Relationships amongst lower tropospheric and column-averaged aerosol properties and composition from co-located NOAA and NASA monitoring sites at Appalachian State University

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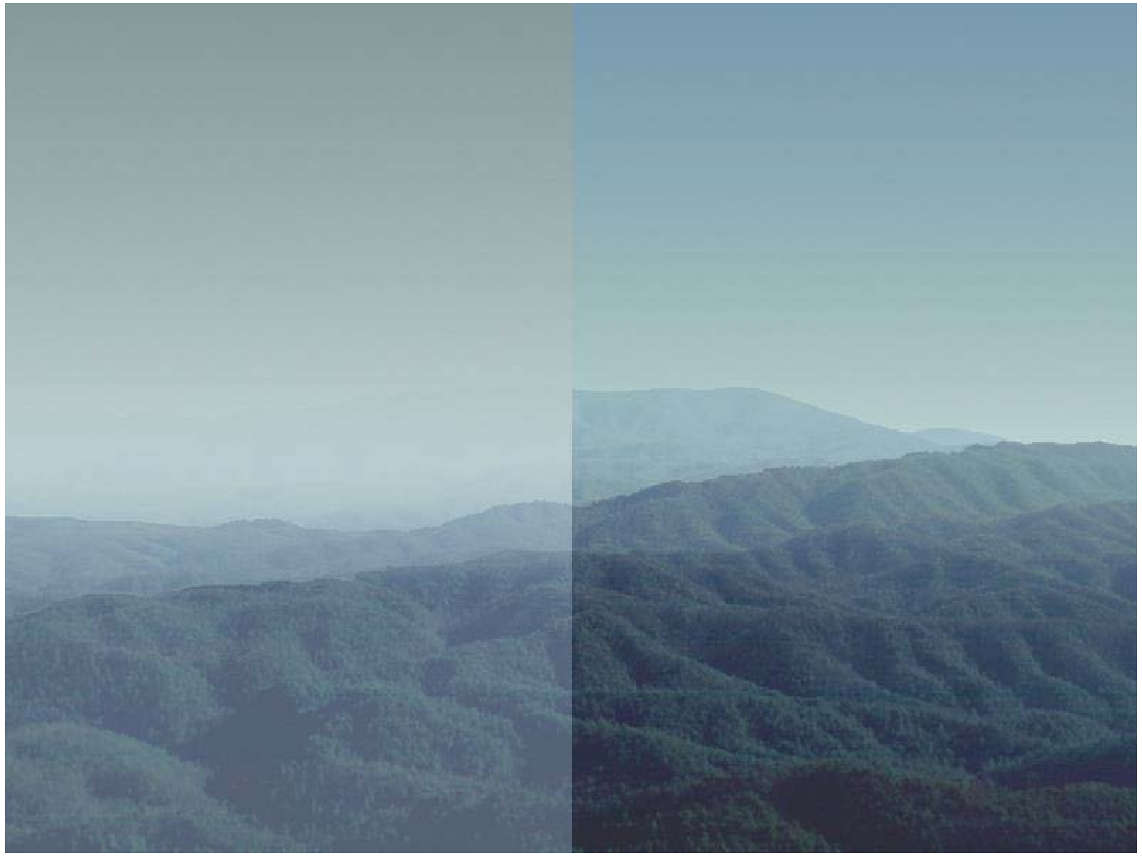
Topics to be Covered

I. Background/Objectives
II. AppalAIR sites and measurements
III. Time series of aerosol properties
IV. Relationships amongst aerosol properties
V. Conclusions and Future Work

I. Background: Relevance of aerosol direct radiative forcing studies in southeastern US



- Southeastern US is home to high warm-season levels of sulfate and biogenic secondary organic aerosol and is one of only a few regions not to have warmed in 20th century
- Simulated mean visibility for 20% of haziest days annually at Great Smoky National Park in 1990 (left) and 2011(right).
- Figure from Hand, et al. *Widespread reductions in haze across the United States from the early 1990s through 2011 2014*, Atmos. Env, 2014
- Could improved air quality in eastern US lead to regional warming????
- Studies of aerosol direct radiative forcing (DRF) from regionally-representative sites could be used to evaluate models and potentially lead to better understanding of degree of influence of changing regional air quality on climate change

I. Background / Objectives

- Studies of aerosol direct effects on solar radiation (***aerosol direct radiative forcing-DRF***) necessitate knowledge of the spectral dependence of the following aerosol optical properties (AOPs)

  (1) **aerosol optical depth (AOD):** vertical integral of aerosol light extinction coefficient

  (2) **Single scattering albedo(SSA):** Relative contributions of scattering and absorption to aerosol light extinction

$$SSA = \frac{\sigma_{scat}}{\sigma_{scat} + \sigma_{abs}} = \frac{\sigma_{scat}}{\sigma_{ext}} \quad 0 \leq SSA \leq 1$$

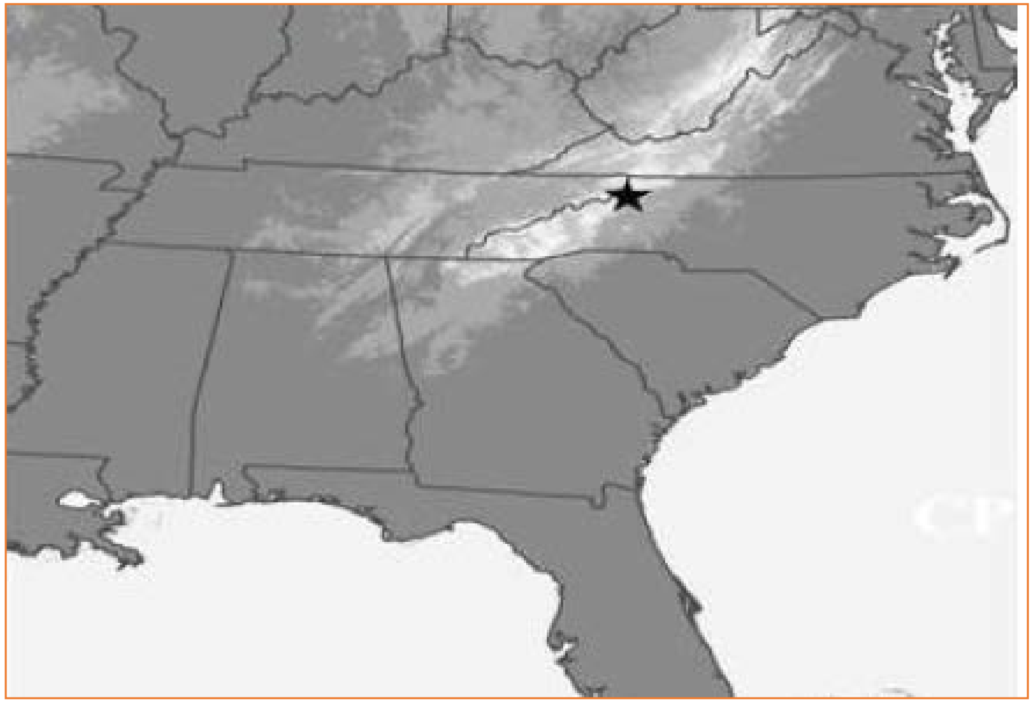
  (3) **Asymmetry parameter (g):** Indicator of directional dependence of light scattering (via the average cosine of scattering angle). Amount of light scattered and directional dependence depend on ratio λ/D, with g approaching 1 for very large particles (droplets) and g~0.50 for very small particles (Rayleigh scattering)

