Cavity Ring-Down Measurements of Aerosol Optical Properties during the Asian Dust above Monterey Experiment and DOE Aerosol Intensive Operating Period

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ABSTRACT

Aerosols have a significant effect on visibility, the radiation balance in the atmosphere, and can be a threat to human health. One of the biggest obstacles to a better understanding of these effects and an improved ability to model climate is an inadequate knowledge of the optical properties and the spatial distribution of atmospheric aerosols. Improvements in our knowledge of aerosol characteristics require improved in situ measurements of extinction coefficient and single-scattering albedo. This paper describes the use of continuous wave cavity ring down technology (CW-CRD) to address this problem. The innovations in this instrument, called Cadenza, are the use of CW-CRD to measure aerosol extinction coefficient with the simultaneous measurement of scattering coefficient. Combining these two parameters, one can obtain the single scattering albedo and absorption coefficient, both important aerosol properties. The use of two wavelengths (675 nm and 1550 nm) also allows us to obtain a quantitative idea of the size of the aerosol through the Ångström exponent. The minimum sensitivity of this instrument is 1.5×10^{-6} m^{-1} (1.5 Mm^{-1}). The small size and fast response time (seconds) of Cadenza makes it an ideal choice for both aircraft and ground-based studies. This instrument has recently successful participated in two missions in April and May 2003 aboard the CIRPAS Twin-Otter aircraft. These missions, Asian Dust Above Monterey (ADAM) and the DOE Aerosol Intensive Operating Period (IOP), had as objectives the study aerosol layers aloft, their transport, and radiative effects. These objectives matched the design goals of Cadenza extremely well and provided comparisons with other aerosol measurement instruments. Results from both missions will be presented in this paper.

INTRODUCTION

Past studies have shown that aerosols can have significant effects on the balance of radiation in the atmosphere and be a threat to human health. These effects can manifest themselves globally as well as locally on climate, the hydrological cycle, and air pollution. One of the biggest obstacles to a better understanding of these effects and an improved ability to model climate is an inadequate knowledge of the optical properties and spatial distribution of atmospheric aerosols. This deficiency is regarded as one of the primary contributors to uncertainty in climate change predictions. The Intergovernmental Panel on Climate Change (IPCC) has identified radiative forcing due to aerosols as one of the most uncertain components of climate change models and as a topic urgently in need of further research[1].

The potential importance of aerosols in earth's climate has been well documented[2-4]. As an example, the global-average direct forcing due to aerosols is estimated to be –0.4 (±0.3) Wm^{-2}, compared with 2.4 (±0.3) Wm^{-2} for green house gases[5]. The indirect direct forcing due to aerosols, through their effect on clouds and precipitation, is estimated at -1.9 (±0.8) Wm^{-2}, nearly of equal magnitude to green house gas forcing. The uncertainty associated with forcing due to aerosols indicates that these effects are not known with sufficient accuracy to define future climate change[1].

Regionally, radiative effects due to aerosols can be much larger than global effects[6,7]. A recent international study, the Indian Ocean Experiment (INDOEX), which culminated in an intensive field campaign in 1999, explored the effects of air pollution at a local and regional level[8]. This study revealed that the pollution haze is transported far beyond the source region. These pollutants scatter and absorb incoming solar radiation and thus reduce up to 10% of the solar energy reaching the ocean and 10 to 20% over the land masses[8]. These findings which have raised serious questions related to the impact of atmospheric pollutants of that magnitude on health, marine life, plant ecosystems, and agriculture[8], have underscored the need for an improved knowledge of aerosol optical properties.

