



Variability in Aerosol Properties Associated with Changes in Relative Humidity as Measured by the Airborne High Spectral Resolution Lidar-2

Richard Ferrare¹, John Hair¹, Chris Hostetler¹, David Harper¹, Shane Seaman¹,
Taylor Shingler¹, Ewan Crosbie², Edward Winstead², Luke Ziemba¹, Lee
Thornhill², Marta Fenn², Marian Clayton², Amy Jo Scarino², Sharon Burton¹,
Anthony Cook¹, Glenn Diskin¹, Rich Moore¹, Claire Robinson², Josh DiGangi¹,
John Nowak¹, Hongyu Liu³, Seethala Chellappan⁴, Paquia Zuidema⁴, Joseph
Schlosser⁵, Armin Sorooshian⁵, Sue van den Heever⁶



¹NASA Langley Research Center

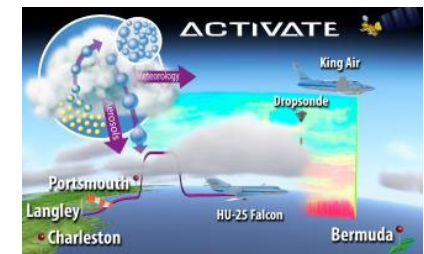
²SSAI/NASA/LaRC

³National Institute of Aerospace/NASA/LaRC

⁴University of Miami

⁵University of Arizona

⁶Colorado State University

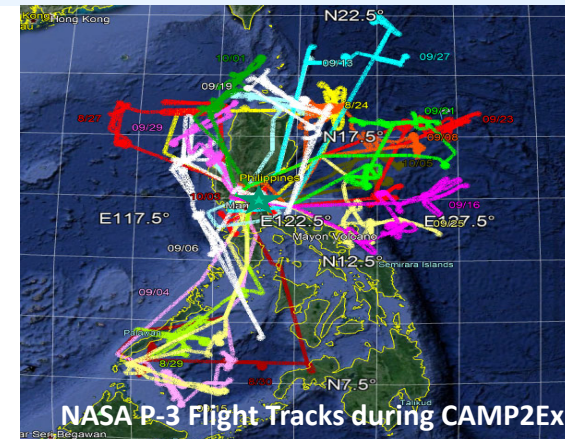


HSRL-2 Data Acquired During NASA CAMP2Ex and ACTIVATE Field Missions



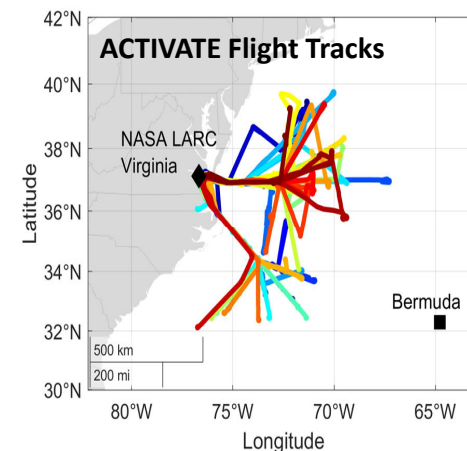
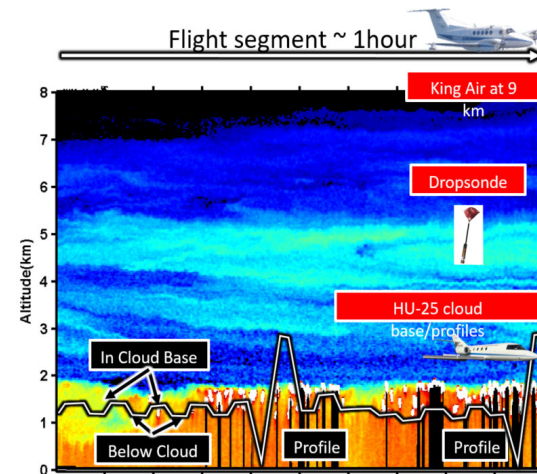
NASA CAMP2Ex (2019)

- CAMP2Ex addresses three NASA focus areas
 - Aerosol and cloud microphysics
 - Cloud and aerosol radiation
 - Aerosol and cloud meteorology
- NASA LaRC HSRL-2 deployed on P-3B aircraft for nadir viewing measurements
- P-3B, based at Clark Air Base, conducted 19 science flights between Aug. 24 and Oct. 5, 2019



NASA EVS-3 ACTIVATE (2020, 2021, 2022)

- Characterize aerosol-cloud-meteorology interactions using systematic and simultaneous in situ and remote sensing airborne measurements with two aircraft
- Focus on marine boundary layer (MBL) clouds of the US Mid-Atlantic Coast
- NASA LaRC HSRL-2 deployed on LaRC King Air aircraft for nadir viewing measurements
- NASA LaRC HU-25 Falcon aircraft simultaneously deployed in situ instruments



Summary of HSRL-2 CAMP2Ex and ACTIVATE Products



Available Archived Data Products

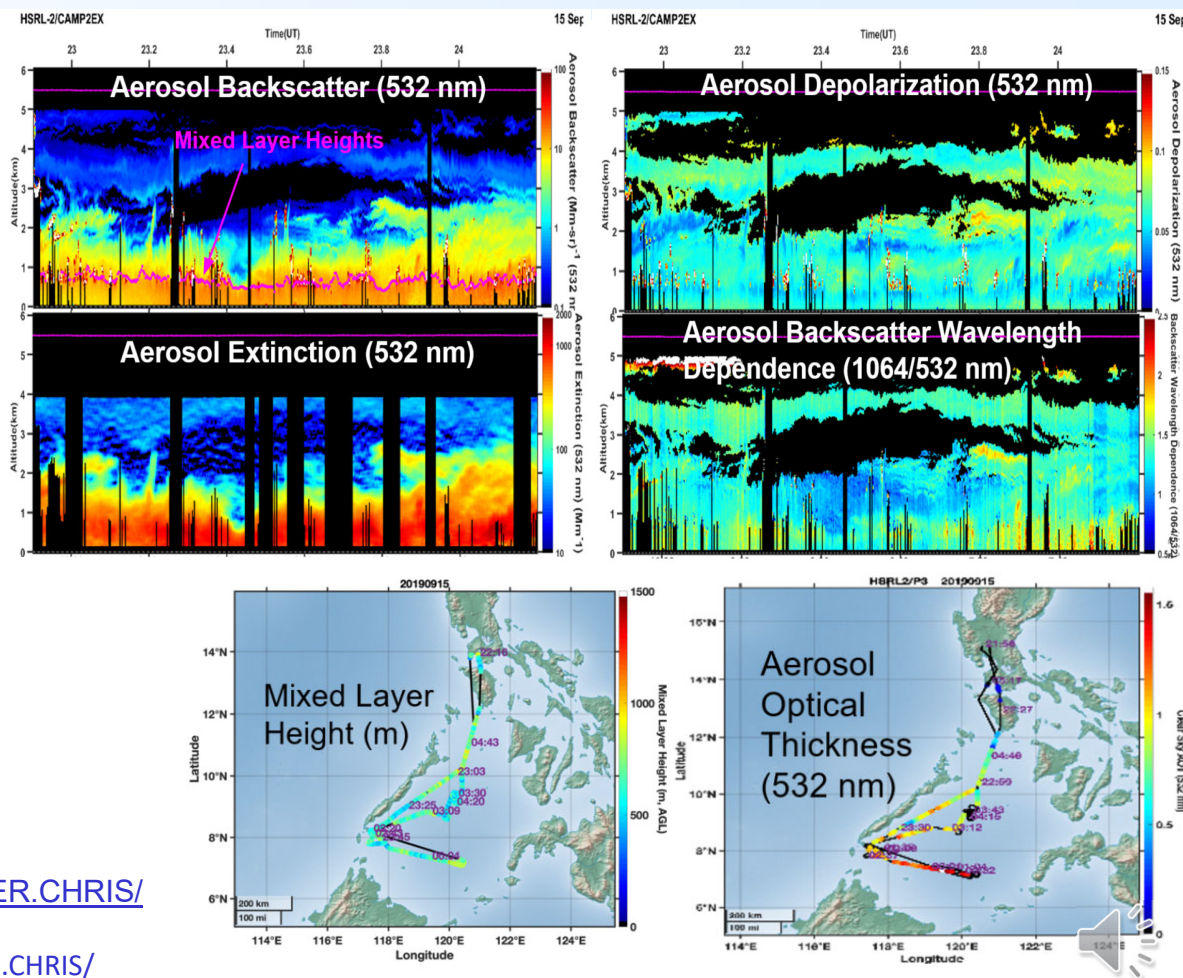
- Aerosol Extensive Measurements
 - Particulate backscatter profiles (355, 532, 1064 nm)
 - Particulate extinction profiles and AOT (355 and 532 nm)
- Aerosol Intensive measurements
 - Particle depolarization profiles (355, 532, 1064 nm)
 - Extinction-to-backscatter ratio profiles (355 and 532 nm)
 - Angstrom exponent profiles
 - Extinction: 355-532
 - Backscatter 355-532, 532-1064
- Aerosol Type
- Aerosol Optical Thickness
- Mixed Layer Heights
- Mean aerosol extinction and backscatter values within and above the Mixed Layer

