

Quantifying and reducing the uncertainties of in situ aerosol absorption measurements with the extinction-minus-scattering technique

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Aerosol light absorption contributes to the earth's radiative forcing budget directly, through the absorption of solar radiation, and indirectly, by altering cloud formation and longevity. Black carbon typically dominates aerosol light absorption in the atmosphere, with significant but poorly constrained influences from brown carbon and mineral dust. Sources of these absorbing aerosols include numerous combustion processes such as biomass burning and power generation, as well as dust entrainment.

Quantitative and accurate measurement of aerosol light absorption is challenging. Filter based techniques are prone to multiple measurement artefacts such as concentration dependence, changes to filter behavior due to particle loading, and scattering cross-sensitivity. In situ absorption measurements include photoacoustic, refractive-index-based, incandescence-based, and extinction-minus-scattering (EMS) techniques. Historically, applications of the EMS technique have most commonly used two separate instruments measuring scattering and extinction; but the relatively recent CAPS PM_{SSA} is one of the first instruments to simultaneously measure ambient extinction and scattering for the same aerosol sample, greatly reducing measurement uncertainties. However, no technique is without its limitations. For example, in EMS, as the particle diameter approaches the wavelength of the laser, the fraction of light scattered at small angles increases in magnitude and more light is lost through the exit of the integrating sphere. This light loss or "truncation effect" increases with particle diameter.

To quantify this truncation effect, we paired a CAPS PM_{SSA} to the Aerodynamic Aerosol Classifier (AAC) which, unlike all other aerosol classifiers, supplies a truly monodisperse aerosol sample. Using this system, we quantified this truncation effect across a wide particle size range (70nm – 5µm) for both polystyrene latex (PSL) and ammonium sulfate aerosols. We explore the reasons behind observed discrepancies between the measurements and a range of different model calculations and present an optimized model, which accounts for angle-dependent light scattering at the internal glass surface of the CAPS PM_{SSA}. Our results serve to reduce uncertainties in EMS measurements for improved ambient monitoring.