

Broadband Measurements of the Mass Absorption Coefficient of Soot Emitted by an Inverted Methane Diffusion Flame

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Emission of soot by combustion systems comprises the primary source of light absorbing carbon-based aerosols, a species which plays an important role in radiative transfer affecting climate change. The starting point of all Global Climate Model (GCM) calculations is the assumption that the wavelength dependence of the mass absorption coefficient (MAC) of soot is λ^{-1} . We present wavelength dependent measurements of nascent soot produced by a methane diffusion flame over a wide spectral range (300-660 nm) in order to determine the validity of this assumption.

For all these measurements, soot was generated using a methane diffusion flame operated at an overall fuel to oxygen ratio of 0.7 produced by an inverted burner. The advantage of the inverted configuration is that the buoyant gases from the downward combustion flow prevent the flame from flickering, producing relatively steady size distributions and concentrations, not only over a short measurement period, but from day-to-day. A small fraction of the soot and combustion gases was sampled using an eluter which serves to heavily dilute the soot sample with nitrogen so as to minimize any coagulation of the soot particles and to keep the relative humidity of the gas phase below 15%. The soot sample then flows through a neutralizer which produces an equilibrium charge distribution on the soot sample and then into a DMA where a nominal particle size can be selected. The output of the DMA is then directed to a Centrifugal Particle Mass Analyzer (CPMA) where a fixed particle mass is then chosen.

The output of the CPMA is then sent to instruments for the measurement of number density (MCPC), mass (CPMA) and the presence of multiply charged particles (SP2). Soot absorption is also measured with three instruments. The first two are photoacoustic-based instruments which directly measure absorption, one a laser-based monitor operating at 532 and 405 nm and the second, a lamp-laser hybrid employing laser operating at 405 nm and 660 nm and an incoherent light source combined with a rotating filter wheel that produces measurements at a total of 8 different wavelength from 301 to 660 nm. The third monitor, a CAPS PM_{ssa} monitor, provides an indirect absorption measurement. It employs cavity attenuated phase techniques to measure the total optical extinction and an internal integrating nephelometer to measure the

scattering component. Absorption is derived from the subtraction of these two quantities. At the low single scattering albedo values measured (0.1-0.2), this approach provides an extremely accurate measurement of the soot absorption.

The results are shown below in Fig. 1. The value of the MAC ranges from $6 \text{ m}^2 \text{ g}^{-1}$ at 660 nm to $19 \text{ m}^2 \text{ g}^{-1}$ at 301 nm. The error bars represent an estimate of the accuracy of the relative absorption (not the MAC) measurement at each point. Errors in the measurement of the total mass loading, estimated at $\pm 7\%$ (which includes contributions from measurement of soot particle number density, per particle mass and the influence of multiply charged particles), cause the entire set of points to move up or down in concert. Plotting $\log(\text{MAC})$ versus $\log(\lambda)$ and using a linear least squares fit produces an Ångstrom absorption exponent (AAE) of 1.25 ± 0.25 where the uncertainty is 2σ in value. The data seem to suggest that the AAE of nascent methane-flame produced soot could be slightly greater than 1.

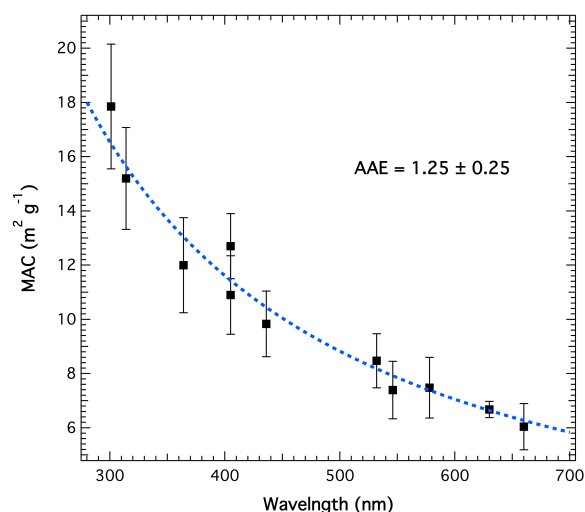


Figure 1. Measurement of the mass absorption coefficient (MAC) of methane flame-produced soot as a function of wavelength.

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