Published in final edited form as:

J Quant Spectrosc Radiat Transf. 2017 January ; 187: 145–149. doi:10.1016/j.jqsrt.2016.09.026.

Light Source Effects on Aerosol Photoacoustic Spectroscopy Measurements

James G. Radney* and **Christopher D. Zangmeister**

National Institute of Standards and Technology, Material Measurement Laboratory, 100 Bureau Dr., Gaithersburg, Maryland, 20899, USA

Abstract

Photoacoustic spectroscopy measurements of flame-generated soot aerosol coated with small amounts of water yielded absorption enhancements that were dependent on the laser used: quasi-continuous wave (Q-CW, ≈ 650 ps pulse duration and 78 MHz repetition rate) versus continuous wave (CW). Water coating thickness was controlled by exposing the aerosol to a set relative humidity (RH). At ≈ 85 % RH, the mass of the soot particles increased by an amount comparable to a monolayer of water being deposited and enhanced the measured absorption by 36 % and 15 % for the Q-CW and CW lasers, respectively. Extinction measurements were also performed using a cavity ring-down spectrometer (extinction equals the sum of absorption and scattering) with a CW laser and negligible enhancement was observed at all RH. These findings demonstrate that source choice can impact measurements of aerosols with volatile coatings and that the absorption enhancements at high RH previously measured by Radney and Zangmeister (2015) [1] are the result of laser source used (Q-CW) and not from an increase in the particle absorption cross section.

Keywords

Photoacoustic Spectroscopy; Aerosol; Absorption Enhancement; Coatings

1. INTRODUCTION

Photoacoustic spectroscopy is a well-documented technique for measuring the absorption of solids, liquids, gases, thin films and aerosols. The first demonstration of aerosol photoacoustic spectroscopy was made in the late 1980's [2, 3], although advancements in laser and microphone technology in the late 1990's [4, 5] caused a rapid increase in the number of aerosol photoacoustic studies.

In aerosol photoacoustic spectroscopy (PAS), intensity modulated light is directed towards aerosol particles suspended in a gas. Absorbed light energy can then evaporate volatile coatings, catalyze chemical reactions, be re-emitted radiatively (fluorescence or phosphorescence) or be re-emitted thermally. In the case of thermal emission, the local temperature increase drives adiabatic expansion of the carrier gas thereby generating a

*Corresponding Author james.radney@nist.gov (J. Radney) Phone: (301) 975-3904. Fax: (301) 975-3670.

pressure wave (i.e. sound) that can be detected by a microphone. Whenever the absorbed energy is dissipated through means other than thermal emission, the photoacoustic response is dampened. In order to generate an acoustic wave, the light source must be intensity modulated; continuous application of light will generate a temperature gradient but not a pressure wave (i.e. maxima and minima are required).

The photoacoustic response of an aerosol sample is dependent upon the strength of the light source used and the absorption coefficient (α_{abs}) of the sample – i.e. the absorption strength per unit propagation distance – with α_{abs} being a function of the number density of absorbing particles (N) and the absorption cross section of those particles (C_{abs}): $\alpha_{\text{abs}} = NC_{\text{abs}}$. Since only the former is controllable in the experimental setup, it is customary to use sufficiently bright sources such as lasers to perform PAS measurements. However, photoacoustic measurement uncertainties related to the type of laser used has received little attention. In previous investigations, pulsed [6–8], quasi-continuous wave [1, 9, 10] and continuous wave [2, 5, 11–14] lasers have been used with the latter being the most common.

Here, we compare the photoacoustic response of monodisperse flame-generated soot exposed to water vapor using a quasi-continuous wave (Q-CW, 650 ps pulse duration with 78 MHz repetition rate) and continuous wave (CW) laser at $\lambda = 660$. Water vapor is used to impart a thin coating on the particles with coating thickness being controlled by the relative humidity (RH). It has been predicted that absorption should increase relative to coating thickness [15] due to refractive index matching between the medium and coating [16]. We present these enhancements as the apparent increase in C_{abs} as a function of RH, and hence coating thickness, using both the Q-CW and CW lasers. We also compare C_{abs} to similar measurements of the extinction cross section (C_{ext} , sum of absorption and scattering cross sections) made using a cavity ring-down spectrometer with a CW laser. We find that at the highest RH measured ($\approx 85\%$), the soot particles uptake approximately a single monolayer of water and C_{abs} is enhanced by 35 % and 15 % for the Q-CW and CW lasers, respectively, while no enhancement was observed in C_{ext} . These findings demonstrate that the choice of light source can affect aerosol PAS measurements, especially when the particles contain volatile coatings as is common when sampling from the ambient atmosphere.

