Airborne HSRL-2 Measurements of Non-Spherical Sea Salt and Implications for CALIOP Aerosol Retrievals

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New ROSES CALIPSO Project
“Assessing and Improving CALIPSO Aerosol Optical Properties”

Ongoing Work as Part of this Project

- Investigate CALIOP aerosol classification and lidar ratios using airborne HSRL measurements
  - Airborne HSRL provides direct measurements of AOD and lidar ratio and aerosol intensive parameters for inferring aerosol type
  - **Assessments include aerosol type (e.g. dust, sea salt) as well as lidar ratio – focus of this presentation**
- Assessments of CALIOP AOD and lidar ratios derived from constrained retrievals
  - AOD constraints from passive sensors (e.g. MODIS, PARASOL) and surface reflectance (e.g. SODA, ODCOD)
  - Lidar ratio recommendations to account for regional and seasonal variability
- Improve Aerosol Classification using Constrained Retrievals

➢ **These activities take advantage of the extensive airborne HSRL measurements (i.e. about 150 airborne HSRL underflights of CALIOP between 2006-2022)**

Relevance

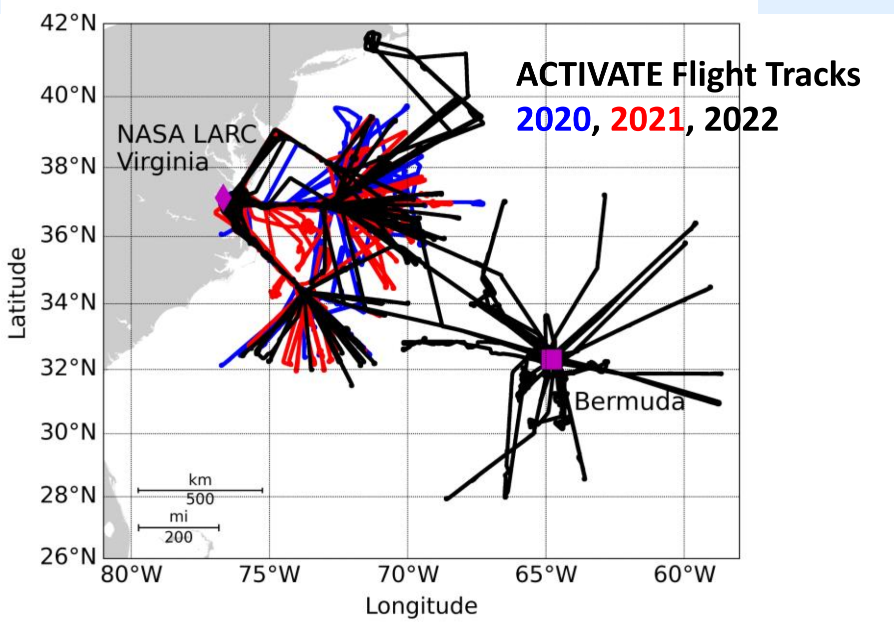
- **CALIOP classification specifies the lidar ratio used by the operational algorithms to compute aerosol backscatter, extinction, AOD**
- **If CALIOP detects elevated depolarization, the aerosol is classified as dusty marine, polluted dust, or desert dust depending on altitude and location**
- **Dusty marine, polluted dust, and desert dust have higher lidar ratios 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**

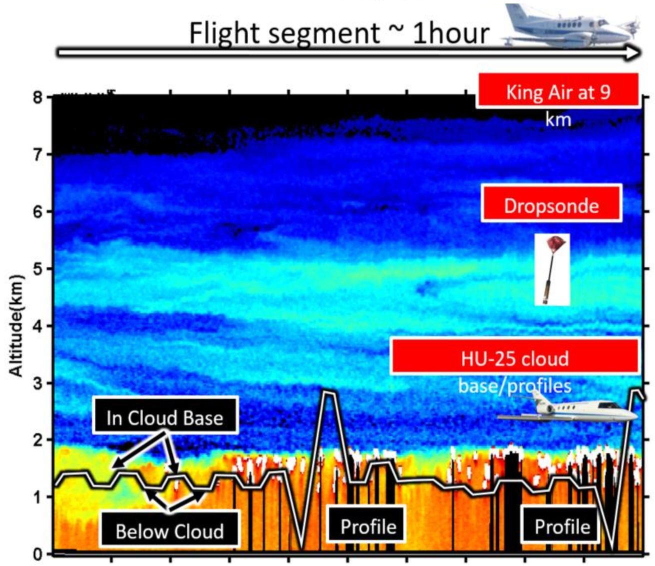
Aerosol Subtype	Version 4 Lidar Ratio (sr)
Biomass Burning	70
Clean Continental	53
Clean Marine	23
Dust	44
Dusty Marine	37
Polluted Continental	70
Polluted Dust	55

