



Atmospheric black carbon deposition and characterization of biomass burning tracers in a northern temperate forest



F. Santos ^{a, b, *, 1}, M.P. Fraser ^c, J.A. Bird ^{a, b}

^a School of Earth and Environmental Sciences, Queens College, City University of New York, Flushing, NY, USA

^b The Graduate Center, City University of New York, New York, NY, USA

^c Global Institute of Sustainability, Arizona State University, PO Box 875402, Tempe, AZ, USA

HIGHLIGHTS

- Black carbon (BC), organic carbon, and molecular markers in fine particulates.
- Ambient PM_{2.5} was mainly influenced by biomass burning and plant waxes.
- Atmospheric BC dry deposition was a minor source of soil BC.

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ABSTRACT

Aerosol black carbon (BC) is considered the second largest contributor to global warming after CO₂, and is known to increase the atmosphere's temperature, decrease the albedo in snow/ice, and influence the properties and distribution of clouds. BC is thought to have a long mean residence time in soils, and its apparent stability may represent a significant stable sink for atmospheric CO₂. Despite recent efforts to quantify BC in the environment, the quantification of BC deposition rates from the atmosphere to terrestrial ecosystems remains scarce. To better understand the contribution of atmospheric BC inputs to soils via dry deposition and its dominant emission sources, atmospheric fine particle (PM_{2.5}) were collected at the University of Michigan Biological Station from July to September in 2010 and 2011. PM_{2.5} samples were analyzed for organic C, BC, and molecular markers including particulate sugars, carboxylic acids, *n*-alkanes, polycyclic aromatic hydrocarbons, and cholestane. Average atmospheric BC concentrations in northern Michigan were $0.048 \pm 0.06 \mu\text{g m}^{-3}$ in summer 2010, and $0.049 \pm 0.064 \mu\text{g m}^{-3}$ in summer 2011. Based on atmospheric concentrations, particulate deposition calculations, and documented soil BC, we conclude that atmospheric deposition is unlikely to comprise a significant input pathway for BC in northern forest ecosystem. The major organic tracers identified in fine particulates (e.g. levoglucosan and docosanoic acid) suggest that ambient PM_{2.5} concentrations were mainly influenced by biomass burning and epicuticular plant waxes. These results provide baseline data needed for future assessments of atmospheric BC in rural temperate forests.

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1. Introduction

Black carbon (BC), formed from the incomplete combustion of plant biomass and fossil fuels (Goldberg, 1985), is released to the

atmosphere at a global rate of ca. 8 Tg ($1 \text{ Tg} = 1 \times 10^{-3} \text{ Gt yr}^{-1}$) (Bond et al., 2004). BC affects climate by directly absorbing sunlight and increasing the temperature of the atmosphere, reducing the reflectivity of snow and ice surfaces, and either warming or cooling the atmosphere when interacting with clouds (Hadley and Kirchstetter, 2012; Bond et al., 2013, and references therein). Airborne BC particles are predominantly between 0.05 and 1 μm (Venkataraman and Friedlander, 1994), with a lifetime thought to vary from 2 to 11 days (Cooke and Wilson, 1996; Schulz et al., 2006). Given the current uncertainties in atmospheric BC concentrations and emission inventories (Bond et al., 2004, 2013), ground-based measurements of BC aerosol are critical to elucidating the

* Corresponding author. School of Earth and Environmental Sciences, Queens College, City University of New York, 65-30 Kissena Boulevard, New Science Building D-216, Flushing, NY 11367, USA.

E-mail addresses: fsantos@gc.cuny.edu, nandaara@hotmail.com, santosf@msu.edu (F. Santos).

¹ Present address: Department of Forestry, Michigan State University, 480 Wilson Road, Natural Resources Building, Room 129, East Lansing, MI, USA.

impacts of BC on regional and global climate. In soils, BC is reported to comprise a significant fraction of the stable carbon (C) pool (Schmidt et al., 1999), and may represent a sizable stable sink for C (Schmidt, 2004; Lehmann, 2007). Rates of BC addition to soils have been predicted to increase globally because of expected increases in wildfire frequency and intensity due to climate change (Westerling et al., 2006; Flannigan et al., 2013). Knowledge of the net contribution of BC to soil C will be needed to predict the long-term effects of forest fires on terrestrial C fluxes. However, BC deposition fluxes from the atmosphere remains a poorly quantified component of net BC additions to terrestrial ecosystems in temperate regions. Moreover, estimations of this total deposition of BC originating from biogenic aerosol sources (i.e. biomass burning) versus anthropogenic aerosol sources (i.e. fossil fuel combustion) are still scarce.

The stability of charred residues in soils can be influenced by the combustion temperature from which BC is generated (Baldock and Smernik, 2002). As charring temperature increases, the initial biomass undergoes physicochemical and structural changes, resulting in BC materials with larger proportions of highly condensed and presumably less degradable carbonaceous compounds (Hammes et al., 2006; Knicker et al., 2008; Keiluweit et al., 2010; Schneider et al., 2010). Thus, BC particles formed under lower combustion temperature (e.g. biomass burning) are expected to degrade faster than those formed under higher temperatures (e.g. fossil fuel combustion; Hammes et al., 2006; Turney et al., 2006). However, whether biomass- and fossil fuel-derived BC emission sources result in distinct degradation dynamics of atmospheric BC in soils remains unclear.