Regional aerosol effects are of interest for several current visibility studies within national parks and wilderness areas of the United States. Visibility, as defined by the Koschmeider equation, is inversely proportional to the extinction coefficient of the ambient aerosols. Two current studies, IMPROVE (Interagency Monitoring of Protected Visual Environments) and BRAVO (Big Bend Regional Aerosol and Visibility Observational study), have provided extensive information concerning the aerosol properties within these regions. Yet, these studies have also acknowledged a further need for information concerning the optical properties of carbon and coarse particles. Unlike sulfur aerosols that have been extensively studied, the mass absorption efficiencies for carbonaceous particles reported in the literature vary by a factor of 4, from 5-20 $m^2 g^{-1}$. This uncertainty plays an especially important role with national forest land, since as a result of expected decreases in the sulfur dioxide emissions, the fraction of extinction associated with sulfates is expected to decrease in the next two decades, while during that same time period carbon emissions are projected to increase due to a dramatic increase (by factors of 5-10) in prescribed fire activity by federal land managers[9].

Ogren[10] and *Heintzenberg and Charlson*[11] have presented overviews of the aerosol properties needed to determine the influence of particles on the atmospheric radiation balance and visibility. The most fundamental of the aerosol optical properties are the aerosol extinction (σ_{ext}), scattering (σ_{scat}), and absorption coefficients (σ_{abs}). From these basic quantities, the single scattering albedo and Ångström exponent can be deduced. Recent studies (c.f. [5, 12]) show that single scattering albedo, together with planetary albedo, determines the sign (cooling or heating) of the aerosol radiative forcing, while the asymmetry parameter of the phase function together with extinction coefficient of the atmospheric aerosol determine the magnitude of the forcing.

Single scattering albedo, ω, is defined as

$$\omega = \frac{\sigma_{sca}}{\sigma_{ext}} = \frac{\sigma_{sca}}{\sigma_{sca} + \sigma_{abs}} \qquad (1)$$

and is a measure of the proportion of incident light scattered and absorbed. Many studies[5,7] have shown that an accurate assessment of aerosol radiative effects requires accurate values of aerosol ω. Recent experimental results have not provided the required accuracy. For example, Russell et al.[7] noted that in both TARFOX and ACE-2, different techniques yielded aerosol ω values differing by as much as 5% (0.90 to 0.95) when attempting to describe the same aerosol. They showed that the radiative effects of such large differences in ω could be very significant climatically (e.g., changing a cooling effect to a heating effect). ω cannot be measured directly. Typically ω is determined in situ from absorption and scattering measurements. In our technique we ratio simultaneous scattering and extinction measurements of the aerosol in the same test section to obtain ω. This should eliminate some of the uncertainty inherent with other methods especially since the measurements of scattering and extinction are correlated to some degree.

The Ångström exponent, å, is a measure of the wavelength, λ, dependence of aerosol optical properties and can be obtained from the relation

$$\mathring{a} \cong -\frac{\log\left(\sigma_{ext,1}/\sigma_{ext,2}\right)}{\log\left(\lambda_1/\lambda_2\right)} \qquad (2)$$

The Ångström exponent is one measure of the optical effective radius, r_e, of the size distribution of aerosol particles. Aerosols with larger r_e, such as dust have an å~0, while aerosol with small r_e, such as smoke, have å~2.

CADENZA

The first CRDS system designed to measure aerosol extinction and scattering, called **Cadenza**, was developed by our team[13]. The in situ measurement of extinction coefficient is particularly difficult because of the low levels of attenuation due to aerosol, on the order of 10^{-1} to 10^{-3} km^{-1} on the surface to 10^{-5} km^{-1} in the stratosphere[14,15]. Currently in situ measurement of aerosol extinction requires very long path lengths and is primarily restricted to measurements of surface visibility[16]. The importance of the problem however has resulted in several attempts to measure extinction in situ on aircraft[17-20]. None of these instruments had sufficient sensitivity to measure typical atmospheric aerosol extinction coefficient.

