



Effect of environmental exposure on charcoal density and porosity in a boreal forest



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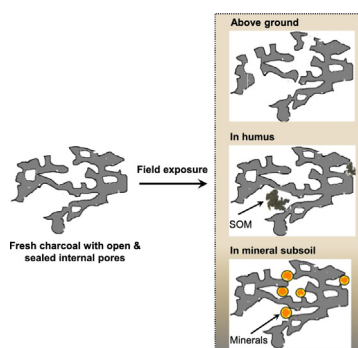
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HIGHLIGHTS

- Charcoal density and porosity changed detectably after 20 months' field incubation.
- The direction of change varied with soil horizon.
- The direction and magnitude of the environmental-driven shifts in charcoal density and porosity are ecosystem-specific.

GRAPHICAL ABSTRACT



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ABSTRACT

Although charcoal's density and porosity shape its environmental roles (e.g. controlling its landscape movement and determining the internal pore space available as microbial habitat), the rate of change of these properties in the environment remains largely unknown. With time, charcoal pores may fill or charcoal particles may shatter, altering the ecosystem services delivered. In this study we examined the effects of environmental exposure on the density and porosity of charcoals pyrolyzed at two different temperatures (350 and 520 °C). Fresh charcoal made from the dominant local tree (*Pinus sylvestris*) was returned in litter incubations to Norwegian boreal forest soils for 20 months in three different placements: (i) aboveground, (ii) in the humus layer, and (iii) under the humus layer in contact with the mineral subsoil. By varying soil horizon placement we were able to separate the effects of infill from the effects of environmental disturbance on charcoal density and porosity. Environmental exposure changed charcoal density and porosity, and the response varied with environmental placement. Charcoal placed in soil layers increased in porosity by ~1–2% after 20 months' incubation. This is likely because open indentations on the charcoal surface became partially occluded, creating more detectable pore space. In contrast, the porosity of charcoal incubated aboveground decreased slightly (~1–2% over 20 months). Because there were no minerals or humic substances to infill the aboveground charcoal samples, this porosity reduction was likely caused by breakage of particles induced by weathering. When charcoal particles cleave through pores, internal pore space is destroyed.

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The small changes observed here indicate that environmental exposure did not trigger rapid shifts in charcoal density and porosity. In addition, these physical properties appear not to have reached equilibrium after 20 months incubation, suggesting that the effect of environmental exposure on charcoal's physical properties occurs on the timescale of years to decades.

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1. Introduction

Charcoal enters the environment as a natural byproduct of fire and plays important roles in the global carbon cycle (Preston and Schmidt, 2006; Bird et al., 2015; Santin et al., 2015). Intentionally made charcoal (biochar) is also added to the environment for agricultural and environmental management purposes due to its potentially beneficial properties (Lehmann et al., 2011). Charcoal is a heterogeneous material exhibiting a wide range of physicochemical properties depending on its feedstock and production processes (e.g., fire temperature, fire exposure time). When produced at low temperatures, charcoal can contain incompletely combusted lignocellulosic fragments, whereas charcoal produced at high temperature can be structurally similar to completely ordered graphite (Hedges et al., 2000; Keiluweit et al., 2010; Wiedemeier et al., 2015). Indeed, charcoal produced from wildfire is often a heterogeneous and complex material that contains a suite of different components from low temperature fire biomarkers (e.g., anhydrosugars) to high temperature condensed aromatics (Singh et al., 2012; Gao et al., 2016). Although much is now understood about the fundamental characteristics of fresh charcoal, we know less about its properties change over time, and how these changes may impact the roles charcoal plays in soils.

To understand charcoal's environmental roles, for example its ability to provide ecosystem services and its record of the spatial scale of fires, we need to better understand charcoal's physicochemical properties and their post-depositional change with time. Such properties determine charcoal's transport in the environment and its role in soil hydrologic and ecological services. The effects of charcoal's chemical properties (e.g., aromaticity, pH, surface chemistry) on its degradation in the environment and vice versa have been extensively studied (e.g., Cheng et al., 2008; Nguyen et al., 2010). In contrast, we know little about charcoal's physical properties, which may also change with production conditions and environmental exposure. Prior studies have reported that charcoal was preferentially eroded from soils by horizontal movement under certain hydrologic conditions (Rumpel et al., 2006, 2009). Indeed, erosion by surface runoff was suggested as the largest mechanism of charcoal loss in soils (Major et al., 2010; Wang et al., 2013), but we do not yet understand the controls on key charcoal properties determining whether it remains in soil or migrates away.