2. EXPERIMENTAL METHODS

2.1. Aerosol Generation and Conditioning

Soot aerosols were generated using a Santoro diffusion flame [17] with ethylene fuel and a 25 L min^{-1} sheath flow around the flame to keep the flame laminar. Ethylene delivery was controlled with a mass flow controller set such that the diffusion flame ran lean in an effort to physically remove larger particles bearing charges greater than +1 at the selected mobility diameter (350 nm) by shifting the size distribution to smaller particles; the reader is referred to the discussion in Radney and Zangmeister (2016) [18] for more details regarding the separation of particles bearing multiple charges using a tandem differential mobility analyzer-aerosol particle mass analyzer (DMA and APM, respectively). Experiments were made across multiple days and the flow of ethylene was adjusted such that soot with similar size distributions and C_{ext} were obtained for each experiment. Particles were aspirated into a 12.7 mm O.D. stainless tube through a 1 mm opening situated 5 cm above flame centerline

and mixed with 5 L min⁻¹ of dry particle-free laboratory air. An ejector pump was situated downstream and operated with 10 L min⁻¹ of dry, particle-free laboratory air. Of the 15 L min⁻¹ of total flow, 0.5 L min⁻¹ was sampled for conditioning and measurement.

Soot was conditioned by passing through a pair of diffusion dryers prior to size-selection via electrical mobility using a DMA operated at a 10:1 sheath:aerosol flow. The DMA was maintained under dry conditions (< 10 % RH) to ensure particles of a constant 350 nm mobility diameter (D_m) were selected. Particles were then passed through a large diameter Nafion [19] dryer/humidifier where the water partial pressure was controlled using a humidity generator that supplied ≈ 11 L min⁻¹ of air parallel to the aerosol flow. The RH of the exiting air stream was monitored by an RH and temperature probe. Prior to measurement, the RH was allowed to stabilize at the desired set-point for 5 min. Humidified particles were then passed through an aerosol particle mass analyzer, either the photoacoustic (PA) or cavity ring-down (CRD) spectrometer and a condensation particle counter (CPC).

2.2. Photoacoustic Spectrometer

The PA used is identical to the one described in Radney and Zangmeister (2015) [1] except that both a diode laser and a supercontinuum laser were utilized to allow for comparison between CW and Q-CW measurements, respectively, at $\lambda = 660$ nm. For Q-CW measurements, the supercontinuum laser was fiber coupled to a tunable wavelength and bandpass filter (TWBF) set to a center wavelength of 660 nm and a bandwidth of 15 nm which produced ≈ 8.8 mW root-mean-squared (RMS) power as measured by a calibrated power meter situated at the exit of the PA cell; the sampling rate of the power meter is 250 kHz, therefore the measured Q-CW power represents an average (650 ps pulse duration with 78 MHz repetition rate). The output of the TWBF was passed in free-space through a mechanical chopper operated at the resonant frequency of the acoustic cavity (nominally ≈ 1.64 kHz in ambient air at 296 K) [20]; as a result, the Q-CW measurements possessed both ultrasonic (78 MHz) and sonic (≈ 1.64 kHz) components. For CW, the diode laser was passed in free space and was similarly modulated by the mechanical chopper and measurements were performed with the laser at full power (≈ 32 mW RMS, normal mode of operation) and low power (≈ 10 mW RMS) for comparison to the Q-CW laser.

2.3. Cavity Ring-Down Spectrometer

The cavity-ring down spectrometer is identical to the one described in Radney and Zangmeister (2016) [18]. Briefly, light from the CW laser was injected into a high-finesse optical cavity until saturation (≈ 100 μ s). The light was then quickly terminated (10s of nanoseconds) using an acousto-optic modulator. The intra-cavity light intensity then decays passively and exponentially due to the scattering and absorption of light by aerosols, gases and the high-reflectivity mirrors ($R > 99.98$ %, transmission ≈ 0.002 %). To determine extinction coefficients, the difference between aerosol-laden and empty-cavity (i.e. HEPA-filtered) is calculated; empty-cavity ring-down times were nominally ≈ 17 μ s.

