A21B-0063

Aerosol Hygroscopicity in an Urban Environment: Airborne Observations during NASA DISCOVER-AQ

LUKE D. ZIEMBA, Andreas J. Beyersdorf, Gao Chen, Suzanne Crumeyrolle, Rich Ferrare, Charlie Hudgins, K. Lee Thornhill, Edward L. Winstead, and Bruce E. Anderson

NASA Langley Research Center

Contact: luke.ziemba@nasa.gov

A21B-0063

Introduction

- Aerosol hygroscopicity (the propensity of particles to uptake water) is an important factor in determining optical properties and cloud activation.
- The relationship between scattering coefficient and relative humidity (f(RH)) is integral for:
  1.) understanding the role of chemical composition on optical properties,
  2.) extrapolating airborne measurements to ambient conditions for evaluation of remote sensing observations, and
  3.) understanding the importance of aerosol water content.
- An empirical relationship is currently used to describe f(RH); validation is necessary for variable atmospheric conditions.
- Data from the DISCOVER-AQ campaign on the NASA P-3B was used to systematically explore aerosol hygroscopicity in the urban Washington, DC/Baltimore, MD area.

Objectives and Impacts

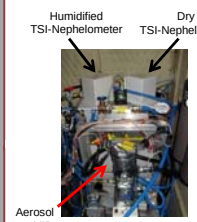
- Document the f(RH) relationship in an urban area as a function of altitude
- Investigate the dependence of hygroscopicity on chemical composition and aerosol size
- Validate the f(RH) model using a combination of in-situ and remote sensing observations
- Explore the temporal and spatial variability in aerosol hygroscopicity

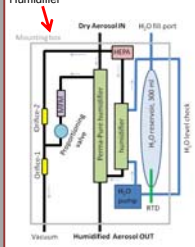
Conclusions and Future Work

- An average f(RH) value of ~1.6 was measured in the DC/Baltimore BL and decreased to nearly 1.2 at higher altitude.
- Hygroscopicity was found to increase throughout the day and with decreasing organic content.
- Correction of dry extinction to ambient humidity resulted in good agreement with an independent, remote sensing measurement.
- Evaluation of the f(RH) model showed a minor RH-dependent bias, especially for highly hygroscopic aerosol.
- Future objectives:
  1. Focus on high humidity conditions
  2. Systematic analysis of spatial variability
  3. Evaluation of hygroscopicity for highway sampling
  4. Development of scanning-RH capability
  5. Relationship with CCN concentration

We would like to thank the EV-1 DISCOVER-AQ project for funding, the science team, and especially the P-3B crew and pilots at NASA Wallops Flight Facility

1. Measurements on the NASA P-3B





- Scattering coefficient (σ_{scat}) was measured at 1 Hz for both humidified and dry conditions.
- Nephelometers were operated in parallel and dried by a Perma-Pure Nafion® column.
- $f(RH)_{in\text{-}situ}$ was calculated by:

$$Y = \frac{\ln\left[\frac{\sigma_{scat,humidified}}{100 - RH_{humidified}}\right]}{\ln\left[\frac{\sigma_{scat,dry}}{100 - RH_{dry}}\right]}$$

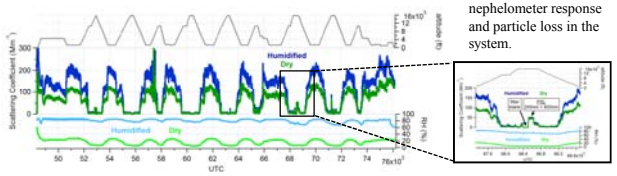
$$f(RH)_{in\text{-}situ} = \frac{\sigma_{scat,80\%}}{\sigma_{scat,20\%}} = \frac{\left[1 - \frac{80}{100}\right]^{(-\gamma)}}{\left[1 - \frac{20}{100}\right]^{(-\gamma)}}$$

- Humidifier was operated with constant sample flow, constant H_2O temperature, and a variable sheath flow through an additional Perma-Pure Nafion® column.
- Humidifier operated autonomously for 8-hour flights; minimal H_2O consumption or effects of variable pressure on system control were observed.

