



Competitive sorption of bisphenol A and phenol in soils and the contribution of black carbon



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ABSTRACT

The sorption of contaminants such as bisphenol A (BPA), an endocrine disrupting chemical associated with various health problems, plays an important role in their transport and bioavailability in soils. This study investigates the sorption of BPA and phenol in two soils, Lu-Kung soil (LK) and Chen-Chu-Lu soil (CCL), as well as in straw-derived black carbon (BC). The sorption of both phenolic compounds in LK is much higher than that in CCL because of the higher organic content in LK (3.5%) as compared to CCL (1.3%). In addition, a higher BC fraction is observed in LK than in CCL. Furthermore, the sorption of BPA is greater than that of phenol because BPA exhibits higher hydrophobicity. For BPA, Freundlich affinity constants, K_f , are 2.62 (Freundlich exponent constant, $n = 0.91$) in LK and 1.54 ($n = 0.92$) in CCL. For phenol, K_f is 0.695 ($n = 1.0$) in LK and 0.0014 ($n = 2.3$) in CCL. For competitive sorption experiments, phenol sorption in LK could be suppressed by BPA, but the reverse is not possible. Chemical hydrophobicity strongly affects their competition. Similar competitive adsorption behaviour is observed on BC. Phenolic compounds' sorption on carbonaceous materials occurs through a bilayer formation. Thus, surface adsorption on carbonaceous materials largely determines the sorption of organic compounds in soils.

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1. Introduction

Environmental pollution by endocrine disrupting chemicals (EDCs) has recently attracted significant attention because these substances have several applications, and thus, their environmental distribution is widespread (Li and Wang, 2015; Pojana et al., 2007). Soils adsorb organic pollutants from the water that decreases the bioavailability of the pollutants in the aqueous phase. Therefore, EDCs' sorption in soils plays an important role in their transport in the environment. It is imperative to investigate the EDCs' sorption behaviors in soils.

Bisphenol A (BPA) is a chemical primarily employed in the production of polycarbonate plastic, epoxy resins, and other materials; it has been classified as an EDC because of its adverse effects on the normal function of the endocrine system in humans. Global demand for BPA surpassed seven million tons in 2013, and an annual growth rate of 4.7% from 2014 to 2020 is predicted (Im et al., 2015; Grand View Research, 2014). It was estimated that approxi-

mately 25% of this quantity will be released into the environment (Lelland, 2010; USEPA, 2010). BPA uptake into the crops through contaminated soils was estimated (Zhang et al., 2015), found in cereal food (Niu et al., 2012), and especially were in high concentrations in some cases (Liao and Kannan, 2013; Noonan et al., 2011). Another aromatic organic pollutant, phenol, is an important industrial feedstock with many applications. It is primarily used to produce phenolic resins and in manufacturing synthetic fibers (Weber and Weber, 2010). Natural organic matter also contains a lot of phenolic compounds (Bohn et al., 2001).

Partition of organic compounds in soil organic matter (SOM) determines the sorption of organic pollutants in soils (Chiou et al., 1983). The slow sorption behavior of organic contaminants was also studied (Pignatello and Xing, 1996; Shih and Wu, 2002; Shih and Wu, 2005) and transport of chemicals in humic acid was investigated through molecular simulation (Shih, 2007). Liu et al. (2008) indicated that in organic compounds with different physicochemical properties, the sorbates' hydrophobicity and the soil organic carbon content contribute to the retention of organic pollutants. In addition, these authors indicated that the solute's molecular structure and the other properties of soil affect sorption behavior (Cornelissen and Gustafsson, 2003; He et al., 2006; Shih and

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Table 1
pH values and compositions of soils.

Soil	pH	Sand (%)	Silt (%)	Clay (%)	Organic matter (%)	Black carbon (%)	Elemental composition (%)			
							C	H	O	N
LK	7.4	31	46	23	3.5	0.33	2.01	1.15	5.12	0.11
CCL	5.2	50	35	13	1.3	0.08	0.78	1.21	4.65	0.07

Wu, 2005). On the other hand, Hou et al. (2010) focused on the competitive interaction among pyrene, BPA, and sulfamethoxazole in sediment sample and its separated fractions. Sun et al. (2010) examined the sorption of 17 α -ethinyl estradiol (EE2) and BPA by nonhydrolyzable carbon, BC, and bulk soils and river sediments to realize the importance of different condensed SOM fractions and their mechanisms. However, only a few studies considered the competitive sorption of EDCs with their related compounds. Deciphering the sorption mechanisms of aromatic organic pollutants in soils will contribute to the prediction of organic chemical fate and transport in the environment.

