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Monitoring optical properties of aerosols with cavity ring-down spectroscopy

Ling Li [a], Jianmin Chen [a,c,*], Hui Chen [a], Xin Yang [a,c], Yong Tang [a], Renyi Zhang [a,b]

[a] Center for Atmospheric Chemistry Study, Department of Environmental Science & Engineering, Fudan University, Shanghai 200433, China
[b] Department of Atmospheric Sciences, Texas A&M University, College Station, Texas, TX 77843, USA
[c] Institute of Global Environment Change Research, Fudan University, Shanghai 200433, China

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ABSTRACT

This article describes the design and performance of a cavity ring-down spectroscopic (CRDS) instrument for measuring extinction coefficients of laboratory and ambient atmospheric aerosols. Through averaging 1000 individual waveforms, a minimum detectable aerosol extinction coefficient of $6.1\times10^{-7}\,m^{-1}$ is achieved. Tests with polystyrene spheres (PSS), we suggested this CRDS system could measure the extinction coefficient of aerosol with uncertainty $<3\%$ under laboratory controlled experimental conditions. The visual range measured with CRDS agrees well with visibility observations from Shanghai Meteorological Bureau. Combined with the TSI integrating nephelometer and NO_x analyzer, CRDS was used to monitor the optical properties of ambient aerosols in the heavy pollution episode. The uncertainty for using the CRDS and TSI nephelometer to measure single scattering albedo (SSA) in an ambient measurement is estimated to be $<12\%$.

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1. Introduction

Aerosol particles are major component of urban pollution. They influence climate directly by scattering and absorbing of an incoming solar radiation and indirectly by acting as cloud condensation nuclei. Recently, much attention has been paid to the global estimate of direct radiative forcing, due to aerosols. The investigations estimate that the light absorbing aerosols yield a warming effect ranging 0.16–0.80 W m^{-2} (Chung & Seinfeld, 2002; Haywood & Shine, 1995; Haywood & Ramaswamy, 1998; Haywood & Boucher, 2000; Jacobson, 2001; Liao & Seinfeld, 2005 Menon, Hansen, Nazarenko, & Luo, 2002; Wang, 2004). This wide range of radiative forcing suggests that there remains a significant uncertainty which limits our ability at present to quantify the effect of human emissions on climate. To reduce the uncertainties for estimating aerosol radiative forcing, accurate measurements of aerosol scattering and absorption are crucial. Unlike scattered light, the absorbed photons should disappear and cannot be sensed directly. Measuring absorption accurately has been proven to be more difficult than measuring scattering. Traditional measurements of an atmospheric absorption, the Particle Soot Absorption Photometer (PSAP) and Aethalometer, measure light reflected from or transmitted through a particle-laden filter (Bond, Anderson, & Campbell, 1999; Sheridan et al., 2005). Most published atmospheric aerosol absorption and single scattering albedo data are based on these measurements with an error in the range 20–35% (Bond et al., 1999;

* Corresponding author at: Center for Atmospheric Chemistry Study, Department of Environmental Science & Engineering, Fudan University, Shanghai 200433. China. Tel.: +86 21 65642298; fax: +86 21 65642080.
E-mail address: jmchen@fudan.edu.cn (J. Chen).

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Magi, Hobbs, Schmid, & Redemann, 2003) and 30–35% (Schmid et al., 2006; Weingartner et al., 2003) respectively. These filter based techniques require many correction factors that limit the quality of the derived data. Recently, photoacoustic spectroscopy has been used to measure absorption for laboratory and atmospheric aerosol particles (Arnott, Moosmuller, Rogers, Jin, & Bruch, 1999; Arnott et al., 2003; Lack et al., 2008). In this technique, light absorbed by particles heats the surrounding air, resulting in an acoustic signal that can be detected with a microphone. This method shows promise for rapid, real-time measurement of absorption, but further development is required to reduce the interference due to heat and mass transfers when volatile species evaporate, especially under high relative humidity (RH) conditions (Arnott et al., 2003; Raspet, Slaton, Arnott, & Moosmuller, 2003).

