



Modification in light absorption cross section of laboratory-generated black carbon-brown carbon particles upon surface reaction and hydration



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HIGHLIGHTS

- Surface reactions and hydration of BC-BrC particles have been simulated.
- Neutralization between base gases and condensed sulfuric acid restructured BC-BrC.
- Restructuring of BC-BrC particles caused reduction in light absorption at low RH.
- Hydrophilic inorganic coating led to hygroscopic growth of internally mixed BC-BrC.
- Hygroscopic growth of processed BC-BrC particles enhanced light absorption at high RH.

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ABSTRACT

Recent reports indicate that the absorption enhancement of black carbon (BC) and brown carbon (BrC) particles is determined by the evolution of morphology and mixing state during the atmospheric processing. In this study, laboratory-generated BC-BrC mixture particles (BC-BrC) were exposed to sulfuric acid, ammonia/triethylamine, and water vapor sequentially to investigate the alteration in light absorption, morphology and mixing state during simulated atmospheric processing. Condensation of sulfuric acid and exposure to ammonia or triethylamine at 5% relative humidity decreased light absorption cross section of BC-BrC by 13%–26%, but subsequent hydration at 85% relative humidity led to an increase by 5%–20%. Our results show that surface reactions and hydration can significantly alter the light absorption cross section of BC-BrC. Our observation suggests that the restructuring of BC-BrC induced by neutralization reactions on the BC-BrC surface was responsible for the reduction in light absorption cross section at low relative humidity. On the other hand, the formation of an aqueous coating through water uptake by the hydrophilic inorganic coating at high relative humidity caused an absorption enhancement.

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1. Introduction

Black carbon (BC) has now been accepted as a strong absorber of visible solar radiation, leading to a positive (warming) radiative forcing that is estimated to be about one quarter to one half of the magnitude of atmospheric CO₂ (Bond et al., 2013; IPCC, 2007; Ramanathan and Carmichael, 2008). Recently, another class of light-absorbing carbon-containing substances (organic carbon, OC) in atmospheric aerosols, namely brown carbon (BrC), arises much

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scientific interests because its intense absorption at the shorter visible and UV wavelength may lead to underestimation of absorption of solar radiation in the troposphere (Andreae and Gelencsér, 2006).

Annually, 8.0 Tg BC and 33.9 Tg OC are estimated to be emitted at a global scale, and the contributions of biomass/biofuel burning to BC and OC are estimated to be 62% and 93%, respectively (Bond et al., 2004). Condensable organic species produced from either biomass burning (Lewis et al., 2008) or diffusion flames (Schnaiter et al., 2006), characterized by absorption in the visible light range with an absorption Ångström exponent (AAE) greater than unity, has been linked to the source of BrC. The presence of both BC and BrC in particulate matters formed from biomass/biofuel burning at flaming and smoldering conditions complicates the estimation of their climate impacts (Chakrabarty et al., 2010; Kirchstetter et al., 2004; Lack et al., 2012).

Atmospheric processing poses additional uncertainties in estimating the radiative forcing of BC particles. Once emitted into the atmosphere, BC particles undergo aging processes including condensation of low volatility species, coagulation with pre-existing particles, and potentially, cloud processing. Over a time-scale of a few hours to a few days, the aging processes lead to an internal aerosol mixture. Nonrefractory coating in the internally mixed BC particle is believed to act like a lens that enhances the light absorption by directing more light towards to the BC core (“lensing effect”), when compared with an equivalent external mixture (Jacobson, 2001). Theoretically, the extent to which absorption of BC can be enhanced depends critically on the ratio of the mass of nonrefractory particulate matter ([NR-PM]) to that of BC ([BC]) in the internally mixed particle ($R_{BC} = [NR - PM]/[BC]$) (Bond et al., 2006). A number of ambient studies on optical properties of aged BC (which usually contained amounts of BrC) measured absorption enhancement (E_{abs}) that differed in a large range from 1.06 to 1.7 with a large scale of R_{BC} (Cappa et al., 2012; Knox et al., 2009; Lack et al., 2012). The discrepancy can be partly explained by the different particle configuration caused by various atmospheric aging processes (Adachi and Buseck, 2013; Schnaiter et al., 2003, 2005).

Previous laboratory studies focused on absorption amplification of BC upon condensation of sulfuric acid and organic materials on BC surface (Cross et al., 2010; Schnaiter et al., 2005; Shiraiwa et al., 2010; Zhang et al., 2008). However, the condensed materials are subject to further atmospheric processing, such as surface reactions with gaseous species (Biskos et al., 2009; Wang et al., 2010) and hydration and dehydration cycles (Khalizov et al., 2009; Zhang et al., 2008). Water soluble coating (such as sulfuric acid and glutaric acid) leads to hydration and morphology modification of BC, both of which further amplify their absorption abilities (Khalizov et al., 2009; Mikhailov et al., 2006).