In addition, one needs to have suitable approximation for **vertical distribution of aerosols** and the **dependence of (1)-(3) on ambient RH**

- One problem is that few sites have all of these measurements and even those that do typically have data gaps (sometimes large) in some of the measurements.
- Another problem is degree to which measurements of aerosol properties measured near the surface are representative of the atmospheric column
- Yet another problem is relating the optical properties to those used by the modelers (aerosol chemical composition and size distributions)
- ***So-Should we just give up?*** 
- ***Probably not!*** Relationships between lower and column-averaged AOPs and (when possible) aerosol chemical composition can provide some information that could be used to fill in the gaps

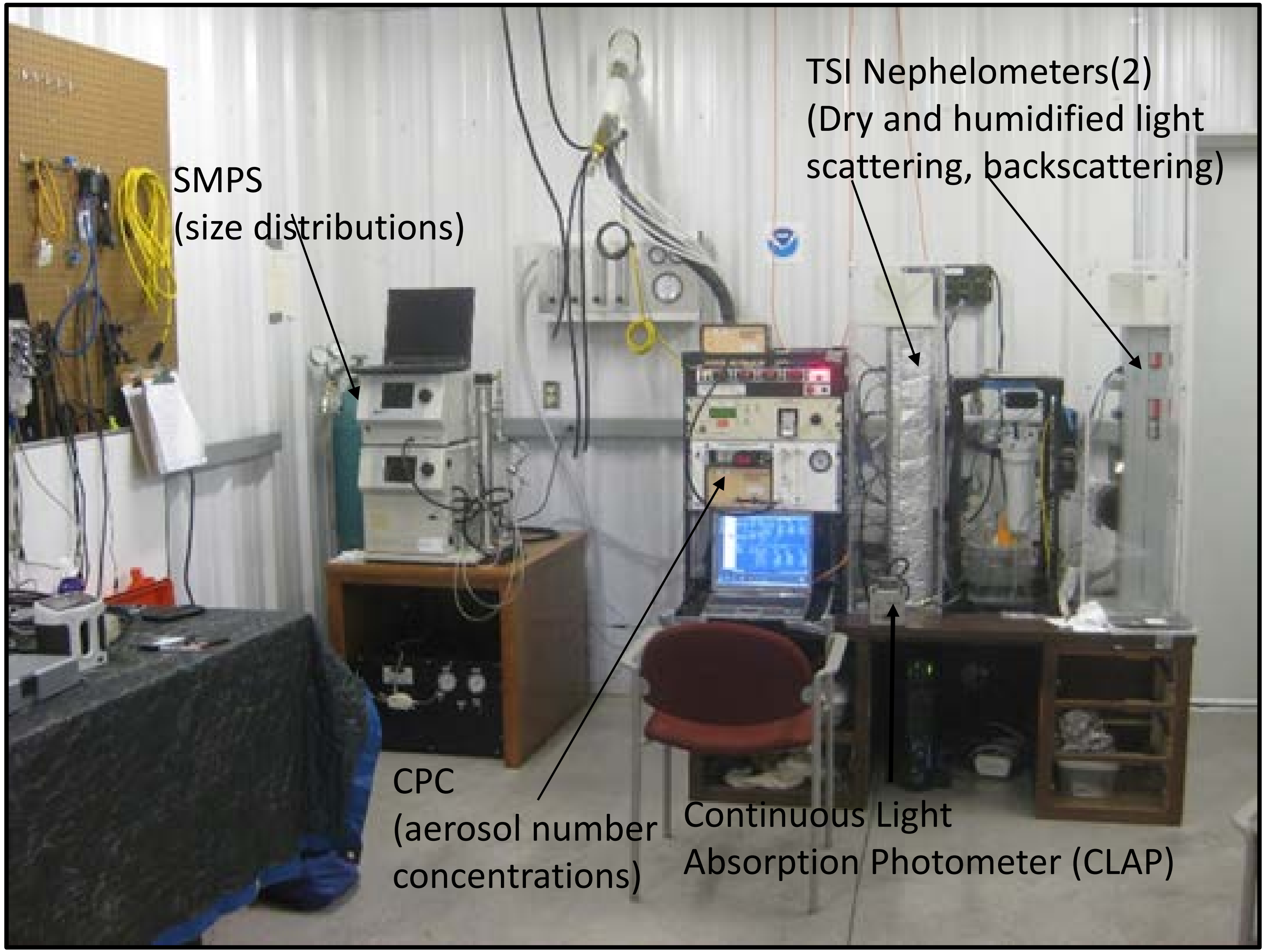
Appalachian Atmospheric Interdisciplinary Research facility at Appalachian State University (AppalAIR)

- *AppalAIR sites in Boone, North Carolina, USA (APP): 36.2N, 81.7W, 1080m* asl) are home to one of only 3 co-located NOAA-ESRL (lower tropospheric aerosol properties), NASA AERONET (column-averaged aerosol properties), and NASA MPLNET (vertical aerosol and cloud profiles) in the US and the only site in the eastern US
- High elevation, small-town location provide ideal location for measuring 'background' aerosol levels in the SE U.S., home to high levels of SOA
- Sites are unique in that it is heavily dependent on undergraduate (and some MS-level) students, along with an over-worked college electronics technician and foolishly ambitious Physics professor



