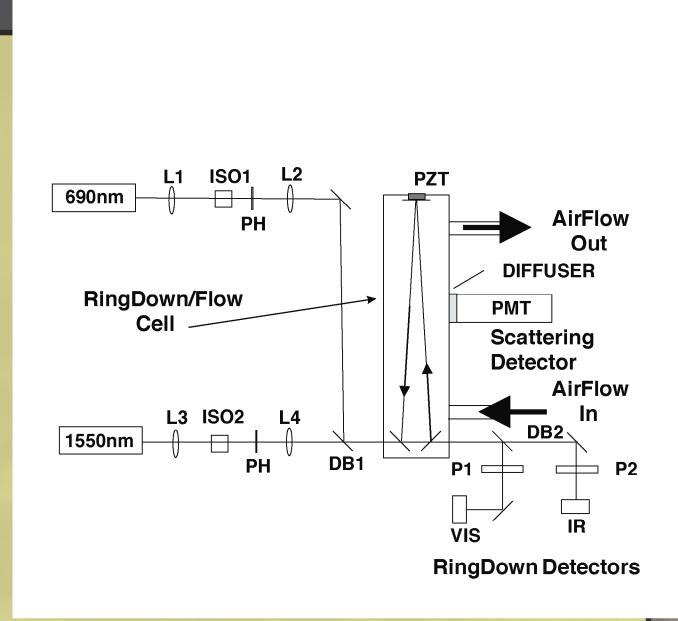
# Continuous-Wave Cavity Ring-Down Measurement of Aerosol Properties

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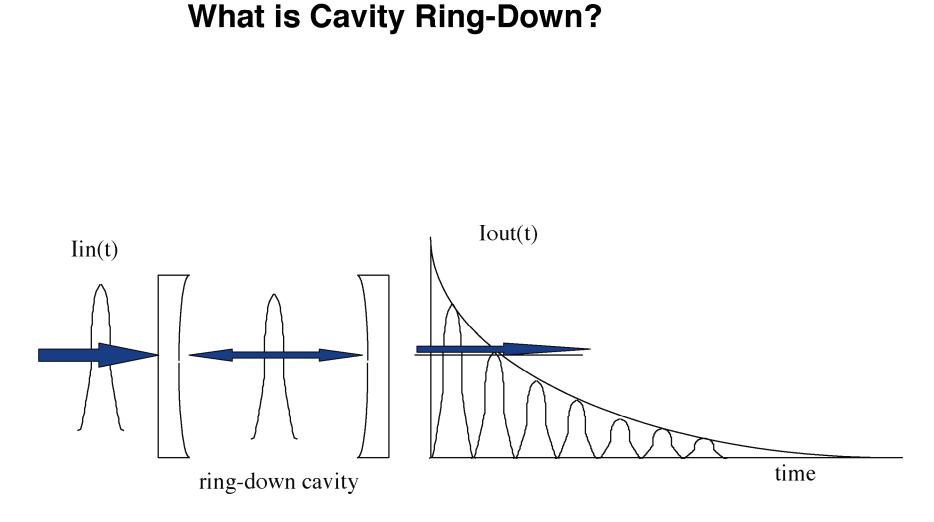
With Contributions From: R. Castaneda<sup>1</sup>, B.A. Paldus<sup>2</sup>, D.S. Baer<sup>3</sup>, J. Ogren<sup>4</sup>, P. Sheridan<sup>4</sup> P. Arnott<sup>5</sup>, H. Moosmüller<sup>5</sup>, S. Varma<sup>5</sup>, D. Covert<sup>6</sup>, A. Virkkula<sup>6</sup>, N. Ahlquist<sup>6</sup>, D. Atkinson<sup>7</sup>

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Schematic of the prototype system

Two CW laser diodes at wavelengths of 690 nm and 1550 nm, are 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. This instrument configuration consists of three mirrors that form a narrow isosceles triangle, unlike the two mirror system described below in the ping pong model. Input and output mirrors are set at 45 deg at one end of the cell and the third mirror is set at the other end of the cell 20 cm away. Light from the output mirror is focused onto the ring-down detectors that are located on the right of the diagram. One wall of the flow cell is made of BK-7 glass. In this configuration, the scattering detectors are located next to the glass wall. Aerosolladen or filtered air enters the cell through 0.64 cm diameter tubing with a flow rate of 1.5 L/min. Air entered the cell at 90° to the optical axis. The Cadenza I and II employed a flow direction transverse to the optical axis. The optical path of the instrument, the path of the laser light through the aerosol-laden flow, was 36 cm. The total size of the instrument is small: 0.46 m x 0.61 m. The electronics associated with the instrument takes up half of an equipment rack. In this CW-CRD application, the back mirror is moved rapidly with a piezo-electric 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 on the order of micro seconds. The ring down signal is then recorded as in pulsed-CRD. Ring-down occurs at a frequency of 50 to 100 Hz in this prototype system and 500 to 1000 shots were averaged over about 10 sec. to achieve one sample.



