

Changing air quality in the SE U.S and potential implications for regional solar radiation budget

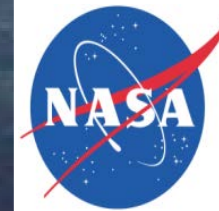
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Figure from Hand, et al. *Widespread reductions in haze across the United States from the early 1990s through 2011*, Atmos. Env, 2014



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Talk Outline

- I. Background-Aerosols in the SE U.S
- II. Aerosol direct radiative effect and radiative properties at APP
- III. Long-term time series of aerosol radiative properties-Possible future implications for DRE
- IV. Conclusions and future work
- V. References

I. Background

- Several studies (Goldstein et al., 2009 and others) have concluded that high warm-season aerosol loading (mainly biogenic SOA and sulfates) in the SE U.S produces a cooling effect (i.e. negative direct radiative effect-DRE) at top of atmosphere and that aerosols may have played a role in the regional 'warming hole' during the 20th century
- Aerosol loading over the eastern U.S has decreased significantly over the last few decades. Will this lead to regional warming?
- Long-term climatologies of aerosol DRE and the aerosol optical properties influencing DRE (aerosol optical depth, single-scattering albedo, and asymmetry parameter) are necessary to improve parameterization of aerosols in models

Aerosol optical properties influencing DRE

- Studies of aerosol direct effects on solar radiation (***aerosol direct radiative effect-DRE***) 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

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

(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 $g \sim 0.50$ 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.

