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Monitoring Atmospheric Particulate Matter through Cavity Ring-Down Spectroscopy

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Cavity ring-down spectroscopy was explored as a means to measure atmospheric optical extinction. Ambient air was sampled through a window on the campus of the University of Florida and transported to a ring-down cell fashioned from standard stainless steel vacuum components. When a copper vapor laser operating at 10 kHz is employed, this arrangement allowed for nearly continuous monitoring of atmospheric extinction at 510 and 578 nm. We have characterized the system performance in terms of detection limit and dynamic range and also monitored a change in atmospheric extinction during a nearby wildfire and fireworks exhibition. The sensitivity and compatibility with automation of the technique renders it useful as a laboratory-based measurement of airborne particulate matter.

Techniques that provide real-time analysis of atmospheric optical extinction are required in many fields of both basic and applied science. For instance, techniques for on-line, real-time analysis may as easily find use in monitoring clean room air for particulate contaminants as well as in measurements of earth's atmospheric extinction in studies of the planet's radiation budget or urban pollution. Such analysis requires the analytical technique employed to be rapid, robust, and extremely sensitive. Aerosol particles are very dilute in the atmosphere, with concentrations of ∼20 $\mu g/m^3$ during clear days in urban settings, but can reach ∼100 $\mu g/m^3$ during severe pollution events.[1] Remote continental sites typically have concentrations as low as ∼5 $\mu g/m^3$.[2] Despite the low-mass concentration of aerosols, they are believed to be an important part of the global radiation balance,[3] which is still not fully understood,[4] and high levels of atmospheric particulate matter have been linked to increased mortality rates.[5,6] Thus, development of reliable instrumentation capable of monitoring atmospheric particulate matter and its effects on atmospheric extinction is a priority.

Traditionally, measurements of atmospheric extinction have been carried out through long-path methods through use of either multipass absorption cells[7,8] or laser-based techniques where path lengths of up to 1 km or more are common.[9−13] Long-path techniques often simplify the measurement as no sampling of the ambient air is generally required and extinction data are averaged over a large distance. However, the laser beam used for these measurements can often wander due to atmospheric turbulence and the slight movement of buildings generally used to house the light source and detector elements can increase noise in the signal as well as confound long-term measurements.

Cavity ring-down spectroscopy (CRDS) is one of a family of analytical techniques aimed at making ultrasensitive absorption (i.e., attenuation) measurements, particularly with gas-phase samples. Traditional absorption measurements require the determination of two parameters, the incident intensity and the transmitted intensity, and the measurement's success hinges on the ability to distinguish between the two. Typically, traditional absorption measurements are hindered by source intensity fluctuations often limiting the minimum detectable absorption per pass to parts per thousand or even parts per hundred levels. Cavity ring-down absorption measurements eliminate the dependence on the source intensity fluctuations by measuring the decay of light in time within a stable optical cavity. When coupled with high-reflectivity mirrors and state-of-the-art detection electronics, this improvement allows for minimum detectable absorptions as low as 10^{-8} m^{-1}.[14] In a typical ring-down experiment, a stable optical resonator of length 0.5−1.5 m is constructed from two (or more) highly reflective (R > 0.999) dielectric mirrors. A pulsed laser is used to introduce light into the cavity, and light that leaks from the cavity is monitored by a photomultiplier tube (PMT). The intensity of light within the cavity decays exponentially as a

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function of time and can be given by

$$i(t) = i_0 \exp(-[(1 - R) + \alpha L](tc/L))$$

where i_0 is the initial light intensity, R is the reflectivity of the mirrors, L is the length between the mirrors (m), c is the speed of light (m/s), and α is the extinction coefficient of the sample (m^{-1}). The extinction coefficient can be found directly through the expression

$$\alpha = \frac{1}{c}\left(\frac{1}{\tau_{\text{sample}}} - \frac{1}{\tau_{\text{empty}}}\right)$$

where τ_{sample} and τ_{empty} are the time constants (s) of the exponential decay for the cavity filled with sample and the empty cavity, respectively. Ring-down measurements are usually performed such that absorption is responsible for attenuation of radiation, although scattering of radiation has also been employed previously in several cases.[15,16]

