Supplemental material for ACPD manuscript “Photoacoustic optical properties at UV, VIS, and near IR wavelengths for laboratory generated and winter time ambient urban aerosols

M. Gyawali[1], W. P. Arnott[1], R. A. Zaveri[2], C. Song[2], H. Moosmüller[3], L. Liu[4], M. I. Mishchenko[4], L.-W. A. Chen[3], M. C. Green[3], J. G. Watson[3], J. C. Chow[3]

[1]Physics Department, University of Nevada, Reno, Nevada System of Higher Education, 1664 N. Virginia Street, Reno, NV, 89557, USA

[2]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA, 99352, USA

[3]Desert Research Institute, Nevada System of Higher Education, 2215 Raggio Parkway, Reno, NV, 89512, USA

[4]NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA

Photoacoustic instrument and calibration

S.1 Instrument description

The photoacoustic instrument (PA) is one of the few devices that provide the fundamental in-situ measurement of light absorption coefficients for suspended particles. The detailed working mechanism, measurement accuracy, evaluation and calibration of PA have been described in the early work (Arnott et al., 2000; Arnott et al., 2005a; Arnott et al., 2005b; Lewis et al., 2008; Moosmüller et al., 1998). Here we describe the basic principle and calibration technique applied for the 355 nm instrument. In the operation of the PA, sample air is continuously introduced into the acoustic resonator section and is illuminated by laser radiation, power-modulated at the acoustic resonance frequency. The radiation is absorbed by particles within the sample air, and is assumed to be immediately transferred to the surrounding air as heat. The associated pressure change contributes to a standing wave in the resonator that is detected by a microphone (Arnott et al., 1999). The simultaneous light scattering measurement is carried out with a cosine-weighted sensor positioned at the centre of the cavity to act as a reciprocal nephelometer (Lewis et al., 2008; Abu-Rahmah et al., 2006). The sensor is fiber coupled to a photomultiplier tube (PMT) for light scattering detection. A piezoelectric transducer positioned at an antinode of the resonator cavity is used periodically to determine the resonance frequency and quality factor of the resonator. The absorption and scattering coefficients measurements have 5% and 15% uncertainty for sub-2.5-micron diameter particles (Lewis et al., 2008).

Light absorption and scattering measurements in this study were made simultaneously with a dual wavelength PA operating at 405 and 870 nm (Lewis et al., 2008), and two other single wavelength PAs operating at 532 and 1047 nm (Arnott et al., 2000; Arnott et al., 2005b). Prior to the ambient measurements, all PAs were calibrated and their performance was evaluated by generating a wide range of absorbing and scattering aerosol in the laboratory as described by Lewis et al. (2008).

The light absorption coefficient, β_{abs}, in dimension of inverse length is obtained from,

$$\beta_{abs} = \frac{P_m}{P_L}\frac{A_{res}}{\gamma - 1}\frac{\pi^2 f_o \cos(\phi_m - \phi_L)}{Q} \quad , \tag{S.1}$$

where P_m is the measured microphone power at the resonance frequency f_o, P_L is the laser power at the resonance frequency, A_{res} is the resonator cross-sectional area, Q is the quality factor, γ is the ratio of isobaric and isochoric specific heats of air, and ϕ_m *and* ϕ_L are the electric phases of the microphone pressure and laser power, respectively. The resonance frequency and quality factor depend on temperature, pressure, and relative humidity. For the sample air, these quantities are measured by pressure, temperature, and relative humidity sensors located in the sample line downstream of the resonator.

The scattering coefficient, β_{sca} in dimension of inverse of length, is obtained from

$$\beta_{sca} = \alpha \frac{|P_{PMT}|}{|P_L|} \quad , \tag{S.2}$$

where α is the calibration factor determined at the time of instrument calibration and $|P_{PMT}|$ and $|P_L|$ are the magnitudes of Fourier transform of the PMT and laser power at the resonance frequency, respectively.