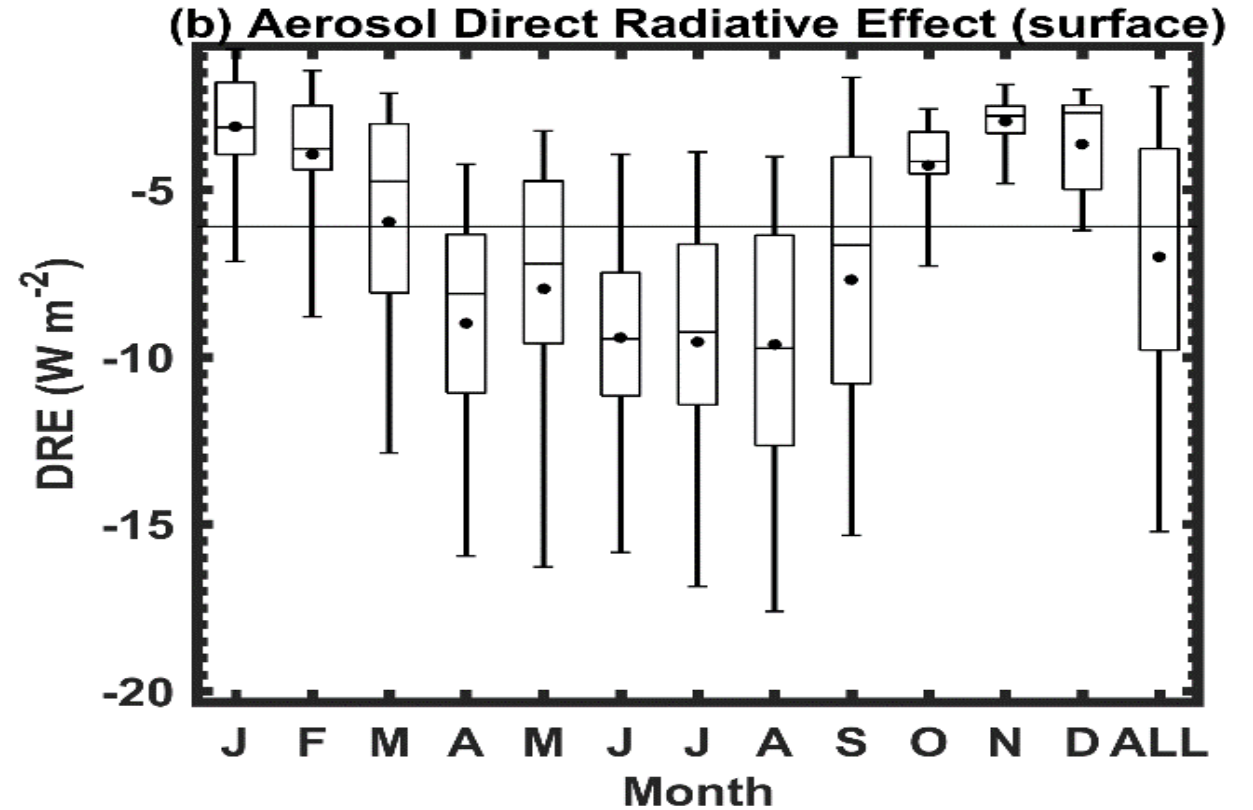
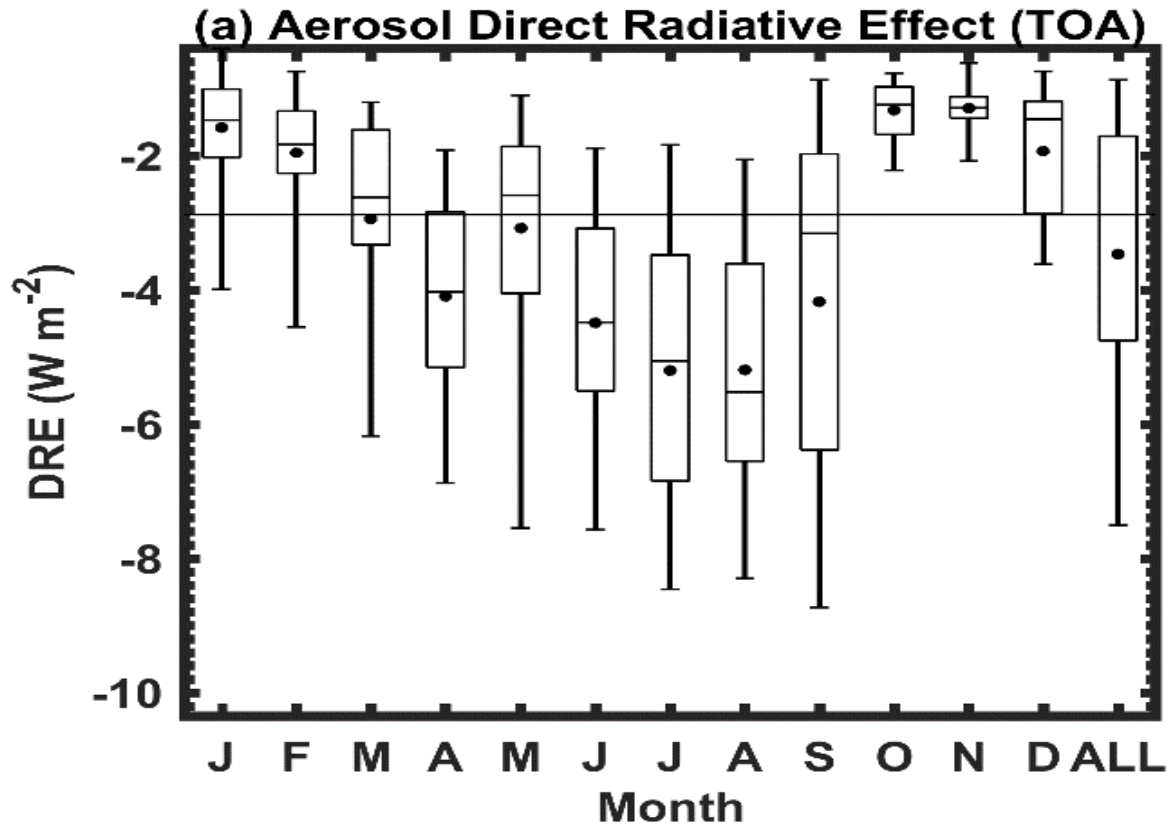
The need for ground-based evaluation of DRE in the SE U.S.

- To date, the few multi-year DRE studies conducted in SE U.S (Alston and Sokolik, 2016; Goldstein et al., 2009) have relied on satellite-based measurements of AOD and SSA and tabulated (or guessed) values for g , which provide a regional picture but also lead to large DRE uncertainties (Sherman, and McComisky, 2017)
- They also used simple radiative transfer equations (Haywood and Shine, 1995), which can lead to a large over-estimation of the aerosol cooling effect (Sherman and McComiskey, 2017)
- Aerosol DRE studies using AOD, SSA, and g from ground-based aerosol network sites (NOAA-ESRL, AERONET, MFRSR) possess small enough uncertainties (Sherman et al., 2015; Eck et al., 1999;) to estimate DRE at TOA and surface with uncertainties of $\sim 1 \text{ W/m}^2$ or better (Sherman and McComiskey, 2017) and can complement the spatial coverage of satellite-based studies

II. Aerosol direct radiative effect and radiative properties at APP

- APP site in Boone, NC (36.21N, 81.69W, 1080m asl) is home to the only co-located NOAA-ESRL, NASA AERONET and (active) NASA MPLNET sites in U.S, with data records (except vertical profiles) of 7-8 years, nearing the ~10 years recommended for studying trends in aerosol radiative properties and DRE
- Initial DRE climatology at APP (Sherman and McComiskey, 2017) shows the same seasonal dependence of TOA DRE as other studies (Alston and Sokolik, 2016) but much less negative summertime aerosol cooling effect, likely due to differences in the method for computing DRE (SBDART radiative transfer model versus 1st-order calculation)

