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Quantifying organic aerosol single scattering albedo over the tropical biomass burning regions☆

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HIGHLIGHTS

- A creative method for determining OA SSA is suggested.
- Observation-based retrieval reveals OA SSA of 0.91.
- Observationally constrained global mean sulfate and nitrate AOD is 0.017.
- The global aerosol direct radiative forcing can be lower than currently believed.

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ABSTRACT

Despite growing evidence of light-absorbing organic aerosols (OAs), their contribution to the Earth's radiative budget is still poorly understood. In this study we derived a new empirical relationship that binds OA single scattering albedo (SSA), which is the ratio of light scattering to extinction, with sulfate + nitrate aerosol optical depth (AOD) and applied this method to estimate OA SSA over the tropical biomass burning regions. This method includes division of the attribution of black carbon (BC) and OA absorption aerosol optical depths from the Aerosol Robotic Network (AERONET) observation and determination of the fine-mode ratio of sea-salt and dust AODs from several atmospheric chemistry models. Our best estimate of OA SSA over the tropical biomass burning regions is 0.91 at 550 nm. Uncertainties associated with observations and models permit a value range of 0.82–0.93. Furthermore, by using the estimated OA SSA and comprehensive observations including AERONET, Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging Spectroradiometer (MISR), we examined the first global estimate of sulfate + nitrate AOD through a semi-observational approach. The global mean sulfate + nitrate AOD of 0.017 is in the lower range of the values obtained from 21 models participated in AeroCom phase II. The results imply that most aerosol models as well as climate models, which commonly use OA SSA of 0.96–1.0, have so far ignored light absorption by OAs and have overestimated light scattering by sulfate + nitrate aerosols. This indicates that the actual aerosol direct radiative forcing should be less negative than currently believed.

1. Introduction

During the last decade, light absorption by atmospheric aerosols has attracted significant attention because of its huge impact on regional climate and the Earth's radiation balance. In the solar spectrum, under cloud-free conditions, direct radiative forcing by atmospheric aerosols is usually negative at the top of the atmosphere (TOA); however, it can become less negative or even positive with increasing aerosol absorption (Boucher et al., 2013; Feng et al., 2013). Studies regarding aerosol light absorption have considered black carbon (BC) as a single light absorbing species. During the last 10 years, laboratory and field experiments have provided strong evidence of the fact that some organic aerosols (OAs) absorb a substantial amount of sunlight, whereas others only scatter sunlight (Kirchstetter et al., 2004; Andreae and Gelencsér, 2006;

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Alexander et al., 2008; Chen and Bond, 2010; Chung et al., 2012; Lack et al., 2012). The evidence for the presence of light absorbing OAs originates from the spectral dependence of light absorption that cannot be explained by BC absorption near specific combustion sources. OAs that absorb sunlight appear brownish owing to their strong solar absorption toward shorter visible and ultraviolet radiation; therefore, such OAs are known as brown carbon (BrC; Andreae and Gelencsér, 2006). The origins of atmospheric OAs are the incomplete combustion of biomass (Reid et al., 2005; Hoffer et al., 2006; Chakrabarty et al., 2010; Lack et al., 2013; Forrister et al., 2015) and fossil fuels (Bond, 2001; Yang et al., 2009; Zhang et al., 2011) and the secondary formation from gas-phase organic compounds (Laskin et al., 2014; Nakayama et al., 2013; Yu et al., 2014). The OAs make up more than 80% of fine-particle mass in the atmosphere in tropical savannas (Magi, 2009) where tens to hundreds of mega hectares are burned every year (Giglio et al., 2013) (Fig. 1). However, there are various OA source materials and chemical composition; therefore, it is difficult to generalize optical properties of OAs (Kanakidou et al., 2005).

The aerosol light absorption can be quantitatively expressed by using single scattering albedo (SSA) which is defined by the ratio of aerosol light scattering to aerosol light extinction (the sum of absorption and scattering). For non-absorbing aerosols the SSA is 1. It is crucial to exactly understand the SSA of aerosol since the sensitivity in aerosol direct radiative forcing is primarily driven by differences in SSA (Abel et al., 2005; Magi et al., 2008) rather than the absolute value of absorption, and hence even small error in its estimation can change the sign of aerosol radiative forcing (Yamamoto and Tanaka, 1972; Takemura et al., 2002; Kanakidou et al., 2005). Large uncertainties exist, however, in the measurement of OA light absorption from both laboratory experiments and field observations, which tend to cause substantial overestimation of OA light absorption. For example, the extraction of laboratory generated aerosol samples with organic solvents prior to thermochemical analysis generally shows greater absorption than those not extracted (Chen and Bond, 2010). In addition, the most commonly used in situ instruments for measuring aerosol light absorption, including the Aethalometer (Magee Scientific Co., Berkely, CA) and the Particle Soot Absorption Photometer (Radiance Research, Seattle, WA), tend to overestimate aerosol light absorption because the absorption is enhanced by backscattering and multiple scattering (Chow et al., 2009).