In addition, EDCs are often present in a mixture in the environment; thus, sorption about multiple solutes in soil cannot be ignored. A number of studies were conducted to examine the competitive sorption of organic pollutants in soils and sediments (Xing and Pignatello, 1998; Xing et al., 1996; Yu and Huang, 2005). They concluded that the competitive sorption mechanism could include hole-filling in highly condensed phases of SOM. In recent years, Cornelissen and Gustafsson (2003) indicated that carbonaceous materials such as black carbon (BC) or biochar are more important for the sorption of organic contaminants than previously expected. These substances exhibit a high sorption capacity for organic contaminants in the environment (Lü et al., 2012; Song et al., 2013). Farmers in Asia often burn the rice straw on site after the rice is harvested. Then straw-derived BCs are commonly generated and remained in farms (Wang et al., 2013; Yang and Sheng, 2003). Thus, the role of BC in modeling the sorption of organic compounds in soils must be considered.

This study aims to investigate the sorption behaviors of two aromatic organic compounds, phenol and BPA, in two Taiwan soils, Lu-Kung soil (LK) soil and Chen-Chu-Llu (CCL) soil. Both LK and CCL soils are typical agricultural soils in Taiwan. Furthermore, this study evaluates the sorption mechanisms of these chemicals in the two soils, to investigate the competitive sorption behaviors of the two compounds in LK soil and straw-derived BC and to elucidate the competitive mechanisms of these two chemicals in soil.

2. Materials and methods

2.1. Characteristics of the two Taiwan soils, black carbon and the two sorbates

Two Taiwan soils, LK soil and CCL soil, were used as sorbents. LK soil and CCL soil were collected from the two agricultural fields in Changhua and Chiayi counties of Taiwan, respectively (Shih and Wu, 2005). These two soils are common soils in Taiwan and included in five representative soil groups of Taiwan. The soil group of LK is slate alluvial soil and CCL is red earth based on their parent materials. The soil orders are Inceptisols and Udisols for LK and CCL, respectively (Chen and Lee, 1995). Soil samples were passed through a 2-mm sieve to remove plant residues, pebbles and rocks, and were then dried for the further analysis. The basic samples' characteristics are presented in Table 1. Soil organic matter, the natural organic matter in soil, makes up much of the characteristic brown color of decaying plant, animal, as well as microbial debris and contributes to the brown or black color in surface soils. Therefore, soil organic matter is composed of a heterogeneous mixture

Table 2
Physicochemical properties of the two organic compounds used in experiments.

Compound	Molecular weight (g/mol)	Water solubility (mg/L)	log K_{ow}	p K_a
Phenol	94.1	83000	1.44	9.98
Bisphenol A	228.3	120.0	3.32	9.6–10.2

of soil organic substances containing acidic functional groups and nitrogen, but excluding artificial organic matters such as charcoal, black carbon, etc. CCL is an acidic soil and LK soil is a neutral soil. Four major elements, C, H, O and N, were determined by using an elemental analyzer (CHN-O-S-Rapid Analyzer, Heraeus). To prepare black carbon used in this study, rice straw from farms was burned and washed with pure water. They were homogenized and then passed through a 100–300 mesh sieve to remove large particles as well as small particles smaller than 43 μ m by a 325 mesh sieve. The particle sizes of obtained black carbon ranged from 43 to 147 μ m. Black carbon was oven-dried at 100 °C overnight and stored before use. Table 2 lists the major properties of the two sorbates, phenol and BPA, both purchased from Acros Organics.

2.2. Black carbon quantification in LK soil

The method proposed by Gustafsson et al. (1996) and Agarwal and Bucheli (2011) was applied to measure the BC content. A 5 g soil sample was weighed and 3 M HCl was added to remove the carbonates. Two stages of thermal treatment were then conducted: the sample was thermally oxidized in a muffle furnace at 375 °C for 24 h; the temperature was then increased to 600 °C at a rate 5 °C/min and hold for 4 h. The sample was weighed at the end of each stages. The weight difference between stage 1 and stage 2 and total loss (dry base) are defined as BC and SOM, respectively.

2.3. Sorption kinetics experiments

Approximately 1.0 g of the soil sample was mixed with 10 mL solution including one sorbate with 100 mg/L sodium azide to avoid biodegradation, and was placed in a 20 mL amber glass vial with a PTFE-lined screw cap. The vials were agitated at 150 rpm at 25 °C. At preselected intervals, the soil solutions were withdrawn, centrifuged and then filtered with 0.45 μ m filters for analysis. All the experiments were repeated three times. The kinetic study showed that 24 h was adequate to reach equilibrium. Simultaneously, to evaluate the loss of chemicals, blank experiments containing chemicals without soil were also conducted. Moreover, these two soils were confirmed not contaminated by BPA and phenol.

2.4. Sorption equilibrium experiments and models

Except for the extended sorption time (24 h), sample preparation, blanks, and analysis methods were the same as the kinetic experiments. The sorbed solute amount in soils was calculated from the difference between the initial and the equilibrium concentrations of the solute. Triplicate experiments were performed. The pH values of the solutions and soil suspensions were also monitored.

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