



Variation in sorption of propiconazole with biochars: The effect of temperature, mineral, molecular structure, and nano-porosity



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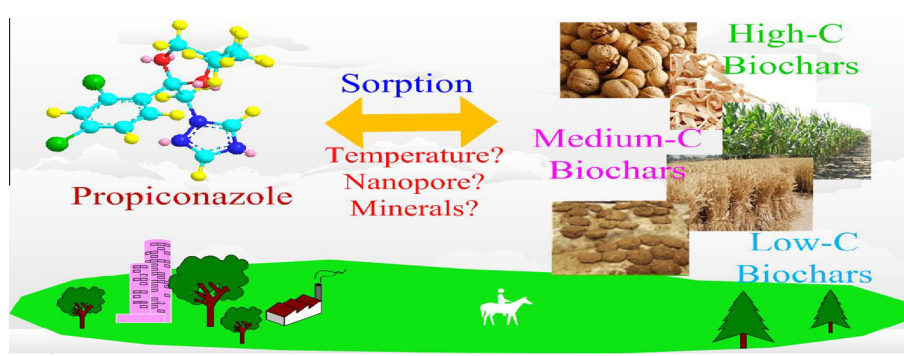
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HIGHLIGHTS

- Surface polar groups mainly consisted of O-containing groups of minerals.
- Pore-filling in aromatic C of biochar control nonlinear propiconazole sorption.
- HTT or C contents of biochar didn't necessarily control propiconazole sorption.
- Minerals may exert certain influence on propiconazole sorption.

GRAPHICAL ABSTRACT



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ABSTRACT

Sorption behavior of propiconazole (PROPI) by plant-residue derived biochars (PLABs) and animal waste-derived biochars (ANIBs) obtained at three heating treatment temperatures (HTTs) (300, 450 and 600 °C) (e.g., BCs300, BCs450, and BCs600) and their corresponding de-ashed BCs450 was investigated. PLABs belonged to high- or medium-C biochars and ANIBs were low-C biochars. Surface C concentrations of the tested biochars were generally higher than their corresponding bulk C. Surface polar groups were mainly composed of O-containing groups of minerals within biochars. The nonlinearity coefficients (n) of propiconazole (PROPI) sorption isotherms ranged from 0.23 to 0.64, which was significantly and negatively related to organic carbon (OC)-normalized CO₂-surface area (CO₂-SA/OC) of biochars. This correlation along with the positive relationship between CO₂-SA/OC and aromaticity indicates that pore-filling in nanopores within aromatic C dominate nonlinear PROPI sorption. HTTs or C contents do not necessarily regulate PROPI sorption. Removal of minerals from BCs450 elevated PROPI sorption because minerals may exert certain influence on sorption via impacting spatial arrangement of polar groups and/or organic matter (OM)-mineral interactions. This study helps to better understand sorption behavior of PROPI to biochars and evaluate the potential role of biochar in water treatment systems.

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1. Introduction

Pesticide use in agriculture has been instrumental in increasing crop yields and efficiency in the food production processes supplying food to an ever-increasing world population. However, the use

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of pesticides has led to a series of toxicological and environmental risks, which has drawn extensive attention from all aspects of society (Li and Randak, 2009). Innovative strategies are needed to reduce the environmental impact of pesticides, while improving agricultural productivity. Biochar is a porous carbon rich charcoal-like substance created by heating waste biomass under low oxygen conditions or anaerobic conditions. Biochar has been used as an increasingly utilized cost-effective soil amendment in agricultural and environmental applications (Sohi et al., 2010; Beesley et al., 2011), which can reduce greenhouse gas emissions, sequester C, as well as improve soil fertility and thus plant growth (Steinbeiss et al., 2009; Atkinson et al., 2010; Woolf et al., 2010). Moreover, biochar has a large surface area and high microporosity, which results in a very high affinity and capacity for sorbing and immobilizing organic contaminants (Keiluweit et al., 2010; Kookana, 2010; Sun et al., 2011a, 2011b). To date, there has been a growing body of literature on the application of biochar in water treatment reviewed by Inyang and Dickenson (2015). In particular, the use of biochar for the removal of persistent organic pollutants from aqueous systems has been addressed (Liu et al., 2012; Zheng et al., 2013; Chen et al., 2015). The sorption behavior of some pesticides and herbicides as well as their removal mechanisms has been previously documented. For an instance, the removal performance of atrazine (Cao et al., 2009), catechol (Kasozi et al., 2010), carbaryl (Zhang et al., 2013), 2,4-Dichlorophenoxyacetic acid (Kearns et al., 2014), fluridone (Sun et al., 2011a), and oxamyl (Taha et al., 2014) by various biochars was 0.02, 20, 1, 0.72, 10 mg g⁻¹, and ~99% sorbed, respectively, and their removal mechanisms (e.g., proposed sorption mechanisms) were mainly partitioning, pore-filling and diffusion, hydrophobic and π - π EDA interaction, surface adsorption, partitioning on amorphous-C, and H-bonding with polar groups, respectively. However, knowledge on the sorption propiconazole (PROPI) by biochar is very limited. PROPI is a hydrophobic systemic triazole fungicide, which is used worldwide for controlling harmful microorganisms, inhibiting fungus attack and preventing foliar or root disease, such as powdery mildew, leaf spot, rust and root rot (Garland et al., 1999). PROPI has been listed as a persistent, potentially toxic compound and possible human carcinogen by the European Union (Adam et al., 2005). Its low mobility and relatively high adsorption in soils rich in OM result in its accumulation in soils and pose a risk for the soil ecosystem (Thorstensen et al., 2001). Organic carbon (OC) has been reported to be the most important factor in adsorption of PROPI by soils (Riise et al., 2001). Therefore, it is hypothesized that rich-OC biochar should have high sorption capacity to PROPI and there is practical application of biochar in water treatment containing PROPI.