Ping-Pong Model of Cavity Ring-Down

An excellent review of the CRD techniques and applications can be found in the collection of papers edited by Busch and Busch [1999]. The principle behind CRD is briefly described here using the 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

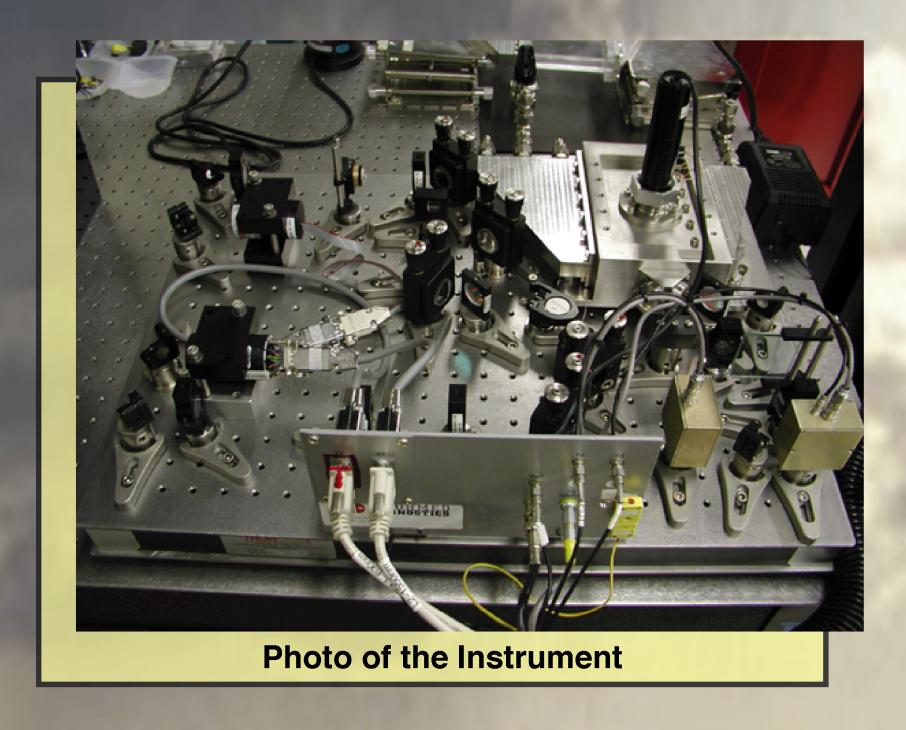
#### $\tau = \frac{L}{c} \left( 1 - R + \sigma_{ext} L + \sigma_{Ray} L + \sigma_{gas} L \right)^{-1}$

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

In the present approach, extinction coefficient is given by the difference between measurements made when the cell contains filtered air and when the cell contains a particulate-laden flow:

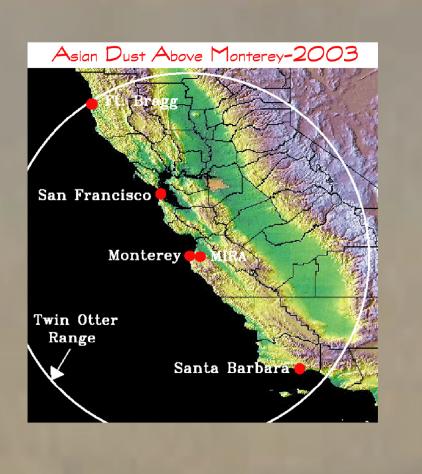
#### $\sigma_{\rm ext} = 1/c(1/\tau_{\rm aer} - 1/\tau_0)$

where  $\tau_{aer}$  is the ring-down time of the aerosol laden flow and  $\tau_0$  is for the filtered air. The minimum detectable absorption of CW-CRD systems is on the order of  $10^{-4}$  to  $10^{-6}$  km<sup>-1</sup>. [Paldus and Zare, 1999] Thus, a measurement accuracy of 1% (to 0.01%) in extinction coefficient is achievable at extinction levels of  $(10^{-4}$  km<sup>-1</sup>)  $10^{-2}$  km<sup>-1</sup>.



