Atmospheric Environment 127 (2016) 347–354

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Light absorption by biomass burning source emissions

Yuan Cheng [a], Guenter Engling [b, *], Hans Moosmüller [b], W. Patrick Arnott [b, c], L.-W. Antony Chen [b, d], Cyle E. Wold [e], Wei Min Hao [e], Ke-bin He [a, f, g]

[a] State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, China
[b] Division of Atmospheric Sciences, Desert Research Institute, Reno, NV, USA
[c] Department of Physics, University of Nevada, Reno, NV, USA
[d] Department of Environmental and Occupational Health, University of Nevada, Las Vegas, NV, USA
[e] Fire Science Laboratory, USDA Forest Service, Missoula, MT, USA
[f] State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing, China
[g] Collaborative Innovation Center for Regional Environmental Quality, Beijing, China

HIGHLIGHTS

- Two distinct types of biomass smoke were identified by the MAE vs. OC/EC dependence.
- They also differed with respect to artifacts in the filter-based b_{abs} measurement.
- Present assessments of biomass burning BC emissions might be largely overestimated.
- Biomass burning emissions with high OC/EC ratios may include some liquid-like OM.
- These liquid-like organic components can complicate filter-based b_{abs} measurements.

ARTICLE INFO

Article history:
Received 21 July 2015
Received in revised form
1 December 2015
Accepted 17 December 2015
Available online 20 December 2015

Keywords:
Biomass burning
Light absorption
Black carbon
Photoacoustic
Liquid-like organics

ABSTRACT

Black carbon (BC) aerosol has relatively short atmospheric lifetimes yet plays a unique and important role in the Earth's climate system, making it an important short-term climate mitigation target. Globally, biomass burning is the largest source of BC emissions into the atmosphere. This study investigated the mass absorption efficiency (MAE) of biomass burning BC generated by controlled combustion of various wildland fuels during the Fire Laboratory at Missoula Experiments (FLAME). MAE values derived from a photoacoustic spectrometer (~7.8 m^2/g at a wavelength of 532 nm) were in good agreement with those suggested for uncoated BC when the emission ratios of organic carbon (OC) to elemental carbon (EC) were extremely low (i.e., below 0.3). With the increase of OC/EC, two distinct types of biomass smoke were identified. For the first type, MAE exhibited a positive dependence on OC/EC, while the overestimation of the light absorption coefficient (b_{abs}) by a filter-based method was less significant and could be estimated by a nearly constant correction factor. For the second type, MAE was biased low and correlated negatively with OC/EC, while the overestimation of b_{abs} by the filter-based method was much more significant and showed an apparent OC/EC dependence. This study suggests that BC emission factors determined by the commonly used thermal-optical methods might be sustantially overestimated for some types of biomass burning emissions. Our results also indicate that biomass burning emissions may include some liquid-like organics that can significantly bias filter-based b_{abs} measurements.

1. Introduction

Black carbon (BC) aerosol plays a unique and important role in the Earth's climate system through absorbing solar radiation, influencing cloud processes, and altering the melting behavior of snow and ice cover (Bond et al., 2013; Lack et al., 2014). As an important short-term climate mitigation target, however, BC has not been unambiguously defined yet due to the multitude of measurement principles. Consequently, reported BC data are dependent on the respective measurement techniques (Andreae

* Corresponding author.
E-mail addresses: ycheng@mail.tsinghua.edu.cn (Y. Cheng), Guenter.Engling@dri.edu (G. Engling).

http://dx.doi.org/10.1016/j.atmosenv.2015.12.045
1352-2310/

and Gelencsér, 2006; Baumgardner et al., 2012; Bond et al., 2013; Petzold et al., 2013; Buseck et al., 2014). In this paper, we adopt the terminology recommended by Petzold et al. (2013) and use the term BC to generally describe the carbonaceous materials in particles that are strongly light-absorbing in the spectral range of visible light, refractory, and insoluble.

Presently, global atmospheric absorption attributable to BC remains highly uncertain. The mass absorption efficiency (MAE; also known as the mass absorption cross section, MAC) is an essential factor converting BC mass concentration (in $\mu g/m^3$) simulated by chemical transport models to the absorption coefficient in the atmosphere (in m^{-1}) required for use in radiative transfer models. MAE of BC cannot be directly measured; instead, it is calculated as the absorption coefficient per BC mass (in this case, the determination of BC mass should be independent of optical absorption measurements).

Historically, the BC mass fraction of carbonaceous aerosol is most frequently quantified by filter-based methods such as thermal-optical measurements. BC mass determined by thermal-optical methods is commonly referred to as elemental carbon (EC). In thermal-optical analysis, EC can survive up to ~900 K in the presence of oxygen, and meanwhile, the removal (e.g., combustion) of EC leads to a rapid increase of the filter transmittance and reflectance which are typically monitored at 632 or 678 nm (Chow et al., 2004; Cavalli et al., 2010). Thus, EC is distinguished from other carbonaceous components (i.e., organic carbon) by its much higher thermal stability and considerably stronger light-absorbing capacity in the spectral range of red light. More recently, there have been growing applications of in-situ techniques to the determination of BC mass. Examples of these in-situ techniques include (1) aerosol mass spectrometric methods (e.g., the Aerosol Time-of-Flight Mass Spectrometer, ATOFMS, Pratt and Prather, 2012; the Particle Analysis by Laser Mass Spectrometry, PALMS, Murphy, 2007), (2) laser induced incandescence (LII) methods (e.g., the High Sensitivity Laser Induced Incandescence instrument, HS-LII, Schulz et al., 2006; the Single Particle Soot Photometer, SP2, Schwarz et al., 2006), and (3) the Soot particle Aerosol Mass Spectrometer (SP-AMS) which is a combination of the aerosol mass spectrometric technique and the LII technique (Onasch et al., 2012).

