A Novel Humidity-Controlled Single Scatter Albedo Monitor to Quantify the Effects of Water on Light Absorption by Black and Brown Carbon

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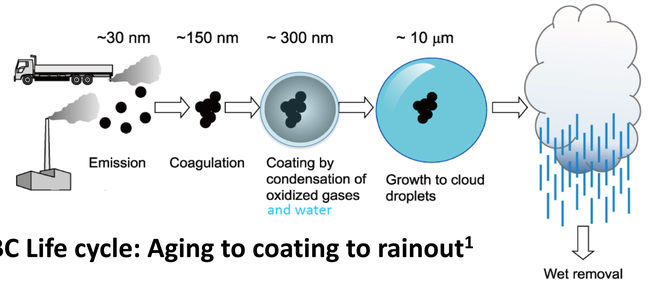
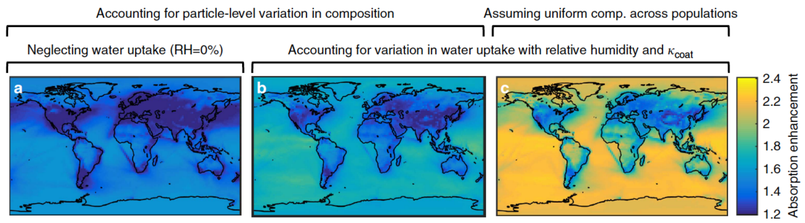
Motivation

Black carbon (BC) emitted by fossil fuel combustion and biomass burning gets coated with organics and water as it ages in the atmosphere and is eventually removed by rain.[1] Transparent organic coatings on BC have been shown to amplify their light absorption cross section by lensing in laboratory and field measurements and simulated by core shell Mie theory. However, the absorption enhancement by water coatings is not well measured due to instrumental challenges since direct techniques like photoacoustic spectroscopy become non-linear due to water desorption at high relative humidity (RH).[2] Climate model simulations show that the absorption enhancement of BC coated by water over the oceans is much greater than that by organics over land, underscoring the need to measure it.



BC Life cycle: Aging to coating to rainout[1]

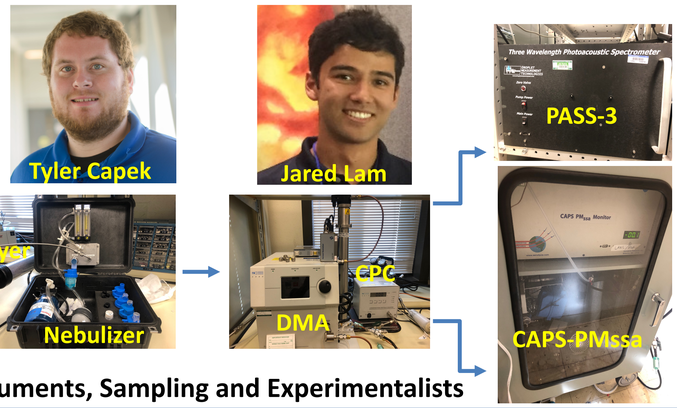


Spatial variation in modeled BC absorption enhancement[3]

We report first results with the calibrated new humidity controlled optical instrument on the the effect of water uptake on absorbing particles.

Humidified Extinctiometer/Nephelometer

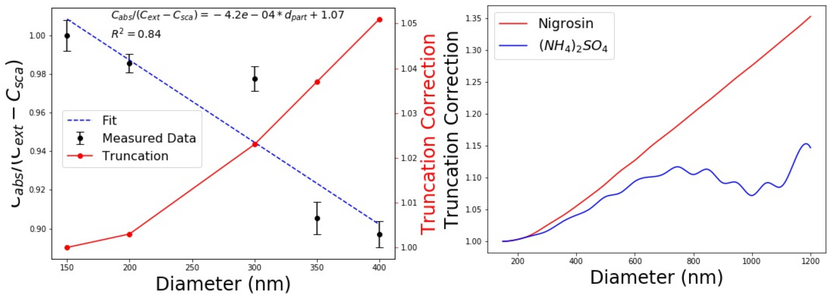
We developed a humidity scanning system for the Cavity Attenuated Phase Shift monitor (CAPS PMssa, Aerodyne Research Inc.) that measures extinction and scattering 450 nm[4] on the same aerosol sample. Absorption is derived as (extinction –scattering) and single scatter albedo (SSA) as scattering/extinction). We demonstrated that our instrument scans RHs from 20% to 85% over a few minutes.



Instruments, Sampling and Experimentalists

Truncation Correction and Experiments

The CAPS-PMssa monitor has a systematic truncation angle effect that reduces scattering by light loss through the ends of the cavity.[4] We compute truncation correction using Mie theory that requires knowledge of the refractive index and size of the particle. We use complex refractive indices of ammonium sulfate and nigrosin of m=1.521+0.002i and m=1.70+0.31i respectively.[5] A truncation correction was calculated for diameters from 150-1200nm. The dry particles measured by the CAPS-PMssa were size selected using a Differential Mobility Analyzer (DMA), which is then used to estimate particle growth in a humid environment with κ-Köhler Theory[6]. For nigrosin and ammonium sulfate these parameters are $\kappa = 0.1656$ and $\kappa = 0.534$ respectively[6]. κ-Köhler estimates particle size at each RH for the truncation correction calculation. The CAPS-PMssa Mie correction has only been calibrated by scattering polystyrene particles. We extended this to absorbing particles by direct light absorption measurements on dry size selected dry nigrosine aerosols with a 3-wavelength photoacoustic (PASS-3 at 405, 532 and 781 nm, DMT)[7] and compared it to CAPS-PMssa absorption.