In a typical extinction or visibility cell, radiation from a light source is passed through a sample. The intensity of the light is measured before, I_0, and after, I, and the extinction coefficient, σ, of the sample can be determined from Beer's Law:

$$\frac{I}{I_0} = e^{-\sigma L} \approx 1 - \sigma L \qquad (3)$$

where L is the sample length. The minimum detectable extinction depends on how accurately one can measure the change in light intensity ($\Delta I = I-I_0$). For typical absorption experiments, $\Delta I/I_0$ is approximately 10^{-3} after one second of averaging. For example, for a one meter cell, the minimum detectable aerosol extinction with a one second sampling rate would be approximately 1000 Mm^{-1}. The accuracy of practical multi-pass systems can be extended to $\Delta I/I_0 \sim 10^{-5}$. However, multi-pass extinction systems suffer from two major drawbacks: they are difficult to align and keep aligned and the sample volume is large leading to poor response time.

Briefly, CRD employs high reflectivity mirrors to achieve a path length of kilometers in a small cell. Since the technique was first demonstrated by O'Keefe and Deacon[21] it has been used primarily for absorption spectroscopy[22]. We expect that this instrument and its successors will help reduce uncertainty in optical properties and spatial and temporal variation of aerosols. Thus it will greatly contribute to visibility studies, aid in our understanding of climate forcing by aerosol, and assist in satellite validation and the validation of aerosol retrieval schemes from satellite data. An excellent review of the CRD techniques and applications can be found in the collection of papers edited by Busch and Busch[23]. The principle behind CRD is briefly described here using a so-called 'ping-pong' model. A pulse of laser light is injected into a cavity that consists of two highly reflective mirrors. The mirror reflectivity is typically better than 99.96%. The laser pulse bounces between the two mirrors inside the ring-down cavity like a ping-pong ball. Each time the pulse interacts with the back mirror, a small amount of light (e.g., 0.04%) leaks out. This light is collected and detected with a photomultiplier or similar detector. The intensity of the light leaking out of the back of the ring-down cavity decreases exponentially. It can be shown that the exponential decay, or ring-down time, is related to the mirror reflectivity and the absorption of the material inside the cavity by the relationship

$$\tau = \frac{L}{c}\left((1-R) + \sigma_{ext}L + \sigma_{Ray}L + \sigma_{gas}L\right)^{-1} \tag{4}$$

where L is the cell length, c is the speed of light, R is the mirror reflectivity, σ_{ext} is the coefficient of extinction due to aerosol, σ_{Ray} the coefficient of Rayleigh scattering, and σ_{gas} the coefficient of absorption due to gaseous species in the cell. (Note that extinction is the sum of scattering plus absorption.)

Extinction coefficient is obtained from the difference between measurements made when the cell contains filtered air and when the cell contains a particulate-laden flow

$$\sigma_{ext} = \frac{1}{c}\left(\frac{1}{\tau_{aer}} - \frac{1}{\tau_0}\right) \tag{5}$$

where τ_{aer} is the ring-down time of the aerosol laden flow and τ_0 is for the filtered air.

While the ping-pong model explains the exponential decay of the signal, it is too simple to account for the fact that only light having frequencies near the cavity resonance mode will resonate in the ring-down cell. Thus, the laser linewidth must be mode-matched to a single cavity mode or multi-mode excitation in the cell will cause excessive noise. In this application a continuous wave (CW) laser source is used which results in several advantages over the pulsed laser technique[24]. Pulsed laser systems are bulky and their sample rate is limited by the repetition rate of the laser, typically about 10 Hz. CW laser diodes can be obtained with very narrow line widths that can be more effectively coupled into the cavity so that the sensitivity of the system is not limited by the laser linewidth. First, the resulting overlap between the laser and cell linewidth results in actual energy build up in the cell. Second, CW lasers diodes can be obtained with very narrow line widths that can be more effectively coupled into the cavity so that the sensitivity of the system is not limited by the laser linewidth. Also, CW laser diodes also have a higher duty cycle than pulsed lasers, which results in faster sampling. Finally, the use of CW laser diodes results in a more compact and rugged instrument suitable for aircraft operations. Pulsed laser systems are larger and their sample rate is limited by the repetition rate of the laser, typically about 100 Hz. The resulting overlap between the laser and cell linewidth results in actual energy build up in the cell, benefiting both the extinction and scattering measurements. For a cell with non-absorbing mirrors and negligible internal losses, the peak circulating intensity is approximately given by