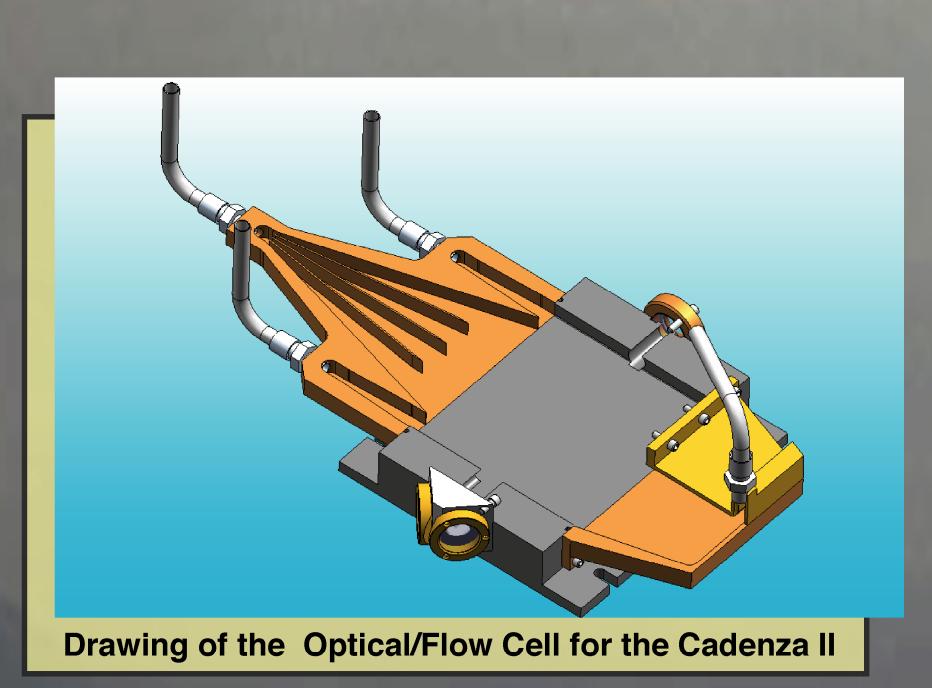
#### Abstract

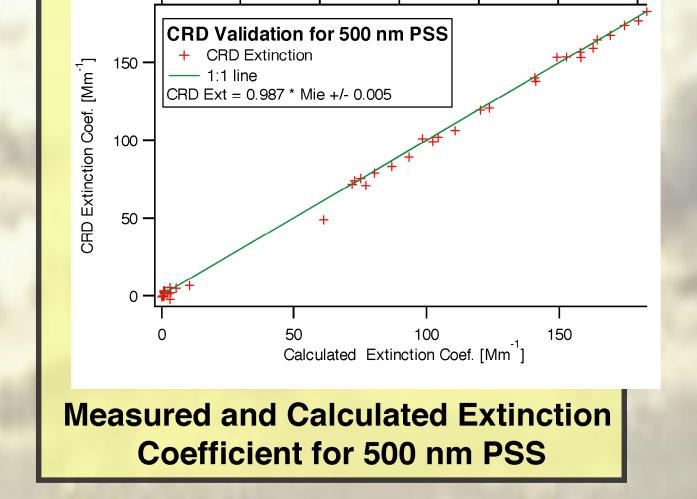
Large uncertainties in the effects that aerosols have on climate require improved in-situ measurements of extinction coefficient and single-scattering albedo. This poster describes the use of continuous wave cavity ring-down (CW-CRD) technology to address this problem. The innovations in this instrument are the use of CW-CRD to measure aerosol extinction coefficient, the simultaneous measurement of scattering coefficient, and small size suitable for a wide range of aircraft applications. Our prototype instrument, Cadenza I, measures extinction and scattering coefficient at 690 nm and extinction coefficient at 1550 nm. The instrument itself is small (60 x 48 x 15 cm) and relatively insensitive to vibrations. Cadenza I has been shown to make accurate and sensitive measurements of extinction and scattering coefficients. Combining these two parameters, one can obtain the single-scattering albedo and absorption coefficient, both important aerosol properties. Minimum sensitivity of Candenza I is 1.5x10<sup>-6</sup> m<sup>-1</sup> (1.5 Mm<sup>-1</sup>). Validation of the measurement of extinction coefficient has been accomplished by comparing the measurement of calibration spheres with Mie calculations. Cadenza I has been tested in our lab and used in the field. Preliminary results from the recent Reno Aerosol Optics Study are presented. Cadenza II, the flight version of this instrument ( $\lambda$ =675, 1550 nm) has recently been integrated into the CIRPAS Twin Otter, and will participate in two aerosol studies ADAM-2003 (Asian Dust Above Monterey) and ARM Aerosol IOP at SGP.