As one of the most abundant secondary chemical components of atmospheric aerosols, sulfate is usually found to be internally mixed with BC. They are believed to alter the morphology, hygroscopicity, and optical properties of BC particles based on the field observation (Fu et al., 2012; Li et al., 2013). However, there were only a few studies on BC internally mixed with ammonium sulfate/bisulfate generated either by coagulation (Schnaiter et al., 2003) or by an “evaporation-condensation” procedure (Stratmann et al., 2010). In the real atmosphere, sulfuric acid efficiently sinks on the BC surface (Riemer et al., 2004; Zhang et al., 2008) and subsequently ammonia/amine is transferred to the particulate phase in an effort to neutralize the acidic components forming ammonium/ammonium sulfate (Seinfeld and Pandis, 2006). Neutralization of the acidic coating by base gaseous species bring additional mass into the nonrefractory coating (i.e. increase in R_{BC}), which can lead to absorption amplification of BC. On the other hand, neutralization,

i.e. transformation of sulfuric acid to ammonium/ammonium sulfate/bisulfate could modify the hygroscopicity, density and phase of the overall coating (Chan and Chan, 2012; Qiu and Zhang, 2012; Tang and Munkelwitz, 1994). The influence of neutralization following the condensation of sulfuric acid on BC surface at different relative humidity (RH) is still unclear.

In this study, BC and BrC mixture particles (BC-BrC) were artificially produced from fuel-rich flames. Polycyclic aromatic hydrocarbons (PAHs), as the main OC components of our BC-BrC, were assigned to BrC because of their light absorption nature in line with previous investigations on particles from both fuel-rich flames (Apicella et al., 2004; Schnaiter et al., 2006) and wood combustion (Chen and Bond, 2010). We exposed our BC-BrC particles to sulfuric acid (H_2SO_4 , SA), ammonia (NH_3 , AM) or triethylamine ($(CH_3CH_2)_3N$, TEA), and water vapor sequentially to investigate the absorption and morphology alteration. The procedure simulated transfer of ammonia/amine to the particulate phase by neutralization with condensed sulfuric acid on the BC-BrC surface and potential hygroscopic growth of internally mixed BC-BrC due to hydrophilic inorganic coating in the atmosphere. Transmission electron microscope (TEM), energy dispersive X-ray spectroscopy (EDX), thermal-evaporation, and Mie calculation using a “core-shell” configuration were employed to investigate mechanisms for observed property variation. In addition, potential bias on absorption enhancement owing to the instrumental uncertainties was discussed.

2. Experimental methods

2.1. Particle generation and characterization

BC-BrC particles were generated in a laminar diffusion burner by combustion of methane (99.999%) at a flow rate of 0.2 L min^{-1} and air (99.999%) of 0.5 L min^{-1} . A flow of 0.05 L min^{-1} was sampled from the chimney and immediately diluted by 20-fold with nitrogen (99.999%), dried to $<0.5\% \text{ RH}$ by a silica gel diffusion drier and a Nafion tube (Perma pure), and then directed to further experimental usage. Authentic polystyrene latex spheres (PSL, Thermo Scientific) particles were generated using an atomizer (TSI 3670).

The extinction spectra and the chemical composition of BC-BrC were measured using an UV-Vis spectrophotometer and a gas chromatography-mass spectrometry (GC-MS), respectively (more details were described in the supplementary information, SI).

Fresh and processed particles were deposited onto a copper grid coated with a carbon film using a single-stage cascade impactor with a 0.5-mm diameter jet nozzle (Li and Shao, 2010), and analyzed by a transmission electron microscope (Jeol JEM-2100F) at an accelerating voltage of 200 KV and by an energy-dispersive X-ray spectrometer (EDX). A collection time of 30 s for each EDX spectrum was employed.

2.2. Processing of BC-BrC particles

Size distributions, mass, and hygroscopicity of particles were measured using a system comprising two differential mobility analyzers (DMA, TSI 3081), an aerosol particle mass analyzer (APM, Kanomax 3601), and a condensation particle counter (CPC, TSI 3771), as shown in the supplementary scheme. Briefly, polydisperse particles were brought to charge equilibrium by a Kr-85 neutralizer (TSI 3077), and then size-selected (D_0) by applying a fixed voltage to DMA1 (TSI, 3081). The obtained monodisperse particles (with a geometric standard deviation of 1.07) were exposed to sulfuric acid vapor in a 100 ml reservoir containing 20 ml 86 wt% H_2SO_4 solution ($25 \pm 0.1 \text{ }^\circ\text{C}$), directed to a 50 cm long 10-cm-i.d. residence chamber (growth reactor), and then through a potassium hydroxide (KOH)

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