$$I_{circ} \approx I_{inc}/T \qquad (6)$$

where I_{inc} is the incident intensity and T is the mirror transmittance[25]. This benefits both the extinction and the scattering measurements. The minimum detectable absorption of CW-CRD systems is on the order of 10^{-7} to 10^{-9} m^{-1} [26]. Thus, measurement accuracy for extinction coefficient of 1% to 0.01% is achievable at extinction levels of 10^{-5} m^{-1}.

Figure 1 shows the optical layout of the ARC system. It uses two CW laser diodes at wavelengths of 675 nm and 1550 nm, located on the left. The laser beams are conditioned with spatial filters, combined with a dichroic beamsplitter, and coupled into a single cavity/flow cell. The cavity/flow cell consists of four mirrors that form a bow-tie configuration, unlike the two-mirror system described in the ping-pong model. In Figure 1 the input mirrors are located at the bottom of the cell and output mirrors are located at the top of the cell. Light from the output mirror is focused onto the ring-down detectors that are located on the right of the diagram. A scattering detector is mounted on the flow cell wall. Aerosol-laden or filtered air enters the cell through 0.64 cm diameter tubing with a flow rate of 3 L/min. The ring-down cell is 20 cm wide with an optical path of approximately 80 cm. The effective pathlength of the system is about 1 km. In this CW-CRD application, the laser current is dithered rapidly while monitoring the light output of the cell. When a resonance occurs, the light energy builds up in the cell and after it reaches a threshold, the laser is switched off rapidly on the order of 50 ns. Ring-down times for this system are tens of μsecs. The ring down signal is then recorded as in pulsed-CRD. Ring-down occurs at a frequency of 100 Hz in this system, and100 shots are averaged over 1 sec to achieve one sample. Minimum sensitivity of the instrument is presently 1.5×10^{-6} m^{-1}. The use of two wavelengths allows us to obtain a quantitative idea of the size of the aerosol through the Ångstrom exponent.

