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IN SITU MEASUREMENTS OF AEROSOL OPTICAL PROPERTIES VIA CAVITY RING DOWN TECHNOLOGY WITH AN EMPHASIS ON SPECTRAL PROPERTIES OF CARBONACEOUS AEROSOLS

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ABSTRACT

Large uncertainties in the effects of aerosol on climate require improved in situ measurement of extinction coefficient, and single scattering albedo. This paper describes Cadenza, a new continuous wave cavity ring-down (CW-CRD) instrument designed to address these uncertainties. Cadenza measures the aerosol extinction coefficient for 675 nm and 1550 nm light, and simultaneously measures the scattering coefficient at 675 nm. The use of Cadenza, or future generations of this instrument, aboard a long duration campaign, such as Sophia, will provide needed information pertaining to spatial variability in aerosol properties. This talk will especially focus on the absorption measurement of carbonaceous species.

INTRODUCTION

Aerosols in the troposphere and stratosphere have both direct and indirect effects on the earth's radiation balance. Uncertainties in these effects are well documented by the Intergovernmental Panel on Climate Change [Houghton et al., 2001]. Hansen et al. estimated the global averaged direct forcing due to aerosols to be -0.4 (± 0.3) W/m^2 and the indirect forcing due to aerosols through changes in clouds to be -1.0 (+0.5/-1.0) W/m^2. Furthermore, induced changes in regional radiative fluxes can be an order of magnitude larger than the global mean forcing by aerosols or greenhouse gases [Krishnan and Ramanathan 2002].

Until the last few years, interest in climatic effects of aerosols has mainly focused on the scattering properties of sulphate aerosols (e.g. Kattenberg et al, 1996). Most recently, the effect of aerosol absorption, largely via carbon, has gained attention (e.g. Haywood et al., 1997, Schult et al., 1997). Carbonaceous species, black carbon (BC) and organic carbon (OC) are responsible for most of the absorption associated with aerosol particles. The amount of radiant energy these aerosols absorbs has profound effects on climate and air quality. For example, BC might be the second largest direct radiative forcing agent, next to CO_2, with a positive (i.e. warming) forcing larger than that of methane [Jacobson, 2001].

INSTRUMENTATION – CADENZA

With an increasing knowledge of the utmost importance of aerosol absorption on global climate, it is ironic that aerosol absorption coefficient is one of the most difficult aerosol properties to measure. A new cavity ring-down (CRD) instrument, called Cadenza (NASA-ARC), measures the aerosol extinction coefficient for 675 nm and 1550 nm light, and simultaneously measures the scattering coefficient at 675 nm. Absorption coefficient is obtained from the difference of measured extinction and scattering within the instrument. Cadenza is an autonomous, fast response instrument designed for long duration airborne or ground-based field measurements.

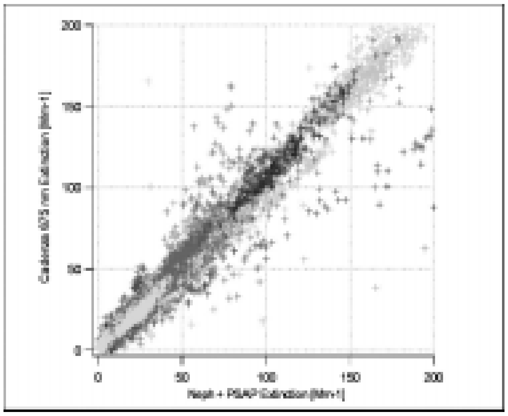

Fig. 1: Cadenza Extinction with TSI Nephelometer Scattering + PSAP Absorption, Overall Comparison of 8-sec. averages for All Flights during Aerosol IOP campaign. Least Squares Bisector =0.982.

Cadenza has repeatedly performed successfully in several recent field missions. In May 2003, Cadenza was deployed in the DOE Aerosol Intensive Operating Period (IOP) in Oklahoma. During these flights Cadenza produced measurements of aerosol extinction in the range of 0.2 to 300 Mm^{-1} with an estimated precision of $0.1Mm^{-1}$ for 1550 nm light and 0.2 Mm^{-1} for 675 nm light. As shown in Figure 1 and 2, Cadenza data from the Aerosol IOP Mission compared favorably with data from other instruments aboard the CIRPAS Twin Otter. The comparison includes TSI Nephelometer and PSAP (Particle Soot Absorption Photometer) data from D. Covert, University of Washington. Recently, Cadenza also productively participated in EVE (Extended MODIS Wavelength Validation Experiment) based in Marina, CA.