Diurnally-averaged clear-sky aerosol DRE above APP (June 2012-Feb 2016)



- Annual DRE cycle (calculated using Santa Barbara DISORT Radiative Transfer model) closely follows annual AOD cycle at APP (not shown here)
- Aerosol cooling effect at TOA is ~ 5 -6 times greater during summer than winter and DRE at surface is ~ 4 -5 times greater during summer

DRE sensitivity to aerosol radiative properties and surface albedo at APP

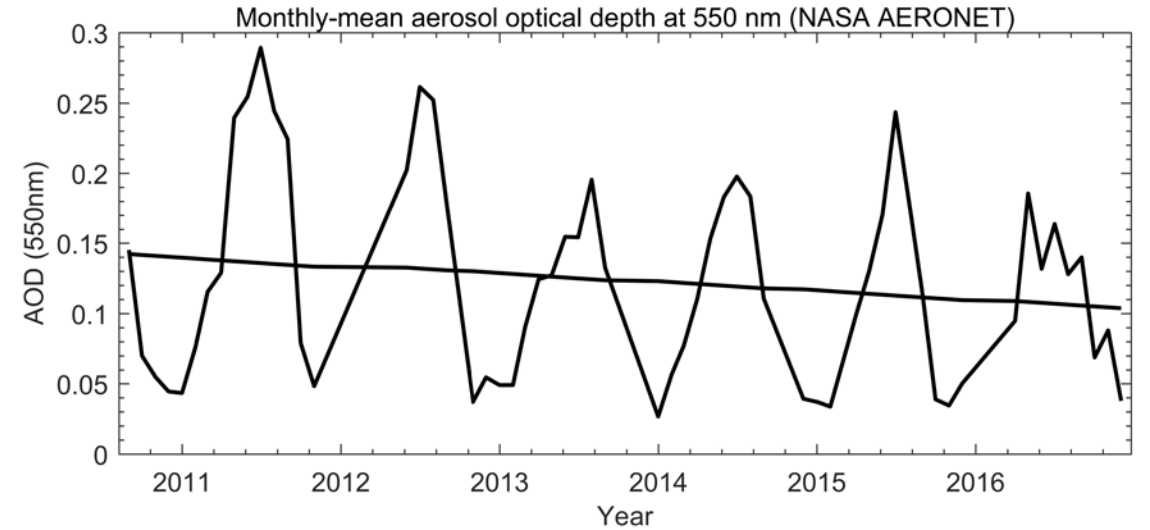
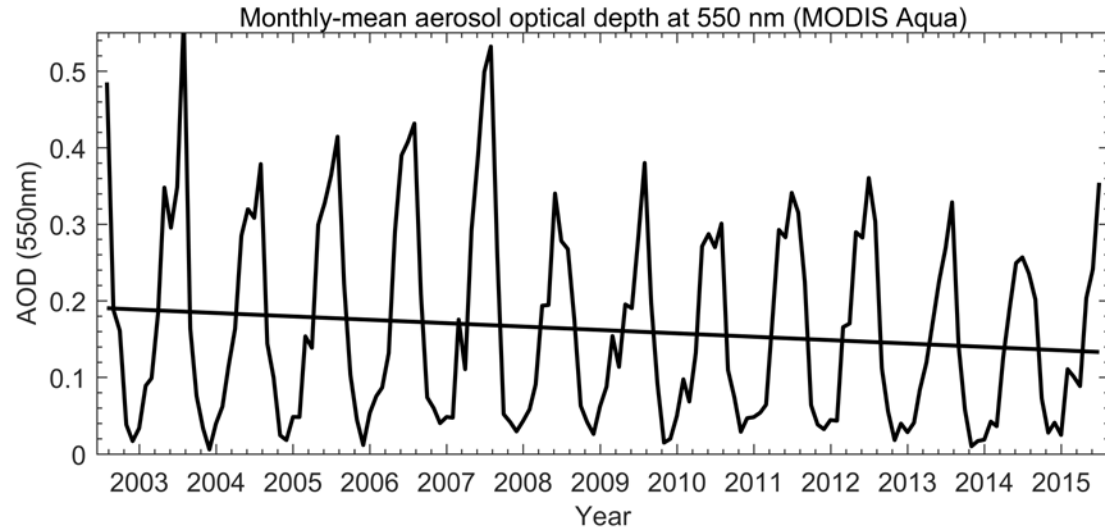
Property p_i	MAR	JUN	SEP	DEC
TOA S_{AOD}	-47	-35	-34	-43
TOA $S_{\omega 0}$	-9.1	-39	-18	-2.6
TOA S_g	5.9	19	12	1.8
TOA S_R	0.92	3.3	1.4	0.22
Surface S_{AOD}	-90	-69	-61	-72
Surface $S_{\omega 0}$	16	54	30	4.3
Surface S_g	6.1	19	12	1.9
Surface S_R	0.77	2.7	1.4	0.20