HSRL-2 data from CAMP2Ex at

<https://www-air.larc.nasa.gov/cgi-bin/ArcView/camp2ex#HOSTETLER.CHRIS/>

HSRL-2 data from ACTIVATE at

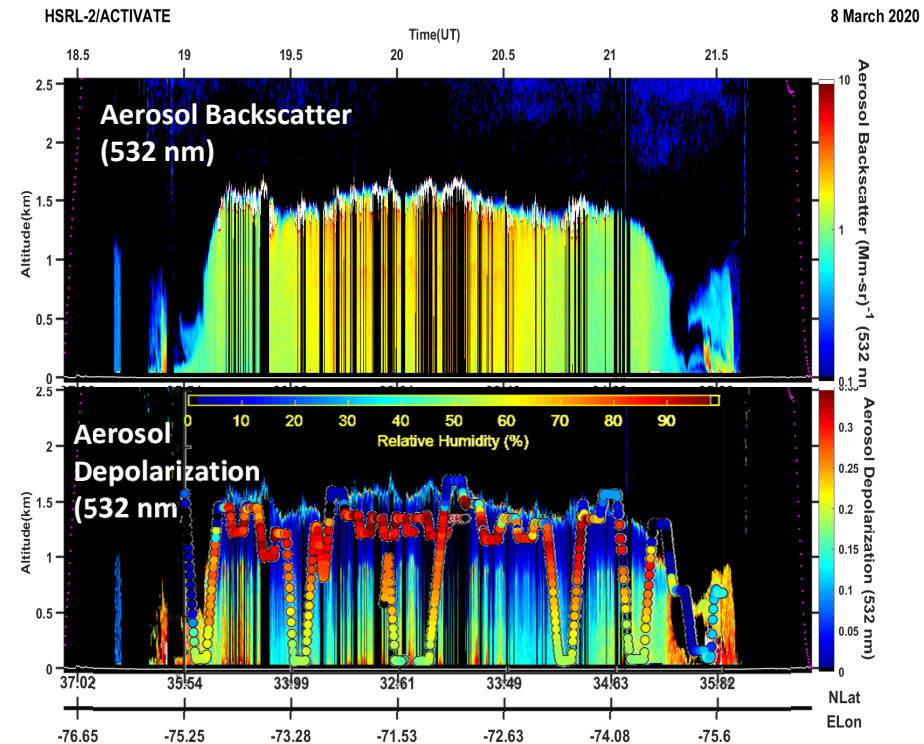
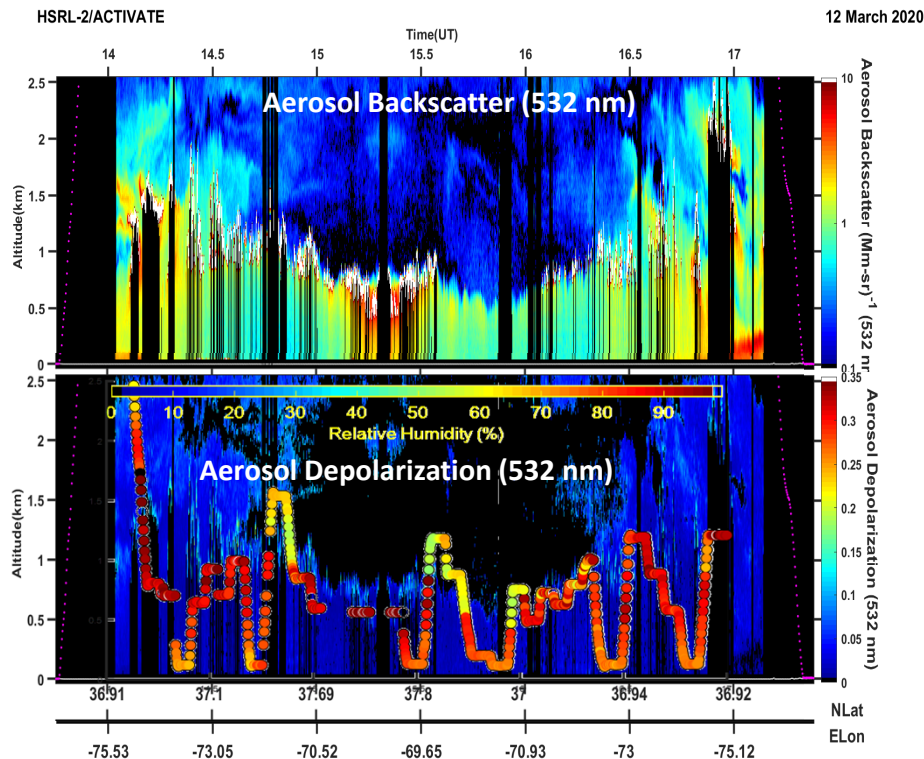
<https://www-air.larc.nasa.gov/cgi-bin/ArcView/activate.2019#HOSTETLER.CHRIS/>



HSRL2 Observations of Low and High Particulate Depolarization over the Ocean During ACTIVATE