In this work, an instrument to monitor atmospheric extinction of ambient air through a fixed-frequency cavity ring-down method is described. We have characterized the instrument's sensitivity and temporal response and determined the extinction coefficient at two wavelengths for ambient air. As a demonstration of the ability of the technique to monitor atmospheric events, we have also monitored the change in atmospheric extinction following smoke from a nearby wildfire and during a local fireworks display.

EXPERIMENTAL SECTION

Sampling System. Experiments were conducted over the period of May 4, 2001–July 3, 2001. Over this period, outside temperatures ranged from 18.3 to 33.3 °C and relative humidity from 35 to 98%. Temperature and humidity within the ring-down cell were monitored by a temperature/humidity probe and remained relatively constant at ~25 °C and ~50%. A schematic of the sampling/analysis system is depicted in Figure 1. Briefly, ambient air was sampled through a second story window in the chemistry laboratory building on the campus of the University of Florida into a 6-m length of 10-cm-diameter vinyl duct in order to transport the air to the ring-down system. The laboratory ventilation system was used to induce air flow through the vinyl tubing. The flow rate was 56 L/min corresponding to a linear flow velocity of 0.38 m/s. An aerosol deposition program[17] was used to compute the transfer efficiency of the tubing system. Figure 2 illustrates the calculated transport efficiency as a function of particle aerodynamic diameter for the system employed. As transport efficiencies of >98% were obtained for particles of <2.5 μm, no corrections were made to account for sampling losses. For the measurement of filtered ambient air, the air was sampled through 0.38-cm-i.d., 0.635-cm-o.d. plastic tubing and passed through an in-line gas filter (Wafergard F, Millipore, Holliston, MA) before entering the measurement chamber. This filter was known to produce 7-log-unit reduction in number density for 30-nm-diameter particles; consequently, air flowing through this filter was assumed to be particulate free.

Cavity Ring-Down System. The cavity ring-down cell is illustrated in Figure 1. The 1.69-m linear cell was constructed from two 60-cm lengths of 3.4-cm-i.d., 3.81-cm-o.d. stainless steel tubing welded to CF flanges at both ends. The tubes were fastened to a six-way stainless steel cross at the center of the cavity (l = 12.5 cm), and three-way crosses at either end (l = 15 cm) which allowed for transport of the ambient air through the tubes length. Initially, it was observed that over the course of several hours particulate matter deposited on the surface of the ring-down mirrors, adversely effecting their reflectivity. Unfortunately, windows cannot be used to contain the measurement cell and

