Title of Grant / Cooperative Agreement:	The missing Middle: An investigation on aerosol properties and lifecycle in the lower to middle free troposphere
Type of Report:	Summary of Research
Name of Principal Investigator:	Dr. Robert Holz
Period Covered by Report:	05/15/2015 - 05/14/2018
Name and Address of recipient's institution:	The Board of Regents of the University of Wisconsin System
NASA Grant / Cooperative Agreement Number:	NNX15AK93G

Reference 2 CFR § 1800.908 or 14 CFR § 1260.28 Patent Rights *as applicable (abbreviated below)*
The recipient may use whatever format is convenient to disclose subject invention required in subparagraph (c)(1). NASA prefers that the recipient use either the electronic or paper version of NASA Form 1679, Disclosure of Invention and New Technology (Including Software), to disclose subject inventions. Both the electronic and paper version of the NASA Form 1679 may be accessed at the electronic New Technology Reporting Web site https://invention.nasa.gov.

A final new technology summary report listing all subject inventions (or a statement certifying there were none) for the entire award period; which report shall be submitted within 90 days after the end date for the period of performance within the designated system noted within the award document."

Have any Subject Inventions / New Technology Items resulted from work performed under this Grant / Cooperative Agreement?	No ●	Yes ○
If yes a complete listing should be provided here: Details can be provided in the body of the Summary of Research report.		

Reference 2 CFR § 1800.907 or 14 CFR § 1260.27 Equipment and Other Property *as applicable* (abbreviated below)
A Final Inventory Report of Federally Owned Property, including equipment where title was taken by the Government, will be submitted by the Recipient no later than 60 days after the expiration date of the grant. Negative responses for Final Inventory Reports are required.

Is there any Federally Owned Property, either Government Furnished or Grantee Acquired, in the custody of the Recipient?	No ●	Yes ○
If yes please attach a complete listing including information as set forth at § 1260.134(f)(1).		

Attach the Summary of Research text behind this cover sheet.

Reference 2 CFR § 1800.902 or 14 CFR § 1260.22 Technical publications and reports *as applicable* (abbreviated below)
Reports shall be in the English language, informal in nature, and ordinarily not exceed three pages (not counting bibliographies, abstracts, and lists of other media).

A Summary of Research (or Educational Activity Report in the case of Education Grants) is due within 90 days after the expiration date of the grant, regardless of whether or not support is continued under another grant. This report shall be a comprehensive summary of significant accomplishments during the duration of the grant.

Final report for "The Missing Middle: An investigation on aerosol properties and lifecycle in the lower to middle free troposphere"

Funding document: NNH15AK93G

Institutional Principal Investigators:	*Business Managers:*
Jeffrey S. Reid Marine Meteorology Division Naval Research Laboratory (NRL) 7 Grace Hopper Ave., Monterey, CA, 93943 (831) 656-4725; jeffrey.reid@nrlmry.navy.mil	**Linda Hazard** Marine Meteorology Division Naval Research Laboratory 7 Grace Hopper Ave., Monterey, CA, 93943 (831) 656-4794; hazard@nrlmry.navy.mil
Robert E. Holz SSEC/CIMSS 1225 W. Dayton St Madison, WI 53706 (608) 263-2566; reholz@ssec.wisc.edu	**Jennifer Hackel** SSEC/CIMSS 1225 W. Dayton St Madison, WI 53706 (608)262-0985; john.roberts@ssec.wisc.edu

Summary goals:

A joint Naval Research Laboratory and University of Wisconsin-Madison effort is exploring the nature of LMFT aerosol and cloud features using: a) SEAC^{4}RS supported ground based lidar data from Huntsville, AL and Singapore; b) in situ observations from SEAC^{4}RS DC-8 research flights; and c) a combined analysis supported by remote sensing and mesoscale modeling with a radar assimilation framework. These components allowed for an examination of the sources and properties of LMFT aerosol layers and their relationship to alto level clouds. While the study was able to characterize synoptic features transported into the region (such as smoke injected to mid-levels from the Pacific Northwest, the focus was on aerosol dynamics in the vicinity of small to deep convection and frontal features. For conditions of small to medium convection, we documented aerosol particle chemistry and microphysics from the PBL, through the entrainment zone and ultimately detrainment into the free troposphere We suspect that the melting level (0^{o}C) has special significance in the injection of PBL air into the free troposphere. Cumulus mediocris and congestus can inject PBL air to 0^{o}C, resulting in the commonly observed upward decay in particle mass from the mixed layer top that is reported in many field campaigns. The resulting transported layers often develop altocumulus tops. Also, at 0^{o}C there is proclivity for aerosol layer formation through detrainment from altocumulus melting level shelves of deep convection. In between the 0^{o}C and thunderstorm outflow levels, a relative barren zone exists as clouds that initiate freezing have enough latent heat to become full thunderstorms. Aerosol layers found in this barren zone are most often a result of large scale lifting.

Summary activity:

This research proposal had two major thrusts for investigating the nature of mid tropospheric layers; 1) the analysis of airborne and model data led by the Naval Research Laboratory, and 2) the development and analysis of a suitable ground, airborne and space based lidar dataset led by the University of Wisconsin. Completed milestones include:

Year 1:

1. Quality assurance of DC8 airborne datasets
2. Completion of a COMAPS mesoscale simulation
3. Generation of a level 2 HSRL dataset for aerosol and clouds

Year 2:

1. Perform comprehensive analysis of HSRL dataset on aerosol layering
2. Wrote and published JGR paper on special issue on aerosol vertical structure

Year 3:

1. Performed analysis of boundary layer aerosol properties and granted a regional baseline analysis for SEAC4RS.
2. Wrote and published paper in JGR examining spatial correlation of aerosol properties in SEAC^{4}RS, and evaluating the representativeness of the study to other years.
3. Wrote and are about to submit a paper on lidar and DC-8 analysis of aerosol outflow into the lower and middle free troposphere.

Analysis in the third year

Of particular note in the third year our the outcomes of regional studies of the boundary layer environment from surface stations and profile data from the DC-8.

Ground analysis: We used the summer 2013 ground network as a baseline for further discussion as to what intensive measurements from aircraft and ground stations really mean, and likewise provide insight into the monitoring of PM from space. Most importantly, we noted that intersite correlations of PM$_{2.5}$ throughout Southeast were better than AOD intersite correlations. In fact, when using surrounding PM$_{2.5}$ measurements to interpolate PM$_{2.5}$ mass at the a site in the middle of the southeast site, we found significant improvement over using the site-specific AOD measurements and regression coefficient derived from the PM$_{2.5}$-AOD linear relationship. This coupled with the findings of Reid et al., (2017) demonstrate why AOD PM2.5 relationships are often so poor-it is the perturbations from the entrainment zone and free troposphere rather than the mixed layer that drives variability in the relationship (e.g., Figure 1). We did find, however, that SEAC^{4}RS is representative of the region for the past five years. But significant changes to PM$_{2.5}$

chemistry and mass were observed over the long-term. The long-term change in $PM_{2.5}$ mass and chemistry had impacts on the aerosol optical properties. The α_{sp} saw a reduction across all seasons between the 1998-2007 and the 2008-2013 period. This, accompanied by the reduction in aerosol hygroscopicity due to decreasing SO_4^{2-} concentrations had a clear impact on the $PM_{2.5}$-AOD regression coefficients calculated using satellite-derived AODs, with a steady decline observed in the regression coefficient from 2000 to 2015. Thus, while intensive field campaigns are necessary to develop an in-depth understanding of aerosol physical and optical properties at a given location, long-term observation stations are needed to accurately quantify trends that impact the $PM_{2.5}$-AOD relationship.