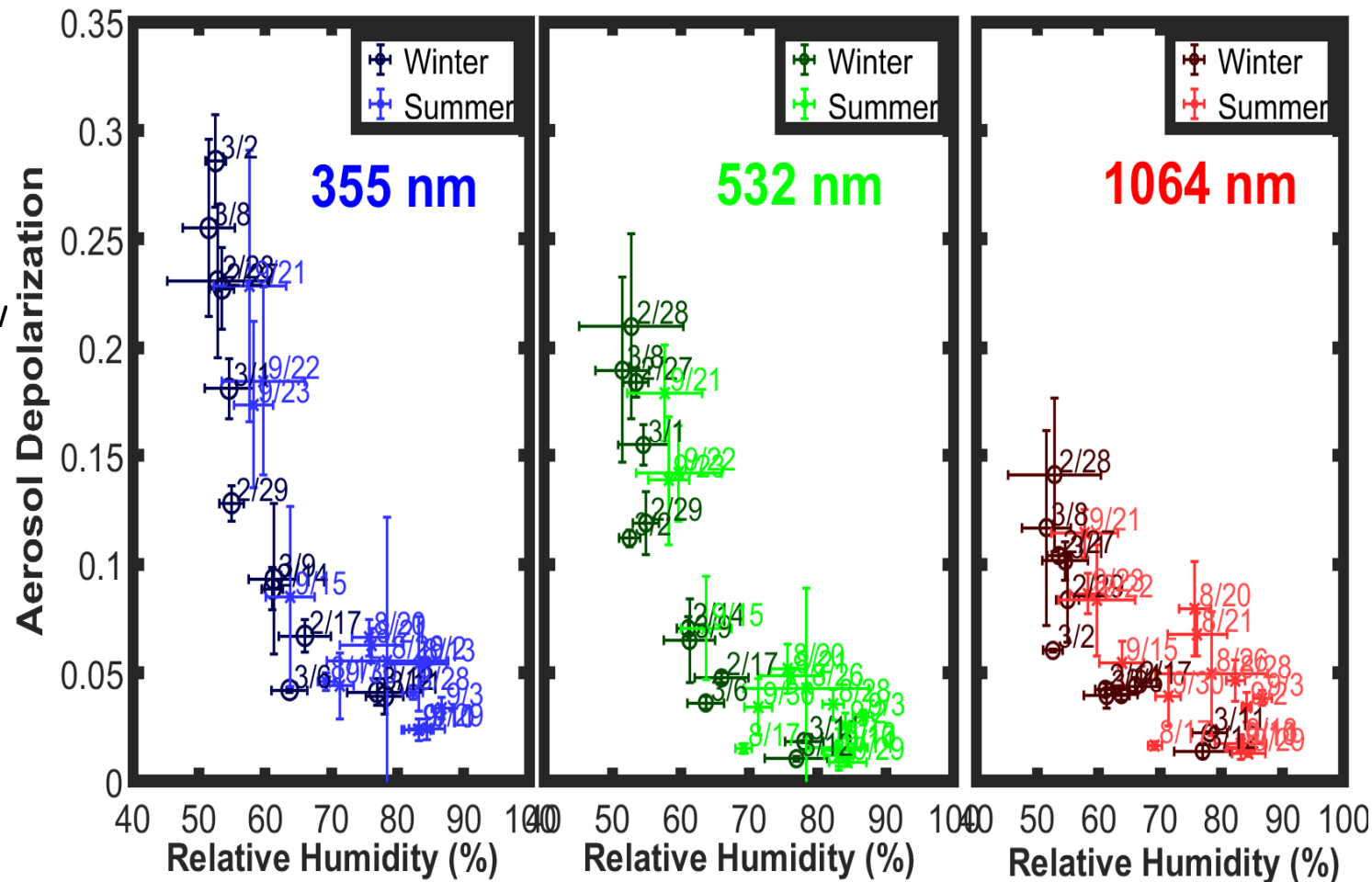
- Typically HSRL2 measures low aerosol depolarization over the ocean. This is associated with spherical sea salt aerosols.
- Note the high (>65%) relative humidity (RH) in the lowest 1 km derived from airborne in situ (Diode Laser Hygrometer-DLH) measurements on the Falcon aircraft
- In contrast, during several ACTIVATE flights during the winter and summer campaigns, HSRL2 measured elevated (>10-15%) aerosol depolarization in the lowest 1 km. Note the lower (<60%) relative humidity (RH) derived from the airborne in situ DLH measurements on the Falcon aircraft



HSRL2 Measurements During ACTIVATE 2020 Show Elevated Aerosol Depolarization when RH is below about 60%



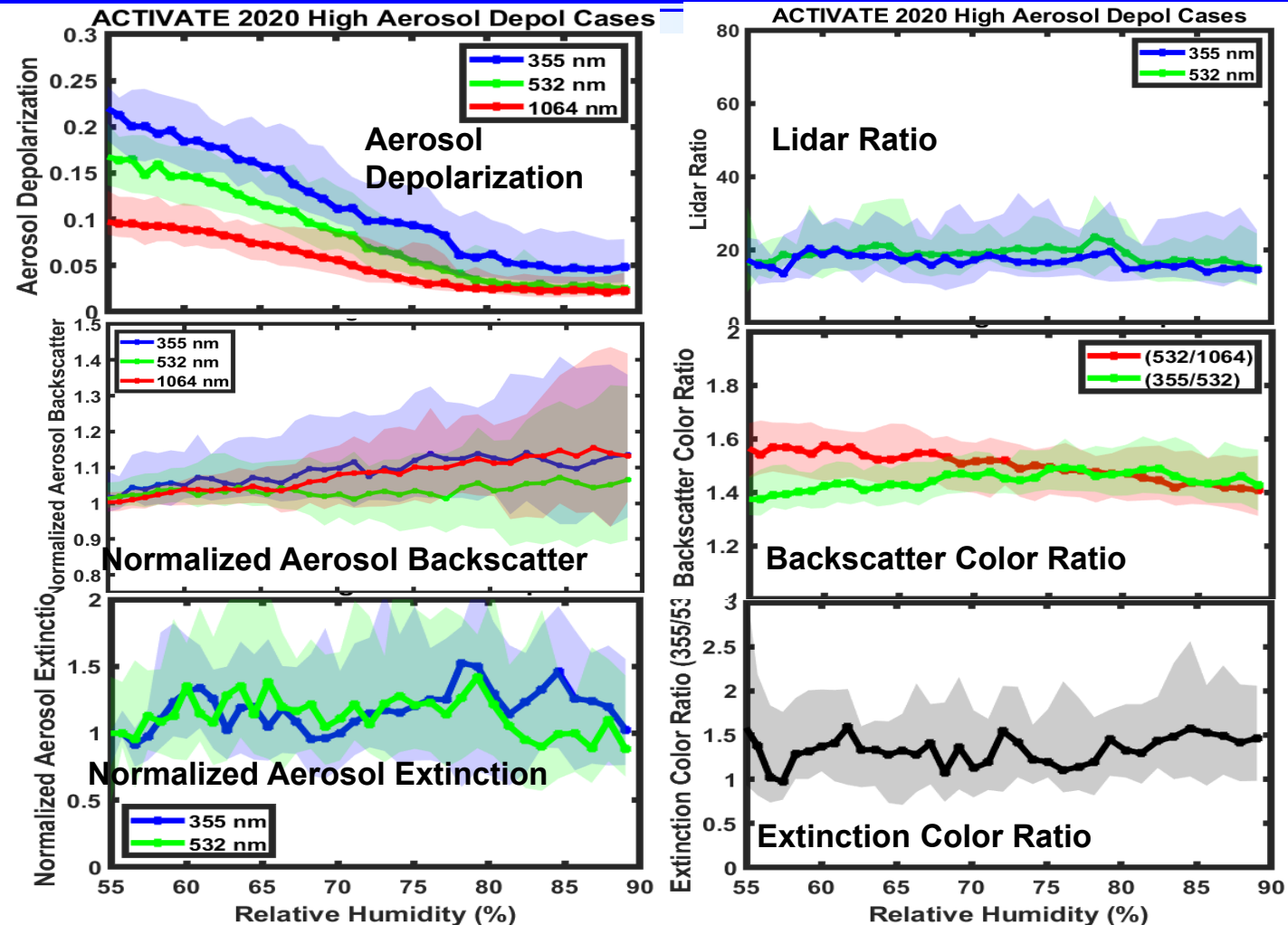
- Average depolarization in lowest 20% of Mixed Layer coincident with dropsondes
- Each point represents average for that date
- High aerosol depolarization is associated with RH below about 60%
- Depolarization decreases with increasing wavelength
- Elevated depolarization during both winter and summer
- HSRL-2 measurements similar to previous lidar observations of nonspherical sea salt (Haarig et al., 2017)



HSRL2 Measurements of Aerosol Optical Properties as a function of RH for Cases of High Aerosol Depolarization during ACTIVATE