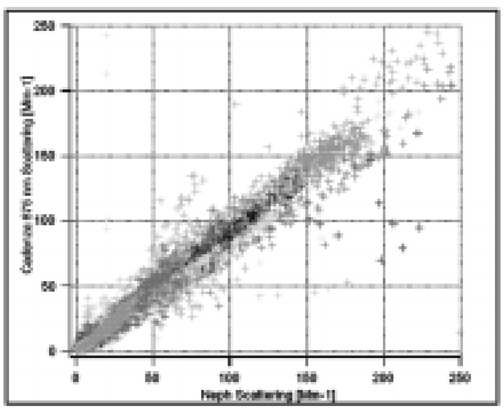

Fig. 2: Cadenza Scattering with TSI Nephelometer Scattering, Overall Comparison of 8-second averages for All Flights during Aerosol IOP campaign. Least Squares Bisector = 0.983.

FUTURE PLANS – DISTINGUISHING BC OR OC; INTERNALLY OR EXTERNALLY MIXED

Typically climate models treat black carbon as the only light-absorbing aerosol component and assume that absorption varies weekly with wavelength [e.g. Bergstrom et al., 2002]. Laboratory studies have generally shown that an aerosol whose only absorbing component is BC general exhibits a small spectral dependence of λ^{-1} with absorption (shown in figure 3). Departures from this λ^{-1} behavior are indicative of organic carbon (OC) or other absorbing species. Of special interest is OC, produced commonly in biomass burning and low temperature combustion processes, which may also contribute significantly to light absorption in the ultraviolet and visible spectral regions [e.g. Kirchstetter and Navakov, 2004; Bond, 2001] (demonstrated in figure 3). The spectral dependence of light absorption in OC compared to BC is clarified with molecular theory. If the spectral dependence of light absorption by aerosols underestimated in models, the aerosol positive radiative forcing will be understated [Bond, 2001]. Furthermore, aerosol extinction of ultra violet sunlight via OC absorption reduced photolysis rates and thus may be responsible for decreases in tropospheric ozone concentrations [e.g. Jacobson, 1999].

Besides composition, the state of mixture (i.e. externally or internally mixed) of BC in atmospheric aerosols is a significant source of uncertainty in quantifying the climatic effect aerosols in models. The state of mixture influences both the direct radiative forcing and the atmospheric lifetime of black carbon. Similar to the distinguishing differences be BC and OC, internal mixtures of BC have a strong absorption feature compared to external mixtures at wavelengths smaller than 600 nm [M.Fiebig, personal communication, 2004].

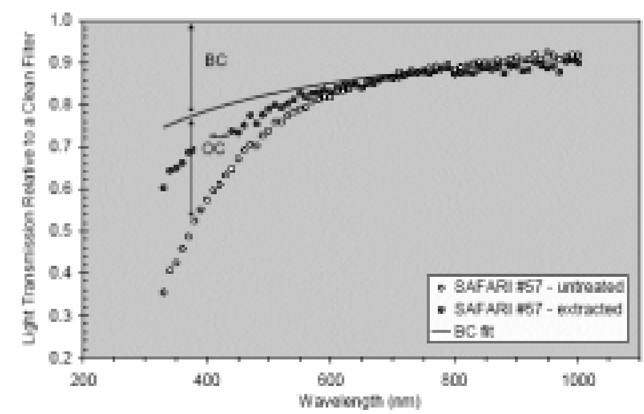

Fig. 3: Taken from TW Kirchstetter et al. 2004 with permission, Illustration of apportionment of light absorption to BC and OC in a biomass smoke sample. Two portions of each filter sample were analyzed: one unaltered and one treated with acetone to extract soluble OC.

Thus to investigate and differentiate carbonaceous species, via production mechanism and mixing states, an extensive data set of in situ measurements of aerosol absorption is need wavelengths below 600 nm. Plans currently exist for a future generation of Cadenza to measure at two shorter wavelengths. This new instrument will measure aerosol extinction and scattering coefficients simultaneously within the same cell in situ at 405 nm and 532 nm. In combination with the current Cadenza, measuring optical properties at 675 and 1550 nm, the spectral response of specific extinction, scattering and absorption coefficients will be investigated thoroughly from the visible to infrared. The extensive flight hours provided within the upper deck of SOFIA could provide an invaluable upper tropospheric data set of aerosol optical properties.

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