HSRL-2 Data Acquired During NASA EVS-3 ACTIVATE Field Missions

NASA EVS-3 ACTIVATE (Feb-Mar, Aug-Sep 2020; Jan-Jun, Dec 2021; Jan-Jun 2022; data used here from 2020-2021)

- 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 off the US Mid-Atlantic Coast
- NASA LaRC HSRL-2 deployed on LaRC King Air aircraft for nadir viewing measurements
- Dropsondes deployed from LaRC King Air aircraft
- NASA LaRC HU-25 Falcon aircraft simultaneously deployed in situ instruments to measure BL clouds and aerosols below King Air



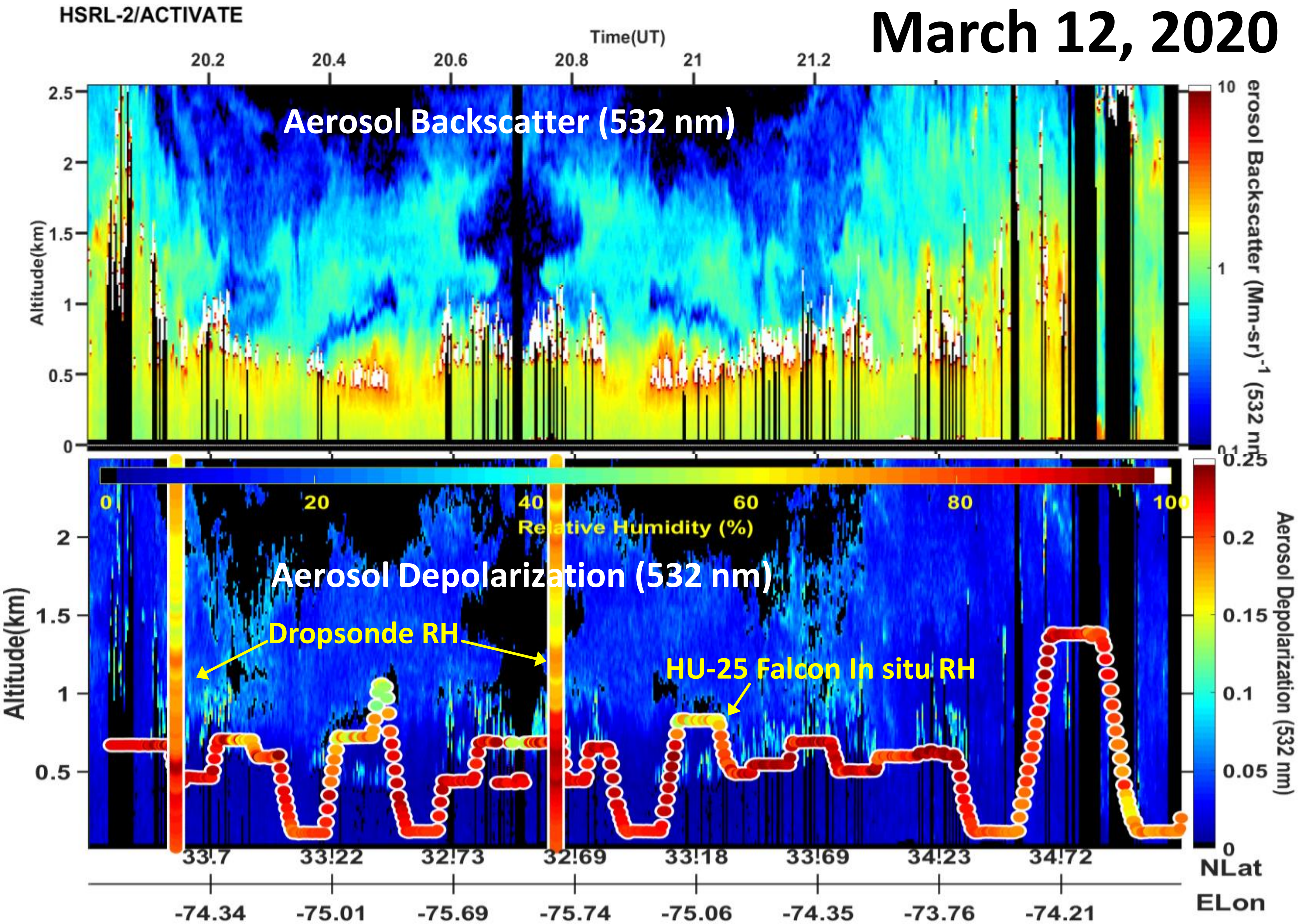


HSRL-2 Data Products

- Particulate backscatter and depolarization profiles (355, 532, 1064 nm)
- Particulate extinction profiles (355 and 532 nm)
- Aerosol Type
- Aerosol Optical Thickness
- Mixed Layer Heights
- Ocean products*