II. AppalAIR facility-NOAA-ESRL lower troposopheric aerosol monitoring site (APP)

34m aerosol sampling tower



II. AppalAIR 2 field site- NASA AERONET, NASA MPLNET, NASA SolRadNET

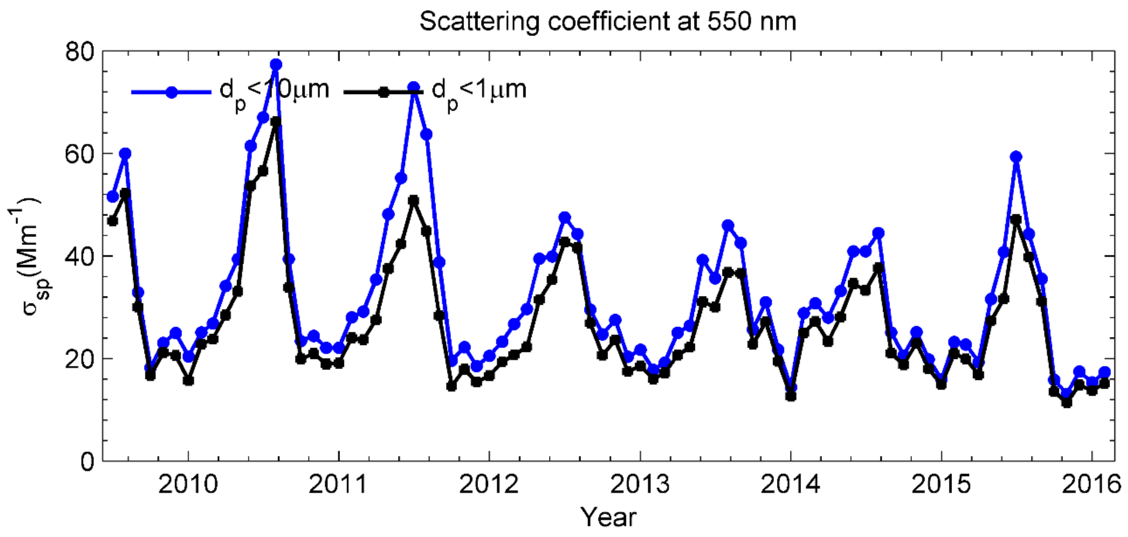
Yankee Scientific All-Sky Imager

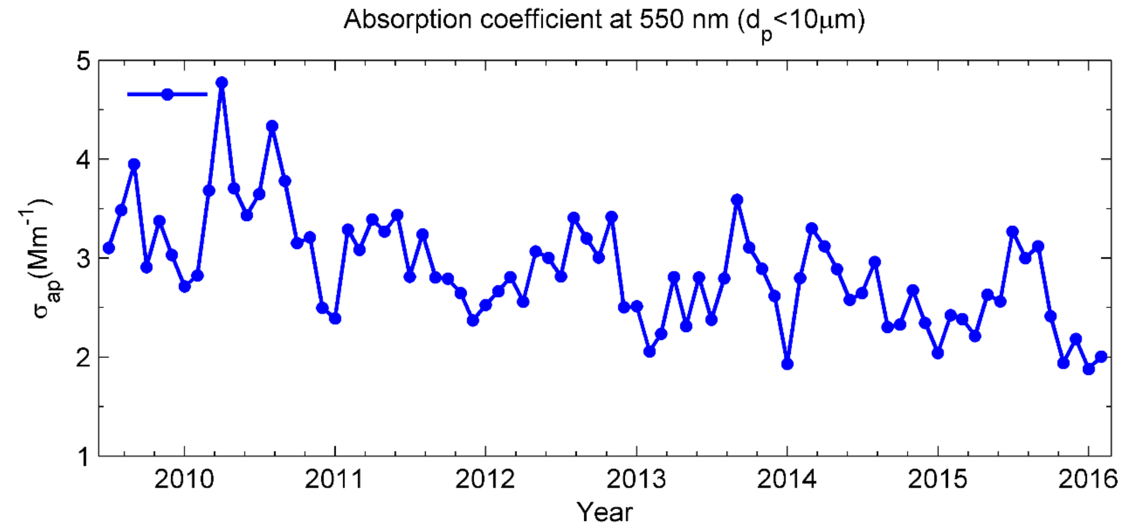
Kipp & Zonen Solar Pyranometer (SolRadNET)

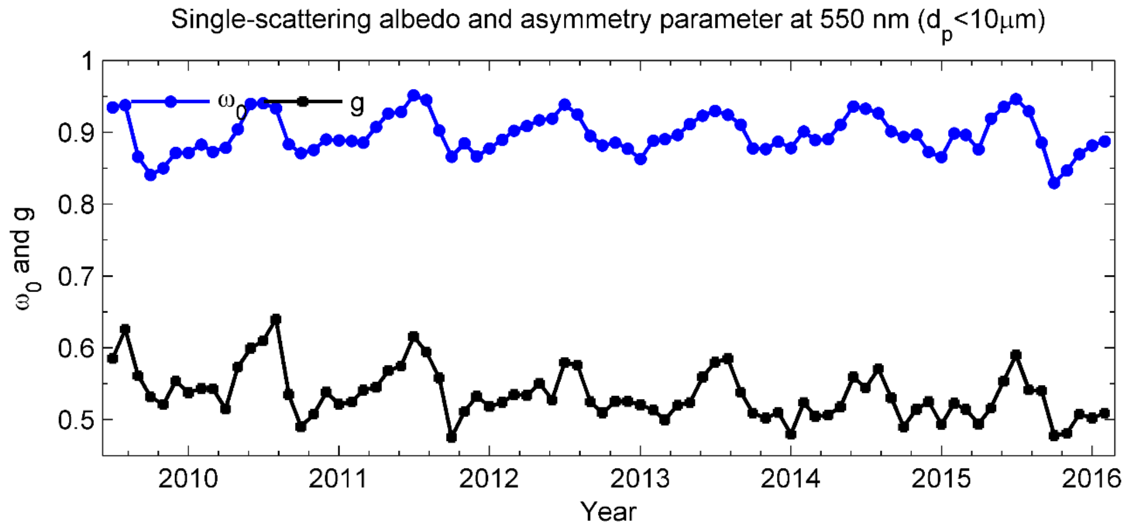
Cimel Sunphotometer (Column-averaged aerosol properties- AERONET

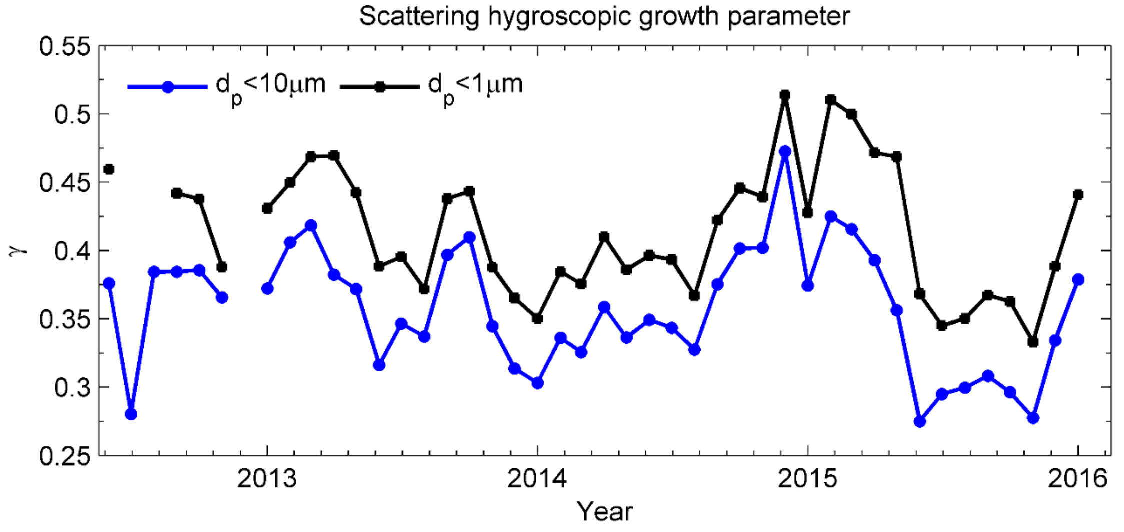
Micro-pulsed lidar (Vertical aerosol and cloud profiles- MPLNET