By providing a thorough examination of different physical, chemical and optical properties of $PM_{2.5}$ in the SEUS region we demonstrate the capabilities of the coefficient of determination relationships, and where we are in need of moving beyond our attempts to optimize this relationship and instead develop new ways of predicting aerosol mass, such as using triangulation methods among existing ground measurements. Our studies suggest that there are large limitations to the use of AOD for prediction of surface-level $PM_{2.5}$ mass. While high-load aerosol events are easily tracked using remote optical observations, more subtle variations in $PM_{2.5}$ are more difficult to predict using only AOD. The $PM_{2.5}$-AOD relationship varies significantly both seasonally and annually due to changes in aerosol mass, chemistry and resulting optical properties. Our results point to challenges of the use of monitoring air quality via optical means. This is not at all to say satellite data is without value. Without a doubt air quality monitoring and forecasting systems will continue to improve and make better use of satellite data. This is especially true in in data void regions. But the variability between sites in meteorology, and aerosol optical properties, vertical distribution demonstrated studied here naturally points to the large uncertainty developers must cope with. Therefore, the baselining of satellite, modeling, and intensive in situ datasets is still crucial rationale for continued and comprehensive surface monitoring.

Airborne studies: The airborne analysis focused on a DC 8 flight on August 12, 2013 from the SEAC^{4}RS campaign. This flight demonstrated the best examples of Altocumulus cloud (Ac), aerosol and water vapor layering phenomena in a convective regime over the southeastern United States (SEUS) for the study. This day was chosen due to proximity of the DC-8 research aircraft to a High Spectral Resolution Lidar (HSRL) at Huntsville Al. The HSRL gives period level perspective on Ac clouds and their observed aerosol “halo” to help interpret in situ DC-8 data. Analysis of the meteorology of the region on this day supported that aerosol was “local” to the SEUS and thus should be taken representative of regionally forced convective environments. A 33 hour sample of lidar data was presented to demonstrate the diurnal cycle of cloud features in this convective environment. The HSRL provided aerosol backscatter and precisions at or better than 5% of Rayleigh, and demonstrated extraordinarily fine aerosol features in the vicinity of altocumulus clouds formed in the outflow of deep convection. This day was in turn compared to a DC-8 profile conducted that afternoon on the downwind side of a developing storm providing in situ data on the middle free troposphere aerosol environment.

The day is best represented in the lidar profile provided in Figure 2. Aside from typical boundary layer development and cirrus outflow, numerous aerosol and Ac decks were identified, many of these Ac produced ice virga. Ac formed at the top of the residual of the previous day’s planetary boundary layer entrainment zone, where air was largely influenced by boundary layer cloud detrainment. This layer formed in the morning hours, and increased in base altitude with the developing boundary layer below it. Such rising may be a result of mesoscale flows or cloud lofting. Above the PBL-top Ac, several other combined aerosol-Ac-water vapor layers were observed. Including 1) a 4 km detrainment layer that we surmise is from the very tops of cumulus mediocris clouds; 2) layers at or just below 4.7 km/0^{o}C melting level representing deep convective detrainment shelves, and 3) 6-7 km layers consistent with that appear to be consistent with detrainment from the tops of congestus clouds. From the HSRL, Ac clouds were associated with clear aerosol "halos", typically with Ac clouds on top. The intensity of aerosol backscatter associated with Ac cloud halos appeared to decrease with height, beyond what would be expected from adiabatic expansion. The lowest Ac clouds associated with PBL entrainment zone have larger returns associated with their proximity to the polluted PBL and large accumulation particle size, and hygroscopicity.

The lidar environment was well sampled by the DC8 on the down shear edge of a thunderstorm (Figure 3). Particles in the near boundary layer were as expected, relaitvley large and organic domnated. However, middle free troposphere layers had markedly smaller accumulation mode sizes with height, but higher CN counts. Aerosol layers above 0^{o}C had the smaller accumulation model sizes and highest CN concentrations. This is consistent with further cloud processing and scrubbing of detraining air at higher altitudes. Indeed particle size and composition data suggest that detraining particles undergo aqueous phase or microphysical transformations, while at the same time larger particles are being scavenged.

Examination of profiles suggest an excess of water vapor and aerosol particles relative to CO within and above the PBL entrainment zone to the melting level, and observations around clouds may reflect multiple entrainment-detrainment events (e.g., Yeo and Romps, 2013). We expected the ratio of water vapor to CO concentration in undiluted ascent should be uniquely determined by the parcel initial properties in the mixed layer, and departures from this ratio within the cloud reflect the action of mixing. Detraining air from deep convection at the melting level provides the strongest local source of water vapor (direct and via evaporated cloud), and also the largest water vapor to CO ratio. We hypothesize that up to the melting level, detrainment is dominated by boundary layer air, whereas above this level air is more mixed involving entrainment/detrainment along the clouds. Water vapor flux to the middle free troposphere may also be enhanced by evaporating precipitation, whereas higher altitude parcels undergo dehydration.

