



Light absorption of organic carbon emitted from burning wood, charcoal, and kerosene in household cookstoves[☆]

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ABSTRACT

Household cookstove emissions are an important source of carbonaceous aerosols globally. The light-absorbing organic carbon (OC), also termed brown carbon (BrC), from cookstove emissions can impact the Earth's radiative balance, but is rarely investigated. In this work, PM_{2.5} filter samples were collected during combustion experiments with red oak wood, charcoal, and kerosene in a variety of cookstoves mainly at two water boiling test phases (cold start CS, hot start HS). Samples were extracted in methanol and extracts were examined using spectrophotometry. The mass absorption coefficients (MAC_λ, m² g⁻¹) at five wavelengths (365, 400, 450, 500, and 550 nm) were mostly inter-correlated and were used as a measurement proxy for BrC. The MAC₃₆₅ for red oak combustion during the CS phase correlated strongly to the elemental carbon (EC)/OC mass ratio, indicating a dependency of BrC absorption on burn conditions. The emissions from cookstoves burning red oak have an average MAC_λ 2–6 times greater than those burning charcoal and kerosene, and around 3–4 times greater than that from biomass burning measured in previous studies. These results suggest that residential cookstove emissions could contribute largely to ambient BrC, and the simulation of BrC radiative forcing in climate models for biofuel combustion in cookstoves should be treated specifically and separated from open biomass burning.

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1. Introduction

Light-absorbing organic carbon (OC), also termed as “brown carbon” (BrC), directly affects the Earth's radiative balance by absorbing incoming solar radiation (Ramanathan et al., 2001; Anderson et al., 2003; Bond and Bergstrom, 2006). Unlike black carbon (BC) – the most efficient light absorbing agent, which absorbs across the spectral range with a weak dependence on wavelength (λ) (Bond, 2001; Bond et al., 2013; Lack and Langridge, 2013), BrC light absorption is mainly observed at near-UV and

shorter visible wavelengths with a strong λ dependence (Kirchstetter et al., 2004; Laskin et al., 2015). Open biomass burning, including uncontrolled wildfire and prescribed agriculture or forest fires (Forrister et al., 2015; Washenfelder et al., 2015; Chakrabarty et al., 2016; Xie et al., 2017b), and biofuel use for household cooking and heating are significant primary sources of BrC (Zhang et al., 2016; Fleming et al., 2018). Additionally, a number of studies have investigated the formation of BrC through gas-phase photo-oxidation of volatile organic compounds (VOCs) from biogenic and anthropogenic sources (Iinuma et al., 2010; Nakayama et al., 2010; Updyke et al., 2012; Lambe et al., 2013; Lin et al., 2014; Liu et al., 2016; Xie et al., 2017a) and aqueous-phase photochemical processes (Powelson et al., 2014; Lin et al., 2015; De Haan et al., 2017), suggesting the existence of secondary BrC

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sources. Other potential BrC sources in urban areas, such as motor vehicle emissions, are rarely investigated (Xie et al., 2017b). Furthermore, the variability in the optical and chemical properties of BrC (e.g., spectral dependence and chromophore composition) from different sources is poorly characterized. Ambient and laboratory observations of BrC showed that compared with the water-soluble fraction of OC, the water-insoluble fraction contained more and stronger light-absorbing chromophores (Chen and Bond, 2010; Liu et al., 2013; Zhang et al., 2013; Cheng et al., 2016). The light absorption of biomass burning BrC is strongly dependent on burning conditions (e.g., temperature) (Chen and Bond, 2010; Saleh et al., 2014; Pokhrel et al., 2016; Xie et al., 2017b), and the majority of water extracts absorption was due to high molecular weight (>500 Da) components (Di Lorenzo and Young, 2016; Di Lorenzo et al., 2017; Wong et al., 2017).

Residential solid fuels (e.g., wood, coal) were burned during cooking and heating activities in developing countries, of which the smoke emissions are a leading human health risk factor (Smith and Mehta, 2003; WHO, 2009, 2014). In Asia and Africa, approximately 60–80% of the BC emissions are from residential solid fuel (e.g., coal and biomass) combustion (Cao et al., 2006; Bond et al., 2013); the emissions from residential solid fuels were estimated to contribute ~20% to the global OC budget (Bond et al., 2004). Solid fuel combustions in cookstoves can emit substantial particulate and gaseous pollutants, including PM_{2.5}, BC, OC, CO, NO_x, and SO₂ (Zhang et al., 2000). The emission factors (EFs) of these pollutants from household cookstoves have been intensively examined (Jetter et al., 2012; Shen et al., 2012, 2017, 2018; Qi et al., 2017; Wathore et al., 2017), aiming to reduce pollutant emissions by using improved cookstoves or clean fuels or a combination of them. However, the contribution of cookstove emissions to residential emissions is poorly constrained, and the BrC absorption from cookstove emissions is rarely studied. Sun et al. (2017) studied the light-absorbing properties of BrC from residential coal combustion, and found that the light absorption of BrC (350–850 nm) accounts for 26.5% of the total absorption (BrC + BC), suggesting the importance of accounting for BrC emissions from household cookstove in climate models.