Despite several inter-comparison studies (e.g., Kondo et al., 2011b; Laborde et al., 2013), consistency of BC mass quantified by different types of measurement principles has not been well addressed. Nevertheless, each kind of technique has its uncertainty and limitations (Baumgardner et al., 2012). As for thermal-optical methods, a major problem is that a substantial fraction of organic species can be transformed into materials whose thermal and optical behaviors are quite similar to EC (these materials are usually termed char OC or pyrolysis OC; Yang and Yu, 2002). Previous studies conducted at urban locations such as Fresno, CA (Chow et al., 2004), Pittsburgh, PA (Subramanian et al., 2006), Milan, Italy (Piazzalunga et al., 2011), and Beijing, China (Cheng et al., 2012) typically suggested that thermal-optical methods tend to underestimate BC mass due to uncertainties introduced by char OC. For example, it was observed that water (Piazzalunga et al., 2011) or organic solvent (Cheng et al., 2012) extraction, which removes a substantial fraction of OC and consequently reduces the char OC formation (Subramanian et al., 2006), could lead to a considerable increase in the measured EC concentration of urban aerosol. However, uncertainties in the thermal-optical determination of BC mass have not been well characterized for primary emissions from combustion sources, limiting our ability to evaluate BC inventories and thus the modeled distribution and climate impacts of BC. This is particularly true for biomass burning which is the largest BC source globally (Bond et al., 2013).

On the other hand, the absorption coefficient is typically quantified by optical instruments, most of which are based on the measurement of the change in light transmittance through a filter due to the deposition of aerosol particles (Moosmüller et al., 2009). Examples of instruments using the filter-based technique include the Aethalometer (Hansen et al., 1984), the Particle Soot Absorption Photometer (PSAP; Bond et al., 1999), the Multi-Angle Absorption Photometer (MAAP; Petzold et al., 2005) and the continuous soot monitoring system (COSMOS; Kondo et al., 2011b). Of these instruments, the Aethalometer and PSAP are most commonly used. The latest version of the Aethalometer (model AE33) measures the absorption coefficient at seven wavelengths (i.e., 370, 470, 520, 590, 660, 880 and 950 nm; Drinovec et al., 2015), while the commercial single-wavelength PSAP has been modified to yield absorption measurements at three visible wavelengths (i.e., 467, 530 and 660 nm; Virkkula et al., 2005). Although having been widely used, most of the filter-based techniques suffer from various artifacts that need to be carefully addressed. The following corrections are likely necessary for correction of measured transmittance change (i.e., attenuation) to light absorption (Moosmüller et al., 2009; Lack et al., 2014): (1) the filter loading correction which compensates for the non-linear response of attenuation to the increase of BC loading (i.e., the shadowing effect), (2) the multiple scattering correction which accounts for the enhanced attenuation caused by the scattering of filter fibers, and (3) the particle scattering correction which corrects the enhanced attenuation due to the scattering of loaded particles. Significant efforts have been made to develop and improve empirical corrections for the filter-based instruments. Representative algorithms include those introduced by Weingartner et al. (2003), Arnott et al. (2005), Schmid et al. (2006), Collaud Coen et al. (2010) for the Aethalometer, and those described by Bond et al. (1999), Virkkula et al. (2005) and Ogren (2010) for the PSAP. More recently, an algorithm compensating for the shadowing effect, which is based on the attenuation measurements by two parallel spots, was incorporated into the latest version of the Aethalometer (Drinovec et al., 2015).

A difficult challenge to the filter-based techniques such as Aethalometer and PSAP is that the empirical parameters in the correction algorithms are highly variable depending on aerosol source, chemical composition, mixing state, etc. (e.g., Collaud Coen et al., 2010). Moreover, determination of these empirical parameters is not easily performed in the field, and therefore general corrections calculated during inter-comparison campaigns (e.g., Arnott et al., 2005) are usually applied to all Aethalometer or PSAP units. This is also the case for the COSMOS, although it could reduce the particle scattering effect by volatilizing a substantial fraction of non-BC components prior to particle collection (this is done by a thermal denuder; Kondo et al., 2011b). In contrast to the other filter-based instruments, the MAAP does not rely on empirical corrections; instead, it uses a radiative transfer model to retrieve the absorption coefficient from simultaneous measurement of light transmitted through and scattered back from a particle-loaded fiber filter (Petzold et al., 2005). The MAAP has been suggested to be the most reliable filter-based instrument for aerosol absorption measurements (Andreae and Gelencsér, 2006). However, a basic assumption used in MAAP, i.e., the particle-loaded filter can be treated as a two-layer system consisting of a particle-loaded filter layer and the particle-free filter matrix, might be invalid under some conditions (e.g., in the presence of liquid-like aerosols which will spread throughout filter fibers).

Artifacts associated with the filter-based measurement of aerosol absorption can be completely overcome by directly measuring light absorption on airborne particles. This technique is being used by the photoacoustic spectrometer (PAS; Arnott et al., 1999; Lack et al., 2006), which has become the “standard” method for aerosol absorption measurements (Lack et al., 2014).