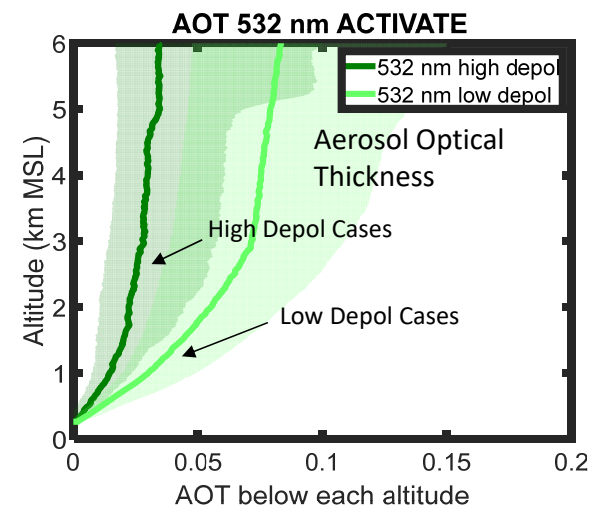
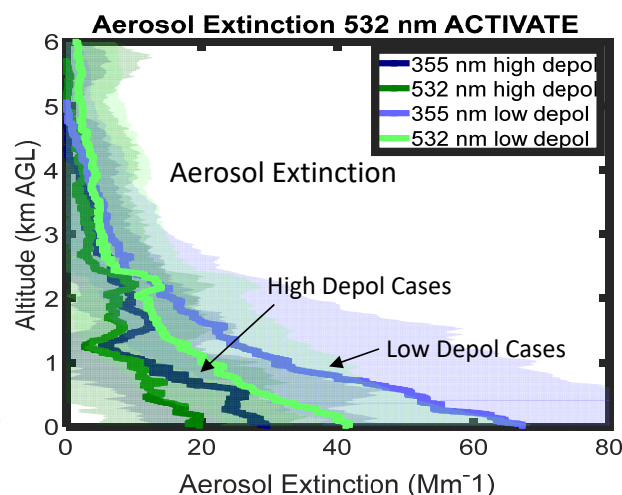
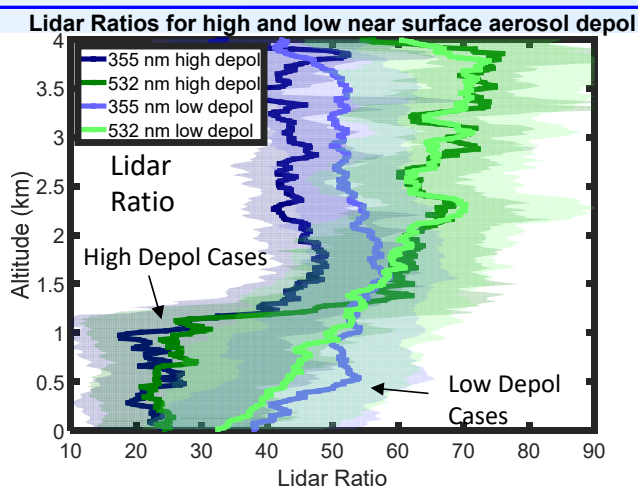
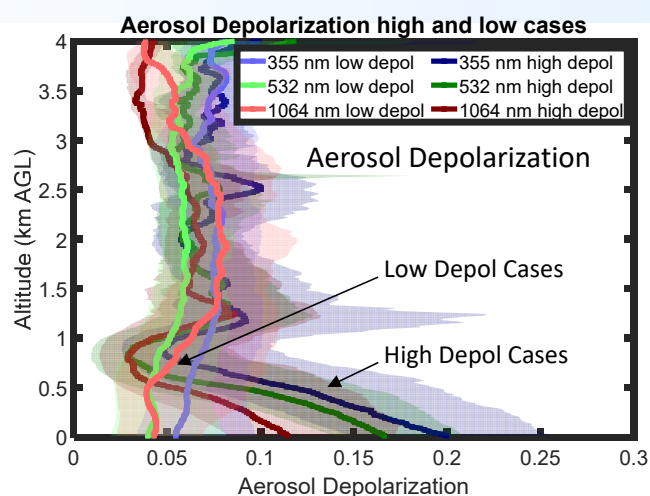
- Cases where aerosol depolarization (532 nm) > 0.1
- Nearly constant water vapor mixing ratio (conservative tracer) to reduce changes due to changes in air mass
- Changes in RH due to changes in temperature
- Significant changes in aerosol shape as shown by changes in aerosol depolarization
- Apparently minimal changes in aerosol size as shown by nearly constant color ratios and lidar ratio



Lower Lidar Ratios, Aerosol Extinction, and AOD associated with the high depolarization, nonspherical sea salt cases



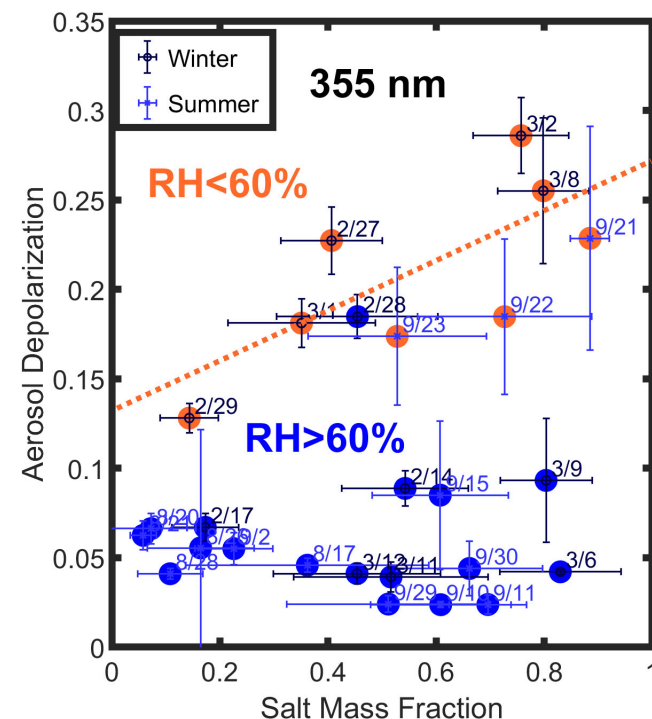
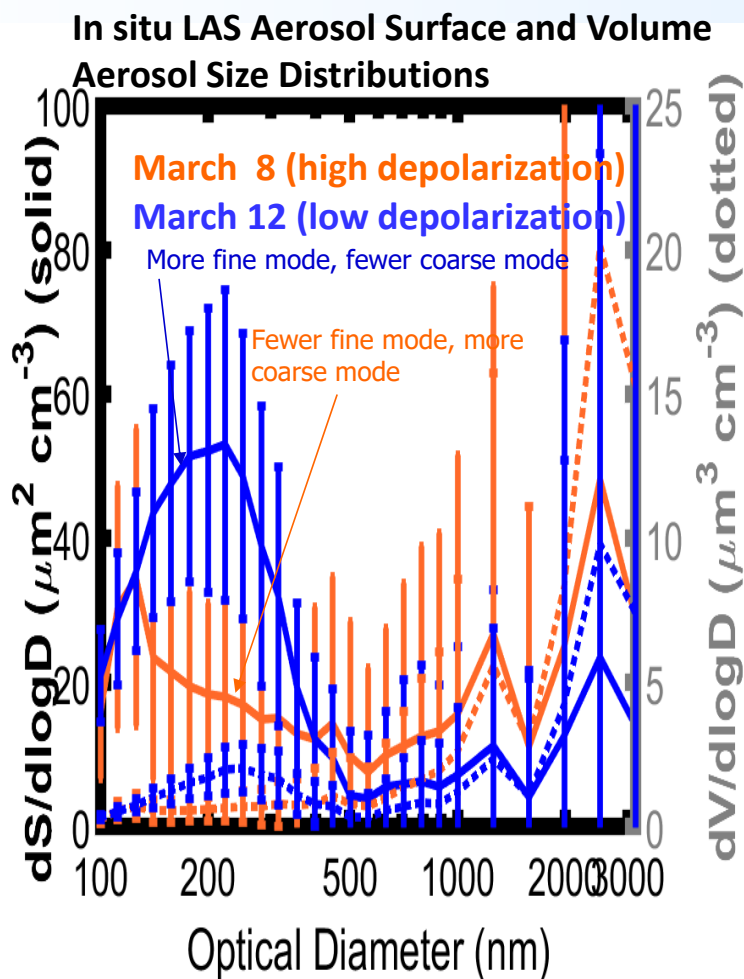
- HSRL2 data divided into high and low near-surface depolarization cases
- High depolarization observed in about 18% of the HSRL2 profiles and during 23 of 33 flights
- Median lidar ratios at 355 and 532 nm were around 20-25 sr, consistent with marine (sea salt) aerosol, during these high depolarization cases
- Higher lidar ratios observed when other aerosol types (e.g. smoke) were present within BL
- Median AOT (532 nm) (0-7 km) for high depolarization cases was about 0.04
- AOT (532 nm) contributed by the nonspherical sea salt particles was small (0.03-0.04)
- Extinction (532 nm) contributed by these particles was small ($\sim 10\text{-}20 \text{ Mm}^{-1}$)