Cadenza II aboard the CIRPAS Twin Otter will participate in two aersol studies ADAM-2003 and ARM Aerosol IOP at SGP





The performance of the Cadenza I instrument is assessed by comparing the measurement of extinction coefficient of calibration polystyrene spheres (PSS) with calculations using a Mie code. The Figure to the left shows this comparison for 500 nm PSS for the 690 nm channel. A TSI Scanning Mobility Particle Sizer (SMPS) was used to obtain a monodisperse aerosol at 500 nm diameter. This was fed into the instrument and the number density was measured using a TSI 3025 CPC. The index of refraction used for PSS in the calculations was (1.45, 0.0). Agreement between measurement and calculation is within 2%.

### The Reno Aerosol Optics Study (RAOS) Preliminary Data

#### Participants:

- NOAA/CMDL (Ogren, Sheridan)
- DRI (Arnott, Moosmüller, Varma)U. Washington (Covert, Virkkula, Ahlquist)
- NASA/Ames (Strawa, Schmid) and Picarro,
- Inc. (Owano, Provencal)

   Thermo Andersen/DLR (Schloesser, Petzold)
- Portland State U. (Atkinson)

#### Objectives:

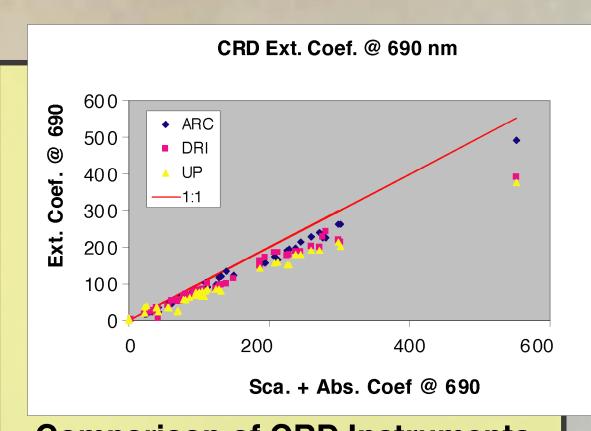
- Characterize new and existing instruments for measuring aerosol light absorption and extinction
- Quantify the uncertainty in the measurements of aerosol light absorption coefficient used by NOAA and ARM
- Derive methods for determining spectral aerosol absorption from multi-wavelength measurements of absorption and (extinction - scattering)

#### Absorption Coefficient Measurements

- Photoacoustic: 532 nm, 1047 nm
- Filter-basedParticle/soot absorption photometer (PSAP)
- #1: 565 nm; #2 460, 540, 660 nm
- Aethalometer: 370, 470, 521, 590, 660, 880, 950 nmCarusso (multi-angle): 670 nm

#### Extinction Coefficient MeasurementsFolded path extinction cell (6.6 m path)

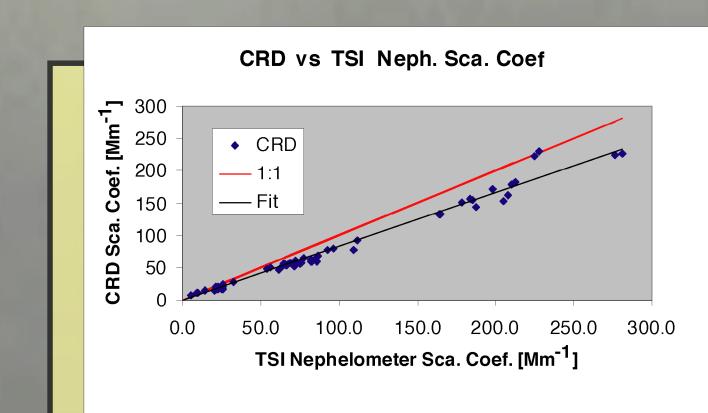
- 460, 540, 660 nm (UW)
- Cavity-ring down (CRD)
  532 nm (DRI)
  690, 1550 nm (Ames/Picarro)
- 532, 1064 nm (PSU)
- Scattering Coefficient Measurements
   TSI 3563 integrating nephelometer 450, 550
- TSI 3563 integrating nephelometer 450, 550, 700 nm
  Radiance M903 integrating nephelometer 530 nm
- DRI integrating sphere nephelometer 532 nm
  Ames/Picarro CRD nephelometer 690 nm



