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## Notes

## Light source effects on aerosol photoacoustic spectroscopy measurements



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## 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,  $\approx 650$  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  $\approx 85\%$  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 [1] are the result of laser source used (Q-CW) and not from an increase in the particle absorption cross section.

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### 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 1980s [2,3], although advancements in laser and microphone technology in the late 1990s [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 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 ( $\alpha_{\text{abs}}$ ) of the sample – i.e. the absorption strength per unit propagation distance – with  $\alpha_{\text{abs}}$  being a function of the number density of absorbing particles ( $N$ ) and the absorption cross section of those particles ( $C_{\text{abs}}$ ):  $\alpha_{\text{abs}} = NC_{\text{abs}}$ . Since only the former is controllable in the experimental setup, it is customary to use

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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  $C_{\text{abs}}$  as a function of RH, and hence coating thickness, using both the Q-CW and CW lasers. We also compare  $C_{\text{abs}}$  to similar measurements of the extinction cross section ( $C_{\text{ext}}$ , 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  $C_{\text{abs}}$  is enhanced by 35% and 15% for the Q-CW and CW lasers, respectively, while no enhancement was observed in  $C_{\text{ext}}$ . 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<sup>-1</sup> 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 [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  $C_{\text{ext}}$  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<sup>-1</sup> of dry particle-free laboratory air. An ejector pump was situated downstream and operated with 10 L min<sup>-1</sup> of dry, particle-free laboratory air. Of the 15 L min<sup>-1</sup> of total flow, 0.5 L min<sup>-1</sup> 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  $\approx 11$  L min<sup>-1</sup> 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 [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  $\approx 8.8$  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  $\approx 1.64$  kHz in ambient air at 296 K) [20]; as a result, the Q-CW measurements possessed both ultrasonic (78 MHz) and sonic ( $\approx 1.64$  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 ( $\approx 32$  mW RMS, normal mode of operation) and low power ( $\approx 10$  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 [18]. Briefly, light from the CW laser was injected into a high-finesse optical cavity until saturation ( $\approx 100$   $\mu$ s). The light was then quickly terminated (10 s 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  $\approx 0.002\%$ ). 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  $\approx 17$   $\mu$ s.

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