Airborne In situ Aerosol Size Distribution and Salt Mass Fraction Measurements When Enhanced Depolarization was Observed

- During March 8 flights (high depolarization) airborne in situ aerosol size distribution measurements show fewer fine mode particles and more coarse mode particles.
- In contrast, flights on March 12 (low depolarization), show more fine mode particles and fewer coarse mode particles.
- During March 8 flights, airborne in situ aerosol size distributions were very similar throughout the mixed layer - this suggests particle shape, not dry size, varied with RH

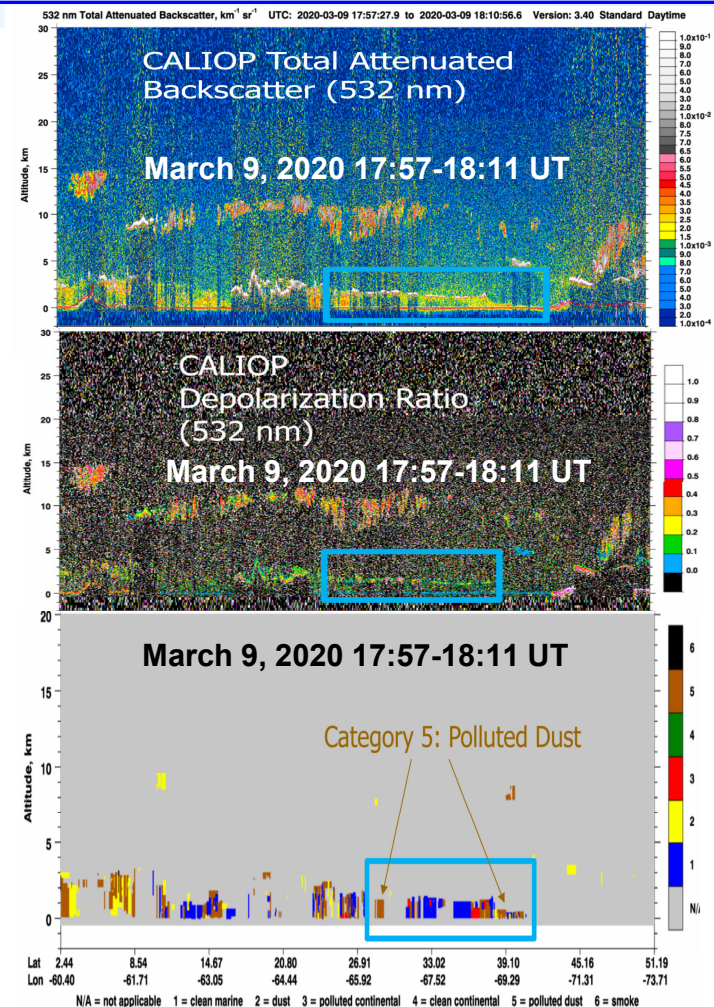


- Elevated depolarization was correlated with salt mass fraction for RH < 60%
- Elevated depolarization was not correlated with wind speed regardless of RH

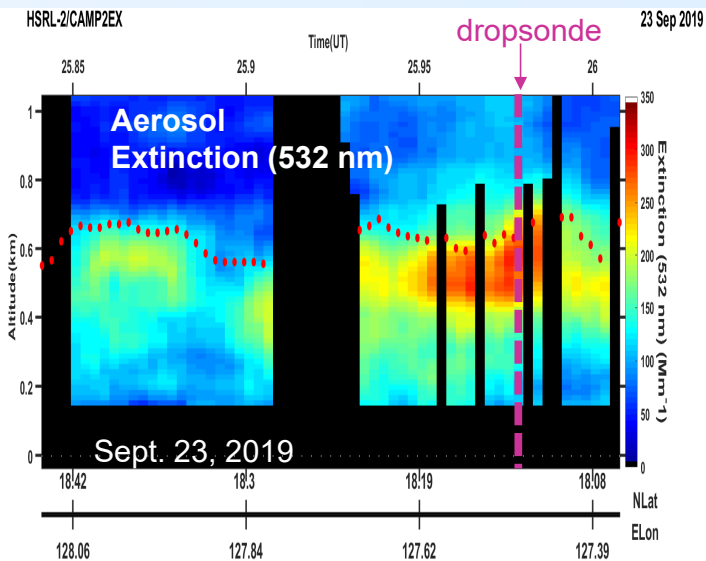
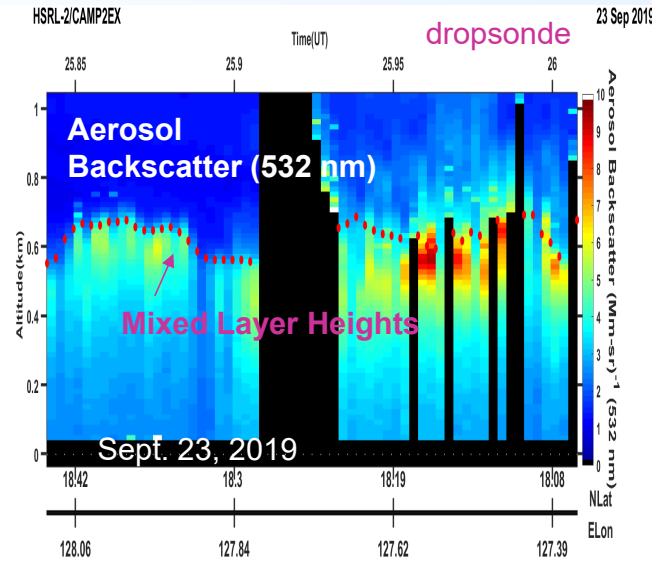
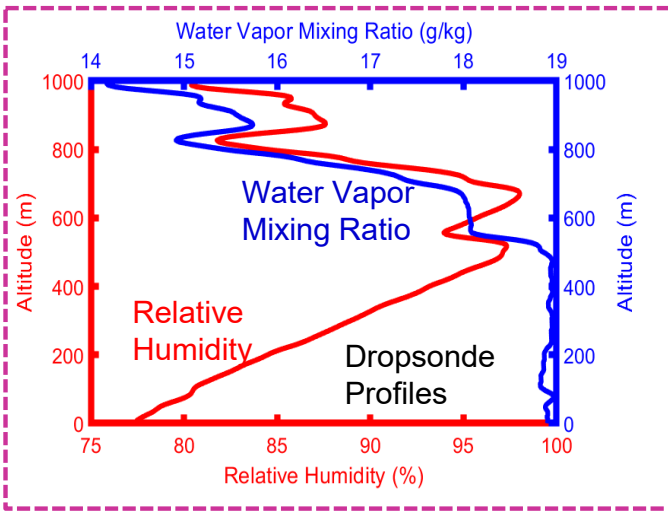
Nonspherical, Depolarizing Sea Salt can lead to Possible CALIOP overestimates of AOD and Aerosol Extinction