This work leads to numerous questions regarding relationships between aerosol layers and the properties of Ac clouds. It has been long hypothesized that increasing trends in aerosol concentrations over the past decades will result in more convective lofting, and then perhaps an indirect effect in associated Ac clouds and perhaps increases in cloud lifetimes (e.g., Parungno et al., 1994). The observation that Ac clouds have visible halos of accumulation mode particles certainly points to Ac as being a coupled with the boundary layer aerosol system. Enhancements in accumulation mode particles near Ac appear to be anti-correlated with CN for this case-likely due to available surface area for secondary mass production and or coagulation. At the same time, explosive nucleation events are visible and expected in the vicinity of clouds. All of this suggests complex CCN-Ac coupling and questions about layer flow dynamics in and around Ac and their associated aerosol layers and/or halos. Does the cycling of air through an Ac feedback into its own CCN budget? Does non-precipitating cycling enhance particle size and hence CCN number for any given supersaturation? In precipitating Ac, where are replacement CCN coming from, and do nucleating CN ever offer a supply? Or, as a hypothesis, perhaps CN events can sustain and enhance CCN populations in Ac clouds. The null hypothesis would then be that CN are consumed in individual droplets and have little overall effect in clouds with such meager updraft velocities and super saturations.

Peer Reviewed Publications

Kaku, K. C., J. S. Reid, J. L. Hand E. S. Edgerton, B. N. Holben, J. Zhang, R. E. Holz (2018), Assessing the challenges of surface- level aerosol mass estimates from remote sensing during the SEAC^{4}RS and SEARCH campaigns: Baseline surface observations and remote sensing in the Southeastern United States, J. Geophys. Res, https://doi.org/10.1029/2017JD028074

Reid, J. S., R. E. Kuehen, R. E. Holz, E. W. Eloranta, K. C. Kaku, S. Kuang, M. J. Newchurch, A. M. Thompson, C. R. Trepte, J. Zhang, J. L. Hand, B. N. Holben, P. Minnis, D. J. Posselt (2017), Ground based high spectral resolution lidar observation of aerosol vertical distribution in the summertime Southeast United States. *J. Geophys Res.*, doi: 10.1002/2016JD025798.

Reid, J. S., D. J. Posselt, R. A. Holz, P. Campuzano-Jost, G. Chen, E. W. Eloranta, J. L. Jimenez, K. C. Kaku, R. E. Kuehn, J. Zhang, B. Anderson, T. P. Bui, G. S. Diskin, P. Minnis, M. J. Newchurch, S. Tanelli, C. R. Trepte, K. L. Thornhill, L. D. Ziemba (2018, to be submitted), Observations of free tropospheric aerosol, water vapor and altocumulus cloud layers in association with a developing convective environment, Atmospheric Chemistry and Physics.