The absorption technique of cavity ring-down spectroscopy was first introduced by O'Keefe and Deacon (1988). For direct absorption spectroscopy, the CRDS has advantages compared with traditional absorption techniques. CRDS is largely immune to shot-to-shot variations of the laser intensity, and also benefits from long effective path length in absorption (Busch & Busch, 1999). It has become a widely used new tool for measurement of trace species (Mercier, Therssen, Pauwels, & Desgroux, 2001; Morville, Romanini, Kachanov, & Chenevier, 2004; Schocker, Höinghaus, & Brockhinke, 2005) in gaseous environments and kinetic studies (Choi, Park, & Lin, 2003; Czyzewski et al., 2002; Nizamov & Dagdigian, 2003). Published results demonstrate a minimum detectable absorption coefficient (noise-equivalent absorption) ranging 10^{-6} cm^{-1} (in the UV range)–10^{-10} cm^{-1}(Berden, Peeters, & Meijer, 2000; Busch & Busch, 1999). CRDS has been recently introduced for measuring extinction coefficients of laboratory and ambient aerosols. (Baynard et al., 2007; Dinar et al., 2008; Moosmuller, Varma, & Arnott, 2005; Khalizov, Xue, Wang, Zheng, & Zhang, 2009; Pettersson, Lovejoy, Brock, Brown, & Ravishankara, 2004; Timothy, Johanna, & Andrew, 2007; Xue, Khalizov, Wang, Zheng, & Zhang, 2009Zhang et al., 2008). Typically, it consists of two highly reflective plano-concave mirrors set opposite one to another. A pulse of laser light is injected into the cavity, and it bounces back and forth between the mirrors. A photomultiplier (PMT) is placed at the other side of the cavity and measures the exponential decay of the emerging light intensity. The amount of light transmitted through the mirror is proportional to the amount of light still trapped inside the cavity. As a result, the time behavior of the light intensity inside the cavity can be determined by monitoring the rate of the decay of light exiting the mirror.

In the case of a cavity free of scattering or absorbing particles, the light intensity within the cavity will decrease exponentially according to the following relationship

$$i(t) = i_0 \exp\left[-(1-R)\frac{tc}{L}\right], \tag{1}$$

where i_0 is the initial light intensity, $i(t)$ is the light intensity after time t, c is the velocity of light, R is the reflectivity of the mirrors, and L is the optical length of the cavity. Considering now a cavity that contains an absorbing or scattering medium, the total loss in the cavity will be given by

$$i(t) = i_0 \exp\left\{-\left[(1-R)+\alpha_{ext} l\right]\frac{tc}{L}\right\}, \tag{2}$$

from which the time constant τ for the decay will be given by

$$\tau = \frac{L}{c[(1-R)+\alpha_{ext} l]}, \tag{3}$$

where α_{ext} is the extinction coefficient of particles inside the cavity and l is the effective sample path length. The extinction coefficient α_{ext} can be given directly through the expression

$$\alpha_{ext} = \frac{L}{lc}\left(\frac{1}{\tau}-\frac{1}{\tau_0}\right), \tag{4}$$

τ_0 is the time constant for the cavity filled with particle free gas, which is obtained by passing the sample through a filter that removed the aerosol. In this work, we discuss the design and performance of a cavity ring-down spectroscopic instrument for measuring aerosol extinction coefficient. Combined with a TSI intergrating nephelometer and NO_x analyzer, this system was used to measure the optical properties of the ambient atmospheric aerosols.

2. Experiment

2.1. Aerosol generation and classification

PSS particles are nebulized with an atomizer (Model 3076, TSI Inc.) using pure nitrogen. The aerosol flow enters a silica gel diffusion dryer to remove bounded water, resulting in a flow with RH $<$ 10%. The dry aerosol passes through a neutralizer to obtain an equilibrium charge distribution and then through a differential mobility analyzer (DMA, Model 5080, TSI Inc.) to select a designed particle size. The size-selected monodisperse aerosol flow is directed to the CRDS cavity for extinction coefficient measurement. A condensation particle counter (CPC, Model 3776, TSI Inc.) was connected to the outlet of the CRDS cavity to measure particle number concentration.

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