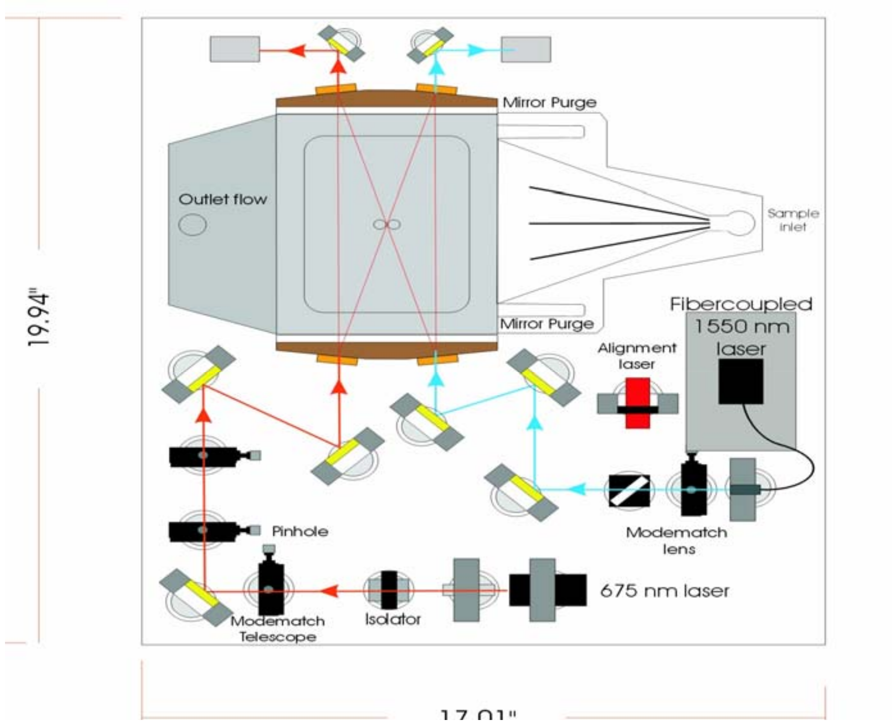


Figure 1. A schematic of the NASA-ARC CW-CRD aerosol extinction cell

Figure 2. CIRPAS Twin-Otter

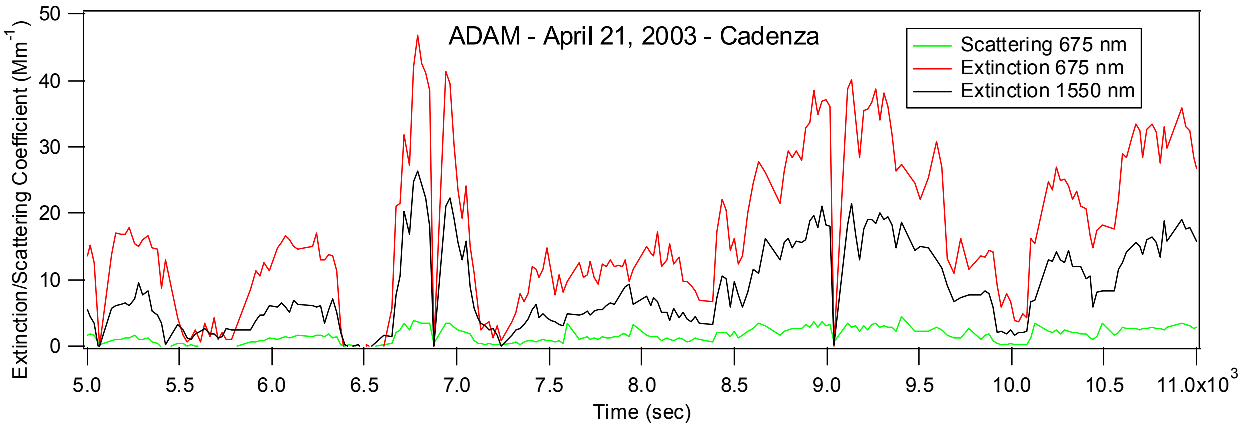
RESULTS

Cadenza was developed by our team and flown on two missions in April and May 2003 aboard the Center for Inter-Disciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin-Otter aircraft (see Figure 2). These missions, Asian Dust Above Monterey (ADAM) and the DOE Aerosol Intensive Operating Period (IOP), had as objectives the study aerosol layers aloft, their transport, and radiative effects. These objectives matched the design goals of Cadenza extremely well and provided comparisons with other aerosol measurement instruments. Cadenza is a CRDS cell with a bow-tie configuration and employed the 'inverse' nephelometer technique for scattering coefficient measurement introduced by[27]. It featured flow geometry transverse to the optical axis that allowed us to very accurately determine the aerosol path and had a fast, 1 second, response time.

Figure 3a illustrates Cadenza measurements of extinction coefficient at 675 nm and 1550 nm and scattering coefficient at 675 nm made during ADAM on April 22, 2003. The variability in signal is illustrative of the natural variability in the atmosphere. Figure 3a shows aerosol optical properties as a function of flight time. Perhaps a better representation of this data is in the form of an altitude profile of extinction as shown in Figure 3b. The aircraft flew a vertical spiral from 6000 to 7500 sec (see Figure 3a) and recorded several aerosol layers aloft. These are represented in Figure 3b. The distinct layer structure seen at approximately 2.5 km profile is likely Asian dust transported across the Pacific.