- Sensitivities are in units of W/m² per unit change in the aerosol or surface property (from Sherman and McComiskey, 2017)
- DRE is most sensitive to AOD, followed in order by single-scattering albedo (SSA or ω_0), scattering asymmetry parameter (g) and surface reflectance
- Thus, low uncertainty in AOD (~ 0.01 - 0.02) such as that at ground-based AERONET or MFRSR network sites is necessary to derive DRE with uncertainty of ~ 1 W/m² or less (Sherman and McComiskey, 2017).....but these sites lack the necessary spatial coverage in many regions
- Uncertainties in MODIS-derived AOD are at least 0.05 and MODIS Collection 5.1 AOD possessed a low bias over Southern Appalachian mountains (Sherman et al., 2016)

III. Long-term time series of aerosol radiative properties-Possible future implications for DRE

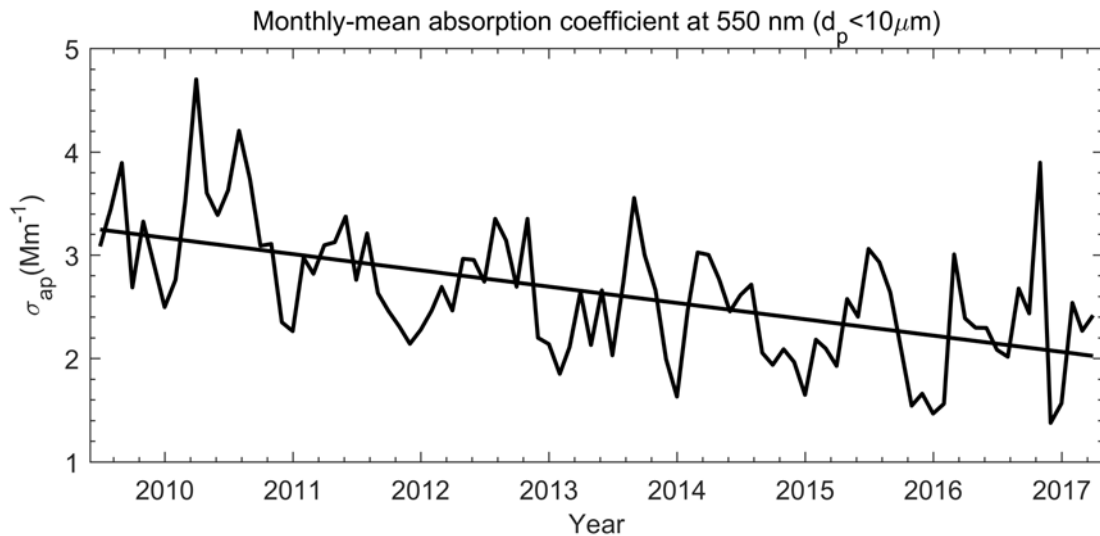
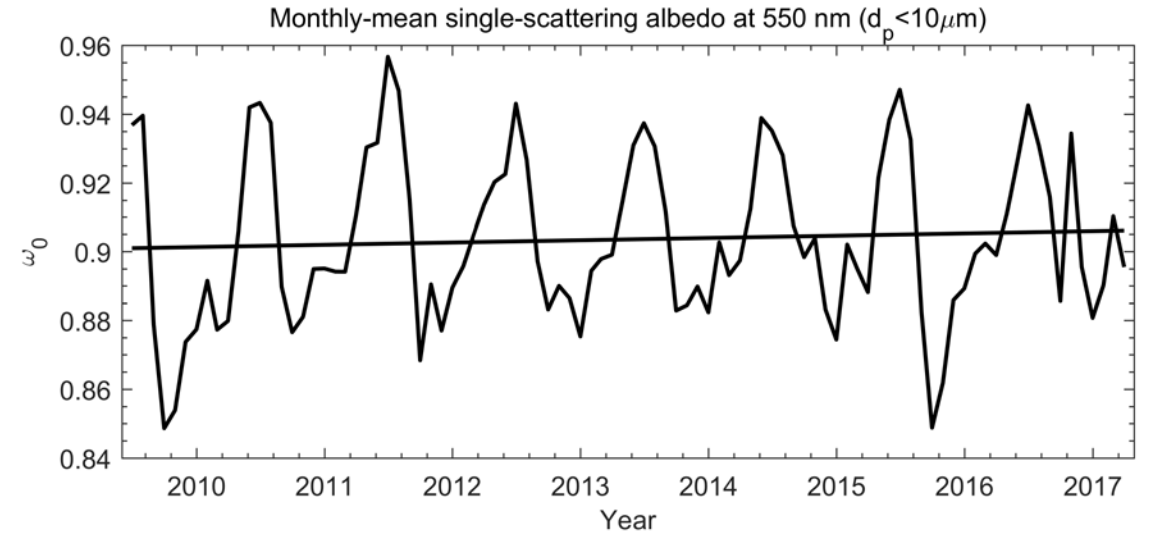
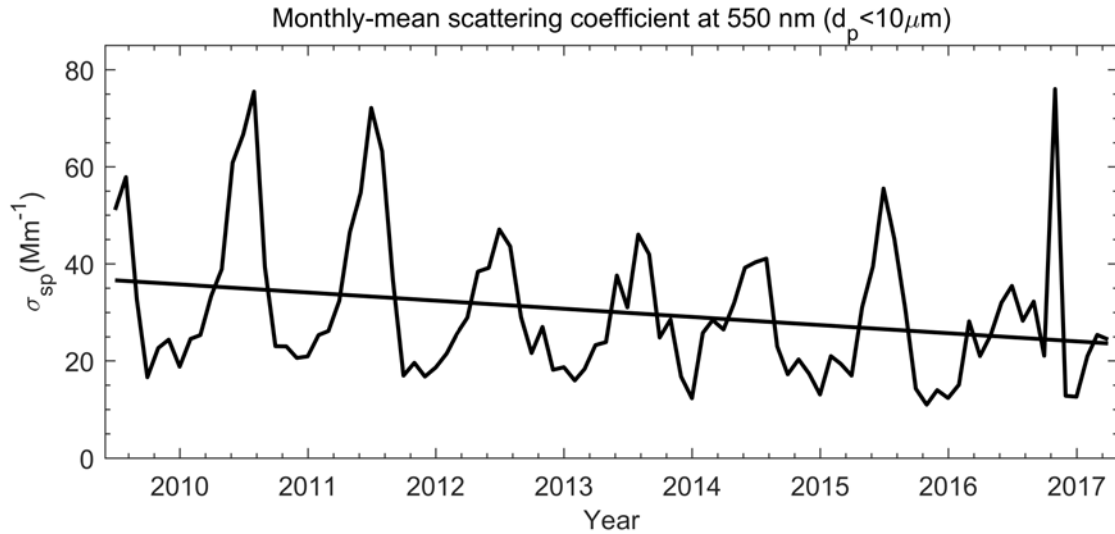
- The time series at APP for the NOAA-ESRL aerosol optical property(AOP) measurements is 8 years and that of AERONET measurements is nearing 7 years so trend studies in AOPs and DRE will soon be possible
- However, the time series at APP do reveal some apparent and potential trends in AOPs which can be used along with the ~15 year MODIS AOD time series and the DRE sensitivities (previous slide) to qualitatively understand long-term changes in DRE