Among its fundamental physical properties, density and porosity play a central role in charcoal's landscape behavior and fate in the environment. Charcoal can be a highly porous material (with total pore volume on the order of 55–85%), with pore size ranging from sub-nanometers to tens of micrometers depending on the feedstock and pyrolysis temperature (Brewer et al., 2014; Hardie et al., 2014). Macropores (>50 nm) that are important for water retention and microbial habitats often account for >95% of charcoal's total porosity, whereas mesopores (2–50 nm) and nanopores (<2 nm) that are important for charcoal's active surface area and nutrients retention only represent <5% of its total porosity (Warnock et al., 2007; Brewer et al., 2014; Gray et al., 2014; Hardie et al., 2014). Dry fresh charcoal with pores filled with air often has a bulk density < 1.0 g/cm³, suggesting that it will float when initially exposed to water. As water infiltrates into charcoal's air-filled pores, its bulk density gradually increases and it can eventually become denser than water. The time scale for this change varies from minutes to days depending on charcoal's total porosity, pore size distribution, and surface chemistry (Sorrenti et al.,

2016). This evolution of charcoal bulk density may significantly impact its transport in the environment. Charcoal dominated by small and/or poorly connected pores is likely to require a longer time to become saturated, remaining mobile longer and traveling farther, whereas charcoal dominated by large, well-connected pores may become saturated faster, remaining closer to its source of production.

In addition, charcoal's porosity and pore size distribution also impact its hydrological and ecological services in soils (Barnes et al., 2014; Hardie et al., 2014; Jeffery et al., 2015; Liu et al., 2016). Charcoal with higher pore volumes (total porosity) is more likely to increase soil total porosity and water holding capacity when introduced into the soil. Charcoal amendment not only increased the total soil water holding capacity, but also the plant available water (PAW) due to its wide range of pore size distribution (Liu et al., 2016). Larger pores cannot retain water under unsaturated conditions whereas small nanometer-scale pores retain water so strongly that is beyond plants' access (Herath et al., 2013; Brewer et al., 2014). Only water retained in pores ranging from ca. 0.2–9.0 μm in soils, corresponding to the water content between permanent wilting point and field capacity respectively, is ecologically relevant for plant survival and growth (Carter and Ball, 1993). Charcoal pore size distribution is also important for its suitability as an ecological niche for soil microbes (Herath et al., 2013; Liu et al., 2016). Micron-sized pores, which are abundant in biomass-derived charcoal and inherited from partially preserved plant cellular structures, are ideal to accommodate soil microbes, including most bacteria and many fungi (Warnock et al., 2007; Hardie et al., 2014).

Charcoal's physical, chemical, and biological properties may be altered with environmental exposure, causing long term shifts in its hydrologic, microbial, and nutrient ecosystem services (Zackrisson et al., 1996; Cheng et al., 2008; Spokas et al., 2014). Even though charcoal is considered a recalcitrant form of organic carbon (OC) with a half-life up to several centuries (Hammes et al., 2008; Bird et al., 1999; Kuz'yakov et al., 2009; Spokas, 2010; Santos et al., 2012; Singh et al., 2012; Fang et al., 2013; Maestrini et al., 2014), it is subject to environmental alterations and biodegradation in soils and its physicochemical properties may significantly change over time (Zackrisson et al., 1996; Cheng et al., 2008). For instance, charcoal's bulk density from that of fresh char with air-filled pores (ca., 0.3 g/cm³) may shift to values above that of water (i.e., >1.0 g/cm³) in soils over time (Sorrenti et al., 2016). When charcoal bulk density becomes >1.0 g/cm³ it is no longer expected to float and its landscape mobility is likely to be greatly reduced. Similarly, as pores infill, charcoal's ability to serve as an ecological niche for roots and microbes is reduced. After entering the environment, charcoal's density and porosity are likely substantially altered by multiple processes, including (i) chemical and physical weathering (Spokas et al., 2014), (ii) infilling of pores with soil minerals and/or soil humic substances (Bird et al., 2008; Lou et al., 2012), and (iii) biological alteration such as plant roots and microbial growth inside charcoal pores (Hockaday et al., 2006; Kasin and Ohlson, 2013; Jaafar et al., 2014).

Despite of its importance, few studies have examined these fundamental physical properties relative to the large volume of literature on charcoal chemical properties (Brown et al., 2006; Keiluweit et al., 2010; Mukherjee et al., 2011; Harvey et al., 2012). In particular, field studies are lacking that examining how charcoal's density and porosity evolve in the natural environment. This basic information is necessary before we can understand the role of charcoal's transport in redistributing its ecosystem services. One exception to this lack of information on

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