3. RESULTS & DISCUSSION

The absorption enhancement of soot was measured for particles with a dry ($RH < 10\%$) D_m of 350 nm. Soot particle mass versus RH is shown in Fig. 1a; error bars represent the 1σ width of the Gaussian-shaped distribution [18]. The multiple points at low ($\approx 5\%$) and high ($\approx 80\%$) RH represent data collected across multiple days; solid gray line corresponds to average mass at low RH. The low RH to high RH average mass increased from 2.96 ± 0.13 fg to 3.11 ± 0.12 fg (2 times uncertainty of the mean), a statistically significant change ($p < 0.05$). If we assume, based on previous measurements in our laboratory [21], that the average diameter of a soot monomer produced by the flame is 17 nm and has a density of 1.8 g cm^{-3} , the dry particles contain ≈ 639 monomers. Thus, the mass increase in going from low to high RH (0.15 fg) corresponds to a final monomer diameter of 17.5 nm which is marginally greater than a single monolayer of water. This simple calculation neglects any impact of particle necking and overlap in monomer contact areas, which would decrease the total surface area and increase water coverage.

The C_{ext} and C_{abs} of soot as a function of relative humidity were measured at $\lambda = 660 \text{ nm}$ and are shown in Fig. 1b as the black circles and colored squares, triangles and diamonds, respectively. The squares (red), triangles (green) and diamonds (blue) correspond to C_{abs} measured using the Q-CW and the CW laser at full ($\approx 90 \text{ mW}$ peak-to-peak) and low power ($\approx 28 \text{ mW}$ peak-to-peak), respectively. The data points correspond to the mean measured values while the error bars represent the 34th and 68th percentiles from all measurements spanning multiple days. The solid grey line corresponds to the average C_{ext} at low RH. As shown in the figure, C_{ext} and C_{abs} have similar values at the lowest humidity, independent of laser source or power for the photoacoustic measurement, with a co-albedo ($C_{\text{abs}}/C_{\text{ext}}$) of $0.97^{+0.08}_{-0.06}$ calculated from the average C_{ext} and the full power CW C_{abs} spanning all days. We attribute the high co-albedo in part to the day-to-day variability of the measured soot cross sections (see discussion in ensuing paragraph). As humidity increases, C_{ext} remains nearly constant, while C_{abs} shows a slight increase when using the CW laser and a larger increase when using the Q-CW laser. As a diagnostic, the soot particles were dried to $RH < 10\%$ after passing through the humidification tube ($RH \approx 85\%$) and aerosol particle mass analyzer and the measured C_{abs} was identical to the value obtained for particles that were not exposed to high RH.

We note that the reported uncertainties are dominated by inter- versus intra-day variation; relative uncertainties in the uncoated (dry) C_{ext} and C_{abs} (full power CW) are 15 % versus 2 % and 7 % versus 4 %, respectively. For PAS measurements, the absolute uncertainty is dominated by microphone noise and is independent of signal causing the lower power measurements to exhibit larger uncertainties. Comparable inter- and intra-day variability in C_{abs} at $\lambda = 405 \text{ nm}$ has been observed for uncoated soot using a similar Santoro diffusion flame in Havey, et al. (2010) [12], Bueno, et al. (2011) [22] and Radney, et al. (2014) [21].

From C_{abs} , the humidity dependent absorption enhancement ($f_{\text{abs}}(RH)$) was calculated as

$$f_{\text{abs}}(RH) = \frac{C_{\text{abs},RH}}{C_{\text{abs},\text{dry}}}$$

and is shown in Fig. 1c. The quantities $C_{\text{abs},\text{dry}}$ and $C_{\text{abs},RH}$ are the absorption cross sections measured at low ($< 5\%$ RH) and a higher RH, respectively. The humidity dependent extinction enhancements ($f_{\text{ext}}(RH)$) are defined similarly and are also shown in Fig. 1c. Similar to C_{abs} and C_{ext} in Fig. 1b, the data points in Fig. 1c correspond to the mean $f(RH)$ values while the error bars represent the 34th and 68th percentiles from all measurements spanning multiple days.