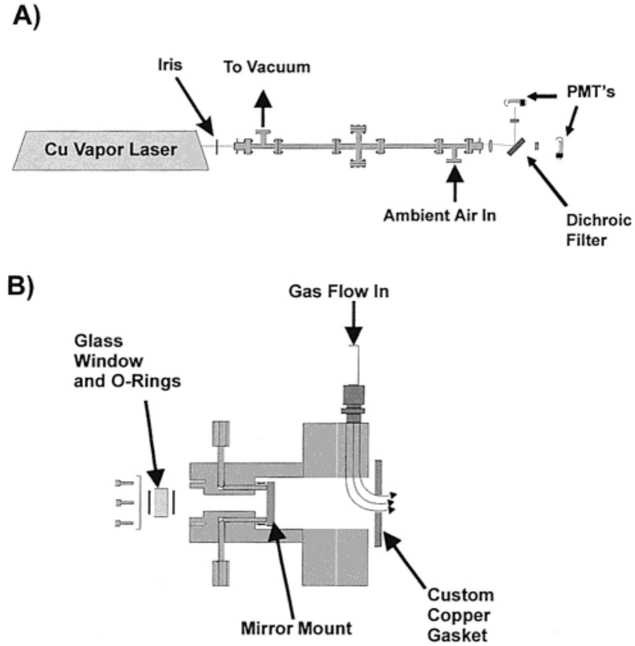

Figure 1. (A) Illustration of the cavity ring-down instrument. The beam from the copper vapor laser is passed through a 1-mm iris before being introduced into the ring-down cell. (B) Detailed depiction of the mirror mounts and custom copper gasket used to prevent deposition of particulate matter on the surface of the highly reflective mirrors. Without the gasket and airflow, the mirror reflectivities were observed to decrease over the first few hours of analysis.

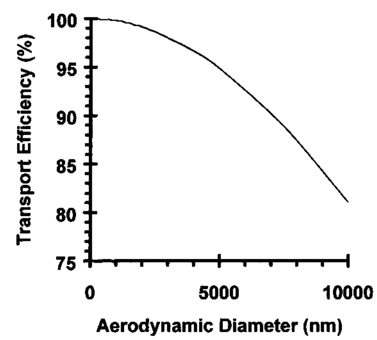

Figure 2. Transport efficiency as a function of particle aerodynamic diameter for the sampling system employed. As transport efficiencies of >98% are obtained for particles of <2.5 μm, no corrections were made to account for sampling losses.

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protect the mirror's surface due to reflection losses at the window surfaces. To combat the deposition problem, we fabricated custom copper gaskets for use at both ends of the tube for the CF flange connections. These gaskets were 4.82-cm o.d. with a 0.508-cm-diameter hole centered on the gasket to allow the laser beam to pass between mirrors. We also have added a 4 L/min air flow within the mirror housing such that the flow is forced through the 0.508-cm hole in the gasket with a linear velocity of ~330 cm/s. The combination of the gasket and gas flow proved effective, as particle deposition on the surface of the mirror was eliminated, and consequently, monitoring atmospheric extinction over long periods of time was possible.

Our 1.69-m ring-down cell employed a symmetric resonator consisting of two 2.032-cm-diameter, 0.635-cm-thick, 6-m-radius of curvature highly reflective dielectric mirrors (Los Gatos Research, Mountain View, CA.). Mirror reflectivities at 510 and 578 nm were 0.999 25 and 0.999 35, respectively, giving rise to empty cavity ring-down times of 7.6 and 8.7 μs. These corresponded to effective path lengths of 2.26 and 2.6 km, respectively. The cavity was aligned by removing the front cavity mirror, passing the laser light through a 1-mm-diameter iris, and reflecting the light off the rear cavity mirror back through the iris. The front mirror was then replaced, and the reflection off its surface aligned with the iris. At this point, fine adjustment of the mirrors was necessary to provide the longest ring-down. The 3σ limit for minimal detectable loss for our ring-down system after averaging of 1500 individual waveforms over 150 ms was found to be 10^{-6} m^{-1}.

Laser System. A copper vapor laser simultaneously emitting in the green (510.6 nm) and yellow (578.2 nm) was employed in this study (model CU-15A; Oxford Lasers). The laser operated at 10 kHz with pulse energies of ~133 μJ at 510 nm and ~66 μJ at 578 nm. The laser pulse width was ~30 ns.