S.2 Calibration

A high concentration of laboratory-produced scattering (table salt) and absorbing (kerosene soot) aerosols were used for scattering and absorption calibration of PA, which resulted in a measurable attenuation of the laser beam intensity in the resonator. The salt was dissolved in water and nebulizers were used to produce airborne droplets from the salt solution. Dried salt aerosols were introduced into the resonator for scattering calibration. The extinction of light passing through aerosol can be expressed using Bouger-Lambert's law,

$$I = I_o \exp(-\beta_{ext} * L_{res}) \ , \qquad \text{(S.3)}$$

where I and I_o are the laser power with and without salt aerosol in the resonator, β_{ext} is the extinction coefficient of the aerosol, and L_{res} is the length of the resonator, 24.86 cm. The aerosol extinction coefficient calculated from equation (S.3) is compared with the scattering measurements to obtain the scattering calibration factor of equation (S.2). Fig. S (a) shows the linear regression between the measured extinction and scattering in which the near unity value of R^2 depicts the excellent correlation between these quantities. Also note the slope of the regression line is near unity, which means the extinction was overwhelmingly due to scattering and no further adjustments are needed for the scattering calibration. After the scattering calibration, the process was repeated with kerosene soot for absorption calibration. Figure S (b) shows the linear regression between the absorption measurements and extinction-scattering calculation. The slope of the regression line indicates that measured absorption coefficients at 355 nm should be increased by a factor of 1.29.

References:

Abu-Rahmah, A., Arnott, W. P. and Moosmüller, H.: Integrating nephelometer with a low truncation angle and an extended calibration scheme, Meas. Sci. Technol., 17, 1723-1732, 2006.

Arnott, W. P., Zielinska, B., Rogers, C. F., Sagebiel, J., Park, K., Chow, J., Moosmüller, H., Watson, J. G., Kelly, K., Wagner, D., Sarofim, A., et al.: Evaluation of 1047-nm Photoacoustic Instruments and Photoelectric Aerosol Sensors in Source-Sampling of Black Carbon Aerosol and Particle-Bound PAHs from Gasoline and Diesel Powered Vehicles, Environ. Sci. Technol., 39, 5398-5406, 2005a.

Arnott, W. P., Hamasha, K., Moosmüller, H., Sheridan, P. and Ogren, J.: Towards aerosol light-absorption measurements with a 7-wavelength Aethalometer: Evaluation with a photoacoustic instrument and 3-wavelength nephelometer, Aerosol Sci. Tech., 39, 17-29, 2005b.

Arnott, W. P., Moosmüller, H., Rogers, C., Jin, T. and Bruch, R.: Photoacoustic spectrometer for measuring light absorption by aerosols: Instrument description, Atmos. Environ., 33, 2845-2852, 1999.

Arnott, W. P., Moosmüller, H. and Walker, J.: Nitrogen dioxide and kerosene-flame soot calibration of photoacoustic instruments for measurement of light absorption by aerosols, Rev. Sci. Instrum., 71, 4545-4552, 2000.

Arnott, W. P., Walker, J., Moosmüller, H., Elleman, R., Jonsson, H., Buzorius, G., Conant, W., Flagan, R. and Seinfeld, J.: Photoacoustic insight for aerosol light absorption aloft from meteorological aircraft and comparison with particle soot absorption photometer measurements: DOE Southern Great Plains climate research facility and the coastal stratocumulus imposed perturbation experiments, J. Geophys. Res., 111, D05S02, doi:10.1029/2005JD005964, 2006.

Lewis, K., Arnott, W. P., Moosmüller, H. and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, J. Geophys. Res., 113, D16203, doi: 10.1029/2007JD009699, 2008.