The ability of biochars to sorb organic pollutants depends greatly on their physical and chemical properties, which vary dramatically with heating treatment temperatures (HTT) and feedstock sources (Chen et al., 2008; Sun et al., 2012). Recently, Keiluweit et al. (2010) proposed a multiphase model to describe the physical-chemical transitions in char as HTT is increased. They found four distinct categories in biochars derived from grass and wood residues: transition char, amorphous char, composite char, and turbostatic char, which exhibited different molecular structure as well as surface characteristics. Their differences in these properties influenced their sorption properties of phthalic acid esters (PAEs) and fluorinated herbicides, moreover, the grass biochars generally exhibited higher sorption to PAEs and herbicides than the wood biochar from the same temperature (Sun et al., 2011a, 2012). On the other hand, previous studies on the sorption of hydrophobic organic contaminants (HOCs) generally used low-mineral biochars produced from relatively pure plant residues (Smernik, 2009). Although some of these contain considerable ash (e.g. grass biochars), the sorption properties of the ash

component have been little considered (Smernik, 2009; Sun et al., 2012). For sorption of HOCs, it is reasonable to assume that the OM still would dominate the sorption of mineral-rich biochars, however, the presence of minerals is likely to have at least a secondary influence (Smernik, 2009). Information about effects of minerals on the sorption of biochars to pesticides, as well as the related OM-mineral interaction mechanism, is rather limited to date. In addition, earlier studies dealt with the role of bulk polarity of biochars on their sorption of HOCs (Chen et al., 2008; Sun et al., 2012), while the impact of their surface polarity on pesticides sorption was seldom investigated, especially for PROPI. It was hypothesized minerals and components (e.g., aromaticity, ash content, bulk or surface polarity) within biochars (exogenous OM of soils) should influence sorption of PROPI.

This study was designed to fill this knowledge gap. Twelve feedstocks from plant residues and animal-wastes that contained various contents of ash were used to produce biochars at three HTTs (300, 450, 600 °C). In addition, the six selected biochars were de-ashed to investigate the influence of the minerals on sorption of PROPI. The primary objectives of this study were to: (1) investigate the effect of both HTT and feedstock on the biochar characteristics and, in turn, their effect on the sorption behavior of PROPI; (2) gain insight into the principal factors or components (e.g., aromaticity, ash content, bulk or surface polarity) controlling sorption of PROPI by each kind of biochar; and (3) identify the impacts of minerals on the sorption of PROPI by biochars.

2. Materials and methods

2.1. Chemicals

PROPI (with a purity > 98%) was purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The molecular structure and selected physicochemical characteristics of PROPI are presented in Table S1, Supplementary data. A stock solution of 100,000 mg/L of PROPI was prepared in methanol and stored at 4 °C in the dark.

2.2. Sorbents

Biochars were produced from two types of feedstocks (9 plant residues: walnut shell, pine wood, poplar leaf, cotton straw, bean straw, potato straw, rice straw, wheat straw, maize straw; and 3 animal wastes: manure of chicken, swine, and cow) at three different HTTs to obtain 27 plant-derived biochars (PLABs) and 9 animal waste-derived biochars (ANIBs). In addition, six biochars produced at 450 °C were also de-ashed. Detailed descriptions of the production procedures of biochars as well as their de-ashed samples can be found in Sun et al., 2013a; Jin et al., 2014; Qiu et al., 2014. The biochar samples are referred to as WALX, PINX, POPX, COTX, BEAX, POTX, RICX, WHEX, MAIX, CHIX, SWIX, and COWX, according to the first three capital letters of source material name, where X is substituted with the appropriate HTT (300, 450 and 600 °C). For comparison, fresh raw material (X = 000) was also included in some analyses. A "D" preceding the sample name is used to identify the de-ashed counterparts. The groups of biochars produced at 300, 450 and 600 °C are also referred to as BCs300, BCs450, and BCs600, respectively. Some of the biochars in this study have been used in previous studies in order to investigate (a) the impact of deashing treatment on biochar structure properties and potential sorption mechanisms of phenanthrene by six biochars from plant residues (rice, wheat, maize), and animal waste (chicken, swine, cow) at 450 and 600 °C (Sun et al., 2013a), (b) the competitive sorption of phenanthrene and dibutyl phthalate by five BCs450 produced from grass straws (soybean, rice, and cotton),

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