



Fast and slow adsorption of carbamazepine on biochar as affected by carbon structure and mineral composition



Jian Chen^a, Di Zhang^{a,*}, Huang Zhang^b, Saikat Ghosh^a, Bo Pan^a

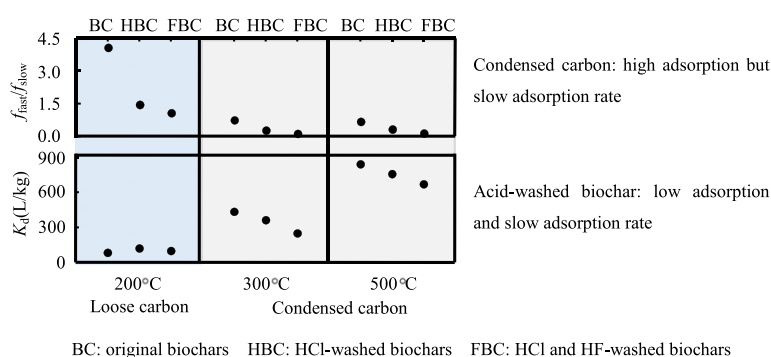
^a Faculty of Environmental Science & Engineering, Kunming University of Science & Technology, Kunming 650500, China

^b Yunnan Institute of Food Safety, Kunming University of Science & Technology, Kunming 650500, China

HIGHLIGHTS

- Hydrophobic and π - π interactions are the dominated adsorption mechanism of CBZ on biochar.
- The fast adsorption of CBZ is attributed to the loose carbon and minerals of biochar.
- The slow adsorption of CBZ is attributed to the condensed carbon of biochar.
- The hydrogen bonds between CBZ and -OH on minerals are likely the fast adsorption mechanism.

GRAPHICAL ABSTRACT



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ABSTRACT

The influence of carbon structure and mineral composition of biochar on fast adsorption and slow adsorption of ionic organic contaminants (IOCs) is still unclear. This study used carbamazepine (CBZ) as adsorbate, peanut-shell-derived biochars produced at different charred temperatures, and the corresponding acid-washed biochars as adsorbents, to investigate the adsorption kinetics as affected by carbon structure and mineral composition of biochar. The adsorption of CBZ on amorphous (loose) carbon was lower than aromatic (condensed) carbon, but the former mainly contributed to the fast adsorption of CBZ. The hydrophobic and π - π interactions were likely the predominant adsorption mechanisms of CBZ on biochar. The ratio of CBZ fast and slow adsorption fractions (f_{fast}/f_{slow}) significantly reduced after acid-wash treatment of the biochars. The X-ray diffraction and Fourier transform infrared spectra indicated that minerals, including CaCO_3 , KAlO_2 , and quartz, were likely responsible for the fast adsorption of CBZ by the hydrogen bonds between CBZ and -OH on the surface of minerals. These results are useful for better understanding of the environmental behavior and prediction of the environmental risks of IOCs in biochar-amended soils.

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1. Introduction

In the last decade, pharmaceuticals have been widely used to treat diseases of both humans and animals, but their residues and

degradation products may cause some adverse effects on environmental ecology and human health, such as chronic toxicity to aquatic organisms (Hazelton et al., 2013), endocrine disruption (Hazelton et al., 2013), and teratogenic effects (Li et al., 2013). Carbamazepine (CBZ), an antiepileptic drug, is one of the most frequently detected pharmaceuticals in soil and aquatic environments (Zhang et al., 2008). It has been indicated that CBZ exhibits the persistence characteristic of

* Corresponding author.

E-mail address: zhangdi2002113@sina.com (D. Zhang).

organic contaminants, potentially leading to long-term environmental risks (Chefetz et al., 2008). CBZ has been used as a marker for assessing anthropogenic influences on aquatic systems (Clara et al., 2004). Understanding the environmental behavior of CBZ in aquatic systems is vital to assess and control its environmental risks.

As a kind of porous carbon produced by heating biomass with limited oxygen (Keiluweit et al., 2010), biochar has received widespread attention due to its ability to enhance soil fertility (Lehmann et al., 2006), facilitate sequestration of atmospheric carbon dioxide (Yang and Sheng, 2003), and improve adsorption of organic pollutants (Cao and Harris, 2010). Many studies focused on the application of biochar in water treatment and soil amendment due to its large surface area and high microporosity (Lehmann et al., 2006; Kookana, 2010). The pore-filling, partitioning, hydrophobic, and π - π electron donor-acceptor (EDA) interactions have been generally recognized as the main adsorption mechanisms of hydrophobic organic contaminants (HOCs) on biochar (Kasozi et al., 2010; Zheng et al., 2013; Chen et al., 2015). These adsorption mechanisms varied with the structure and properties of the biochar (e.g., aromaticity, polarity, surface area, pore volume), depending on the pyrolytic temperature and feedstock sources (Keiluweit et al., 2010). For CBZ, an ionic organic contaminant (IOC), its adsorption mechanism on biochar was likely complex, in comparison to HOCs, due to its species diversity. Previous studies mainly focused on the apparent equilibrium adsorption isotherm of CBZ on biochar (Nielsen et al., 2015), which was invalid for describing the adsorption process (Pignatello and Xing, 1995). Adsorption kinetics study is a useful method for understanding the adsorption process, which could be mathematically divided into fast adsorption and slow adsorption compartments by the two-compartment first-order adsorption model (Pignatello and Xing, 1995; Altfelder et al., 2000). However, few scholars focused on the fast and slow adsorption characteristics of CBZ on biochar. For example, Jung et al. (2013) found that the adsorption kinetics of CBZ on biochar was affected by the π - π interaction, but the characteristics of fast adsorption and slow adsorption were ignored (Jung et al., 2013). Nielsen et al. (2014) explored the adsorption kinetics rate of CBZ on biochar, but the adsorption kinetics mechanism was not involved in this study (Nielsen et al., 2014).