Biogenic (e.g. biomass burning) and anthropogenic sources (e.g. diesel fuel) of BC can be distinguished using source-specific molecular tracers (Simoneit, 1999). Among specific tracers for biogenic emission sources is levoglucosan, an anhydrous saccharide formed from the thermal degradation of cellulose (Simoneit et al., 1999; Fraser and Lakshmanan, 2000; Sullivan and Ball, 2012). Levoglucosan has been successfully used as a tracer for biomass burning worldwide (Zdráhal et al., 2002; Claeys et al., 2004; Jordan et al., 2006; Medeiros et al., 2006; Jia et al., 2010; Giannoni et al., 2012; Rada et al., 2012). Among the organic compounds that have been used to characterize fossil fuel emissions are *n*-alkanes that lack odd-C isomers enrichment found in contemporary biomass, petroleum biomarkers (e.g. cholestane), and polycyclic aromatic hydrocarbons (Simoneit, 1986; Fraser et al., 2000, 2002; Simoneit, 2002; Katianová et al., 2008; Wang et al., 2009). Bond et al. (2004) reported that fossil fuel/biofuel and open burning in North America may account for 5% and 1.5% of the global BC emissions, respectively. However, the relative contribution of plant biomass and fossil fuels to the total amounts of atmospheric BC deposition to soils remains poorly documented.

In this study we measured atmospheric BC concentration in fine particulates at a rural forested site in northern Michigan to estimate atmospheric BC deposition inputs to forest soils during summer 2010 and 2011. We calculated BC deposition based on measured BC to estimate the potential flux of BC to the local ecosystem. Finally, we quantified particle bound molecular marker compounds to identify the dominant emission sources of measured BC aerosol.

2. Materials and methods

2.1. Ambient aerosol sampling

Particulate matter with an aerodynamic diameter smaller than 2.5 μm ($\text{PM}_{2.5}$) was collected in summer 2010 and 2011 at the United States Department of Agriculture UVB measurement site (45.56 N and 84.68 W) located at the University of Michigan

Biological Station (UMBS) in Pellston, Michigan. The UMBS is located approximately 350 km and 400 km north from Detroit (MI) and Chicago (IL), respectively. The established sampling site is an open field (1 ha) surrounded by a mixed hardwood forest that represents the interface of northern temperate and boreal forest. Soil BC was quantified in a previous study using a benzene polycarboxylic extraction method, and averaged 3.3% of soil C (Yarnes et al., 2011).

In 2010, air samples were collected for 54 days between July 8 and September 15, while in 2011 samples were collected for 41 consecutive days between July 22 and September 1. Samples of $\text{PM}_{2.5}$ were collected on ashed (550 $^{\circ}\text{C}$, 8 h) and pre-weighed quartz-fiber filters (Whatman, 8 \times 10 in) by a high volume (1132 L min^{-1}) air sampler (Thermo Fisher Scientific, Franklin, MA, USA) with an air inlet located at 3 m above the ground (230 m above sea level). We collected field filter blanks (installed in the air sampler, but without air flow) to account for potential OC and BC background contamination. The air sampler was equipped with a size-selective inlet (Model 340 HVVI, MSP Corp, Shoreview, MN, USA) to only collect $\text{PM}_{2.5}$. In 2010, collection time per sample varied from 24 to 216 h periods, while in 2011 collection time varied from 24 to 72 h periods. After collection, samples were weighed and stored at -20°C until extraction. Average mass concentration of $\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$) was calculated dividing the mass of $\text{PM}_{2.5}$ (μg) by the total volume of air sampled (m^{-3}).

2.2. Organic carbon and black carbon

Organic carbon (OC) and elemental carbon (EC; a method-specific measurement related to the quantity of BC in the sample) were measured on a Thermal–Optical Carbon Transmittance analyzer (Sunset laboratory Inc., Tigard, OR) at Arizona State University following the method described by Birch and Cary (1996). Briefly, a small portion (1.5 cm^2) of the filter sample was subsampled and heated stepwise in a He atmosphere (i.e. 310 $^{\circ}\text{C}$ for 60 s, 475 $^{\circ}\text{C}$ for 60 s, 615 $^{\circ}\text{C}$ for 60 s, and 870 $^{\circ}\text{C}$ for 200 s), and then separately heated under an oxidizing atmosphere (10% O_2 , and He) at 550 $^{\circ}\text{C}$ (45 s), 625 $^{\circ}\text{C}$ (45 s), 700 $^{\circ}\text{C}$ (45 s), 775 $^{\circ}\text{C}$ (45 s), 850 $^{\circ}\text{C}$ (45 s), and 870 $^{\circ}\text{C}$ (120 s). At each temperature step, C was oxidized to carbon dioxide and then reduced to methane for quantification using a flame ionization detector. The precision of the method has been reported by Clements et al. (2014). The operational definition of OC includes C volatilized during heating in an inert atmosphere, which includes C pyrolysed to EC; while EC (hereafter referred to as BC) is operationally defined as C evolved during heating in an oxidizing atmosphere and corrected for pyrolysed OC. Average mass concentration of OC and BC ($\mu\text{g m}^{-3}$) was calculated we described in Section 2.1.

2.2.1. Atmospheric black carbon deposition

Dry deposition fluxes of atmospheric BC were estimated by multiplying BC concentration in filter sample and dry deposition velocity:

$$F = C \times V_d \quad (1)$$

where F is the dry deposition flux ($\mu\text{g m}^{-2} \text{s}^{-1}$), C is BC concentration ($\mu\text{g m}^{-3}$), and V_d is the dry deposition velocity (m s^{-1}), which is the inverse sum of the atmospheric aerodynamic, quasi-laminar sublayer, and surface or canopy resistances. We selected a lower- and an upper-limit V_d values of 7×10^{-4} and $1.5 \times 10^{-2} \text{ m s}^{-1}$, respectively, to represent a typical range of V_d for forest and grasslands (Pryor et al., 2008).

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