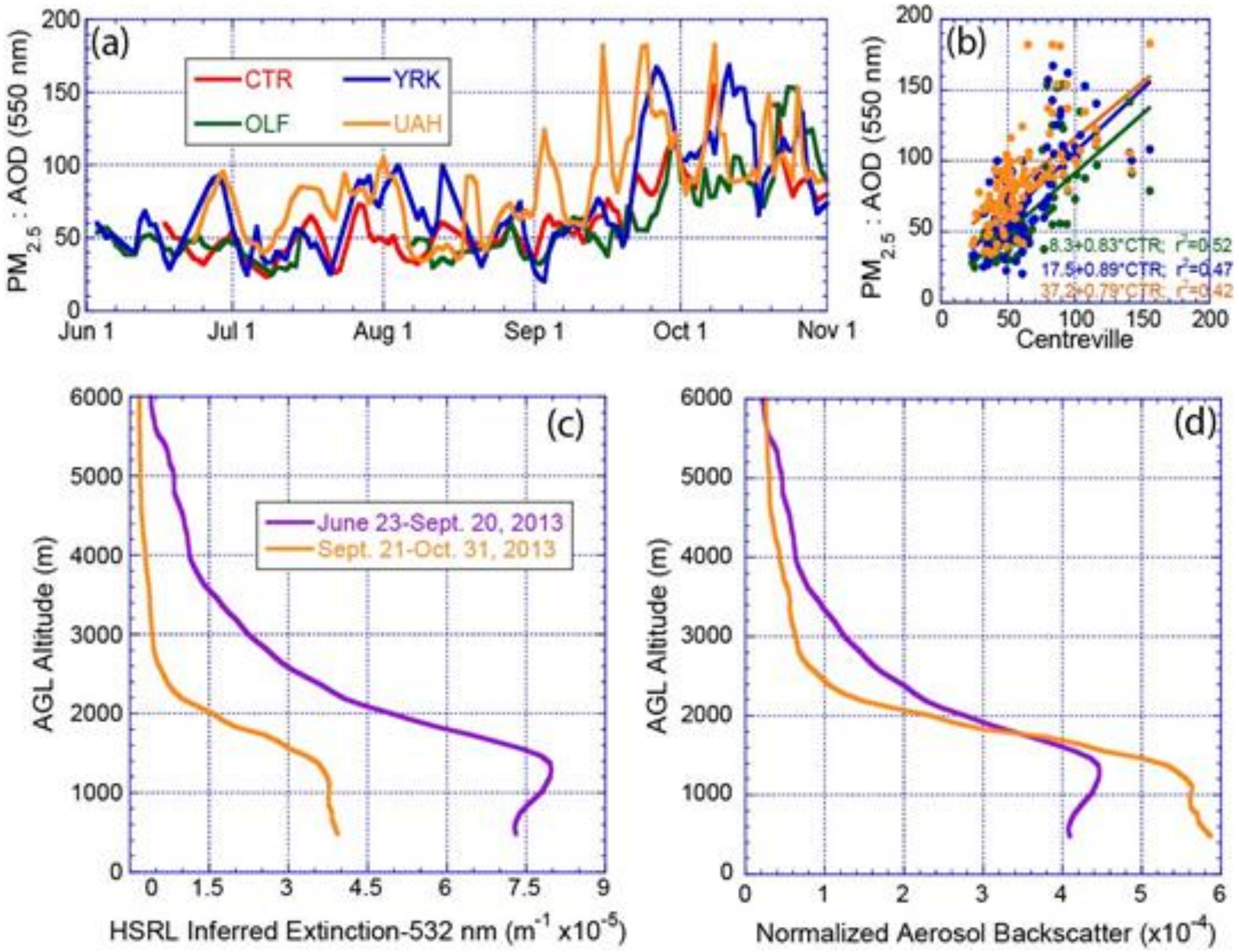

Figure 1. Influence of seasonality on the PM$_{2.5}$ to AOD relationship including (a) time series of the 5 day boxcar average ratio of PM$_{2.5}$ to AERONET 550 nm AOD, (b) the PM$_{2.5}$ to AOD ratio between sites, including linear regression statistics, (c) the average UAH HSRL inferred extinction distribution for cloud free cases for the summer and autumnal time period and (d) backscatter profiles normalized to total integrated backscatter.