Aerosol Optical Depth (AOD) from MODIS Aqua and APP AERONET site



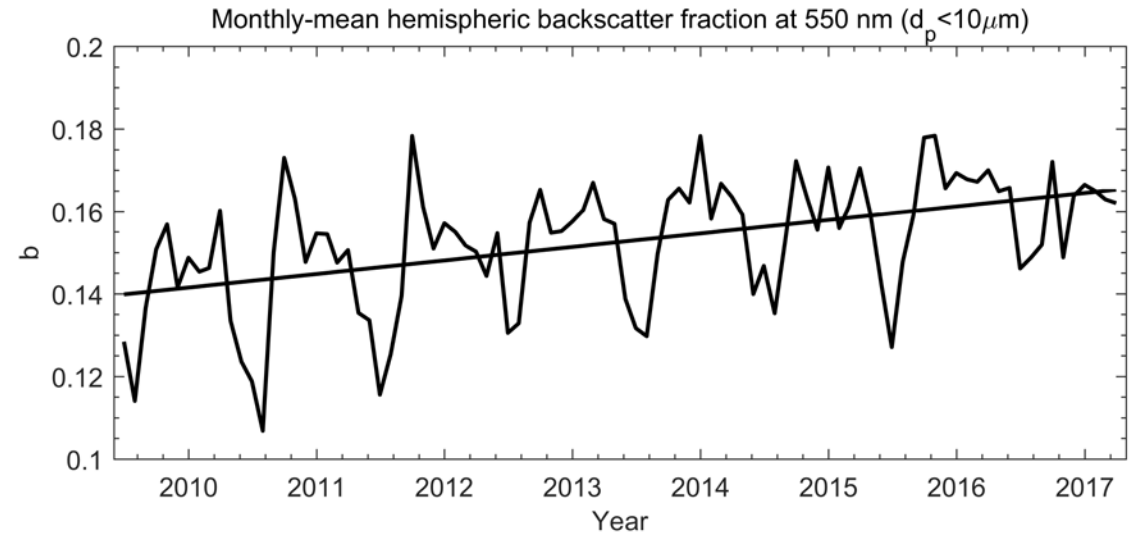
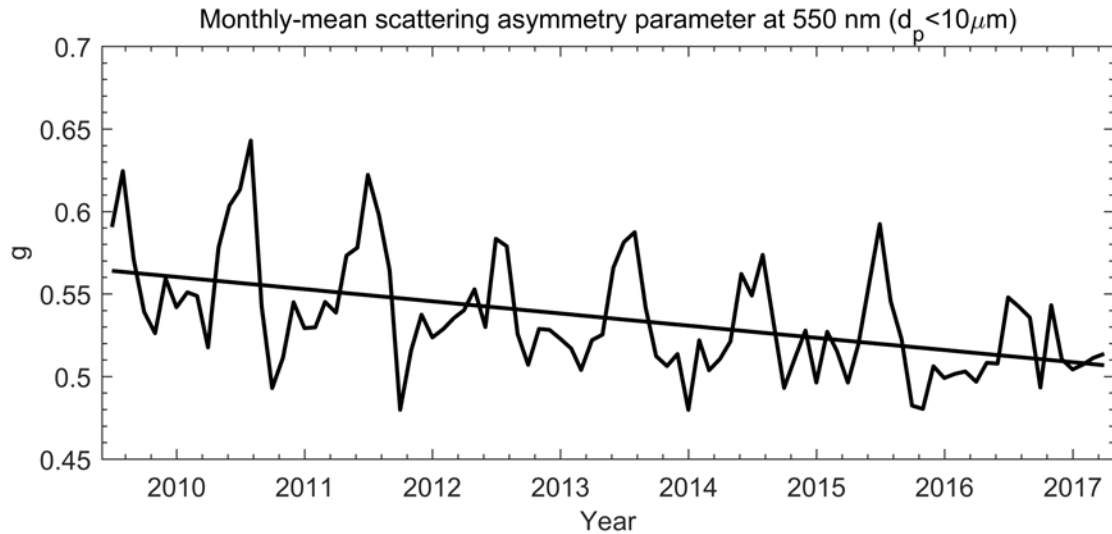
- Decreasing AOD trends in eastern U.S reported in other studies (Alston and Sokolik, 2016; Yoon et al., 2011....)
- Since DRE is most sensitive to AOD, this likely implies that DRE is becoming less negative (smaller cooling effect)