## Comparison of CRD Instruments at 690 nm

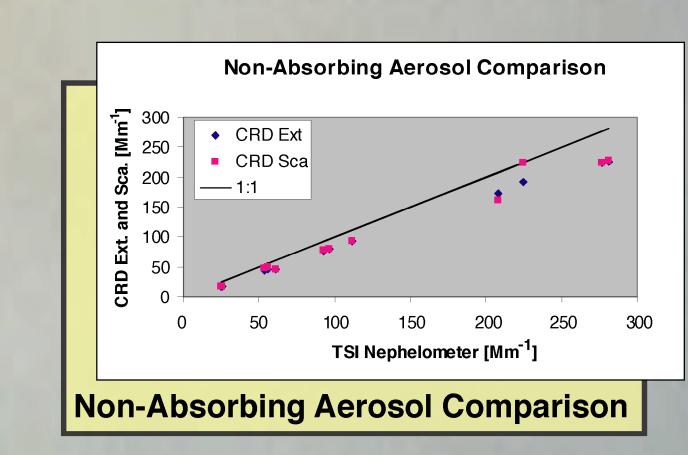
Extinction Coefficients measured by the three CRD instruments are compared to TSI Nephelometer scattering plus DRI Photoacoustic absorption coefficients. The extinction and scattering coefficients are corrected to 690 nm wavelength using an Angstrom Coefficient derived from the 550 nm and 700 nm channels of the nephelometer. The absorption coefficient is corrected using the 532 nm and 1047 nm photoacoustic measurements.

Linear regressions of the measurements yield slope and  $r^2$ : ARC, slope = 0.835,  $r^2$  = 0.96; DRI, 0.774, 0.983; PSU, 0.71, 0.98. Note that all of the CRD extinction measurements are lower than the scattering plus absorption measurements. Correlation coefficients are high.

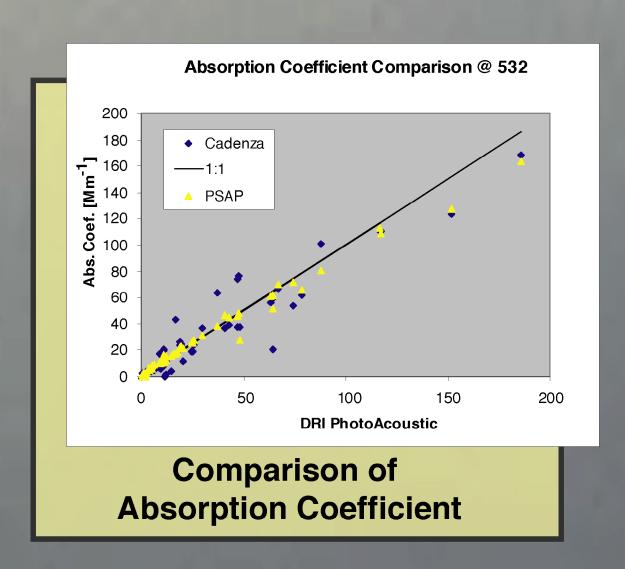


### Comparison of Cadenza and CMDL Nephelometer Scattering Coefficient

Cadenza scattering coefficient is calibrated to the extinction coefficient measured for the non-absorbing aerosol above. This plot shows a comparison of Cadenza vs CMDL Nephelometer scattering for all aerosol tests. A linear regression shows that the Cadenza scattering is below the nephelometer scattering by 17% with a correlation coefficient of 0.98.



CRD extinction and scattering measurements are compared to the TSI nephelometer corrected to 690 nm. Note that the CRD scattering coefficient is calibrated by setting it equal to the CRD extinction measurement for a non-absorbing aerosol, in this case Ammonium Sulfate. The CRD measurements fall below the nephelometer measurements by about 18%, however. This difference is unexplained at this time. The CRD measurement of extinction is a first principle measurement. The only uncertainty is in the length of the optical path through the aerosol laden flow. In this instrument that length is known because the flow is perpendicular to the optical axis. Our confidence in this value and in the CRD extinction measurement is derived from comparisons of measurements to theoretical extinction of calibration spheres described elsewhere in this poster.



Absorption from the CRD instrument and from the CMDL PSAP are compared to that measured by the DRI Photoacoustic, The Ames/Picarro CRD (Cadenza) absorption was obtained by subtracting the measured scattering from the extinction in the CRD cell. The PSAP measurements are lower than the photoacoustic measurements by 9%, with a correlation coefficient of 0.98. The CRD measurements are lower than the photoacoustic measurements by 9% also, with a correlation coefficient of 0.87.