III. Time series of some key lower tropospheric aerosol properties (NOAA-ESRL)





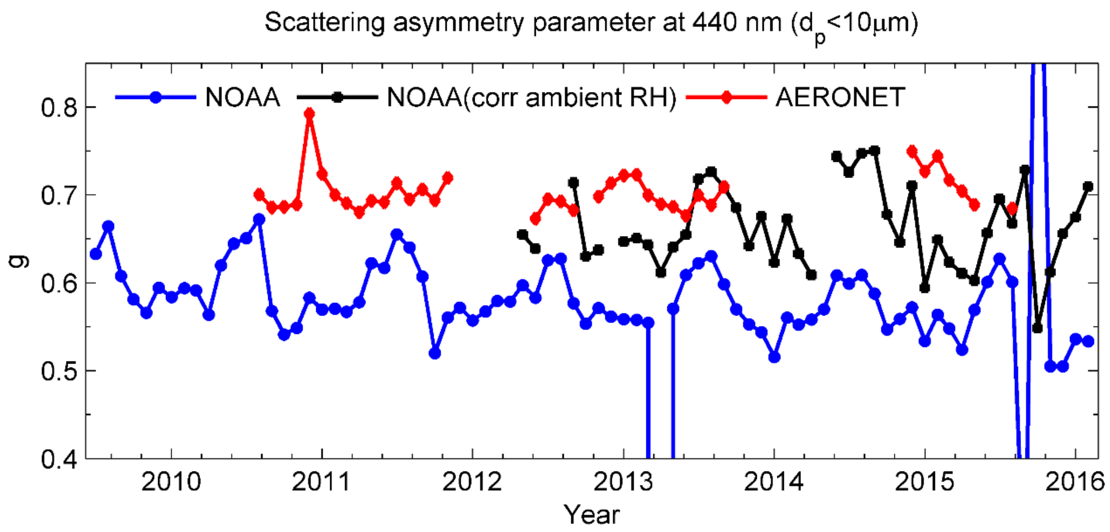


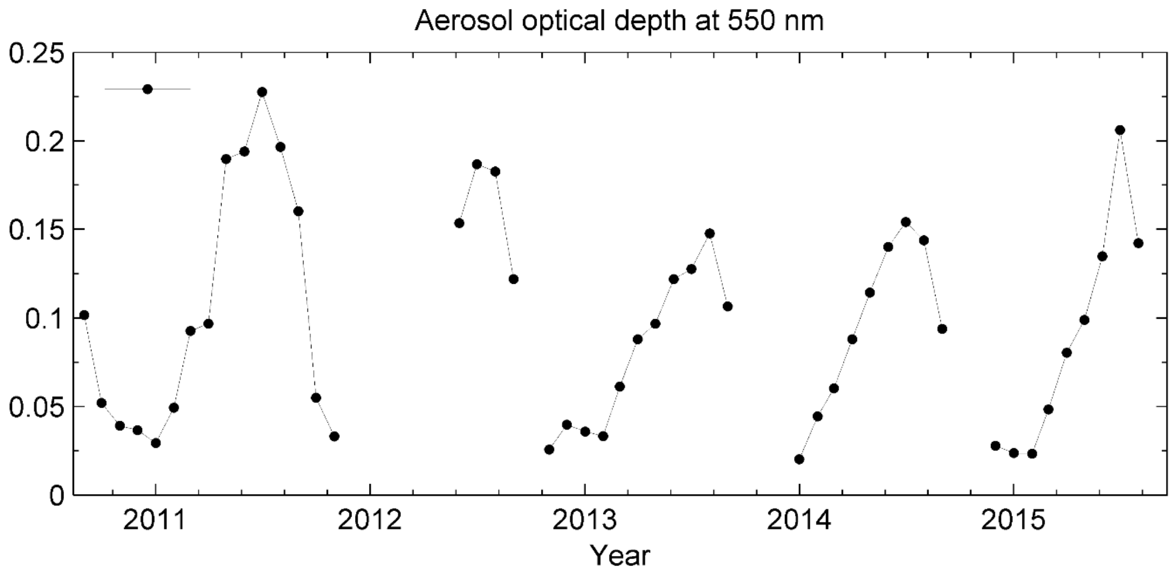


III. Time series of some key column-averaged aerosol properties (NASA AERONET)

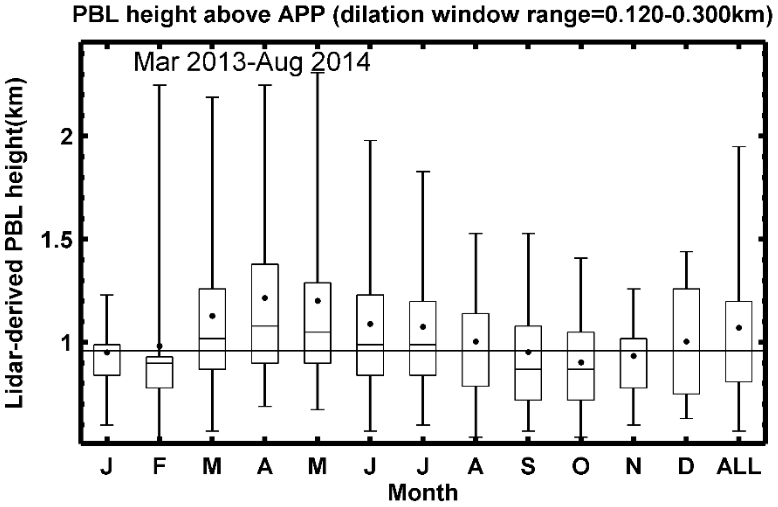
- Unlike NOAA-ESRL in situ, these are made at ambient RH but require unobstructed view of the sun
- Note the absence of AERONET single-scattering albedo, which can only be accurately derived (to within 0.03) for high-AOD conditions (AOD>0.40, from Dubovik et al., 2000)
- Note the data gaps (yearly calibrations, instrument problems)

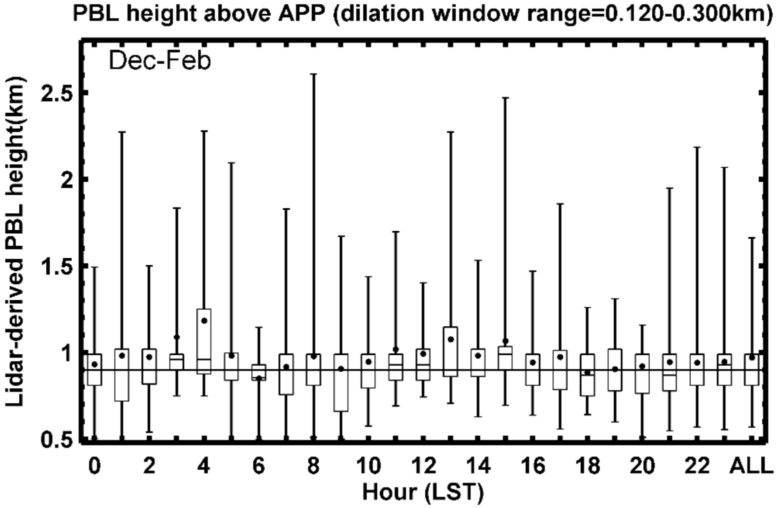
Sky	AOD at 550 nm
Very Clear	<0.05
Clear	0.05-0.10
Somewhat hazy	0.10-0.25
Hazy	0.25-0.50
VERY Hazy	>0.50

