



Sorption of hydrophobic organic compounds to a diverse suite of carbonaceous materials with emphasis on biochar



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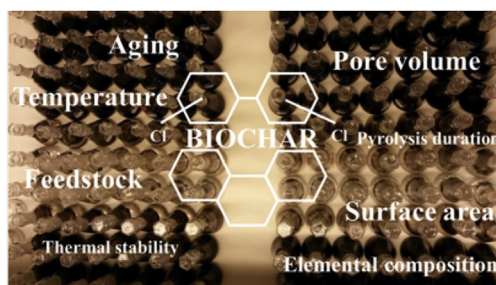
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HIGHLIGHTS

- Biochar plays a crucial role in transport and fate of organic contaminants.
- The multiple factors involved in sorption of HOCs to biochar are investigated.
- Variations in sorptive ability are related to physico-chemical properties of biochars.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbonaceous materials like biochars are increasingly recognized as effective sorbent materials for sequestering organic pollutants. Here, we study sorption behavior of two common hydrophobic organic contaminants 2,2',5,5'-tetrachlorobiphenyl (CB52) and phenanthrene (PHE), on biochars and other carbonaceous materials (CM) produced at a wide range of conditions and temperatures from various feedstocks. The primary aim was to establish structure–reactivity relationships responsible for the observed variation in CM and biochar sorption characteristics. CM were characterized for their elemental composition, surface area, pore size distribution, aromaticity and thermal stability. Freundlich sorption coefficients for CB52 and PHE (i.e. $\text{Log}K_{F, \text{CB52}}$ and $K_{F, \text{PHE}}$, respectively) to CM showed a variation of two to three orders of magnitude, with $\text{Log}K_{F, \text{CB52}}$ ranging from 5.12 ± 0.38 to 8.01 ± 0.18 and $\text{Log}K_{F, \text{PHE}}$ from 5.18 ± 0.09 to 7.42 ± 1.09 . The highest $\text{Log}K_F$ values were observed for the activated CM, however, non-activated biochars produced at high temperatures (>700 °C) sorbed almost as strongly (within 0.2–0.5 Log units) as the activated ones. Sorption coefficients significantly increased with pyrolysis temperature, CM surface area and pore volume, aromaticity, and thermal stability, and decreased with H/C, O/C,

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(O + N)/C content. The results of our study contribute to the understanding of processes underlying HOC sorption to CM and explore the potential of CM as engineered sorbents for environmental applications. © 2015 Elsevier Ltd. All rights reserved.

1. Introduction

Carbonaceous materials (CM), often referred to as “black carbon”, represent a continuum of heterogeneous materials ranging from partly charred plant material to char and charcoal to soot and graphite (Schmidt and Noack, 2000). Carbonaceous materials are produced by incomplete combustion of fossil fuels and vegetation and they are ubiquitous in nature, especially in soils and sediments (Larsson, 1985). CM play an important role in the global carbon cycle and constitute an important carbon sink due to their stability and inertness to microbial and chemical degradation (Schmidt and Noack, 2000). When produced from biological sources and purposely added to soil, CM is often termed “biochar” (Lehmann and Joseph, 2009). The application of biochar to soil has been shown to contribute positively to the mitigation of climate change, energy production, and improvements in the agricultural properties of soil (Lehmann and Joseph, 2009). Furthermore, it has been shown that biochar and other CM addition to soils and sediments enhances sorption of hydrophobic organic compounds (HOC), thereby potentially playing a crucial role in controlling transport, bioavailability, fate and health risk of organic contaminants (Ahmad et al., 2014).

Since most CM are not fully carbonized and thus quite heterogeneous, the primary mechanisms responsible for sequestering HOCs by CM include both hydrophobic partitioning/adsorption to the non-carbonized fraction and adsorption to the carbonized fraction (Chen et al., 2008). The sorption of HOCs to CM, assisted by π - π electron donor-acceptor interactions, has been shown to occur via sorption onto exterior surfaces, which proceeds further and more slowly by a pore-filling mechanism (Nguyen et al., 2007). In addition, polar organic pollutants containing N, O, and F atoms in their structure can form hydrogen bonds with H donor functional groups on the surface of the sorbent (Sun et al., 2012).