Measured Parameter	Instrument	Size Range (μm)
Dry Aerosol Size Distributions, Aerosol Volume	DMT UHSAS	0.06 – 1
Dry Total Scattering Coefficient at 450, 550, and 700 nm	TSI-3563 Nephelometer	< 5
f(RH) for Scattering at 450, 550, and 700 nm	TSI-3563 Nephelometer w/ 80 % humidification	< 5
Total Absorption Coefficient at 467, 530 and 660 nm	PSAP	< 5
Aerosol water-soluble organic carbon concentration	PILS w/Sievers TOC	<5
Aerosol inorganic ion concentration	PILS / offline IC	<5
Black carbon mass concentration	SP2	0.1 – 0.5

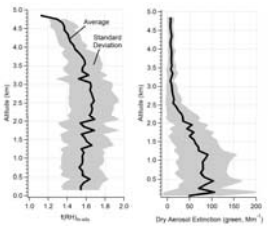
UHSAS – Ultra-High Sensitivity Aerosol Spectrometer
PSAP – Particle Soot Absorption Photometer
PILS – Particle Into Liquid Sampler
TOC – Total Organic Carbon
IC – Ion Chromatography
SP2 – Single Particle Soot Photometer

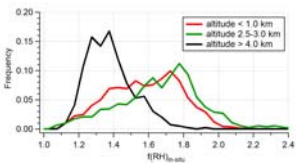
- An example of raw data is shown below.
- In-flight calibration aerosol (PSL, 200nm + 600nm diameter) was used to check nephelometer response and particle loss in the system.



2. Overview of Vertical and Diurnal Variability

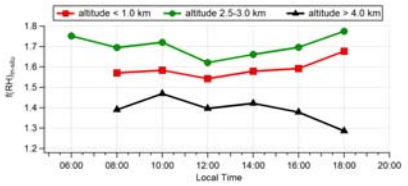
- The dry extinction coefficient profile shows that the majority of aerosol loading was confined to the lower 2.5-km of the atmosphere.
- $f(RH)_{in\text{-}situ}$ showed a general increasing trend up to 2.5 km, from 1.55-1.65 with a constant range of ± 0.2.
- $f(RH)_{in\text{-}situ}$ decreased above this level to an average value of less than 1.4 at 4.7 km.





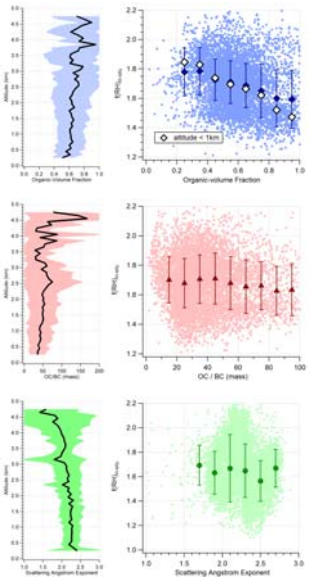
- Decreased $f(RH)_{in\text{-}situ}$ values at high altitude may be the result of preferential wet-depositional removal of more water-soluble particles.
- Frequency histogram analysis further illustrates the altitude dependence of $f(RH)_{in\text{-}situ}$.

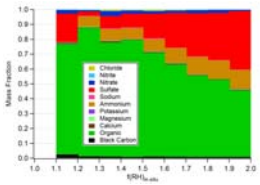
- For both the boundary layer (BL) and 2.5-3.0-km altitude bins, $f(RH)_{in\text{-}situ}$ increased from 12:00 to 18:00 local time, consistent with photochemical processing yielding more hygroscopic aerosol.
- The opposite effect was observed at high altitude.