Data Acquisition and Signal Processing. Light exiting the optical cavity was collected with a 5-cm-diameter, 15.4-cm-focal length glass lens and imaged onto a long-wave-pass dichroic filter to separate the 510- and 578-nm beams. The 510-nm beam was reflected at 90°, passed through a 510 ± 10-nm band-pass filter and imaged onto an end-on type PMT (R647, Hammamatsu). The 578-nm beam was transmitted through the dichroic filter, passed through a 580 ± 10-nm band-pass filter, and imaged onto an identical photomultiplier tube. The photomultiplier tubes were biased at 1000 V with a high-voltage power supply (model 226; Pacific Photometric Instruments) and exhibited response times of ~4 ns. Both PMTs and the dichroic filter were housed in a 40 cm × 35 cm × 22.4 cm aluminum box to reduce stray light. Current from each photomultiplier tube was converted to a voltage through a 50-Ω terminator, and the decay transients were digitized by a 2-channel, 8-bit, 500-MHz digital oscilloscope (TDS 520A, Tektronix) and transferred by either a GPIB interface or diskette to a personal computer. Each acquired waveform typically was the average of 1500 individual ring-downs. Although averaging ring-downs on the oscilloscope eliminated some of the advantage of independence from source intensity fluctuations, it was found to be a simple method of data acquisition with adequate sensitivity. Before data were collected, PMT linearity for both channels was checked with a neutral density filter. The initial portion of the waveform exhibited multiexponential decay associated with excitation of multiple spatial modes. These higher-order spatial modes by definition decayed much faster than the fundamental and typically were not observed after the first few microseconds. Therefore, the first 3 μs of the ring-down signal was deleted from the waveform. The remaining waveform was then fitted to a first-order exponential decay and the ring-down decay constant determined (Microcal Origin v 6.0). To determine the cavity fundamental decay time constant, the optical cavity was flushed with He gas for ~15 min. Helium is not known to absorb at the wavelengths we employ and has Rayleigh losses of 2.46×10^{-7} and 1.48×10^{-7} m^{-1} for the wavelengths employed, which are below the limit of detection for our system. Therefore, we used He to flush the cavity of air in order to determine the fundamental "empty" cavity time constant. It should be noted that it was necessary to flush the cavity with gas for an extended period of time in order to remove all other gases from the system.

Table 1. Comparison of Rayleigh Scattering Extinction Coefficients Determined by the Ring-Down Method Compared with Literature Values at 510 nm[a]

gas	measured value (m^{-1})	literature value (m^{-1})	% error
N_2	1.6×10^{-5}	1.61×10^{-5}	0.3
O_2	1.5×10^{-5}	1.48×10^{-5}	1.0
Ar	1.54×10^{-5}	1.57×10^{-5}	1.9
CO_2	4.07×10^{-5}	4.09×10^{-5}	0.5
SF_6	1.05×10^{-4}	1.07×10^{-4}	1.18

[a] The measured values represent the average of three trials with five measurements per trial for each gas. Literature values adapted from refs 16 and 26−28 and corrected for wavelength by the well-known $1/\lambda^4$ dependence of wavelength on Rayleigh scatter.

Table 2. Comparison of 3σ Minimal Detectable Extinction Coefficients Obtained by the Ring-Down Method Compared to That Obtainable by a Single-Shot Method and a Dual-Beam Approach To Correct for Source Fluctuations

λ (nm)	single pulse (m^{-1})	dual beam (m^{-1})	cavity ring-down (m^{-1})
510	7.7×10^{-2}	2.6×10^{-2}	3.6×10^{-5}
578	1.9×10^{-1}	2.7×10^{-2}	3.1×10^{-5}

RESULTS AND DISCUSSION

After construction of the ring-down system, initial experiments were aimed at evaluating the quantitative nature of the method. To accomplish this, we measured Rayleigh extinction coefficients for five gases at atmospheric pressure. Table 1 illustrates our results. As observed, the measured extinction coefficients agree within 2% of the literature values at 510 nm; thus, we feel confident about the quantitative nature of our results.