Moosmüller, H., and Arnott, W. P., Rogers, C. F., Chow, J. C., Frazier, C. A., Sherman, L. E. and Dietrich, D. L.: Photoacoustic and filter measurements related to aerosol light

Figure Caption:

Fig. S. Plots for 355 nm calibration curves for (a) scattering measurements of negligibly absorbing salt aerosol, and (b) absorption measurements of kerosene soot.

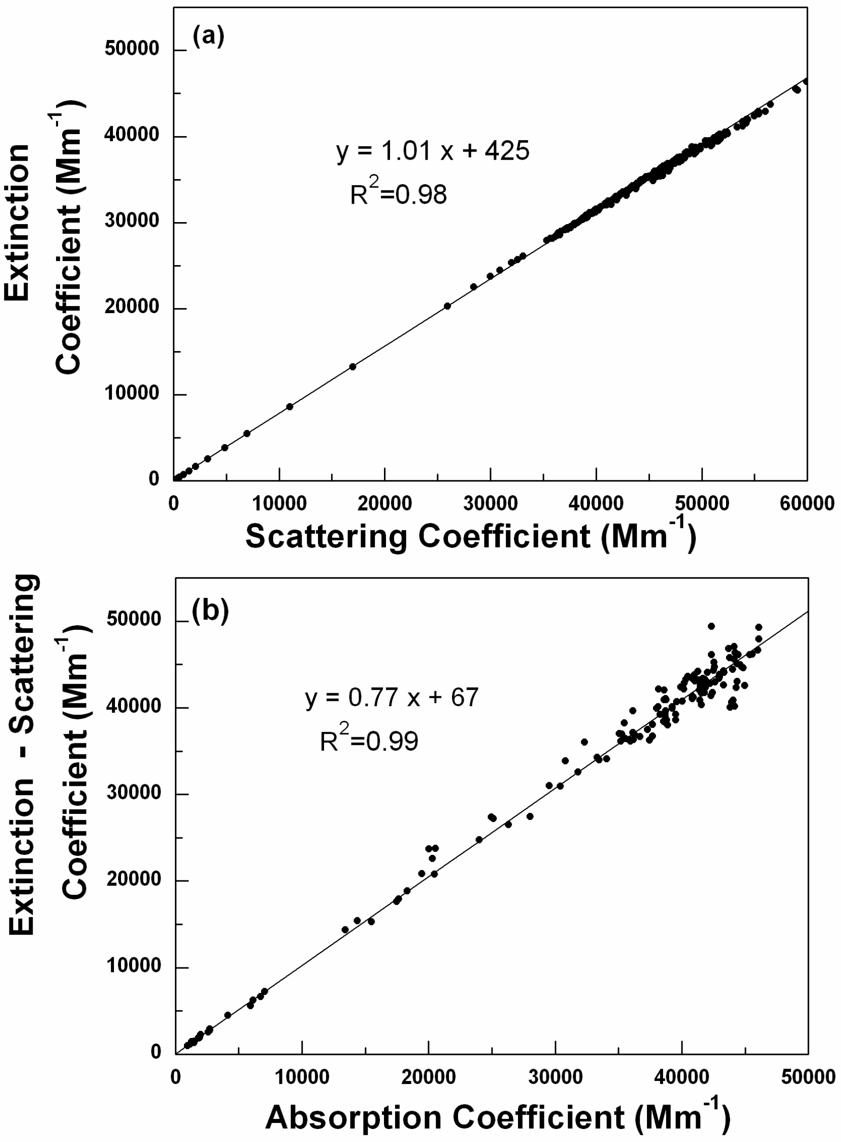
(a)
y = 1.01 x + 425
R^2=0.98
Extinction
Coefficient (Mm^-1)
Scattering Coefficient (Mm^-1)
0
10000
20000
30000
40000
50000
60000
(b)
y = 0.77 x + 67
R^2=0.99
Extinction - Scattering
Coefficient (Mm^-1)
Absorption Coefficient (Mm^-1)
0
10000
20000
30000
40000
50000

Fig. S