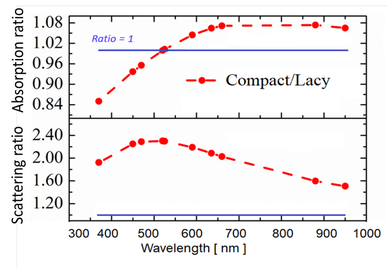


Observed Truncation **Mie Truncation correction**

The ratio of the measured PASS-3 and CAPS-PMssa derived absorptions for 150nm nigrosine particles agree to within the calibration errors. Since truncation errors are negligible at small sizes this ratio is scaled to 1. The ratio falls with increasing size due to truncation and is consistent with the independently calculated Mie truncation correction. The truncation correction for nigrosin is larger than for ammonium sulfate but small (<10%) for particles smaller than 400nm and we limit absorbing water uptake studies to this range. The κ values we determine for the truncation correction for ammonium sulfate (0.25-0.45) and nigrosin (~ 0.1) are consistent with published measurements.

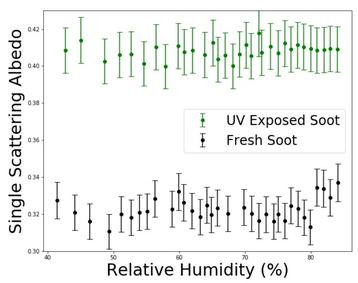
UV irradiation of soot (propane burner)

Restructuring
Compaction
Enhanced Scattering
Reduced Absorption



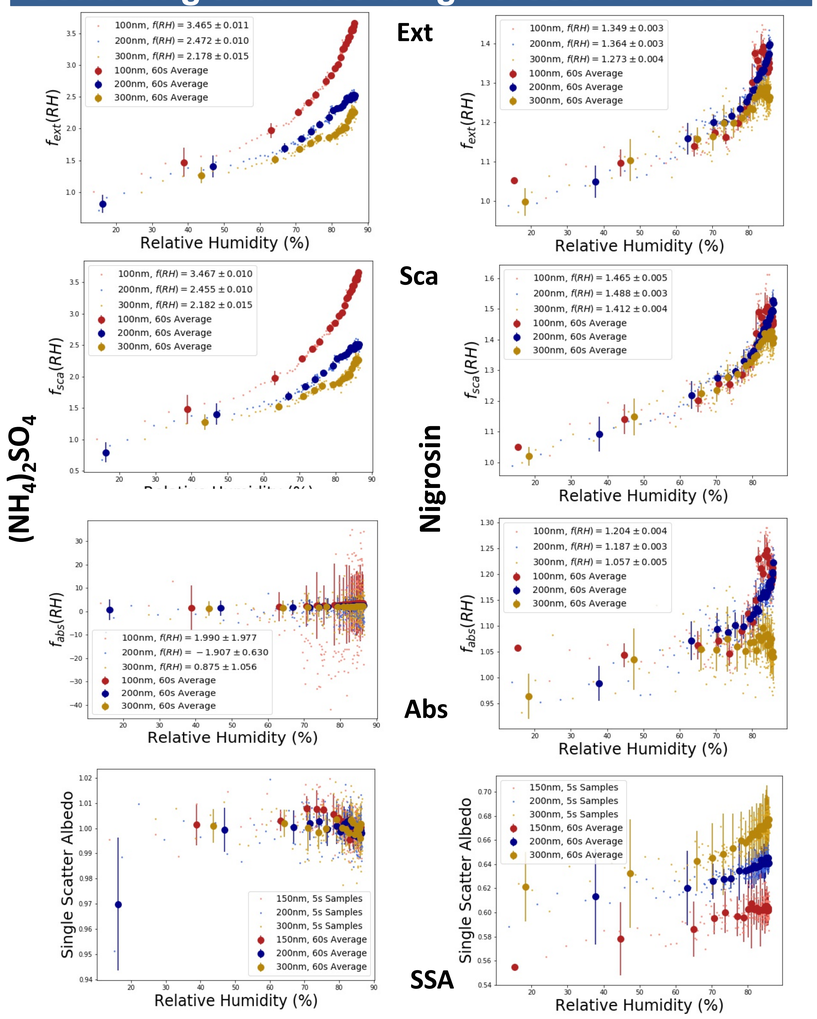
T-Matrix calculations

CAPS-PM$_{SSA}$ Measurements
No water uptake
SSA increase upon UV exposure
Consistent with compaction



S. China, et al., *Geophys. Res. Lett.* **42**, 1243-1250 (2015)

Scattering and Absorbing Aerosol RH Results



Conclusions

- We built and calibrated a sensitive RH scanning CAPS-PMssa dedicated to measuring the effect of water coatings on light absorbing aerosol optics.
- We developed a Mie and κ-Köhler Theory algorithm to compute the truncation error and validated it for absorption by dry PASS-3 measurements.
- Our scattering and extinction enhancements as a function of RH for ammonium sulfate are consistent with extensively published results.
- We observed 20-25% absorption enhancements for 150 and 200 nm nigrosin (12% at 300nm) and a 10% rise in SSA (0.62 to 0.68 at 150nm) at high RH.
- *We will investigate RH effects on organic nitrate BrC, aged laboratory BC and BC in Houston for ARM-TRACER campaign to validate optical models.*

References

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