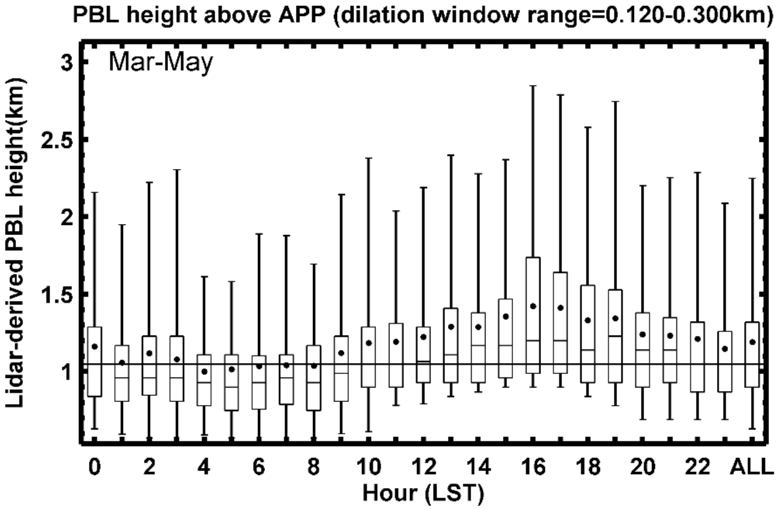




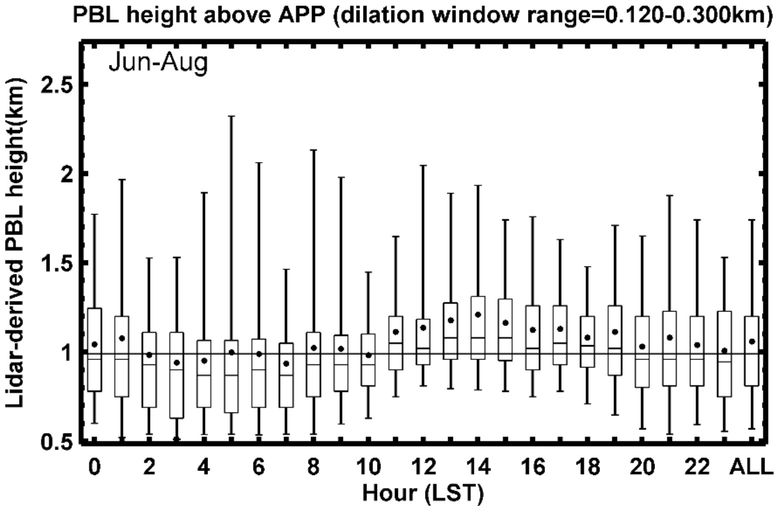
III. Annual and diurnal cycles of lidar-derived planetary boundary layer heights

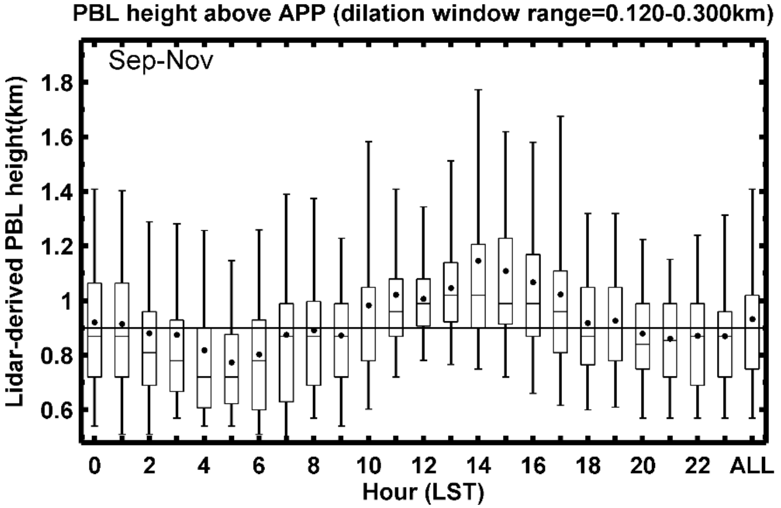






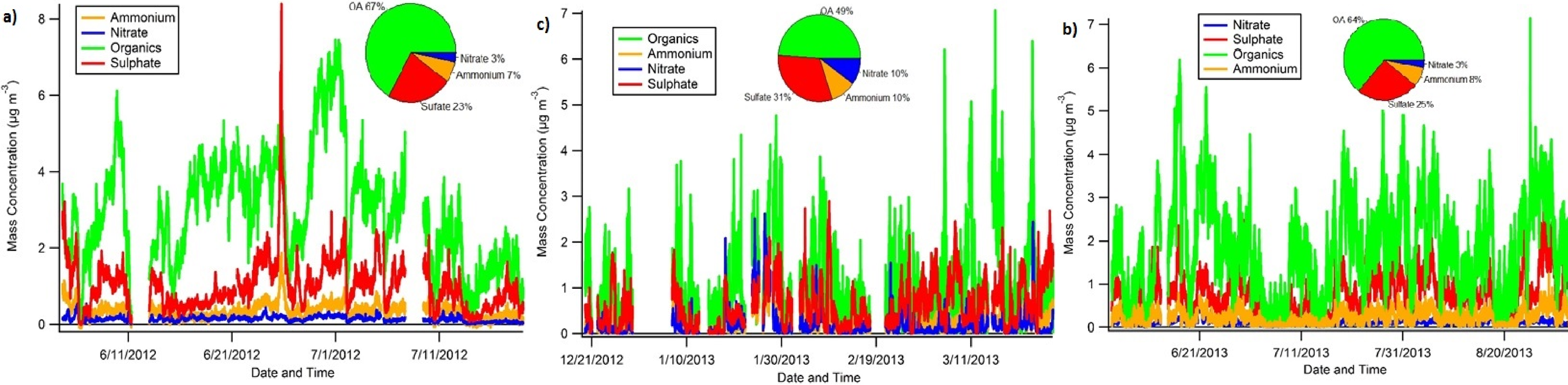
- Lidar only operational during Feb 2013-August 2014 and since March 2016 (when added to NASA MPLNET)
- PBL heights calculated using wavelet covariance transform algorithm implemented by ASU grad student Ben Madison and compared to radiosonde launches in summer 2013