For the measured extinction, negligible enhancement is observed with increasing humidity. For absorption, a moderate enhancement is observed for the CW laser, independent of incident power (Mann-Whitney $p > 0.05$): $f_{\text{abs}}(RH) = 1.11^{+0.04}_{-0.01}$ and $1.17^{+0.09}_{-0.07}$ for the full power and low power measurements at 85 % RH, respectively; uncertainties represent the 34th and 68th percentiles spanning multiple days. At low humidity the absolute C_{abs} values differ by 5 % between the low and high power CW measurements and likely a result of measurement uncertainty. As shown in Fig. 1b, this causes C_{abs} to vary by 11 % at high humidity, even though the relative enhancements (Fig. 1c) do not exhibit a statistically significant difference. A larger and statistically significant enhancement is observed for the Q-CW laser: $f_{\text{abs}}(RH) = 1.36^{+0.10}_{-0.07}$ at 85 % RH.

The absorption enhancement by nanomaterials with thin transparent coatings arises from improved refractive index matching between the particle and air by the water coating [16]; in other work this effect is incorrectly referred to lensing [23, 24] (a geometric optics effect) where the transparent coating acts as a lens directing incident radiation towards the absorbing core. Refractive index matching can moderately increase the observed absorption, but is unlikely to account for the magnitude of enhancement observed in this case. This can be demonstrated by assuming that each soot monomer behaves as an independent subunit of the whole (i.e. Rayleigh-Debye-Gans approximation, RDG) and has a refractive index of $1.77 + 0.8i$ at $\lambda = 660$ nm – the value determined for flame-generated black carbon at $\lambda = 660$ nm in You, et al. (2016) [10]. Therefore, each dry monomer contributes $2.23 \times 10^{-17} \text{ m}^2$ to the measured C_{abs} ; for the roughly 639 monomers in an aggregate, this would correspond to an aggregate C_{abs} of $1.42 \times 10^{-14} \text{ m}^2$ which is within reason of the values shown in Fig. 1. Upon humidification to 80 % RH, which imparts a water shell of 0.5 nm (refractive index = 1.33 at $\lambda = 660$ nm), the C_{abs} of a monomer increases to $2.27 \times 10^{-17} \text{ m}^2$, a difference of 2 % and within the experimental uncertainty of the C_{ext} data, but well outside of the measured C_{abs} . Different values of the soot refractive index change the calculated $f_{\text{abs}}(RH)$ but not to the magnitude observed here. Further, as the CRD did not measure an extinction enhancement, it is likely that another mechanism is affecting the photoacoustic data. Importantly, the measured C_{abs} are larger than the corresponding C_{ext} values for $RH > 25\%$.

In solution, nanoparticle photoacoustic response enhancements have been observed using short-pulse (femto- to nano-second pulses) [25–31] and CW lasers [32, 33] and have been attributed to explosive vaporization of water at the particle/water interface. Notably, water

efflux scales as the inverse of pulse duration [34]; shorter pulses impart energy to the particle at a higher rate than the medium can conduct it away thereby creating a temperature jump at the interface, overheating the water and resulting in explosive vaporization [35]. If a similar mechanism is occurring presently, the PA could not separate the contributions from the explosive vaporization of water and the inherent photoacoustic response as explosive vaporization occurs on the timescale of the heat conduction (MHz) which is much faster than the response of the microphone used (kHz); i.e. Nyquist theorem.

4. Conclusions

In the present investigation, soot coated with water exhibited apparent absorption enhancements of 15 % and 36 % at 85 % RH using CW and Q-CW lasers, respectively. Negligible enhancement was observed in the corresponding extinction measurements. These results demonstrate that understanding the physical response of an aerosol to the source used can impact data interpretation. In reference to Radney and Zangmeister (2015) [1], we hypothesized that judicious choice of laser pulse duration could potentially negate signal dampening when performing aerosol PAS measurements under humidified conditions. Instead, the present data clearly illustrate that the previously observed enhancements were a function of the particle's response to the laser used. Further investigations may be required to elucidate if the observations are also dependent on laser power.