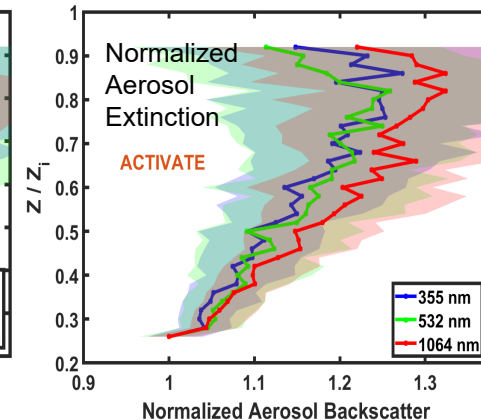
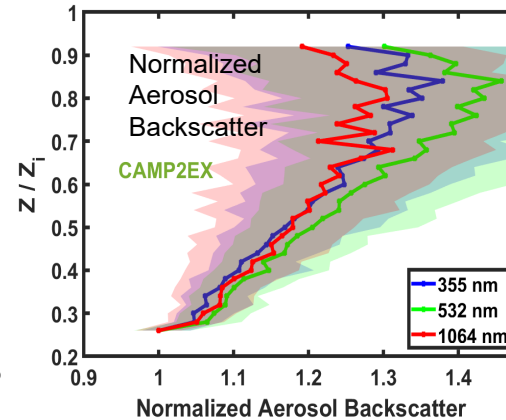
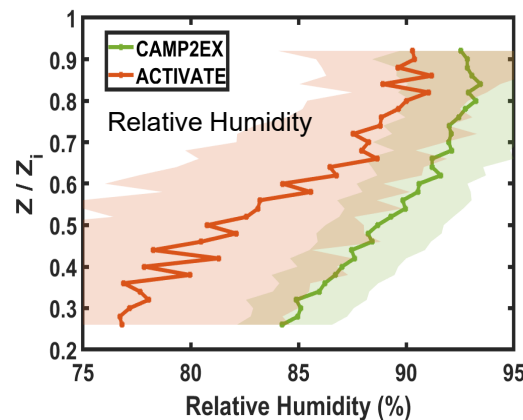
- If CALIOP detects elevated depolarization, the aerosol is classified as dusty marine, polluted dust, or desert dust depending on altitude and location
- This classification specifies the aerosol extinction/backscatter ratio (i.e. lidar ratio) which is used by the CALIOP operational algorithms to compute aerosol backscatter, extinction, and aerosol optical depth (AOD)
- The dusty marine, polluted dust, and desert dust have lidar ratios much greater than marine (37, 55, 44 vs. 23 at 532 nm) so misclassification of sea salt as dust will lead to significant high biases in aerosol backscatter, extinction, and (AOD)
- Examination of CALIOP measurements during several Cold Air Outbreak episodes (see images to right for March 9, 2020 case in ACTIVATE region) reveal that CALIOP also measured enhanced depolarization during such cases and that these typically also had low (<60%) RH – suggests possible (probable?) misclassification and overestimates of AOD and aerosol extinction



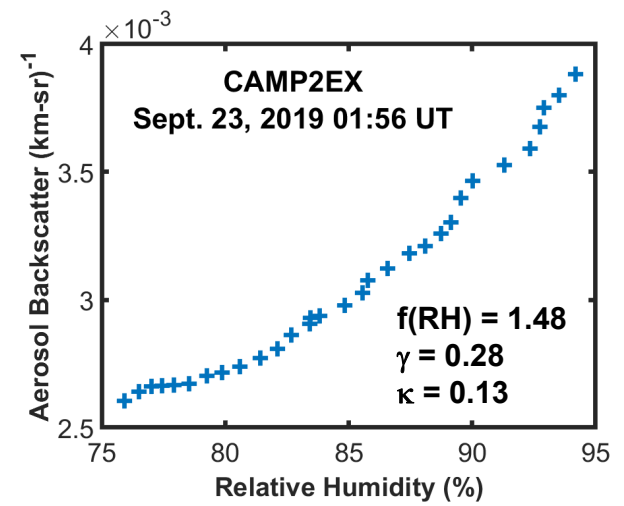
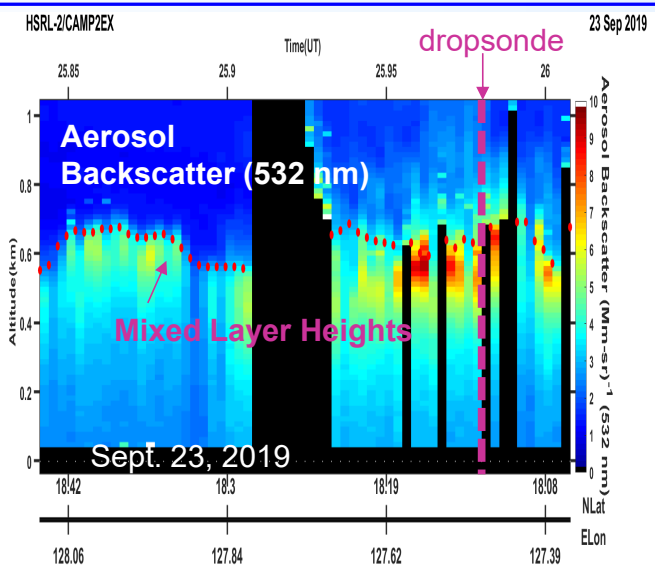
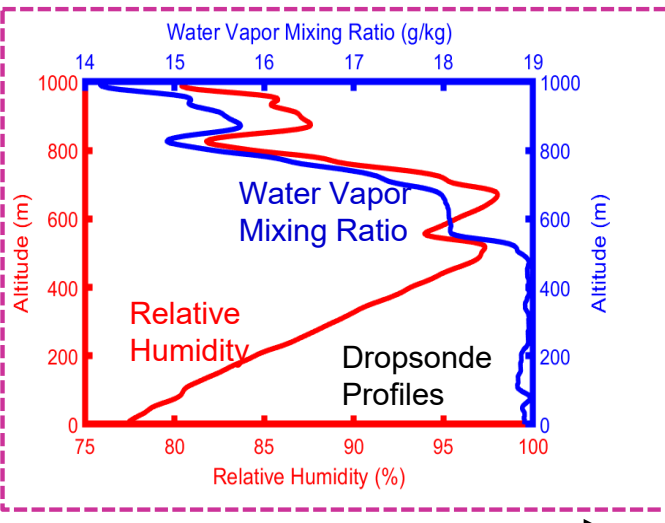
HSRL-2 Measures Increases in Aerosol Backscatter and Extinction within Mixed Layer Due to Increase in Relative Humidity (RH)



- Dropsondes provide relative humidity profiles
- Mixed Layer Height (Z_i) derived from HSRL2 aerosol backscatter profiles
- Restrict cases to nearly constant mixing ratio to select cases where aerosol properties vary with RH and not due to changes in concentration
- As RH increases with height within Mixed Layer, fine mode hygroscopic particles take on water, so aerosol backscatter and extinction increase.
- These plots show behavior averaged over all of CAMP2Ex and ACTIVATE



Quantifying the Aerosol Enhancement Factors Associated with the Increase in Relative Humidity (RH) using HSRL-2 and Dropsondes



- As RH increases with height within Mixed Layer, fine mode hygroscopic particles take on water, so aerosol backscatter and extinction increase.
- To quantify this increase, we compute **aerosol enhancement factor** $f(RH)$, **gamma** (γ), **kappa** (κ) within the mixed layer (i.e. $Z/Z_i < 1$)
- Aerosol backscatter profiles from HSRL2
- RH profiles from dropsondes
- Mixed Layer Height (Z_i) derived from HSRL2 aerosol backscatter profiles
- Restrict cases to nearly constant water vapor mixing ratio so aerosol properties vary with RH and not due to changes in concentration
- Values in the comparisons are for $f(RH=80\%/RH=20\%)$

$$f(RH) = \frac{\beta(RH)}{\beta(RH_0)} = \left[\frac{(100 - RH_0)}{(100 - RH)} \right]^\gamma$$