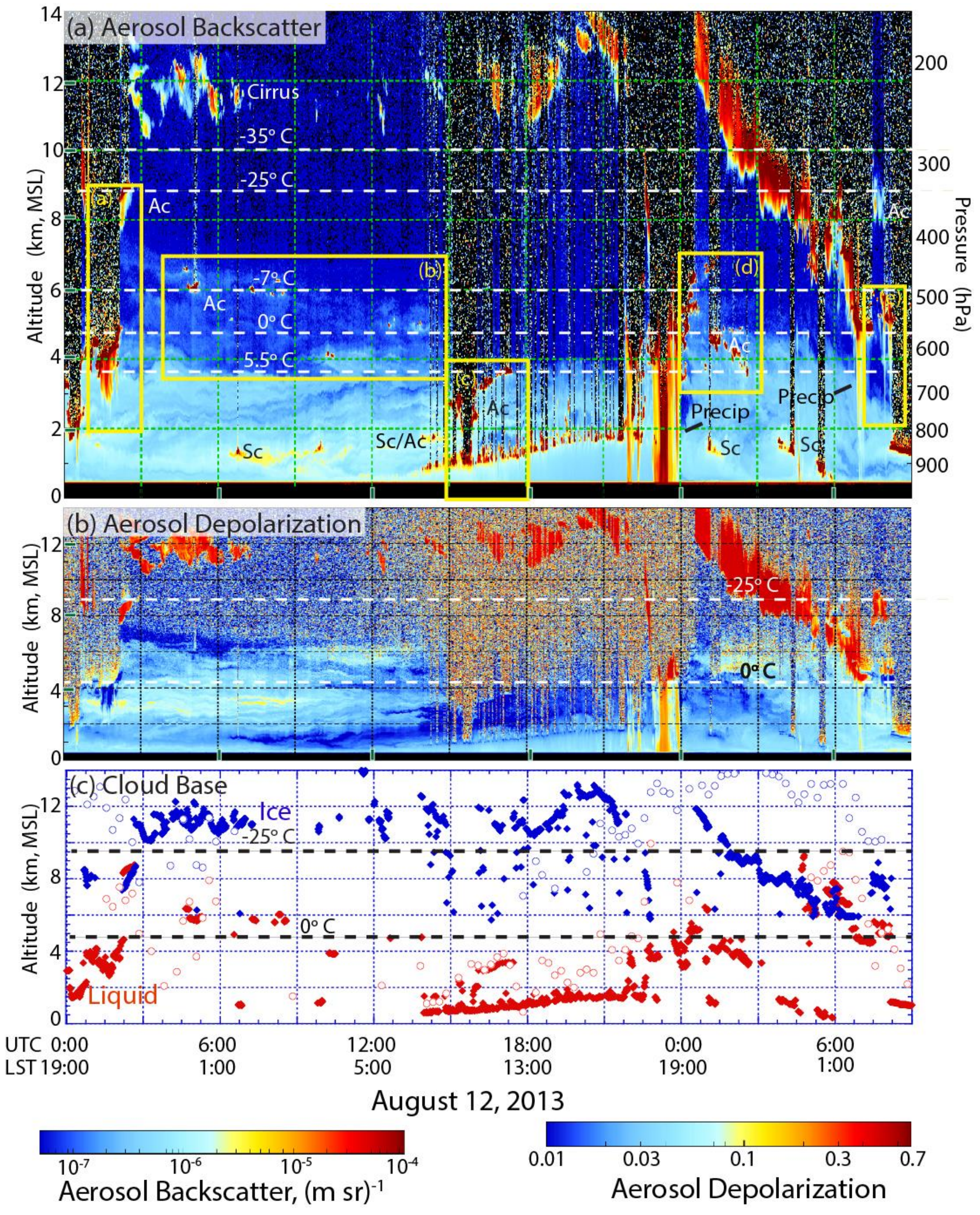

Figure 2. Example lidar data for August 12, 2013. UW HSRL aerosol (a) backscatter and (b) depolarization from the surface to 14 km AGL. Listed are cloud types, phenomenon and from a 13:30 radiosonde release, key temperature isopleths. Also shown in (c) are liquid and ice cloud bases (solid) from the ground based HSRL, and liquid and cloud tops from GOES-13 (open). To convert from AGL to MSL, subtract 220 m.

