Determining Aerosol Hygroscopicity through Airborne In-situ and Remote Sensing Observations in an Urban Environment during NASA DISCOVER-AQ

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ABSTRACT: The DISCOVER-AQ (Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) mission conducted its first field deployment in Washington D.C./Baltimore region. The overarching goal is to better understand the remote sensing column measurements for diagnosing the near surface air quality. To this end, aerosol optical properties (scattering and absorption) are the basis for complex remote sensing retrievals and their relationships with chemical composition can bridge the gap between satellite observations and air quality measurements. Aerosol hygroscopicity, the propensity of a particle to take up water, is an important parameter for understanding how particles scatter visible light. Both hygroscopicity and absorption enhancements are inherently related to the chemical composition of particles; changes to that composition can result in increased hygroscopicity and absorption. This results in a complex vertical and spatial distribution of aerosol properties in urban environments.

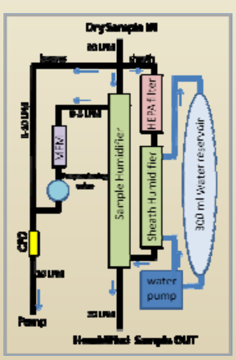
To achieve the science objectives, the DISCOVER-AQ project adopted a sampling strategy involving two NASA aircraft, i.e., P-3B and UC-12, for highly coordinated vertical profiling. The P-3B was dedicated for in-situ observations and repeatedly spiraled over the 6 MDE (Maryland Department of the Environment) ground sites. The UC-12 was used to conduct remote sensing measurements over the region of the interest in coordination with the P-3B sampling. The P-3B aircraft was equipped with comprehensive aerosol measurements for microphysical, optical and chemical properties. We present aerosol scattering, absorption, and hygroscopicity observations from the P-3B airborne observations. The dry aerosol scattering coefficient was measured by a TSI nephelometer and the absorption coefficient was determined by a particle soot absorption photometer (PSAP). A closure analysis has shown high level of consistency between the observed dry scattering coefficient and Mie-theory calculation based on independently observed aerosol number size distribution. The aerosol hygroscopicity is derived from dry (relative humidity (RH) < 40%) and humidified (RH > 75 %) scattering coefficients measured by two additional nephelometers. The derived hygroscopicity was then assessed by comparing the in-situ observed aerosol extinction coefficients (sum of scattering and absorption coefficients) corrected to ambient humidity with the extinction profile measured by HSRL (high spectral resolution lidar) system, which is conducted under ambient conditions. Comparisons are made over for vertical profiles in close proximity in space and time with the HSRL measurements. Among the nearly 250 NASA P-3B vertical profiles flown during the DISCOVER-AQ field deployment, a total of 85 cases were indentified for the comparison. The cases cover an AOD range of less than 0.1 to 0.7 and RH value range from 2 to 96%. A close agreement was found in the comparison which provides a high level of confidence for both the hygroscopicity model and for our ability to accurately measure AOD.

DISCOVER-AQ Aerosol Instrumentation

Summary of P-3B in-situ aerosol measurements

Parameter	Technique/Instrument	Time Resolution	Size Range (µm)
Total Particle Number Density	Condensation Particle Counters (TSI 3025)	1 s	0.003 – 1
Particle Number Density	Condensation Particle Counters (TSI 3010)	1 s	0.010 – 1
Nonvolatile Particle Number Density		1 s	0.010 – 1
Aerosol Particle Size Distribution	TSI Scanning Mobility Particle Sizer	120 s	0.01 – 0.3
	DMT Ultra-High Sensitivity Aerosol Spectrometer	5 s	0.08 – 1.0
	Aerodynamic Particle Sizer (TSI 3321)	5 s	0.5 – 10
Scattering Coefficient	Nephelometer (TSI 3563)	1 s	<10
Absorption Coefficient	Particle Soot Absorption Photometer	5 – 120 s^{b}	<10
Black Carbon Density	DMT Single Particle Soot Photometer	5 s	0.01 – 1.0
Particle Chloride Concentration*	Particle into Liquid Sampler/Ion Chromatograph	300 s	0.01 – 1.0
Particle Nitrate Concentration*			
Particle Sulfate Concentration*			
Particle Ammonium Concentration*			
Particle Sodium Concentration*			
Particle Potassium Concentration*			
Water Soluble Organic Carbon (WSOC) concentration	Particle into Liquid Sampler/Total Organic Carbon	30 s	0.01 – 1.0

P-3B f(RH) Measurement:



The aerosol hygroscopicity is assessed through measurement of the scattering growth factor, f(RH), defined as:

$$f(RH) = \frac{\sigma_{scat,wet}}{\sigma_{scat,dry}}$$

Where $\sigma_{scat,\ wet}$ and $\sigma_{scat,\ dry}$ were measured by two TSI nephelometers. The instrument "dry" and "wet" conditions were set to RH = 40% and 80%, respectively. The "wet" condition was achieved through the device illustrated on the left.