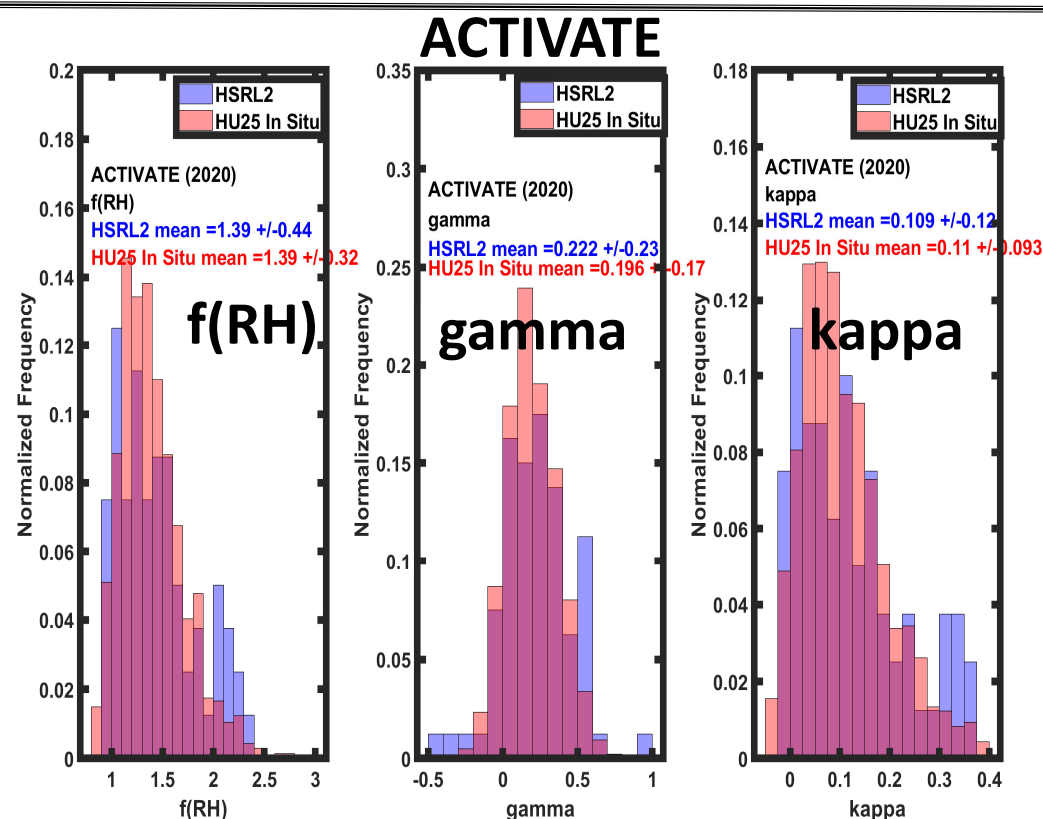
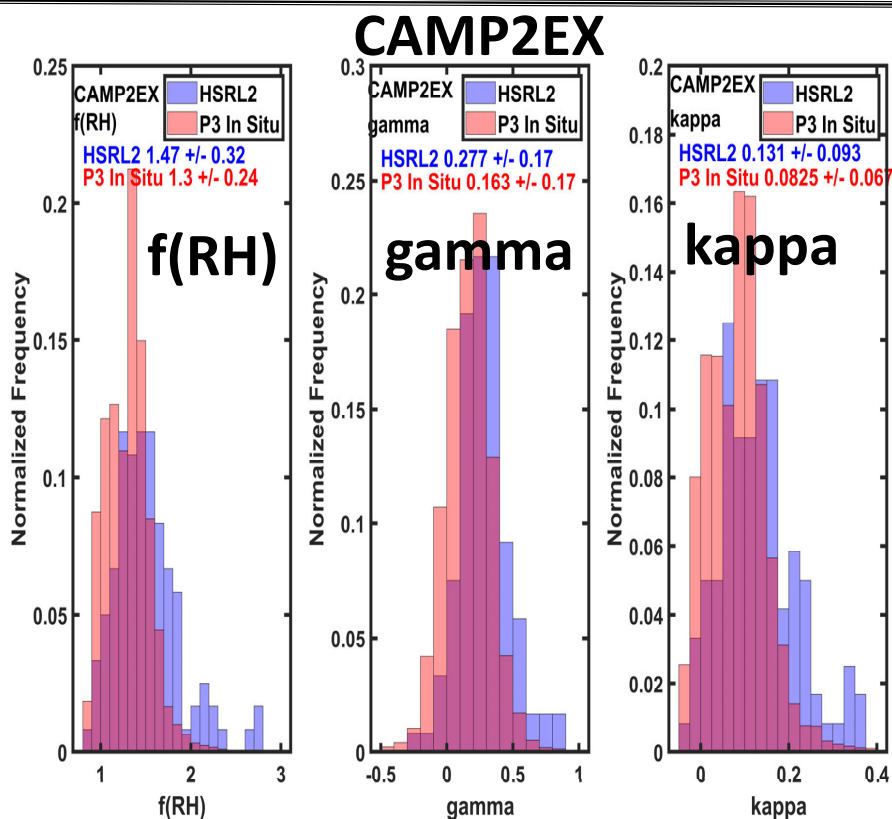
$$\cong 1 + \kappa_{bsc} \left[\frac{RH}{100 - RH} \right]$$

- $f(RH)$, γ , κ (HSRL-2) for aerosol backscatter and extinction are similar

HSRL-2 and In Situ Aerosol Enhancement Factors Show Increase in Aerosol Scattering with Increase in Relative Humidity (RH)



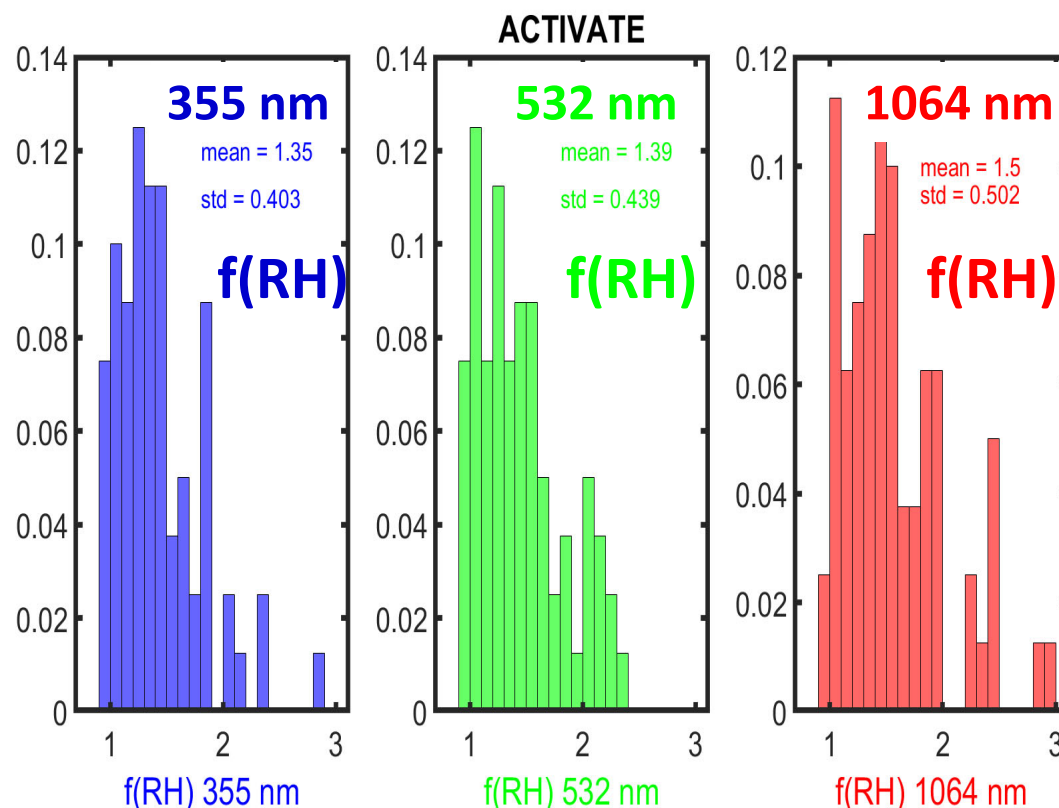
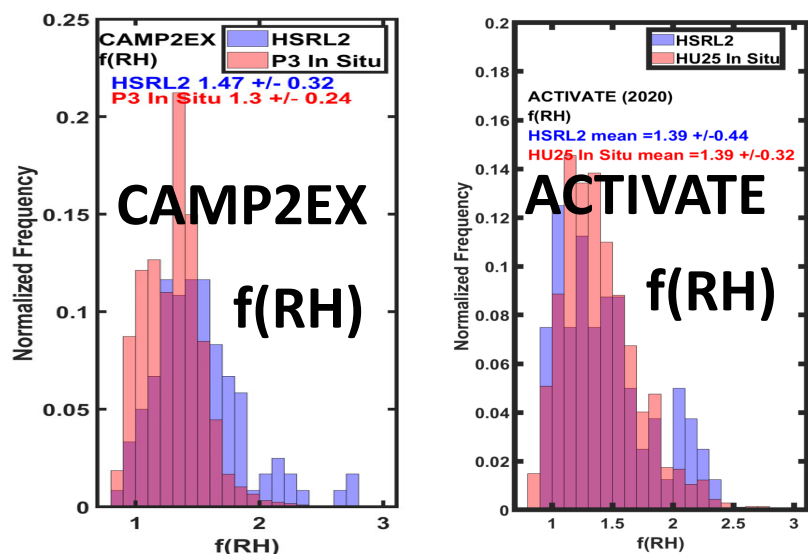
- In situ enhancement factors derived from humidified nephelometer, predominantly for submicron aerosol
- During CAMP2Ex and ACTIVATE, HSRL2 and in situ enhancement values are similar