III. Sub-micron aerosol chemical composition

- Studies conducted using an AMS by former Chemistry researcher Dr. Yong Zhou and former ASU Chemistry undergrad Michael Link during summers 2012-2013 and winter 2013 (Link et al., 2015)

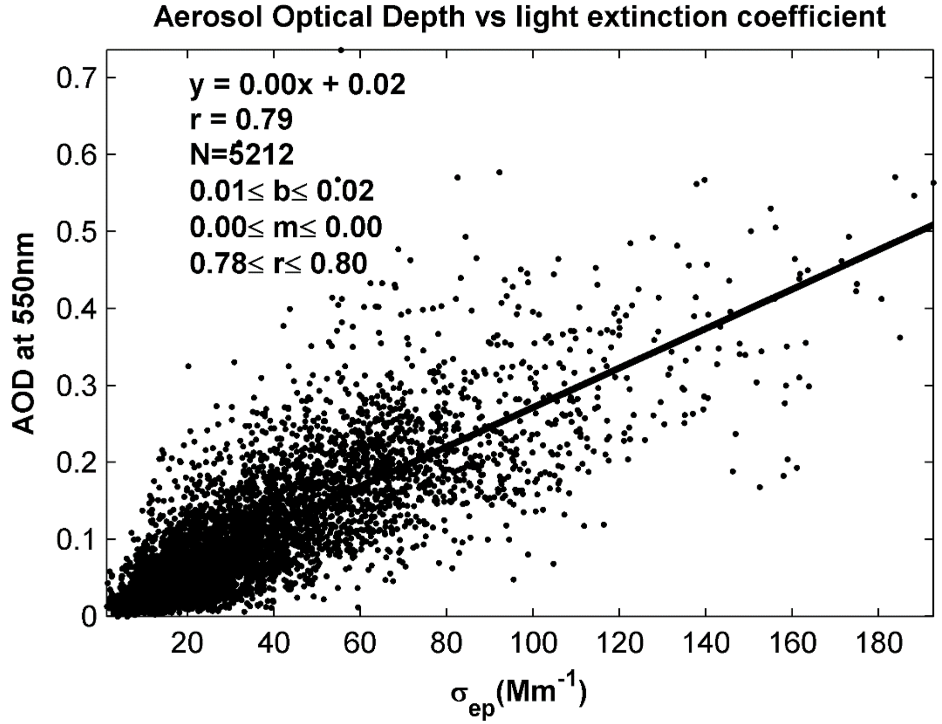


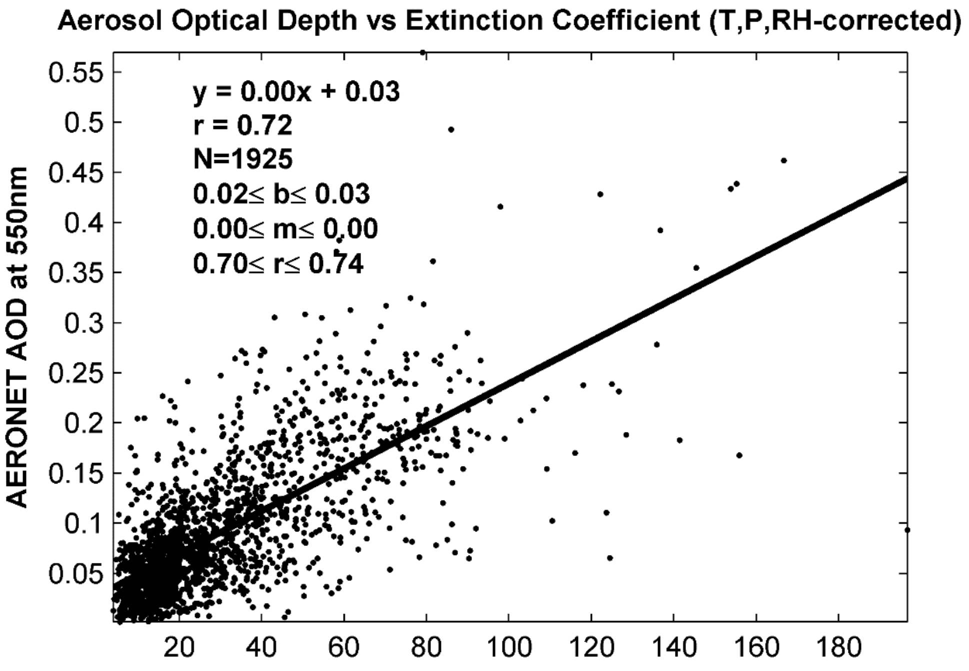
Above figures are from the Supplement to **Link, M.F**., Zhou,Y., Taubman, B.F., Sherman, J.P., Sive, B.C., Morrow,H., Krintz, I., Robertson,L., Cook, R., Stocks, J., West, M, and Sive, B..: A characterization of volatile organic compounds and secondary organic aerosol at a mountain site in the Southeastern United States , J. Atmos. Chem, 01/2015; DOI:10.1007/s10874-015-9305-5, 2015.

IV. Relationships amongst aerosol properties

- Lower tropospheric light scattering demonstrates modest correlation with g (r=0.51) and SSA (0.44), consistent with earlier plot associating high scattering with large, highly reflective particles and low scattering with small, less reflective particles. This is in agreement with observations from four North American NOAA-ESRL sites (Sherman et al., 2015)
- Some interesting examples of relationships (or lack thereof) amongst lower tropospheric aerosol properties and column-averaged properties and aerosol chemistry shown on next few slides

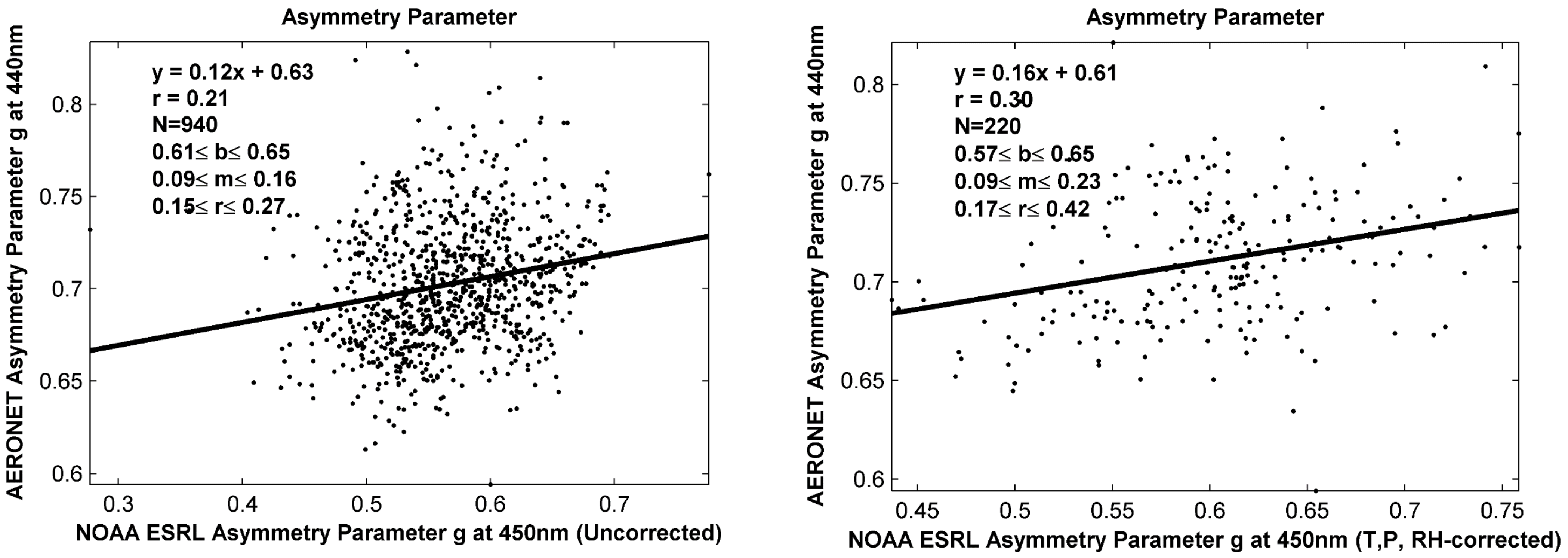
Near-surface aerosol light extinction versus AOD