- ✓ Dry sample was split into a sheath, bypass flow, and sample flow to maintain a constant system flow rate.
- ✓ Sample is not diluted, losses of submicron aerosol are negligible.
- ✓ System is controlled automatically and adjusts for pressure/temperature variability
- ✓ an insulated RH sensor is used (Vaisala, model HMP60).
- ✓ Dry RH varied typically between 10 and 30 % and humidified sampled was typically 80 ± 4 %

Summary of UC-12 remote sensing measurements

Parameter	Instrument	Time Resolution	Spatial Resolution
Aerosol Backscatter (532 & 1064 nm)	High Spectral Resolution Lidar	10 s	~1 km hor. 60 m vert.
Aerosol Extinction (532 nm)		60 s	~6 km hor. 300 m vert.
Depolarization (532 & 1064 nm)		10 s	~1 km hor. 60 m vert.
Aerosol Optical Depth (532 nm)		60 s	~6 km hor.

Science Objective: **to assess the in-situ observation of aerosol hygroscopicity through extensive comparison of the derived in-situ extinction and direct remote sensing measurements under ambient humility conditions**

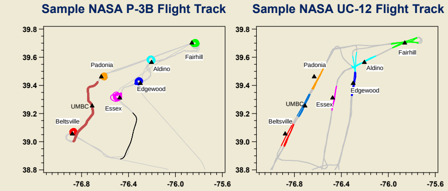
General Approach:

1. **Assess the dry scattering and size distribution measurements by conducting the Mie-scattering closure test**
2. **Derive ambient extinction by applying observed hygroscopicity to dry scattering coefficient and combining with the observed absorption coefficient**
3. **Identify coincident profile measurements from P-3B and UC-12**
4. **Compare in-situ and remote sensing ambient extinction measurements at a variety of environmental conditions**

DISCOVER-AQ Sampling Strategy

DISCOVER-AQ sampling strategy requires repeatedly profiling over the six MDE ground monitoring sites by both NASA P-3B and UC-12 aircraft. During the 14 P-3B flights, a total of 247 spirals were flown. Among these, 109 P-3B vertical profiles were coincident with the UC-12 remote sensing profiles, within 15 km and 15 min.

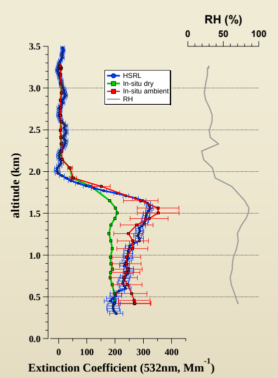
Sample NASA P-3B Flight Track **Sample NASA UC-12 Flight Track**



Summary of Conditions for P-3B Profile Coincidental with UC-12

RF	July Date	Coincident Profiles	f(RH)	Ambient RH (%)	Sulfate (µg m^{-3})	WSOC (µg m^{-3})	CO (ppbv)
1	1	10	1.28 (0.09)	46 (10-81)	NA	1.43 (0.22)	138 (6)
2	2	13	1.35 (0.06)	46 (2-66)	NA	NA	173 (25)
3	5	14	1.45 (0.07)	61 (7-86)	NA	2.03 (0.39)	166 (14)
4	10	8	1.44 (0.07)	56 (3-76)	1.07 (0.45)	4.53 (1.07)	187 (15)
5&	11	5	1.49 (0.04)	67 (21-84)	3.50 (0.39)	6.73 (0.46)	225 (16)
6	14	12	1.31 (0.06)	50 (3-74)	0.18 (0.18)	1.64 (0.17)	117 (5)
7	16	0	1.39 (0.03)	56 (8-74)	0.61 (0.25)	2.44 (0.42)	134 (12)
8	20	6	1.91 (0.07)	73 (8-86)	5.36 (1.32)	5.00 (0.85)	184 (29)
9	21	7	1.70 (0.06)	70 (11-80)	8.46 (2.08)	7.35 (1.57)	218 (29)
10	22	7	1.76 (0.05)	70 (26-88)	6.26 (1.27)	4.83 (0.65)	199 (19)
11	26	11	1.73 (0.06)	51 (5-84)	1.76 (0.85)	1.41 (0.62)	128 (15)
12	27	7	1.59 (0.05)	55 (2-64)	0.55 (0.15)	2.39 (0.24)	115 (3)
13	28	0	1.67 (0.05)	67 (31-89)	3.32 (1.17)	4.79 (0.89)	182 (28)
14	29	9	1.74 (0.05)	72 (11-83)	6.53 (0.86)	5.62 (0.69)	183 (9)