Table 2 compares the detection limit obtained with the ring-down technique compared with that of a 1-m path length non-ring-down transmittance measurement and a dual-beam transmittance measurement corrected for source intensity fluctuations for a single laser pulse. For this analysis, the sample was air at atmospheric pressure. Pulse-to-pulse source intensity fluctuations characteristic of pulsed laser systems limit minimal detectable

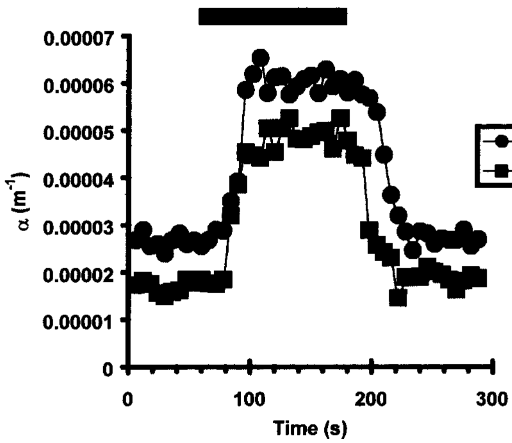

Figure 3. Temporal response of the ring-down system following introduction of a dry NaCl aerosol at 56 L/min. The particles were introduced into the cavity during the time marked with the black bar. Temporal resolution was limited to ~30 s based on the time required to flush the ring-down cell of all particulates.

losses to parts per hundred levels over our 1-m path length. If a dual-beam experiment is used to help correct for source variations, detection limits can be reduced by a factor of 3 and 7, respectively. However, the ring-down technique offers an improvement in minimal detectable extinction of a factor of 10^3 over the other approaches considered in this experiment. However, this experiment neglects the difference in effective path lengths between the non-ring-down and ring-down methods, and it is expected that detection limits for the non-ring-down methods would improve over longer path lengths. Nonetheless, path lengths on the order of kilometers are often experimentally prohibitive, and in terms of what can be reasonably achieved within a laboratory setting, the ring-down method offers a significant improvement. However, the ring-down method suffers from a limited dynamic range and can be used only for losses below 1 pph m^{-1}. Interestingly, this limit roughly corresponds to the detection limits we found for the non-ring-down methods.

Figure 3 illustrates the temporal response of our system. In this experiment, dry NaCl particles produced through an ultrasonic nebulizer were introduced into the sample chamber at a carrier gas flow rate of 56 L/min during the period on the chart denoted as a black bar. Meanwhile, ring-downs were recorded via GPIB interface every 6 s, leading to determination of the extinction of the sample. The data indicate that the chamber required ~18 s to fill and ~30 s to wash out after the particles were no longer being introduced. Thus, we estimate the temporal resolution of our instrument to be on the order of 30 s, which is much more rapid than many atmospheric events. It should be noted however, that the ring-down process itself only requires ~20−30 μs, and the high repitition rate of the copper vapor laser (10 kHz) is well suited to measurements of fast dynamic processes. Figure 3 also suggests our quantitative measurements are accurate for the average extinction ratio ($\alpha_{510}/\alpha_{578}$) when NaCl particles are present corresponds with a NaCl particle of diameter ~320 nm through Mie calculations. Measurements of the NaCl particles produced through the nebulization process using a commercial aerodynamic particle sizer yielded a mean diameter of ~360 nm, close to our measured value of 320 nm.