Comparisons between Cadenza measurements and other instruments made during the Aerosol IOP on May 7, 2003 are shown in figure 4. A comparison of Cadenza and nephelometer scattering at 675 nm from DOE Aerosol IOP are shown in figure 4a. Cadenza extinction coefficient with that derived from the sum of TSI nephelometer scattering plus PSAP absorption coefficient measurements on May 6, 2003 is shown in figure 4b. (Note that extinction = scattering + absorption.) D. Covert and R. Elleman of University of Washington operated these instruments. Figures 4a and 4b show the excellent agreement of the measurements. The spike in the data in Figure 4a and 4b from 11260 sec to 11340 sec resulted from the Twin-Otter flying through a localized smoke plume near the surface. Figure 4c shows this brief time segment. Note the excellent temporal resolution obtained by Cadenza relative to the TSI nephelometer. The nephelometer has a large sample volume and a flow residence time of about 20 sec. The flow in the Cadenza sample volume is perpendicular to the optical axis, giving Cadenza a response time of 1 to 2 sec. Also note that Cadenza registers extinction of up to 250 Mm-1 for this brief period while the nephelometer smoothes that peak extinction out over a longer time period. This fast temporal response is one key advantage to Cadenza. This feature can help characterize pollution plumes from sources.

Finally, figure 5 below shows the measured and derived quantities obtained by **Cadenza** at 675 nm for one flight profile. The easily derived quantities are absorption coefficient and single scattering albedo. By subtracting the scattering coefficient from the extinction coefficient, we obtain absorption coefficient. Single scattering albedo is simply the ratio of scattering to extinction.



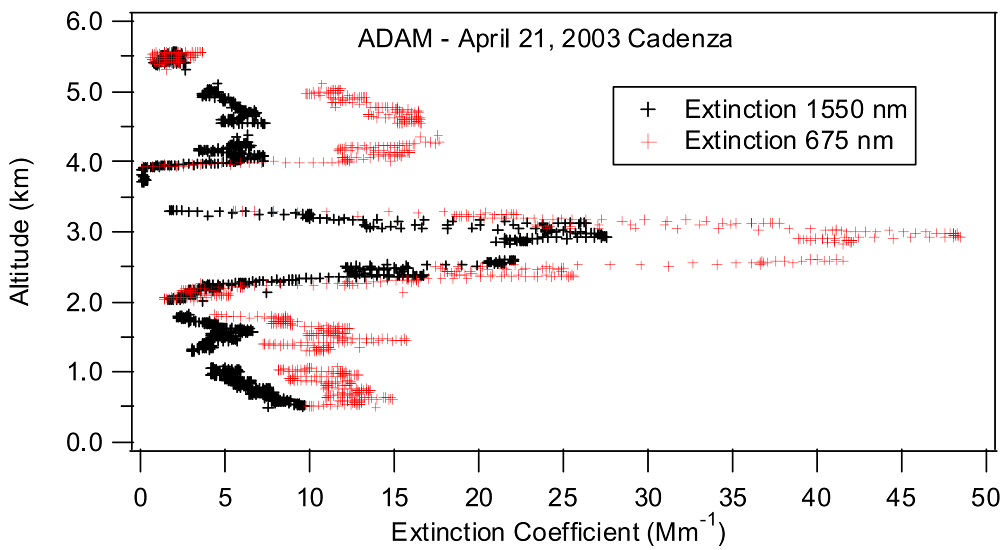


Figure 3. a) Illustrates the three measurements made by Cadenza, extinction coefficient at 675 nm and 1550 nm, and scattering coefficient at 675 nm. b) Altitude Profile of Extinction at two wavelengths as measured by Cadenza.