Thus the sorption strength and mechanism controlling pollutant sequestration by CM is dependent upon molecular (carbonized and non-carbonized fractions) and structural (surface, pore, and bulk properties) characteristics (Zhao et al., 2013). This in turn depends on the CM starting material (feedstock) and pyrolytic conditions (temperature and residence time). Additionally, a sorbent may be activated physically or chemically to form activated carbon with increased macro- and nano-pore surface area and thus increased sorption strength (Molina-Sabio et al., 1996).

Despite many studies observing strong sorption of different compounds onto biochar and other CM, the sorptive behavior of biochar is still not fully understood. For example, it is still unclear what specific properties of biochar, sorbate, or combination of both defines sorption. Unravelling such relationships is crucial to improve current mechanistic understanding and provide a better understanding for discrepancies in the literature, such as the variation in sorption coefficients up to a factor of 1000 for one single compound to biochar (Chen et al., 2008; Gomez-Eyles et al., 2013). Previous studies have focused primarily on the individual effects of temperature (Bornemann et al., 2007), feedstock (James et al., 2005), surface area and pore volume and structure (Graber et al., 2012), and sorbate properties (Zhang et al., 2013) on the sorption of organic compounds to biochars mostly made in the laboratory. Therefore, a systematic investigation of the multiple factors

involved in sorption for a wide range of commercial, field-made (“real-world”) and laboratory-made CM is needed in order to obtain a better understanding and prediction of the fate, transport, and risk of HOC in the environment. Such information will aid in the design and implementation of remediation strategies.

Thus, the main goal of the current study was to investigate sorptive properties of a wide range of biochars and other CM produced from different starting materials, production temperatures and methods, and to relate these variations in sorptive ability to the physico-chemical properties of the sorbate and sorbent.

The current study investigated the sorption of two common HOCs, 2,2',5,5'-tetrachlorobiphenyl (CB52) and phenanthrene (PHE), on 17 different CM with emphasis on biochar with the primary aim of establishing structure-reactivity relationships responsible for the observed variation in CM sorption characteristics. The materials studied were activated and non-activated CM produced from 13 different feedstocks at 250–800 °C, commercially, in the laboratory or in the field via slow pyrolysis (residence time 30 min – 8 h) and fast pyrolysis (residence time 5 s – 10–15 min) in the almost complete absence of oxygen. Also a wildfire charcoal was included. Physico-chemical properties of the sorbent materials were assessed via elemental, surface area and pore size distribution, aromaticity, and thermal stability analyses. Partition and adsorption effects were quantified via the construction of sorption isotherms. To our knowledge this is the first publication in which i) biochar sorption is related to biochar properties in such a comprehensive manner, ii) sorption to in situ “real-world” biochars is compared to sorption to laboratory-made ones and activated carbons, using a passive sampling method.

2. Materials and methods

2.1. Carbonaceous materials

The CM ($n = 17$) represented a wide range of sorbent materials from 13 feedstocks. These included biochars produced from hardwood (HW500, where the number indicates the pyrolysis temperature), digested dairy manure (DDM500), food waste (FW500), paper mill waste (PMW500), pinewood (PW, *Pinus ponderosa* – 250, 350, 500 and 700), switchgrass (SG700, *Panicum virgatum*) (Table 1). One biochar, derived from sustainable forests and produced at 550 °C for 10–15 min (fast pyrolysis biochars), was purchased from Cool Planet, US. Commercial activated carbon was prepared from coconut shell and purchased from Mallinckrodt U.S.P. (cocoAC). One biochar (termed non-activated biochar) was made from mixed wood sievings at 700 °C and a residence time of 20 min with a Pyreg 500W unit at Swiss Biochar, Switzerland. A representative sample of this biochar was vapor activated for 1 h at 900 °C in an activation bombe with a continuous through flow of 900 °C hot water vapor at 0.3 L/h per 100 g of biochar (activated biochar). One “real-world” tropical biochar was produced from maize cobs in a brick kiln in Zambia at a temperature of around 350 °C as monitored by a digital thermocouple. A natural (wildfire) char was collected after a wildfire in Colorado (Hayman Fire) in 2002. Two CM aged for 150 and 2000 years under natural conditions were collected from charcoal kilns at Pejo (46°20'N; 10°38'E) in the Italian Alps and in a soil profile in the Fango Valley (Fango 3,

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