Variation of the Radiative Properties during Black Carbon Aging: Theoretical and Experimental Intercomparison

Cenlin He¹, Kuo-Nan Liou¹, Yoshi Takano¹, Renyi Zhang², Misti Levy Zamora², Ping Yang², Qinbin Li¹, and L. Ruby Leung³

1. Department of Atmospheric and Oceanic Sciences and Joint Institute for Regional Earth System Science and Engineering, UCLA, CA 90095, USA
2. Department of Atmospheric Sciences, Texas A&M, College Station, TX 77845, USA
3. Pacific Northwest National Laboratory, Richland, WA 99352, USA
cenlinhe@atmos.ucla.edu

1. Motivations

- Black carbon (BC) atmospheric aging: freshly emitted BC aggregates coated by soluble materials through condensation, coagulation, and chemical oxidation.
- Laboratory experiments found substantial variations of radiative properties during BC aging, depending on coating material, thickness, and structures (e.g., Zhang et al., 2008; Shinina et al., 2010; Gao et al., 2012).
- Atmospheric observations showed complex structures for BC coated by other aerosol components (Adachi and Buseck, 2013; China et al., 2013), significantly affecting BC radiative properties (Cappa et al., 2012; Lack et al., 2012).
- This study develops a theoretical BC aging model to quantitatively understand the evolution of BC optical properties during aging and compares it with experiments.

2. Methods

- Theoretical BC Aging Model

- Laboratory Experiment

- BC aging after exposure to sulfuric acid vapors at different relative humidity (Zhang et al., 2008; Khatiwada et al., 2006).
- (1) BC mass: aerosol particle mass (APM) analyzer; (2) BC size: tandem differential mobility analyzer (TDMA); (3) BC extinction (532 nm): cavity ring-down spectrometer (CRDS) (4) BC scattering (532 nm): integrating nephelometer.

3. Results

- Observed optical cross sections of fresh BC aggregates are much more sensitive to fractal dimension than the size of primary spherules. Aggregate structure enhances BC absorption and scattering compared with mass-equivalent sphere.
- The off-center core-shell structure shows up to 30% less absorption and scattering than the concentric core-shell structure for thick coating.
- The open-cell structure tends to have weaker absorption and stronger scattering, while the reverse is true for the closed-cell structure.
- The partially encapsulated and externally attached structures have substantially smaller absorption and scattering.

- The absorption enhancement during aging varies by factors of 2-3, depending on BC size, coating structure, and aging stage.
- The increase in scattering is much stronger than absorption, ranging from a factor of 3 to 24.

- This study shows that BC absorption and scattering are highly sensitive to coating states (e.g., coating thickness, morphology, and composition), suggesting the evolution of BC coating states during aging in the real atmosphere could exert significant impacts on BC radiative properties and thus its climatic effects.
- Thus, a reliable estimate of BC radiative effects requires the incorporation of a dynamic BC aging process accounting for realistic coating amount, structure, and composition in climate models.

Fig. 1. A theoretical BC aging model that accounts for three evolution stages. Six typical structures for coated BC are considered based on observations.

Fig. 2. A graphical representation of the geometric-optics surface-wave (GOS) approach for the calculation of light absorption and scattering of coated BC aggregates.

Fig. 3. Laboratory measurements and theoretical calculations of BC extinction, absorption, and scattering cross sections (532 nm) for three aging stages with different initial BC diameters. The measured optical cross sections are generally captured (differences <30%) by theoretical calculations for fresh BC aggregates and coated BC (Stages II and III).

Fig. 4. Sensitivities of BC structures to optical properties at Stages I, II, and III with different initial sizes based on theoretical calculation.

Fig. 5. Enhancement in BC absorption and scattering cross sections during aging from Stage I to II and III.