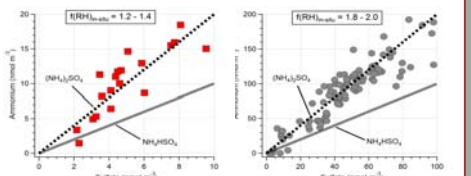
3. Hygroscopicity and Chemical Composition

- Chemical composition measurements from the PILS showed that organic compounds dominatedthe resolved mass.
- The distribution of resolved mass shifts from nearly 90% organic at $f(RH)_{in\text{-}situ}$ of 1.25 to less than 50 % at 1.95, suggesting that the organic compounds present are considerably less hygroscopic than the sulfate component.
- A comparison of the organic volume fraction (the ratio of PILS-derived organic volume to the total volume measured by the UHSAS) yields a similar trend that is exacerbated in the BL.



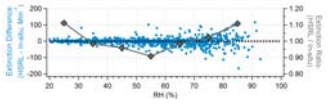


- On the contrary, the mass ratio OC/BC showed no discernible trend in $f(RH)_{in\text{-}situ}$ suggesting that any variation in the source of organic aerosol did not affect hygroscopicity.
- Similarly, no $f(RH)_{in\text{-}situ}$ dependence was observed with scattering Angstrom exponent (AE_{scat}, a proxy for particle size).
- Very little variability was observed in AE_{scat}, consistent with a predominance of pollution aerosol, especially in the lowest 2.5 km.
- The sulfate aerosol observed during DISCOVER-AQ appears to be fully neutralized by ammonium.
- The presence of ammonium sulfate is consistent for both the low $f(RH)_{in\text{-}situ}$ and high $f(RH)_{in\text{-}situ}$ cases.



- $f(RH)_{in\text{-}situ}$ is used to relate dry observations to ambient RH conditions for comparison directly with remote sensing measurements and to better understand aerosol water content.
- Ambient scattering ($\sigma_{scat,\ ambient}$) and extinction coefficients ($\sigma_{ext,\ ambient}$) are calculated as follows:

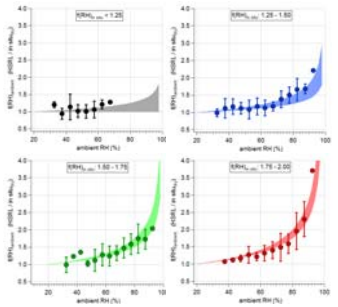
$$\sigma_{scat,ambient} = \sigma_{scat,20\%}\left[\frac{1 - \frac{RH_{ambient}}{100}}{1 - \frac{20}{100}}\right]^{(-\gamma)} \qquad \sigma_{ext,ambient} = \sigma_{scat,ambient} + \sigma_{abs}$$



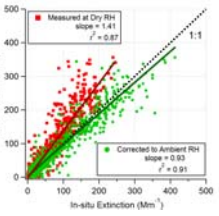
- HSRL and in-situ observations together allowed a unique assessment of the f(RH) model relationship:

$$f(RH)_{ambient} = \frac{\sigma_{ext,HSRL}}{\sigma_{ext,dry}}$$

- A comparison of observations with model predictions (shaded areas) reveals generally good agreement.
- Potential bias was observed for high-f(RH) aerosol; in-situ extinction systematically exceeded HSRL at RH between 50-80%

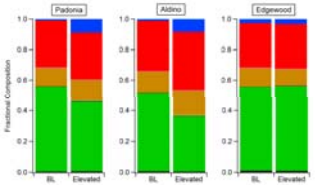


4. Ambient Extinction: Comparison with HSRL



- Good agreement was observed between co-located, coincident observations with the NASA High Spectral Resolution Lidar (HSRL); correction improves both slope and scatter of the relationship.
- Discrepancies showed some humidity dependence and varied no more than ± 10%.
- For the full campaign, no dependence was observed for the extinction bias with chemical composition.

5. Case Study – Flight 10 – 22 July 2011



- A consistent elevated layer was observed at 3 sites by both in-situ and HSRL platforms.
- Extinction coefficient discrepancy differs between the BL and the elevated layer.
- Nitrate-enhancements seem to coincide with HSRL/in-situ-difference trend in elevated layer suggesting a dependence on chemical composition.