (i.e., deprotonated form) when pH is higher than $\text{p}K_{a1}$. The solubility and $\log K_{ow}$ may vary by several orders of magnitude depending on solution pH (Figs. S1 and S2) (Kuramochi et al., 2008). This structural property suggests that the adsorption behavior of TBBPA might be different from nonionic BFRs. A recent study has suggested that TBBPA sorption is pH-sensitive and is favored under acidic conditions (Han et al., 2013). They also suggested that SOM alone dominates the sorption of TBBPA in soils, while clay fractions show minor contribution to the adsorption. This was determined by testing the sorption affinity of TBBPA on montmorillonite, which was found to be quite low in their study. This result might be due to the rather low surface area of the montmorillonite sample they used ($13 \text{ m}^2 \text{ g}^{-1}$). In addition to layered silicates (e.g., montmorillonite), metal oxides are also important soil components and thus should also be included in order to evaluate the contribution of clay fractions to TBBPA sorption.

The influence of some environmental factors, such as dissolved organic carbon (DOC) and heavy metal cations, on the sorption process of TBBPA remain unknown. In addition, studies on the desorption behavior of TBBPA are limited. Sun et al. (2008) found desorption hysteresis of TBBPA from two alkaline soils when measuring the CaCl_2 background electrolyte. This is the only report to date that addresses the desorption properties of TBBPA. However, because the aqueous solubility of TBBPA greatly depends on pH, the fate of TBBPA in soils needs to be clarified for cases when soils are irrigated with alkaline water, such as eutrophicated river/lake water.

The objective of this study is to investigate the effects of soil components and various environmental factors (DOC, heavy metals) on the sorption behavior of TBBPA in soils. In addition, the desorption process of TBBPA in the presence of alkaline river water is also studied. The results will provide more comprehensive information on the transport and bioavailability of TBBPA in soil or sediment environments.

2. Materials and methods

2.1. Sorbents and chemicals

Two surface soils (0–20 cm) — Red soil (RS, Ferrosols) and Black soil (BS, Histosols) were sampled from Yingtan County, Jiangxi Province and Hailin City, Heilongjiang Province, respectively. To characterize the contribution of soil components to TBBPA sorption, the SOM was removed using NaOCl at room temperature. This method better maintains the original type and surface area of the soil amorphous oxide minerals when compared to an H_2O_2 method (Kaiser and Guggenberger, 2003). Following this step, a portion of SOM-removed soils were treated with dithionite–citrate–bicarbonate (DCB) to remove free ferric (hydro)oxides (Bigham et al., 1978; McCarty et al., 1998). Although we used mild treatments to remove SOM and oxides in this study, it is important to note that soil mineral surfaces might still be altered to some extent.

Illite was obtained from Fithian, Illinois and the montmorillonite (SWy) used was from Upton, Wyoming (distributed by Ward's Natural Science Establishment Inc., Rochester, NY, USA). The $< 2\text{-}\mu\text{m}$ -diameter fraction was separated according to the method described by Gu and Evans (2007). Goethite ($\alpha\text{-FeOOH}$) was synthesized according to the method of Hiemstra et al. (1989). Hydrous ferric oxide (HFO) was synthesized according to the method of Lovley and Phillips (1986). The physicochemical properties of these samples were collected in Table 1.

Table 1
Physicochemical properties of various sorbents in this study.

Sorbent ^a	pH	CEC ^b (cmol kg^{-1})	SOM ^c (g kg^{-1})	DCB Fe ^d (g kg^{-1})	AAO Fe ^e (g kg^{-1})	Surface area ^f ($\text{m}^2 \text{ g}^{-1}$)	Texture (%)		
							Clay	Silt	Sand
RS	4.50	9.66	12.4	44.49	2.08	36.17	6.65	30.98	62.37
RS ₁	6.14	6.24	4.5	44.75	3.06	36.66	16.34	41.11	42.56
RS ₂	8.39	8.66	7.9	0.61	–	16.64	12.34	42.00	45.66
BS	7.45	18.66	53.6	9.49	2.30	29.02	1.31	11.25	87.43
BS ₁	9.35	5.47	15.0	9.12	2.06	10.46	8.64	40.83	50.54
BS ₂	9.30	7.53	16.0	0.99	–	8.90	5.34	29.38	65.29
Illite	–	12.06	–	–	–	48.71	–	–	–
MMT	–	90.8 ^g	–	–	–	46 ^h	–	–	–
Goethite	–	–	–	–	–	63.53	–	–	–
HFO	–	–	–	–	–	600 ⁱ	–	–	–

^a RS and BS = the original red soil and black soil; RS₁ and BS₁ = the NaOCl treated RS and BS; RS₂ and BS₂ = the DCB treated RS₁ and BS₁; MMT = montmorillonite; HFO = hydrous ferric oxide.

^b Measured by $\text{Ba}^{2+}/\text{NH}_4^+$ exchange at pH 8 (Sparks, 1996).

^c Measured by dichromate oxidation (Sparks, 1996).

^d Determined using the DCB digestion method (Sparks, 1996).

^e Acid ammonium-oxalate digestion in the dark (Sparks, 1996).

^f Measured by N_2 -BET method (ASAP 2020, Micromeritics, USA).

^g Data from Gu et al. (2010).

^h External surface area (Gu et al., 2010).

ⁱ Estimated and recommended for use (Dzombak and Morel, 1990).

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