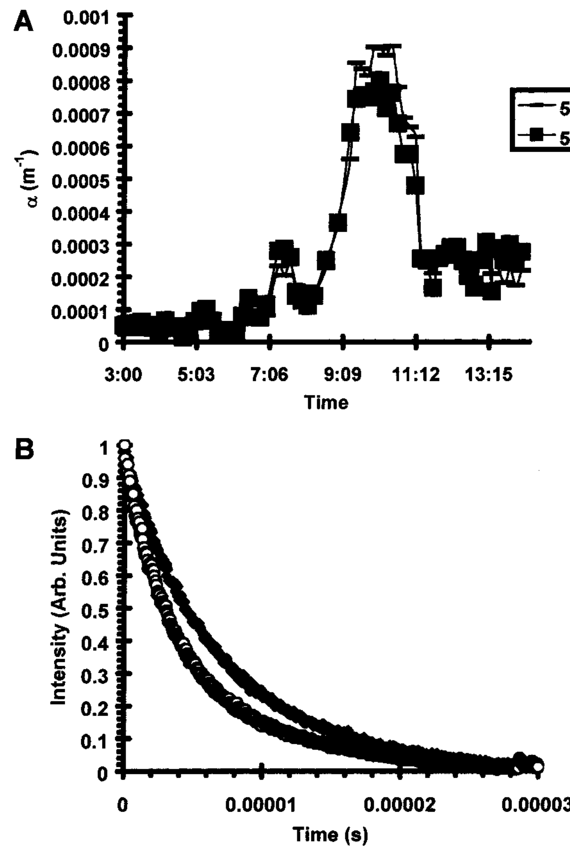

Figure 4. (A) Trace of atmospheric extinction coefficient in time during the morning of May 26, 2001. The increase in extinction is due to smoke from a nearby wildfire. (B) Comparison of ring-down traces at 510 nm for a cell that contains smoke from the fire (○) and an empty cell (●).

Figure 4a illustrates the change in extinction of ambient air following smoke from a nearby wildfire being blown into the vicinity during the morning of May 26, 2001. Westerly winds blew smoke across the state from the Mallory Swamp fire in Lafayette County, FL, about 60 miles northwest of Gainesville. This is illustrated in the satellite image presented in Figure 5. The fire burned more than 60 000 acres over a two-week period, beginning May 14, 2001, after the blaze was sparked by a lightning strike. During the sampling period, smoke was both visible and easily detectable by smell. It is observed that the extinction of the atmosphere increased by a factor of 1735 and 1550% for the wavelengths employed in this study as the smoke was blown into the vicinity. The maximum extinction coefficients of 9.01×10^{-4} and 7.99×10^{-4} m^{-1} for this period exceed the limits representing a polluted urban environment as outlined by Horvath.[18] Figure 4b illustrates a ring-down trace for an empty cavity as well as when smoke from the fire was present within the cell. As observed in the figure, the presence of the smoke causes a more rapid decay of the signal observed.

Figure 6 illustrates the increase in local atmospheric extinction at 510 nm throughout the evening of July 3, 2001. The black bar above the data indicates the approximate time and duration of the University of Florida's annual fourth of July fireworks demonstration. As observed, atmospheric optical extinction in-

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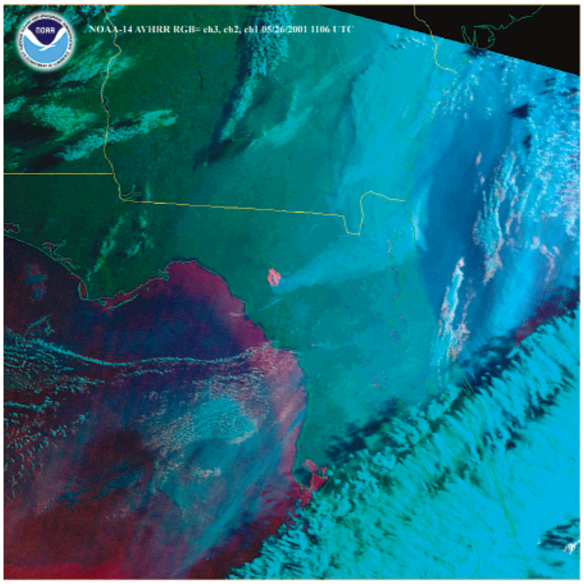

Figure 5. Satellite image of smoke from the Mallory swamp fire at 11:00 a.m. on the morning of May 26, 2001. The trail of smoke from the fire on the satellite image can be seen extending into southern South Carolina. Image courtesy of the National Oceanic and Atmospheric Administration (NOAA).