Aerosol Enhancement Factors Measured by HSRL-2 During CAMP2EX and ACTIVATE



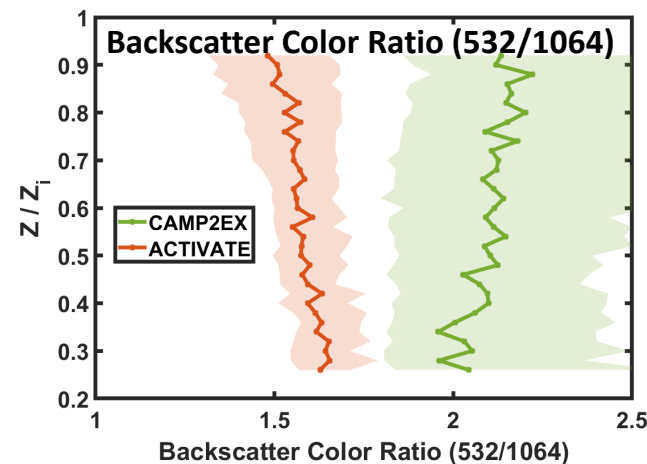
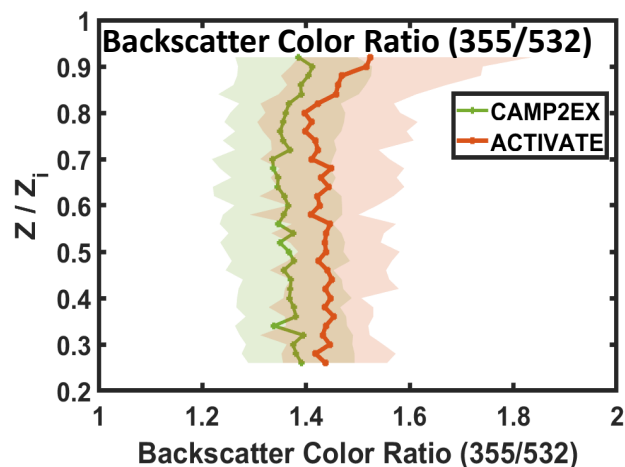
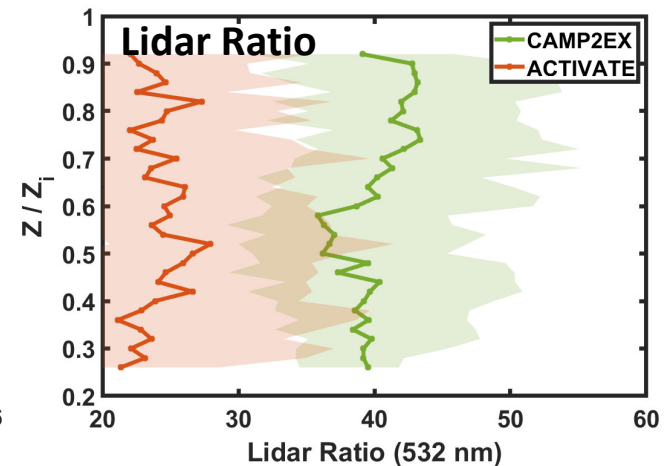
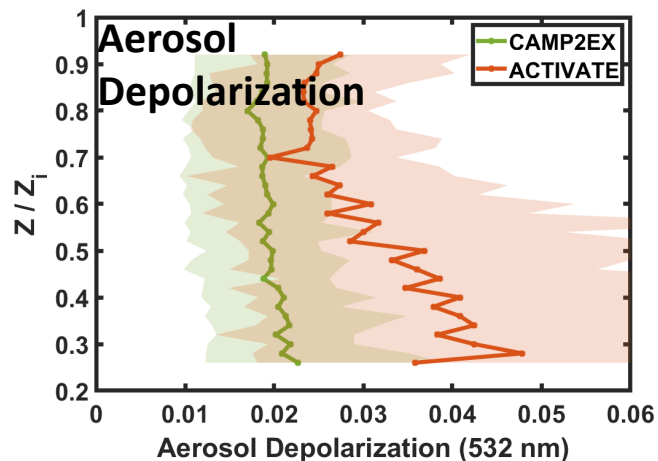
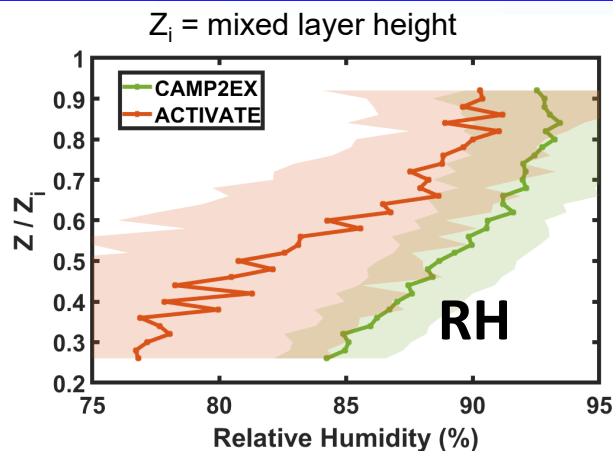
- HSRL2 & in situ $f(\text{RH})$ values are relatively low; appear consistent with biogenic/background aerosol
- HSRL-2 $f(\text{RH})$ increases with wavelength, perhaps associated with larger sea salt particles observed during ACTIVATE



Shingler et al., JGR, 2016 (in situ)

$f(\text{RH}=80\%)$	1.08 ± 0.13	0.99 ± 0.06	1.41 ± 0.13
	BB: Agric.	BB: Wildfires	Biogenic
	1.86 ± 0.36	1.64 ± 0.19	1.41 ± 0.20
	Marine	Urban	Background
			Free Trop.

Median Variability of Aerosol Intensive Parameters with Relative Humidity Within the Mixed Layer



Entire CAMP2EX and ACTIVATE datasets.

Overall, (on average), as RH increases near the top of mixed layer:

- Slight decrease in aerosol depolarization as particles become more spherical; also due in part to low near-surface RH during ACTIVATE
- Little change in color ratios and lidar ratios with RH suggest relatively little change in size

Summary



- Airborne HSRL-2 measurements acquired over the western North Atlantic Ocean during several winter and summer ACTIVATE flights revealed enhanced (>15-20%) particulate linear depolarization in the lowest levels of the marine boundary layer. These cases were associated with Cold Air Outbreaks.
- The strong correlation of elevated depolarization with low (<60%) relative humidity (RH), low (20-25 sr at 532 nm) aerosol extinction/backscatter (i.e. lidar) ratio, coincident airborne in situ size and composition measurements, and aerosol transport models indicate that the elevated depolarization is associated with crystalline sea salt, consistent with previous lidar observations.
- HSRL-2 measurements during CAMP2EX and ACTIVATE show increased aerosol backscatter and extinction near the top of the mixed layer associated with high relative humidity.
- Enhancement factors (e.g. $f(\text{RH})$) quantifying this increase in aerosol backscatter and extinction derived using the HSRL-2 measurements are similar to aerosol scattering enhancement factors derived from coincident airborne in situ data.
- These enhancement factors appear to be more consistent with biogenic aerosols than marine aerosols.
- On average, HSRL-2 measurements of lidar ratios and backscatter and extinction color ratios generally show little change with relative humidity.