Example of aerosol extinction comparison: P-3B vs. UC-12



- HSRL extinction at 532 nm reported at 300 m resolution with an uncertainty of ~ 22%
- In-situ extinction converted to ambient condition at 532 nm
- In-situ ambient extinction averaged into 300 m interval
- The propagated in-situ ambient extinction uncertainty is ~ 20%.
- The dry in-situ extinction uncertainty is ~ 10%
- The estimated f(RH) uncertainty is about 17%.
- RH derived from P-3B Hygrometer measurements

Deriving of $\sigma_{ext,\ ambient}$(532nm): in-situ ambient extinction at 532nm

$$\sigma_{ext,ambient}(532nm) = \sigma_{scat,ambient}(532nm) + \sigma_{abs,dry}(532nm)$$

$$\sigma_{scat,ambient}(532nm) = \sigma_{scat,ambient}(550nm)\left(\frac{550}{532}\right)^{\alpha_{ambient}}$$

$$\sigma_{scat,ambient}(550nm) = \sigma_{scat,dry}(550nm)\left(\frac{1-\frac{RH_{dry}}{100}}{1-\frac{RH_{ambient}}{100}}\right)^{\gamma}$$

$$\gamma = \frac{\ln\left[\frac{\sigma_{scat,wet}}{\sigma_{scat,dry}}\right]}{\ln\left[\frac{100-RH_{dry}}{100-RH_{wet}}\right]} \qquad \alpha_{ambient} = \frac{\ln\left(\frac{\sigma_{scat,ambient}(550nm)}{\sigma_{scat,ambient}(450nm)}\right)}{\ln\left(\frac{450}{550}\right)}$$

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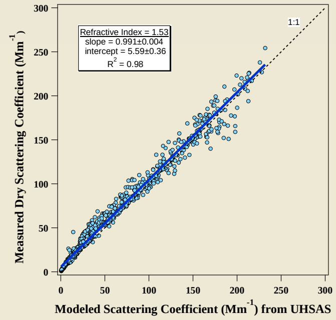
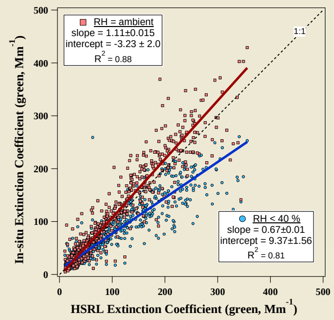


Mie-Scattering Closure Test:

- UHSAS size distribution based on ammonium sulfate calibration
- Aerosol refractive index assumed at 1.53
- Test based on all available data and scattering coefficient at 550 nm
- Regression line based on orthogonal distance regression method



HSRL vs. In-situ Comparison:

- A total of 663 points involved in the Correlation Analysis
- In-situ extinction converted to 532 nm
- In-situ extinction conversion to ambient conditions based on f(RH) measurements
- The in-situ ambient extinction is consistent with the HSRL measurements within the estimated uncertainty

Tentative Conclusions

- Closure test showed high level consistency in measurements of aerosol microphysical and optical properties
- The extensive comparison based on DISCOVER-AQ observations shows that the derived in-situ ambient aerosol extinction is consistent with the HRSL direct measurements well within the combined uncertainties, which validates the in-situ f(RH) measurements
- This study has also provided a quantitative assessment of the empirically-derived hygroscopicity relationship
- The estimated f(RH) measurement uncertainty is in the order of 17%.

Detailed Comparison between HSRL and in-situ ambient aerosol extinction