Providing long-term, continuous, and readily available aerosol observation, the ground-based worldwide network of automatic sun- and sky-scanning measurements, Aeronet Robotic Network (AERONET; Holben et al., 1998, 2001), has been used throughout the literature to determine aerosol optical properties near specific source regions (Russell et al., 2010; Giles et al., 2012). However, these observations yield column-integrated measurements and therefore include different aerosols and mixing states. To make the best use of AERONET data, there is a need for additional work in characterizing light absorption by individual aerosol species. However, due to the difficulties in separating contribution of individual species to total light absorption, only few studies tried to partition OA light absorption using AERONET (Bahadur et al., 2012; Chung et al., 2012).

The main purpose of this study is the development of an algorithm to derive OA SSA empirically and the determination of OA SSA over the tropical biomass burning regions. In particular, we develop an algorithm that binds OA SSA with sulfate + nitrate AOD mostly by using ground-based AERONET data. The algorithm partly employs the ensemble mean of model simulations. We apply this empirical algorithm to determine the OA SSA over the tropical biomass burning regions by using the sulfate + nitrate AOD measured during the Southern African Research Initiative field campaign in August and September 2000 (SAFARI-2000) (Swap et al., 2003). The uncertainties from observations and the retrieval algorithm are extensively considered in estimating the range of OA SSA. Our method is different from that of Bahadur et al. (2012) in that there is no assumption of OA light absorption at higher wavelengths, and the OA SSA is determined from the amount of measured sulfate + nitrate AOD. It should be also pointed out that there are large differences in individual aerosol composition among models, and the model diversity of each of aerosol component will introduce large uncertainties to the aerosol-associated solar direct radiative forcing (Kinne et al., 2006). The biases provide an additional motivation for examining observation-based retrieval of individual aerosol species such as light scattering sulfate and nitrate aerosols. We further provide the first global estimate of sulfate + nitrate AOD constrained mostly by observations using the empirically derived OA SSA.

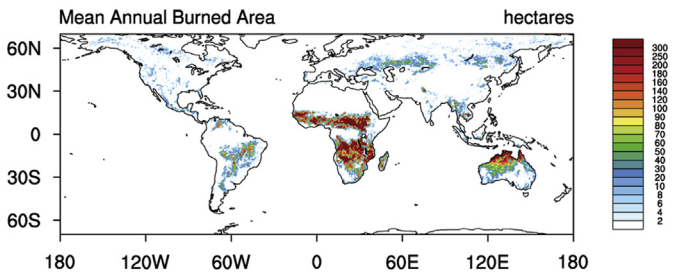

Fig. 1. Mean annual burned area in units of hectares at 0.25° spatial resolution derived from the January 1996 to December 2013 data of the monthly Global Fire Emissions Database fourth version (GFED4; Giglio et al., 2013). The annual mean was computed by the unweighted sum of 12-month data that were then averaged over 18 years.

2. Data and methods

2.1. Data

In this section, we describe the data obtained from the observations and global aerosol simulations. All data are monthly means and are interpolated to the T42 resolution, approximately 2.8° × 2.8°. Unless otherwise noted, we use optical variables at a wavelength of 550 nm. Table 1 shows a summary of the symbols and acronyms used in this paper.

2.1.1. AERONET

AERONET is a ground-based remote sensing maintained by the National Aeronautics and Space Administration (NASA) Goddard Space Flight Center (Holben et al., 1998, 2001). Although this automatic sun- and sky-scanning measurement does not cover the whole globe, its wide angular and spectral measurement provides most reliable and continuous aerosol optical properties in key locations (Dubovik et al., 2002). The AERONET AOD is known to be the most accurate global-scale product, and the uncertainty range

Table 1
Summary of acronyms and symbols used in this study.

Symbol	Acronym	Description
τ	AOD	Aerosol optical depth (from surface to TOA)
τ_a	AAOD	Absorption aerosol optical depth (=(1 − SSA) × AOD)
τ_f	fAOD	Fine-mode aerosol optical depth
τ_c	cAOD	Coarse-mode aerosol optical depth
α	AE	Ångström exponent for AOD
β	AAE	Absorption Ångström exponent
ω	SSA	Single scattering albedo