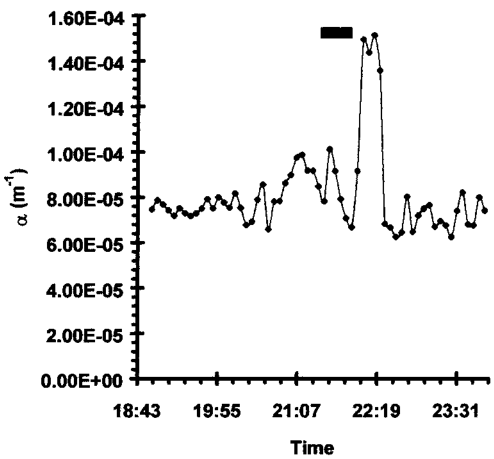

Figure 6. Trace of atmospheric extinction at 510 nm during the evening of July 3, 2001. The black bar above the data indicates the approximate time and duration of the annual University of Florida fireworks demonstration. Data were sampled every 5 min.

creased by ~200% almost immediately after the show. It is unclear why the extinction measurement does not begin to rise until after the demonstration is complete, although this time may represent the time necessary for the particulate matter to travel the ~1-km distance between the fireworks demonstration and the chemistry building.

We also determined the extinction coefficient of particulate-free ambient air by passing it through a filter capable of removing nearly all particulate matter. It was observed that air passed through the filter exhibited an average extinction coefficient of 3.45×10^{-5} and 3.11×10^{-5} m^{-1} at 510 and 578 nm, respectively. Table 2 illustrates the proposed budget of atmospheric extinction at the wavelengths employed in this study. As observed, between 35 and 43% of extinction can be attributed to absorption and scattering by atmospheric gases with the aerosol responsible for the remaining portion. Our model for extinction in particulate-free air is based on extinction coefficients from a variety of sources,[19−24] and local concentrations of trace gases were provided by Alachua county's ambient air monitoring stations whose data are available on the Web at http://www.ees.ufl.edu/air_quality/. As observed, the majority of the observed atmospheric extinction in filtered air can be accounted for by a simple model including Rayleigh scatter from air and absorption from four atmospheric gases. Rayleigh scatter appears to be responsible for the majority of the light extinction at 510 nm while absorption by water vapor is the primary cause at 578 nm. Interestingly, the 578-nm line of the copper vapor laser lies near the maximum absorption of the $X^3\Sigma_g^+ + X^3\Sigma_g^+ \rightarrow a^1\Delta_g + a^1\Delta_g$ $(v = 1)$ band of the O_2-O_2 collision complex which has been previously observed in the earth's atmosphere.[25]

Local visual range can be determined through the Koschmeider equation:

$$x = 3.912/\alpha$$

where x is the local visual range (m) and α is the prevailing atmospheric extinction coefficient (m^{-1}). This equation determines the distance at which a black object has a standard 0.02 contrast ratio against a white background. The average values for atmospheric extinction observed at our location are reported in Table 2 and were 8.04×10^{-5} and 8.70×10^{-5} m^{-1} for 510 and 578 nm, respectively. These extinction coefficients fall within the range of values for clean continental air as given by Horvath[18] and correspond to average visual ranges of 48.6 and 44.9 km, respectively. This visual range is reasonable as all experiments were conducted on clear days (with the exception of the wildfire) during which the National Weather Service reported unlimited visibility at the Gainesville weather monitoring station.

CONCLUSION

As presented, cavity ring-down provides a sensitive, laboratory-based method for determination of atmospheric optical extinction nearly in real time. Through averaging 1500 individual waveforms on an oscilloscope, minimum detectable extinction coefficients of 10^{-6} m^{-1} were achieved. This level of sensitivity is 1 order of magnitude higher than that necessary for detection of Rayleigh scatter from air at the wavelengths employed. Through the use of custom-fabricated copper gaskets and a curtain of air, the ring-down mirrors remain free from particulate debris and monitoring ambient air over long periods of time becomes possible. The extinction coefficients that are determined can be used in conjunction with the Koschmeider equation for determination of local visual range.

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