This study aims to measure the light absorption of BrC from a variety of fuel-cookstove combinations. Specifically, the mass absorption coefficients (MAC, m² g⁻¹) of methanol-extracted OC in PM_{2.5} samples from cookstove emissions were compared across three types of fuels: red oak, charcoal, and kerosene. No previous study has investigated the light absorption of OC from the combustion of charcoal and kerosene – two important fuels used for cooking and lighting in developing countries. The light absorption of gas-phase OC positive artifact (i.e., OC adsorbed on a backup quartz filter downstream of a polytetrafluoroethylene membrane filter for PM_{2.5}) is also presented, which might partly represent the light-absorbing properties of semivolatile gas-phase OC (Chen and Bond, 2010). This study will help increase the understanding of sources of light-absorbing OC emitted to the atmosphere. It also supports estimates of the direct radiative effect due to BrC from cookstove emissions.

2. Methods

2.1. Cookstove emissions test facility

The cookstove emission tests were conducted at the U.S. EPA cookstove test facility in Research Triangle Park, NC. Details of the facility design, test protocol, and pollutant measurement were provided elsewhere (Jetter and Kariher, 2009; Jetter et al., 2012). Briefly, cookstove emissions were routed into a stainless steel hood connected to a primary and secondary dilution tunnel. An induced-

draft blower was used to provide filtered dilution air and hood air flows and maintain negative pressure through the entire system.

2.2. Fuels and cookstoves

As shown in Table S1, Supplementary information, three fuels (red oak (*Quercus rubra*) wood, lump charcoal (not briquettes), and 1-K kerosene) were burned in cookstoves designed to burn each specific fuel. Wood and charcoal are typical solid fuels in developing countries (Bonjour et al., 2013); kerosene has been an important liquid household fuel since the mid-19th century, and is widely used in developing countries for cooking and lighting (Lam et al., 2012). In this work, red oak is selected as representative biomass fuel and burned in five stoves. Both low (~10%) and high (~30%) moisture (on a wet weight basis) oak fuels were burned in a Jiko Poa stove; only low moisture oak fuel was combusted in the four other wood stoves. The wood stoves tested in this work (Table S1) ranged from the traditional 3-stone fire to a forced-draft fan type cookstove (EcoChula XXL) (Jetter et al., 2012, Shen et al., 2017, 2018). Low moisture charcoal was combusted in five charcoal stoves, and Grade 1-K kerosene (ASTM, 2013) was burned in two cookstoves (Butterfly Model 2412 and 2668). Each fuel-cookstove combination was tested in triplicate.

2.3. Water boiling test (WBT) protocol

The WBT protocol (version 4) (Global Alliance for Clean Cookstoves, 2014) was applied to determine cookstove power, energy efficiency and fuel use. The WBT protocol includes three test phases: (1) cold-start (CS) high power phase begins with the cookstove, pot and water at ambient temperature; (2) hot-start (HS) high power phase immediately follows the CS phase with the cookstove hot but the pot and water at ambient temperature; and (3) low-power simmer phase (SIM) begins with the cookstove hot and the water temperature maintained 3 °C below the boiling point. A modified protocol was used for the charcoal stove, which was described in detail by Jetter et al. (2012). Continuous pollutant emission measurements and filter sampling were performed during each phase of the WBT protocol.

2.4. Emissions measurement and sampling

Gaseous pollutants, including CO, CO₂, CH₄, and total hydrocarbons (THC), were monitored continuously at the primary dilution tunnel using infrared and flame ionization detector (FID) analyzers (models 200, 300-HFID and 300MFID; California Analytical; Orange, CA). Background concentrations of gaseous pollutants were measured with real-time instruments at least 10 min before and after each test and were subtracted from test concentrations. Particulate matter with aerodynamic diameter ≤2.5 μm (PM_{2.5}) was sampled isokinetically on quartz-fiber filters (Qf) and polytetrafluoroethylene (PTFE) membrane filters positioned in parallel and downstream of PM_{2.5} cyclones (URG; Chapel Hill, NC) at a flow rate of 16.7 L min⁻¹. The filter-based PM sampling was conducted in the primary dilution tunnel for low-emission fuel-cookstove combinations (e.g., forced-draft biomass stove), and in the secondary dilution tunnel for those with high emission (e.g., natural-draft biomass stove) to avoid overloading filters. In this study, PM_{2.5} samples for red oak-EcoChula XXL and kerosene stoves were collected in the secondary dilution tunnel. A backup quartz-fiber filter (Qb) was installed downstream of the PTFE filter, to estimate the adsorption of gas-phase OC (a measure of “positive artifact”) on the PM_{2.5} sample. PM_{2.5} mass (range 0.023–1.48 mg) was determined by measuring the PTFE filters gravimetrically with a microbalance (MC5, Sartorius, Germany) before and after sampling.

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