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Impact of black carbon on the bioaccessibility of organic contaminants in soil

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HIGHLIGHTS

- BC sorbs hydrophobic organic compounds extremely strongly in soil.
- Sorption of organic contaminants to BC reduces toxicity and biodegradability in soil.
- BC has a significant impact on the bioavailability and bioaccessibility of contaminants to soil biota.
- The application of BC to soil may reduce the risk associated with organic contaminants.

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ABSTRACT

The ability of carbonaceous geosorbents (CGs) such as black carbon (BC) to extensively sorb many common environmental contaminants suggests that they potentially possess qualities useful to the sequestration of harmful xenobiotics within contaminated land. Presently, however, there is limited understanding of the implications for the bioaccessibility, mobility and environmental risk of organic contaminants while sorbed to BC in soil and sediment, in addition to the inherent toxicity of BC itself to terrestrial flora and fauna. We review both the processes involved in and factors influencing BC sorption characteristics, and ultimately consider the impacts BC will have for bioavailability/bioaccessibility, toxicity and risk assessment/remediation of contaminated land. We conclude that while the application of BC is promising, additional work on both their toxicity effects and long-term stability is required before their full potential as a remediation agent can be safely exploited.

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1. Introduction

Extensive research has greatly developed our understanding of the fate and behaviour of organic contaminants in soils and sediments [1–4]. Recently, there has been a focus on the influence of black carbon (BC) on the transformation of such compounds [5–8]. BC is a chemically heterogeneous class of carbon produced largely by the incomplete combustion of fossil fuels, wood and biomass, and is now found ubiquitously throughout the environment due to its widespread production and chemical and microbiological recalcitrance [9,10]. BC can be an exceptionally strong sorbent to a wide range of common environmental contaminants [10]; however, presently there is only limited understanding of the implications for the bioaccessibility, mobility and environmental risk of these contaminants while sorbed to BC in soil [10,11].

With increased biomass burning, coupled with amplified fossil fuel consumption, BC production has increased dramatically over the last century [12–14], with an estimated 50 million tonnes produced annually; the largest fraction (>80%) derived from burnt vegetation [15]. Previous studies assessing the implications of BC on the availability and mobility of toxic compounds in the environment have primarily focused on (i) BC-contaminant interactions, most notably sorption kinetics and sequestration [16–19] and (ii) bioavailability and bioaccumulation by benthic organisms [5,20–22] and plants [23,24]. However, only few investigations have attempted to quantitatively assess the impact of BC upon the bioavailability of organic contaminants in soil [7,10]. Such research may have potential implications for the risk assessment and bioremediation of contaminated land and is therefore essential to explore. As a result, this review intends to achieve three objectives: (i) to summarize the current understanding of the fate and behaviour of organic contaminants with particular emphasis on sorption and desorption; (ii) to evaluate the key findings of BC-contaminant interactions and (iii) to consider the impacts

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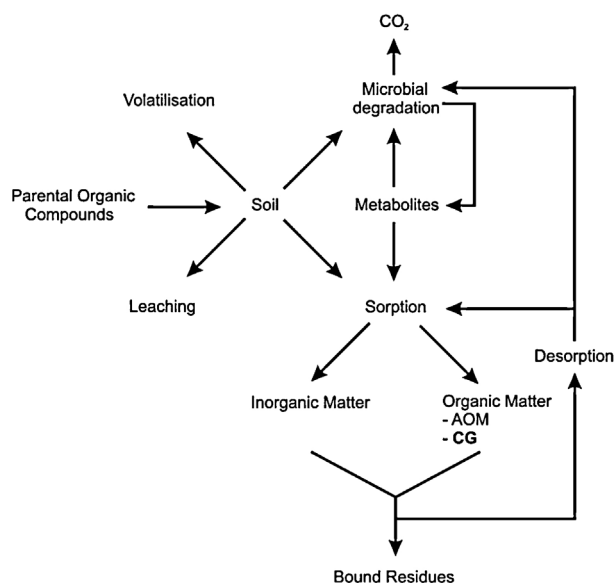


Fig. 1. Summary of the fate and behaviour of organic contaminants in soil. AOM: amorphous organic matter; CG: carbonaceous geosorbents.

Figure adapted from Stokes et al. [3] and Barraclough et al. [95] with the permission of John Wiley and Sons and Elsevier, respectively.

BC will have for bioavailability/bioaccessibility, toxicity and risk assessment/remediation of contaminated land.

2. The fate and behaviour of organic contaminants in soil

Organic compounds are exposed to a variety of processes in soil (Fig. 1), which ultimately determine their fate. Leaching, volatilization and degradation lead to the rapid loss of the readily available or labile fraction (typically in the aqueous phase). In contrast, contaminants may persist in soil and lead to the formation of the recalcitrant and/or irreversibly bound fractions [3,25]. The sequestration of compounds has been demonstrated to increase with time; a process termed ageing [26]. Such intra-soil processing is understood to lead to contaminant immobilization, diminished toxicological risk, and a reduction in bioavailability and chemical extractability [27–30]. These findings have therefore prompted investigations to elucidate the processes that cause these physical interactions.