Single-scattering albedo



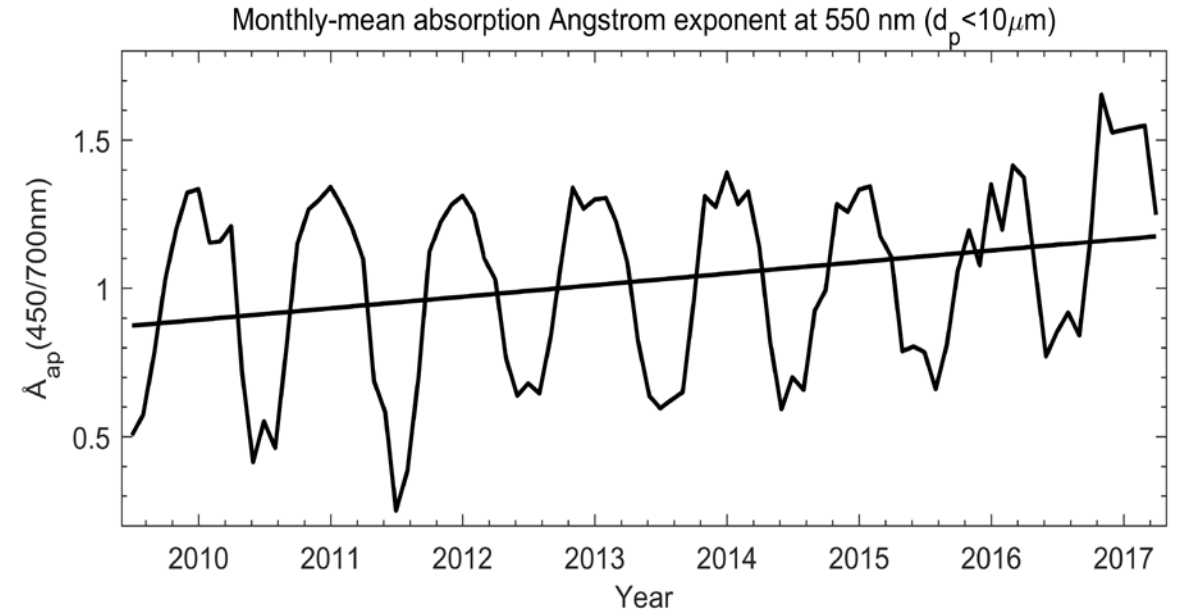
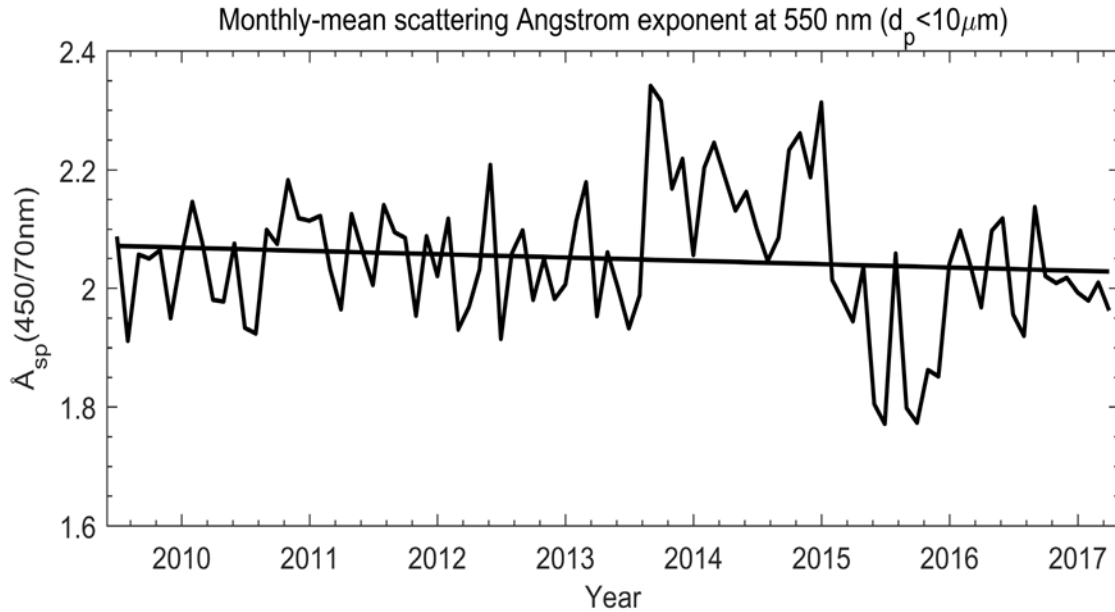
- Aerosol light scattering and absorption are decreasing at similar rates, leading to very small (if any) changes in single-scattering albedo
- The time series is modulated by months with influence from wildfires (June 2016, November 2017)