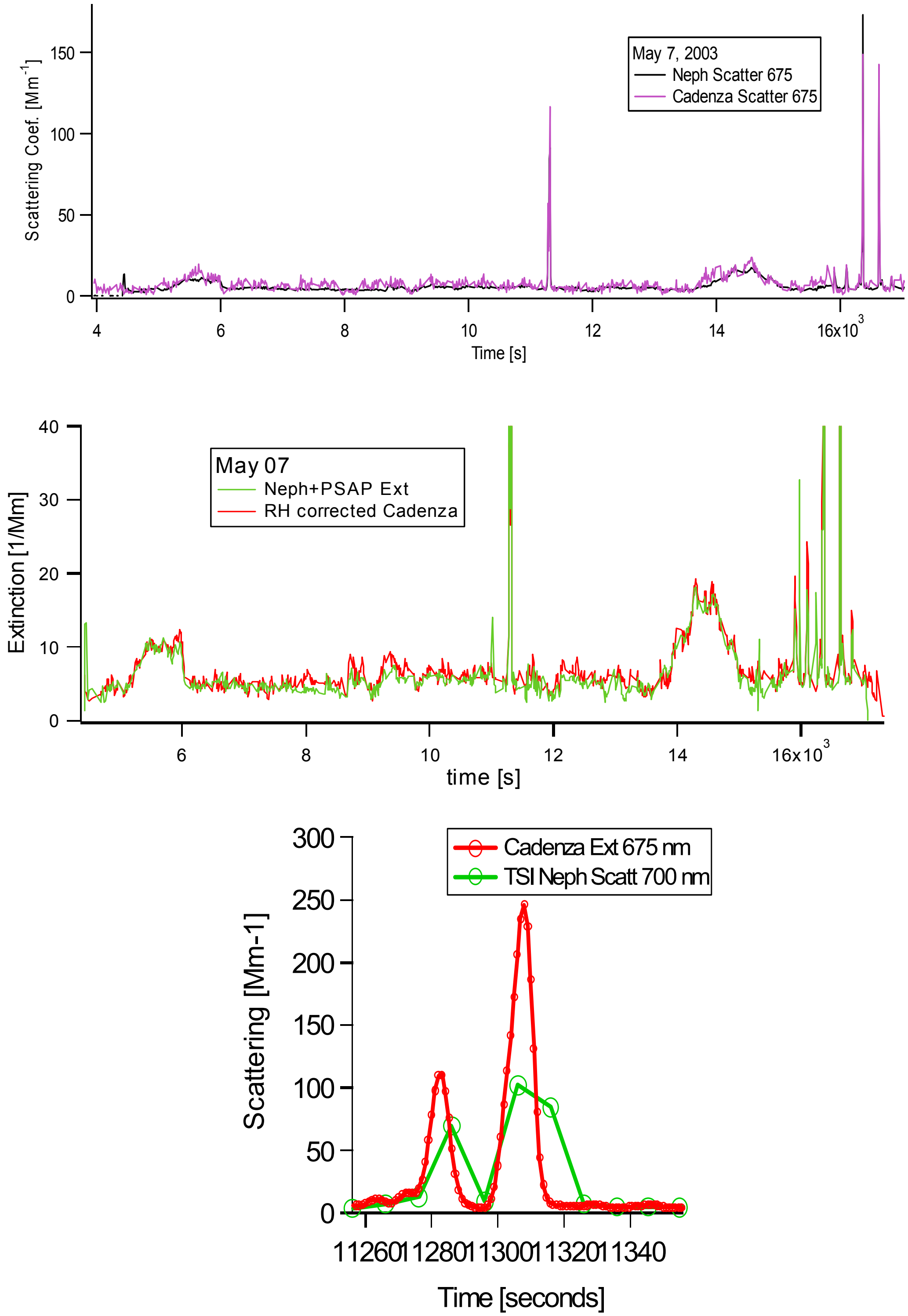


Figure 4. a) Comparison of scatter by the nephelometer and Cadenza both at 675nm. b) Summation of the scatter from the nephelometer and absorption from the PSAP compared to the extinction measured by Cadenza. c) Comparison of Cadenza extinction and nephelometer scattering made while sampling a smoke plume from 11260 sec to 11340 sec. Note the faster response time of Cadenza gives a better representation of the size and magnitude of the plume.