Sequestration is believed to be a combination of (i) diffusion into inaccessible soil micropores and (ii) sorption to the soil matrix [31–33]. The latter process is considered to control ageing [25] and therefore is important for the fate and toxicological risk of organic contaminants [34]. Mechanisms of sorption include both physical (van de Waals forces) and chemical (chemisorption) interactions (a more extensive review is provided by Gevaio et al. [35]). Organic compounds typically show multiple stages in sorption and desorption profiles [36]. Rapid sorption is followed by slower stages that correlate with the recalcitrant and irreversibly bound fractions [3]. Similarly, desorption rates are initially rapid (i.e. hours to days) but then decrease to ‘slowly’ (weeks to months) and ‘very slowly’ (months to years) desorbing rates [37–39]. For example, Cornelissen et al. [40] found that the slowly desorbing fraction of PAHs, PCBs and chlorobenzenes had rate constants of 10^{-3} to 10^{-2} h $^{-1}$, whereas the very slowly desorbing fraction had rate constants of 10^{-5} to 10^{-4} h $^{-1}$. Sorption kinetics may be influenced by sorbate properties, for example sorption can be promoted in certain circumstances by increasing sorbate hydrophobicity [32], or aqueous concentration [41]. Soil pH and other characteristics are also crucial, in particular the organic matter (OM) fraction, which has long

been established as the most important binding site for organic pollutants [29]. Greater OM concentrations have been demonstrated to increase the size of the slowly desorbing pools of both PCBs and chlorobenzenes [42]. However, it is becoming increasingly evident that it is not only the OM size but type that determines sorption–desorption kinetics.

Reviews by both Cornelissen et al. [34] and Koelmans et al. [6] described the theoretical development of the ‘dual-mode’ sorption concept, whereby sorption mechanism varies as a function of OM type. Amorphous organic matter (AOM) is a relatively homogenous, lipophilic, gel-like matrix thought to consist of partly degraded or reconstituted biopolymers, amino acids, lignin and lipids and is thought to be responsible for compound absorption, whereby dissolution into the organic matrix occurs simply by diffusion [34]. In contrast, adsorption (i.e. accumulation on particle surfaces and interphases), has been experimentally demonstrated to occur onto ‘glassy’ and condensed types of OM, which Cornelissen et al. [34] collectively termed as ‘carbonaceous geosorbents’ (CG). As soil and sediment OM is chemically heterogeneous, both processes are likely to occur simultaneously, hence the ‘dual-mode’ concept (Fig. 1) [6,17,34].

3. Black carbon-contaminant interactions

One subdomain of these carbon-rich geosorbents is BC, a general term that encompasses unburned coal, kerogen, soot, fly ash, charcoal [34], and biochar (BC produced intentionally through biomass pyrolysis). As a result of the condensed and aromatic structure of BC [6,10], organic compounds including PAHs, PCBs [17,43–46], pesticides [47–50] and other substances of toxicological concern [51–53] have been shown to sorb extensively. The physicochemical properties of BC vary depending upon the source material, combustion conditions, state of weathering and mode of formation [54,55], with different implications for contaminant sorption [56]. However, only few studies have compared the sorptive behaviour among different BC forms and their intrinsic properties [11]. A summary of its main properties is provided in Table 1.

Sorption mechanisms, strengths and capacities of BC have been attributed to the source of BC [20,57] and the temperatures reached during organic matter combustion [56,58]. BC surface area and surface heterogeneity have also been identified as key components in determining sorption capacity [59]. In addition to the characteristics of the sorbent, properties of the sorbate such as molecular size, polarity and planarity also play an important role in BC-hydrophobic organic contaminants (HOC) sorption [60]. Planar compounds, such as PAHs [45] and planar PCBs [61,62], experience strong sorption to BC due to their ability to penetrate narrow pores in which they interact by π – π forces with the aromatic pore walls [60]. Similar bonding forces occur during the sorption of planar compounds to the BC surface [60]. Conversely, bulky atoms of non-planar compounds, such as highly chlorinated PCBs, may experience impaired electronic interactions with the surface and pore walls of the geosorbent due to steric hindrance caused by the small size of pores in BC, lowering overall sorption [34,60,63,64]. However, dipole interactions in some non-planar compounds, such as the pesticide diuron, can provide enhanced ability to achieve planar conformation and hence better sorption to BC [60,65].

In relation to AOM, sorption to BC has been observed to be 10–1000 times stronger [17,34,66], as indicated by elevated soil/sediment–water distribution coefficients [67]. AOM absorption is typically linear, rapid, reversible and mechanistically defined by the organic carbon-normalized partition coefficient (K_{oc}); whereas, BC exhibits slow sorption and desorption [54,67] that can be highly non-linear [17]. Methods to characterize these processes have employed both Langmuir and Freundlich models [19,48,54], as

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