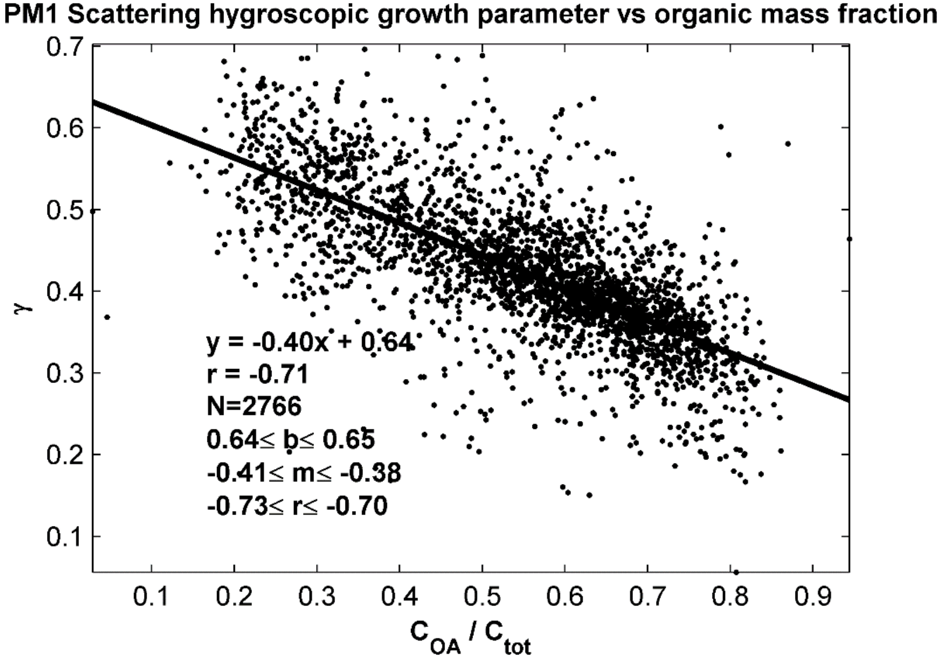
- AOD well-correlated with lower tropospheric extinction coefficient. More study needed to determine why slightly worse when T,P, RH-corrected
- Comparison of box model AOD (calculated as product of extinction coefficient times PBL height) with measured AERONET AOD shows that box model under-estimates AOD by factor of close to 2, even though correlation good (r=0.76). Agreement no better when extinction was T,P, RH-corrected
- Similarly poor agreement also observed at SGP (Lamont, OK) by Bergin et al (2000)
- Further study placed in context of observed lidar extinction profiles is needed but the box model under-estimation of AOD likely implies that surface extinction coefficient measurements and PBL heights not sufficient to estimate AOD and that information on vertical aerosol profiles may also be necessary

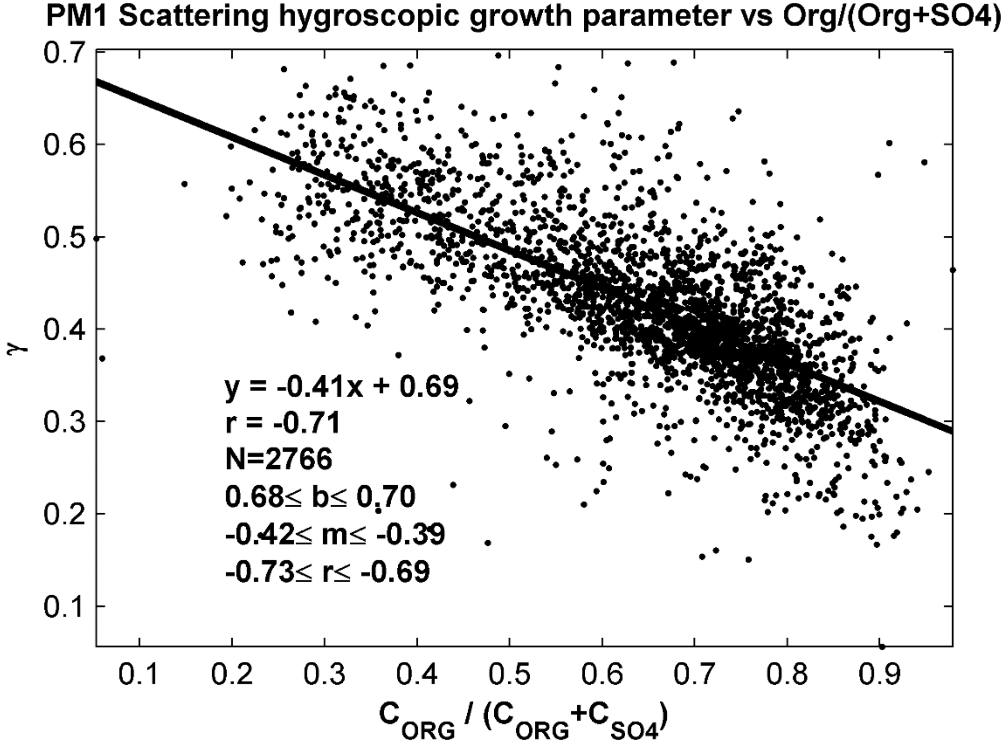
Near-surface versus column-averaged asymmetry parameter