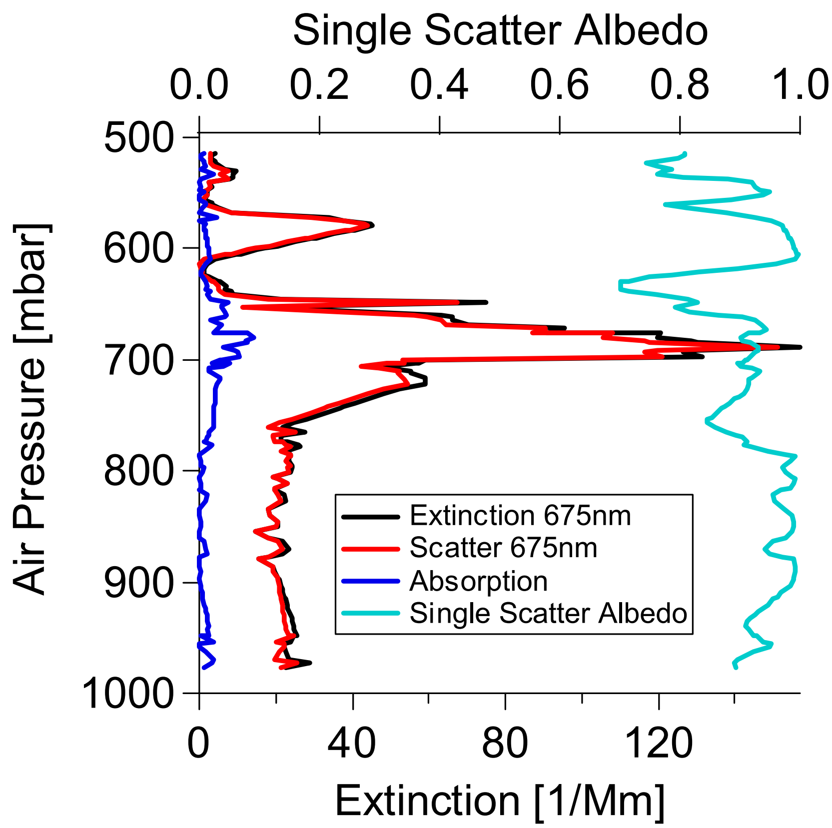


Figure 5. Measured and derived quantities obtained by Cadenza at 675 nm.

CONCLUSION

This paper describes an instrument designed to measure aerosol extinction and scattering coefficients using CW-CRD and results from its deployment in two field missions. The instrument is unique since it is the first application to the measurement of aerosol optical properties using CW-CRD, it is designed for the simultaneous measurement of extinction and scattering at two wavelengths, and its small size and ruggedness make it suitable for application in the field and on airborne platforms. The instrument, called **Cadenza**, has successfully made measurements of extinction and scattering coefficients. Combining these two quantities, one can obtain the single-scattering albedo and absorption coefficient, both important aerosol properties. The use of two wavelengths also allows us to obtain a quantitative idea of the size of the aerosol through the Ångström exponent.

The minimum sensitivity of the instrument is $1.5x10^{-6}$ m^{-1} (1.5 Mm^{-1}). Measurements of scattering coefficient are compared with a state-of-the-art nephelometer and agreement is good. Absorption coefficient and single-scattering albedo deduced from the measurements are reasonable considering the state of the ambient aerosol before and during sampling of a fire plume.

An instrument with this capability will reduce uncertainty currently associated with aerosol optical properties and their spatial and temporal variation. It could contribute to visibility studies, aid in our understanding of climate forcing by aerosol, and assist in satellite validation and the validation of aerosol retrieval schemes from satellite data.

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Key Words

Visibility
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