Scattering asymmetry parameter



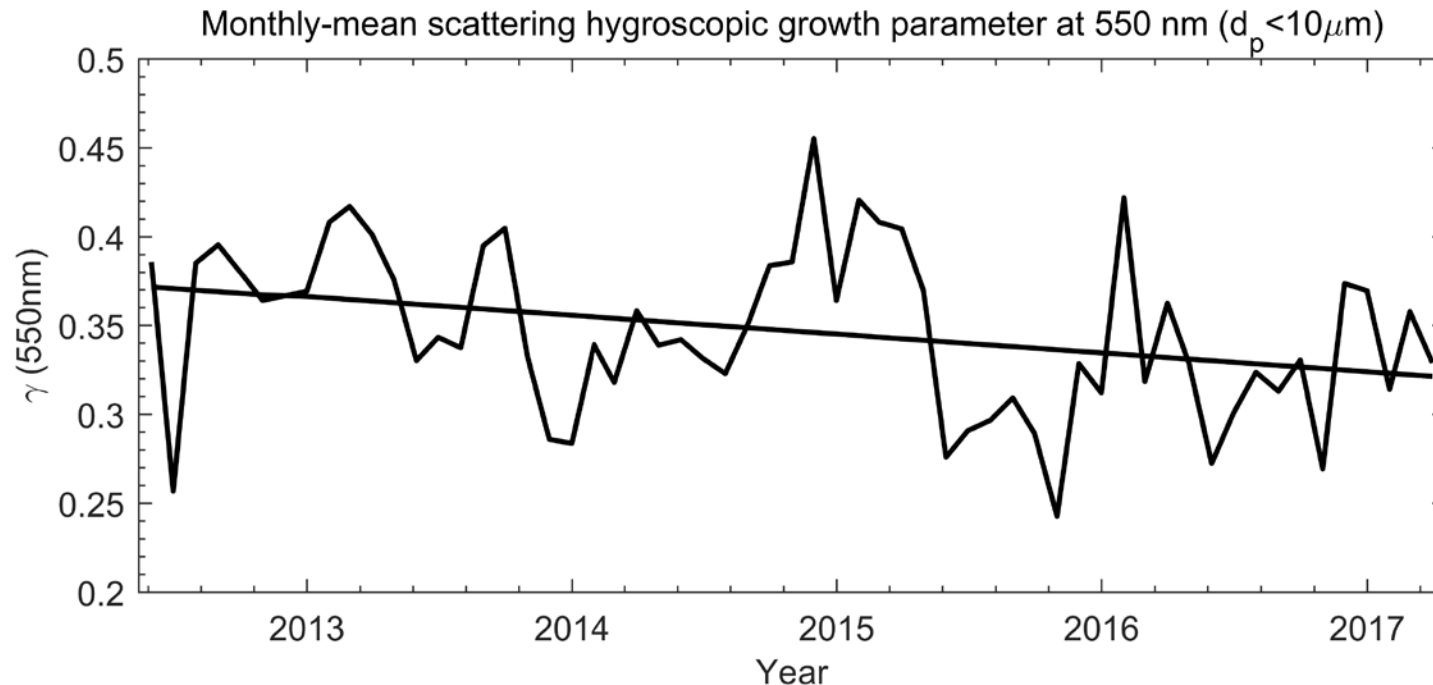
- Asymmetry parameter decreasing, with notable exception of June 2015 (long-distance smoke from Canadian wildfires, along with a few Saharan dust episodes)
- However, scattering and absorption still dominated by sub-micron particles (next slide), indicating a shift toward smaller accumulation-mode particles (i.e. less particle growth)
- Smaller g results in more negative DRE at both the TOA and the surface (by similar amounts), as seen by the DRE sensitivity values at APP. This could (to a small degree) offset the changes in DRE due to AOD decreases

Scattering and absorption Angstrom exponents



- Scattering Angstrom exponent demonstrates little long-term change (other than modulation influenced by smoke/dust events in summer 2015) and more sub-1 micron aerosol influence in 2014 (to be studied) so the fraction of scattering by sub-1 micron particles is not changing
- Absorption Angstrom exponent exhibits 'apparent' increase in time, possibly due to increased influence from biomass burning and (in summer) less coating of soot by transparent coatings such as sulfates and organics (Gyawali, et al. 2009)

Aerosol hygroscopicity



- Data record only 5 years but (for fun) is included to provide some potential insights
- Gamma is highly anti-correlated ($r = -0.71$) with the organic/sulfate mass fraction at APP (not shown) so any long-term decreases could indicate a reduced influence of sulfate aerosols at APP, consistent with studies (Hand et al., 2014) showing large decreases in sulfate in eastern U.S
- Decreasing gamma would reduce light scattering in atmospheric column, which would lower AOD and reduce aerosol cooling effect at TOA and surface (i.e. make DRE less negative)

IV. Conclusions and future work

- Decreasing AOD in SE US most likely is reducing the cooling effect of aerosols at TOA and surface. One hypothesis is that lower levels of SO₂ and NO_x could be leading to less SOA production in region during summer months, coupled with less sulfate due to reductions (for now???) in coal-fired energy production
- Apparent trend toward smaller particles will to a small degree offset the less negative DRE resulting from lower AOD. More detailed studies involving size-segregated aerosol chemical composition are necessary in order to better understand reason for this
- Drier conditions and more wildfires in the SE US may lead to larger increase of episodic wildfires to regional DRE and air quality

Future work will involve

- Long-term aerosol chemistry measurements (hopefully) similar to those conducted using an AMS during 2012-2013
- Detailed trend studies of DRE and AOPs (as datasets near 10 years)

As Dan Jaffe stated this morning: “We welcome collaborators!”

Acknowledgements

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

- Many Appalachian State University (ASU) student researchers who contributed to data collection and instrument maintenance
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