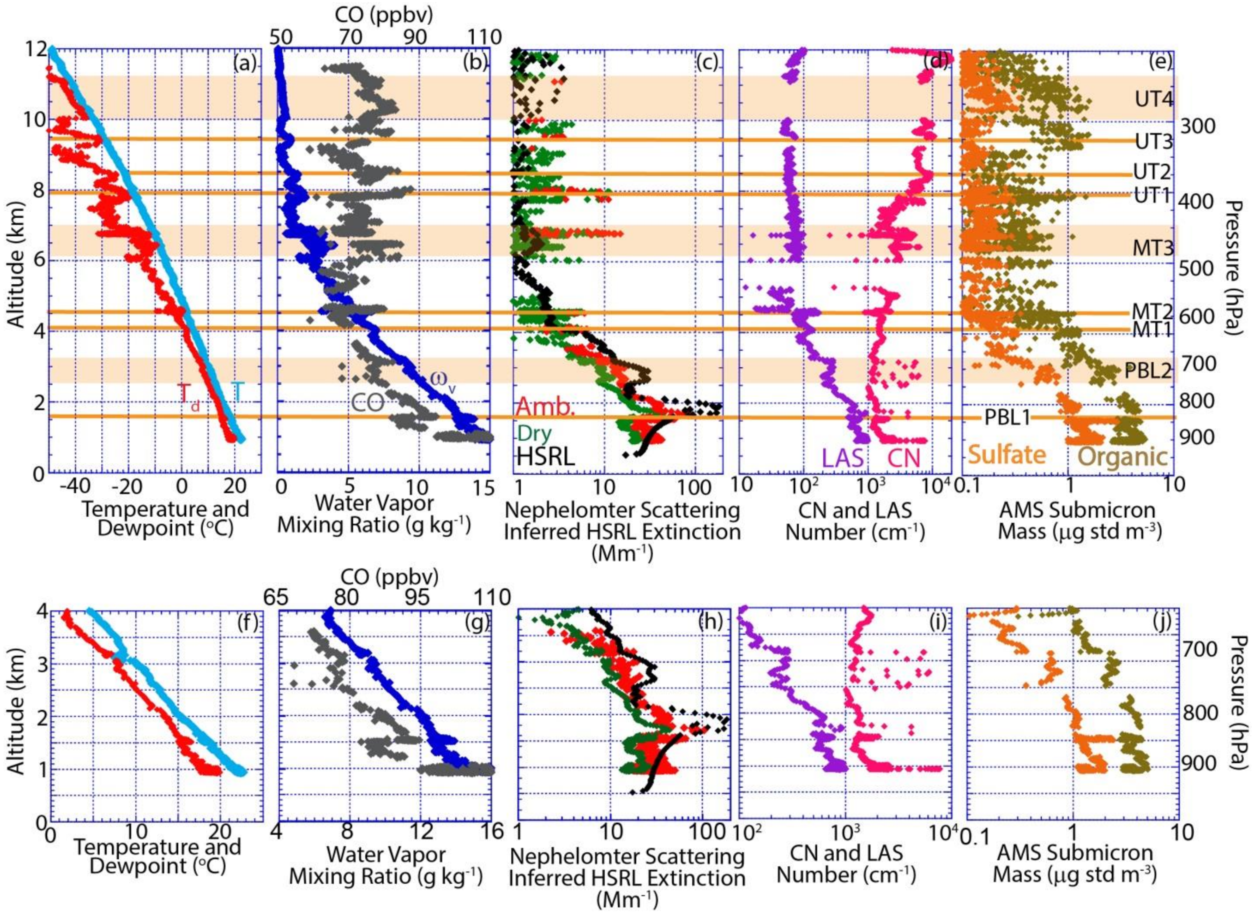

Figure 3. DC-8 aircraft spiral sounding data initiated at August 12, 2013 19:10:30 UTC on the downwind side of a thunderstorm over northwest Alabama. Altitudes are relative to mean sea level, ~ 300 m higher than AGL. Included is (a) Temperature and dew point; (b) Water vapor mixing ratio (ω_v) and CO; (c) DC-8 total ambient and fine dry 550 nm nephelometer with the ground based UW HSRL derived extinction (lidar ratio=55 sr^{-1}) at Huntsville Al; (c) Number concentration from laser aerosol spectrometer (LAS, d_p>0.1 μm) and condensation nuclei (CN, dp>10 nm); (e) Aerosol mass spectrometer organic materials and sulfate. Key moisture and aerosol layers as discussed in the text are marked as orange lines or bands.(f)-(j), same as (a)-(e) expanded in the vertical to enhance PBL feature readability.