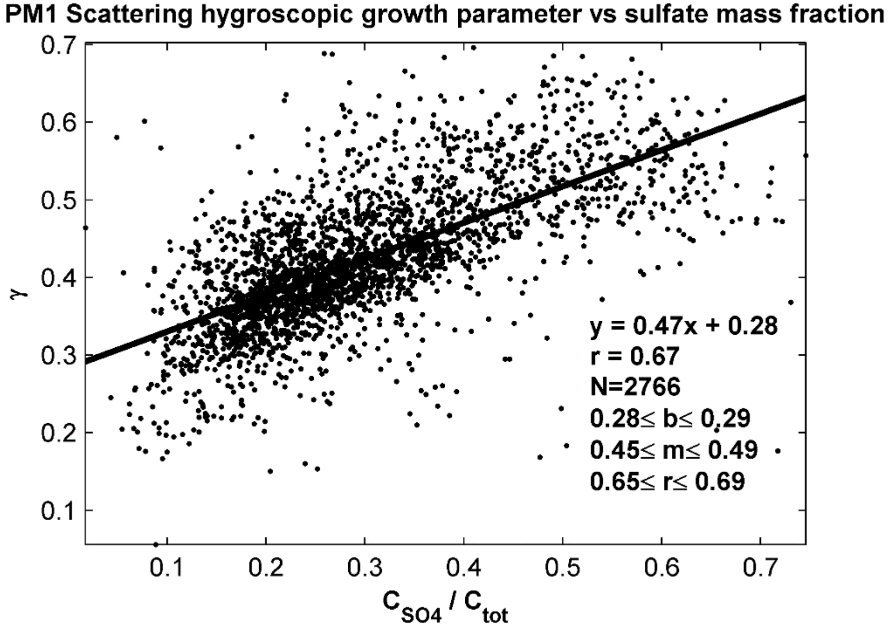


- Asymmetry parameter derived from lower tropospheric and column-averaged measurements are only weakly-correlated (maybe slightly better when NOAA measurements adjusted to ambient RH)
- AERONET g usually larger than NOAA g, even when adjusting to ambient T,P,RH.
- Other lower tropospheric and column-averaged proxies for aerosol size demonstrated weak correlation as well

Scattering hygroscopic growth parameter versus organic and sulfate mass fractions







- Our linear model above is not much different than that reported by Quinn et al. (2004) for ICARTT field campaign off Atlantic coast of US

$$\gamma = -0.50\left(\frac{Org}{Org + SO4}\right) + 0.80 \quad r^2 = 0.42$$

Conclusions and Future Work

- Initial study of relationships amongst lower tropospheric aerosol properties (and aerosol chemistry) produced some interesting results but introduced at least as many questions as answers
- Lower tropospheric light scattering and extinction coefficients display modest to good correlation with SSA and g
- Lower tropospheric extinction coefficient highly-correlated with AERONET AOD (r=0.80) but poor agreement of box-model AOD calculated using extinction and PBL heights with AERONET AOD indicates that it may not be sufficient to estimate AOD
- Poor correlation between lower tropospheric and column-averaged asymmetry parameter (and other indicators of aerosol size)
- Measurements of hygroscopic dependence of aerosol light scattering could possibly be used to estimate organic and sulfate aerosol mass fractions (at least at this site), with similar correlation and linear model parameter to those reported off eastern US coast by Quinn et al (2005).
- Future (current) work includes first measurement-based Aerosol Direct Radiative Forcing (DRF) climatology in SE U.S. (SBDART radiative transfer code). However, a better understanding of factors influencing relationships amongst lower tropospheric and column-averaged is needed. Vertical profiles of aerosols measured by lidar will be used to this end.

Acknowledgements

The aerosol research at AppalAIR would not be possible without the dedicated efforts of…..

- Many Appalachian State University (ASU) student researchers who contributed to data collection and instrument maintenance
- Michael Hughes (ASU College of Arts and Sciences Electronics Technician)
- Dana Greene(ASU College of Arts and Sciences Machinist)
- ASU College of Arts and Sciences (dean Anthony Calamai)
- ASU Department of Physics and Astronomy (Chair Michael Briley)
- NOAA-ESRL Aerosols group (Ogren, Sheridan, Hageman, Andrews…..) for their support in setting up and maintaining the APP site and data processing support
- NASA AERONET, for Cimel instrument calibrations, trouble-shooting, and data processing and web-based display

Appendix

- Appendix 1-Aerosol measurements at AppalAIR

Data Product	Measurement Technique
Aerosol hygroscopic growth: total light scattering & hemispheric backscattering (450nm, 550nm, 700nm)	TSI 3563 Nephelometer operating at a reference RH (≤40%) in series with a second scanning TSI 3563 (40%-90% RH)
Aerosol light absorption (corrected to 450nm, 550nm, 700nm)	Radiance Research Particle Soot Absorption Photometer (PSAP), Continuous Light Absorption Photometer (CLAP)
Aerosol number concentrations	TSI 3783 Environmental Particle Counter and TSI 3760 Condensation Nuclei Counter
Aerosol size distributions	TSI Scanning Mobility Particle Sizer (SMPS) (beginning June 2013)
Aerosol chemical composition (size-resolved, sub-um)	Aerodyne Quadrupole Time-of-Flight Aerosol Mass Spectrometer

Data Product	Measurement Technique
Aerosol Spectral Optical Depth at 8 wavelengths (340,380,440,500,675,870,1020, and 1640nm) Total, coarse, and fine mode components derived	CIMEL 318-EBN Sun/Sky Radiometer
Precipitable water vapor	CIMEL differential extinction 870/936nm
Aerosol size distributions, single-scattering albedo	CIMEL sky radiance measurements (principle plane and almucantor scenerios)
Total broadband irradiance (direct plus diffuse)	Kipp and Zonen CMP22 Pyranomter
Cloud optical depth and cloud fraction	CIMEL and Yankee Scientific TSI-440 All-sky Imager

Data Product	Measurement Technique
Aerosol backscatter profiles used to computer normalized relative aerosol backscatter (NRB) at 532nm and aerosol layer heights	Sigma Space MPL-4B Micro-pulsed Lidar (MPL)
Aerosol Optical Depth (AOD) at 532nm (day and night)	Lidar NRB profiles** , using techniques presented by Welton (1999)
Vertical profiles of aerosol extinction coefficient and extinction-to-backscatter ratio (S)	Lidar NRB profiles**, using custom algorithm based on Marenco (1997)
Boundary layer heights	Lidar NRB profiles **
Cloud base heights	Lidar backscatter profiles

** Requires knowledge of lidar calibration constant C, periodically derived through co-located CIMEL AOD measurements

III. Trace Gases and Meteorology

Data Product	Measurement Technique
C_2-C_{10} NMHCs, C_1-C_2 halocarbons, C_1-C_5 alkyl nitrates, OVOCs, reduced sulfur gases, HCN & CH_3CN	Cryogen free in-situ 5 channel GC/GC-MS system (Sive, Zhou)
CO, CO_2, CH_4, N_2O & SF_6	Greenhouse GC (Sive, Zhou)
VOCs	PTRMS (Sive, Zhou)
O_3	LiCor Ozone Monitor (Neufeld)
Surface meteorology (T, P, RH, winds)	2 MET stations -APP, Beech Mountain (Perry)
Vertical meteorology (T, P, RH, winds)	InterMET Radiosonde Launching System (Perry, Zhou, Sherman, students)