REFERENCES

- [1]. Radney JG, Zangmeister CD. Measurement of Gas and Aerosol Phase Absorption Spectra across the Visible and Near-IR Using Supercontinuum Photoacoustic Spectroscopy. *Anal Chem.* 2015; 87:7356–63. [PubMed: 26098142]
- [2]. Adams KM. Real-time in situ measurements of atmospheric optical absorption in the visible via photoacoustic spectroscopy. I: Evaluation of photoacoustic cells. *Appl Opt.* 1988; 27:4052–6. [PubMed: 20539513]
- [3]. Adams KM, Davis LI Jr, Japar SM, Pierson WR. Real-time, in situ measurements of atmospheric optical absorption in the visible via photoacoustic spectroscopy—II. Validation for atmospheric elemental carbon aerosol. *Atmos Environ.* 1989; 23:693–700.
- [4]. Moosmüller H, Arnott WP, Rogers CF, Chow JC, Frazier CA, Sherman LE, et al. Photoacoustic and filter measurements related to aerosol light absorption during the Northern Front Range Air Quality Study (Colorado 1996/1997). *J Geophys Res: Atmos.* 1998; 103:28149–57.
- [5]. Arnott PW, Moosmüller H, Rogers FC, Jin T, Bruch R. Photoacoustic spectrometer for measuring light absorption by aerosol: instrument description. *Atmos Environ.* 1999; 33:2845–52.
- [6]. Ajtai T, Filep Á, Schnaiter M, Linke C, Vragel M, Bozóki Z, et al. A novel multi-wavelength photoacoustic spectrometer for the measurement of the UV–vis-NIR spectral absorption coefficient of atmospheric aerosols. *J Aerosol Sci.* 2010; 41:1020–9.
- [7]. Haisch C, Menzenbach P, Bladt H, Niessner R. A Wide Spectral Range Photoacoustic Aerosol Absorption Spectrometer. *Anal Chem.* 2012; 84:8941–5. [PubMed: 23035870]
- [8]. Tikhomirov AB, Firsov KM, Kozlov VS, Panchenko MV, Ponomarev YN, Tikhomirov BA. Investigation of spectral dependence of shortwave radiation absorption by ambient aerosol using time-resolved photoacoustic technique. *Opt Eng (Bellingham, WA, U S).* 2005; 44:071203–11.
- [9]. Sharma N, Arnold IJ, Moosmüller H, Arnott WP, Mazzoleni C. Photoacoustic and nephelometric spectroscopy of aerosol optical properties with a supercontinuum light source. *Atmos Meas Tech.* 2013; 6:3501–13.