In previous studies, it was reported that the structure of soil organic matter (SOM) played important roles in adsorption of HOCs (Pignatello and Xing, 1995; Xing and Pignatello, 1997; Pan et al., 2006; Pan et al., 2012). For example, Pignatello and Xing (1995) assumed that fast adsorption was likely related to the outer SOM fractions with loose structure, while slow adsorption was attributed to the inner SOM fractions with condensed conformation (Pignatello and Xing, 1995). Similarly, Keiluweit et al. (2010) divided biochar into four distinct categories, including transition char, amorphous char, composite char, and tubostratic char. It was found that the physicochemical properties of biochar could be altered by increasing the temperature (Keiluweit et al., 2010). Thus, an abundant diversity of fast and slow adsorption

of CBZ might exist on different classified chars. According to the SOM structure concept, we proposed the hypothesis that loose carbon possibly contributed to fast adsorption, and condensed carbon played important roles in slow adsorption. Additionally, summarizing the literatures, some studies pointed out that minerals reduced the adsorption of organic contaminants on biochar by covering some adsorption sites of organic matter (Wang et al., 2008; Zhang et al., 2013). Inversely, it was indicated that H-bonds could occur between organic contaminants and mineral surfaces, leading to enhanced adsorption (Teixidó et al., 2011). Other studies also indicated that the minerals in biochar played a secondary role in adsorption of HOCs, in comparison to organic carbon (Sun et al., 2013a; Sun et al., 2013b; Zhang et al., 2013). These conflicting results indicated that the effects of minerals in biochar on organic contaminants adsorption were still unclear. In these studies, most researchers only focused on the influence of the whole inorganic fractions on the equilibrium adsorption isotherms of organic contaminants (Sun et al., 2013a; Sun et al., 2013b; Zhang et al., 2013; Zheng et al., 2013). However, the effects of the mineral composition of inorganic fractions on the adsorption kinetics of IOCs were generally ignored. Thus, it was speculated that these conflicting findings were possibly resulted from the different mineral compositions of biochars with different origins. To our knowledge, there was not available study on the impact of the mineral composition of biochar on fast adsorption and slow adsorption of IOCs.

Therefore, the objective of this study was to illustrate the underlying mechanisms of fast and slow adsorption of CBZ as affected by varying the carbon structure and mineral composition of biochar. This study will provide important data for understanding IOCs adsorption mechanisms and kinetic processes on biochar.

2. Experimental section

2.1. Materials

CBZ (purity $\geq 99\%$) was purchased from Bio Basic, Inc. and its selected physicochemical properties are listed in Table S1. Peanut shells, collected from a local agricultural field in Kunming, were used as the raw feedstock for the biochars. These peanut shells were ground to pass through a 60 mesh sieve and charred for 4 h at 200, 300, and 500 °C under the condition of limited oxygen. The biochars produced were ground to pass through a 100 mesh sieve. Some biochars were washed using 1 M HCl (labeled HBCX, where X is the temperature) solution or 1:1 (M) HCl/HF (labeled FBCX) solution at a solid to liquid ratio of 1:5, to remove saline minerals, or to remove saline minerals and SiO₂, respectively. Other untreated biochars (labeled BCX) were referred as the control. All of the aforementioned biochars were rinsed with ultra-pure water until reaching neutral pH.

Table 1
The elemental compositions, ash contents, surface area (SA) and pore volume (PV) of biochars.

Samples	Compositions					Atomic ratio		Ash (%)	SA ^a (m ² g ⁻¹)	PV ^b (m ³ g ⁻¹)
	N (%)	C (%)	H (%)	S (%)	O (%)	H/C	(O + N)/C			
BC200	0.988	46.105	5.264	0.404	36.414	1.370	0.611	10.827	1.827	0.010
HBC200	0.679	47.120	5.469	0.000	37.948	1.393	0.616	8.784	1.619	0.010
FBC200	0.763	49.395	5.688	0.031	37.593	1.382	0.584	6.530	1.435	0.010
BC300	1.572	54.945	3.517	0.259	24.844	0.768	0.364	14.862	7.088	0.018
HBC300	1.428	56.913	3.864	0.047	27.333	0.815	0.382	10.415	3.098	0.012
FBC300	1.128	58.321	4.113	0.046	28.421	0.846	0.381	7.970	2.693	0.011
BC500	1.603	59.448	2.710	0.264	19.117	0.547	0.264	16.857	64.728	0.057
HBC500	1.467	66.339	2.814	0.020	17.870	0.509	0.221	11.489	72.251	0.061
FBC500	1.407	69.090	3.002	0.052	18.189	0.521	0.215	8.260	85.072	0.069

^a Surface area measured by N₂ adsorption using BET technology.

^b Pore volume measured at P/P₀ = 0.2.

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