- [10]. You R, Radney JG, Zachariah MR, Zangmeister CD. Measured Wavelength-Dependent Absorption Enhancement of Internally Mixed Black Carbon with Absorbing and Nonabsorbing Materials. *Environ Sci Technol*. 2016; 50:7982–90. [PubMed: 27359341]
- [11]. Lack DA, Lovejoy ER, Baynard T, Pettersson A, Ravishankara AR. Aerosol Absorption Measurement using Photoacoustic Spectroscopy: Sensitivity, Calibration, and Uncertainty Developments. *Aerosol Sci Technol*. 2006; 40:697–708.
- [12]. Havey DK, Bueno PA, Gillis KA, Hodges JT, Mulholland GW, van Zee RD, et al. Photoacoustic Spectrometer with a Calculable Cell Constant for Measurements of Gases and Aerosols. *Anal Chem*. 2010; 82:7935–42. [PubMed: 20804170]
- [13]. Wiegand JR, Mathews LD, Smith GD. A UV–Vis Photoacoustic Spectrophotometer. *Anal Chem*. 2014; 86:6049–56. [PubMed: 24905953]
- [14]. Krämer L, Bozoki Z, Niessner R. Characterisation of a Mobile Photoacoustic Sensor for Atmospheric Black Carbon Monitoring. *Anal Sci*. 2002; 17:s563–s6.
- [15]. Bond TC, Habib G, Bergstrom RW. Limitations in the enhancement of visible light absorption due to mixing state. *J Geophys Res*. 2006; 111:D20211.
- [16]. Yablonoivitch E. Statistical ray optics. *J Opt Soc Am*. 1982; 72:899–907.
- [17]. Santoro RJ, Semerjian HG, Dobbins RA. Soot particle measurements in diffusion flames. *Combust Flame*. 1983; 51:203–18.
- [18]. Radney JG, Zangmeister CD. Practical limitations of aerosol separation by a tandem differential mobility analyzer–aerosol particle mass analyzer. *Aerosol Sci Technol*. 2016; 50:160–72.
- [19]. NIST Technical Disclaimer: Certain commercial equipment, instruments, or materials (or suppliers, or software, ...) are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.
- [20]. Gillis KA, Havey DK, Hodges JT. Standard photoacoustic spectrometer: Model and validation using O₂ A-band spectra. *Rev Sci Instrum*. 2010; 81:064902–13. [PubMed: 20590262]
- [21]. Radney JG, You R, Ma X, Conny JM, Zachariah MR, Hodges JT, et al. Dependence of Soot Optical Properties on Particle Morphology: Measurements and Model Comparisons *Environ Sci Technol*. 2014; 48:3169–76. [PubMed: 24548253]
- [22]. Bueno PA, Havey DK, Mulholland GW, Hodges JT, Gillis KA, Dickerson RR, et al. Photoacoustic Measurements of Amplification of the Absorption Cross Section for Coated Soot Aerosols. *Aerosol Sci Technol*. 2011; 45:1217–30.
- [23]. Lack DA, Cappa CD. Impact of brown and clear carbon on light absorption enhancement, single scatter albedo and absorption wavelength dependence of black carbon. *Atmos Chem Phys*. 2010; 10:4207–20.
- [24]. Lack DA, Cappa CD, Cross ES, Massoli P, Ahern AT, Davidovits P, et al. Absorption Enhancement of Coated Absorbing Aerosols: Validation of the Photo-Acoustic Technique for Measuring the Enhancement. *Aerosol Sci Technol*. 2009; 43:1006–12.
- [25]. Andreev SN, Vovchenko VI, Samokhin AA. Study of explosive boiling of transparent liquid on metal substrate exposed to nanosecond laser pulses. *Phys Wave Phen*. 2007; 15:182–5.
- [26]. Samokhin AA. Estimate of pressure produced during explosive boiling of a liquid film on a substrate heated by laser pulses. *Quant Electron*. 2006; 36:684–6.
- [27]. Metwally K, Mensah S, Baffou G. Fluence Threshold for Photothermal Bubble Generation Using Plasmonic Nanoparticles. *J Phys Chem C*. 2015; 119:28586–96.
- [28]. Kang B, Dai Y, Chang S, Chen D. Explosion of single-walled carbon nanotubes in suspension induced by a large photoacoustic effect. *Carbon*. 2008; 46:978–81.
- [29]. Beveridge AC, McGrath TE, Diebold GJ, Karabutov AA. Photoacoustic shock generation in carbon suspensions. *Appl Phys Lett*. 1999; 75:4204–6.
- [30]. Kang B, Yu D, Dai Y, Chang S, Chen D, Ding Y. Cancer-Cell Targeting and Photoacoustic Therapy Using Carbon Nanotubes as “Bomb” Agents. *Small*. 2009; 5:1292–301. [PubMed: 19274646]

- [31]. Chen A, Xu C, Li M, Zhang H, Wang D, Xia M, et al. Photoacoustic “nanobombs” fight against undesirable vesicular compartmentalization of anticancer drugs. *Sci Rep.* 2015; 5:15527. [PubMed: 26483341]
- [32]. Lei H, Mustafa Y, Nico RV, Michel O. Explosive formation and dynamics of vapor nanobubbles around a continuously heated gold nanosphere. *New J Phys.* 2015; 17:013050.
- [33]. Baffou G, Polleux J, Rigneault H, Monneret S. Super-Heating and Micro-Bubble Generation around Plasmonic Nanoparticles under cw Illumination. *J Phys Chem C.* 2014; 118:4890–8.
- [34]. Lukianova-Hleb EY, Volkov AN, Lapotko DO. Laser Pulse Duration Is Critical For the Generation of Plasmonic Nanobubbles. *Langmuir.* 2014; 30:7425–34. [PubMed: 24916057]
- [35]. Dou Y, Zhigilei LV, Winograd N, Garrison BJ. Explosive Boiling of Water Films Adjacent to Heated Surfaces: A Microscopic Description. *J Phys Chem A.* 2001; 105:2748–55.

Figure 1.

a) Plot of soot mass as a function of relative humidity (RH) for particles with a dry mobility diameter of 350 nm; uncertainties represent the width of the mass distribution (see discussion in text). b) Extinction and absorption cross sections of soot as a function of RH and laser. c) Extinction and absorption $f(\text{RH})$.