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

- Typically HSRL-2 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 dropsondes and airborne in situ (Diode Laser Hygrometer-DLH) measurements on the Falcon aircraft



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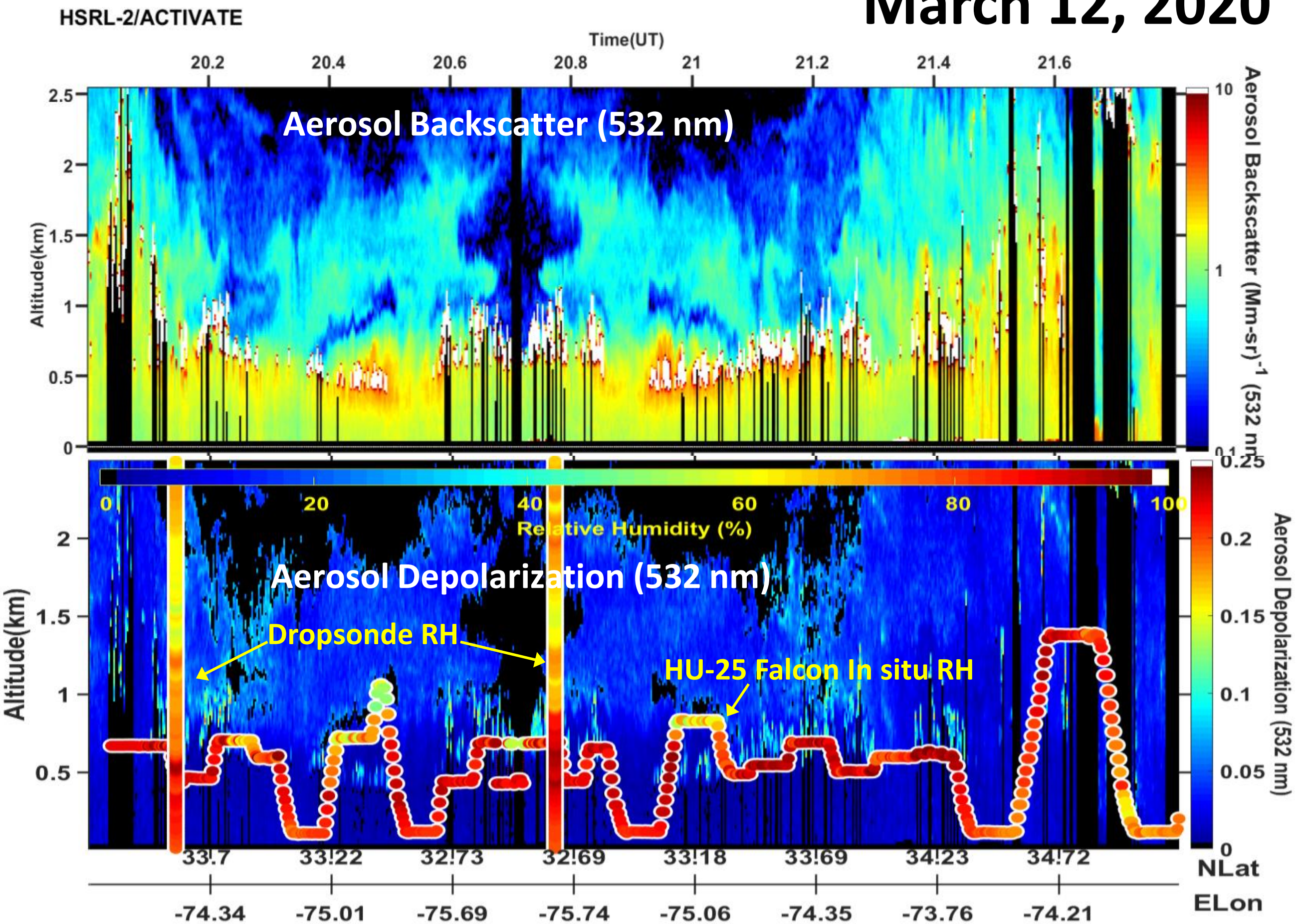
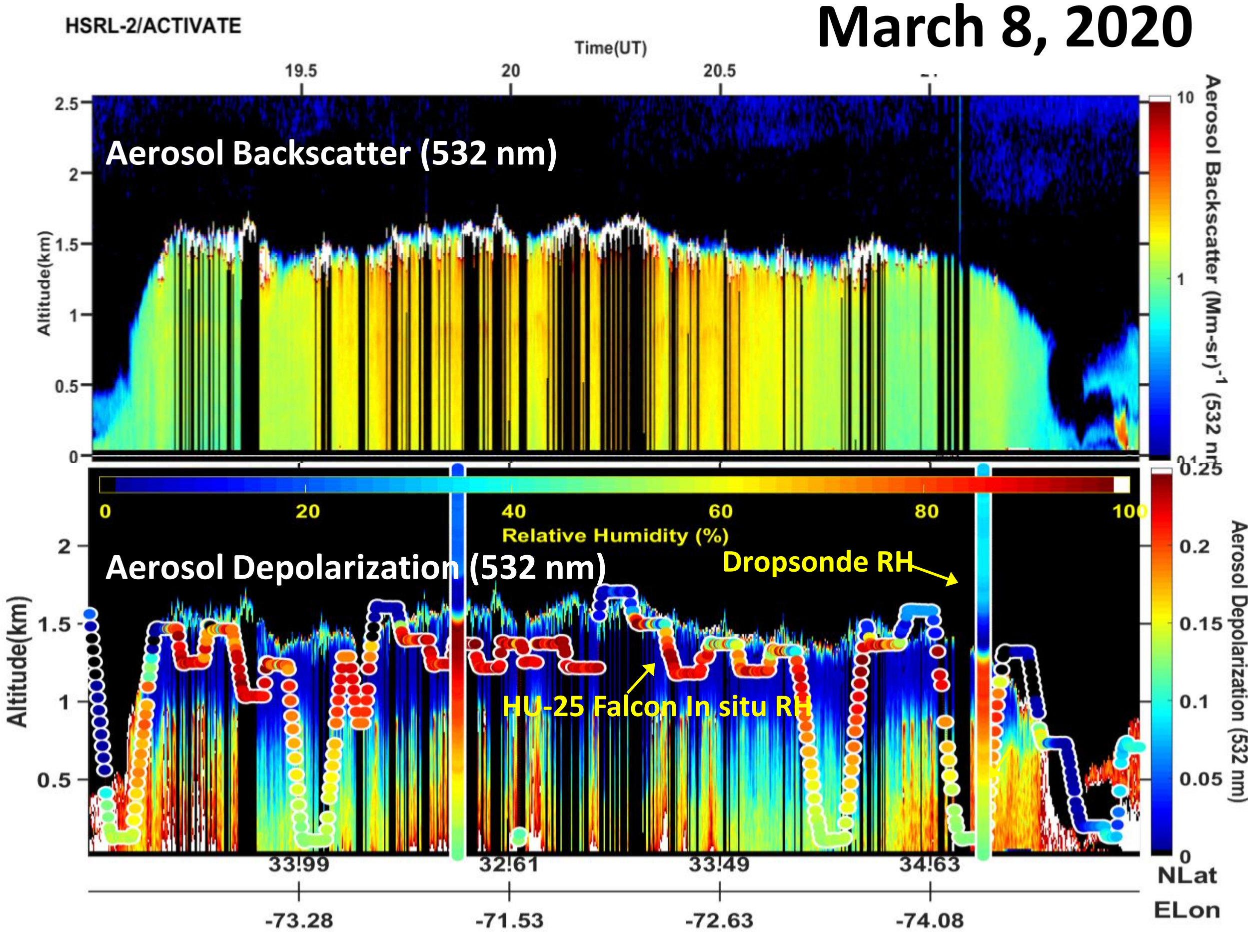
- Note the high (>65%) relative humidity (RH) in the lowest 1 km derived from airborne in situ (Diode Laser Hygrometer-DLH) measurements on the Falcon aircraft

- In contrast, during several ACTIVATE flights, HSRL-2 measured elevated (>10-15%) aerosol depolarization in the lowest 1 km. Note the lower (<60%) relative humidity (RH) derived from dropsondes and the airborne in situ DLH measurements on the Falcon aircraft

March 12, 2020

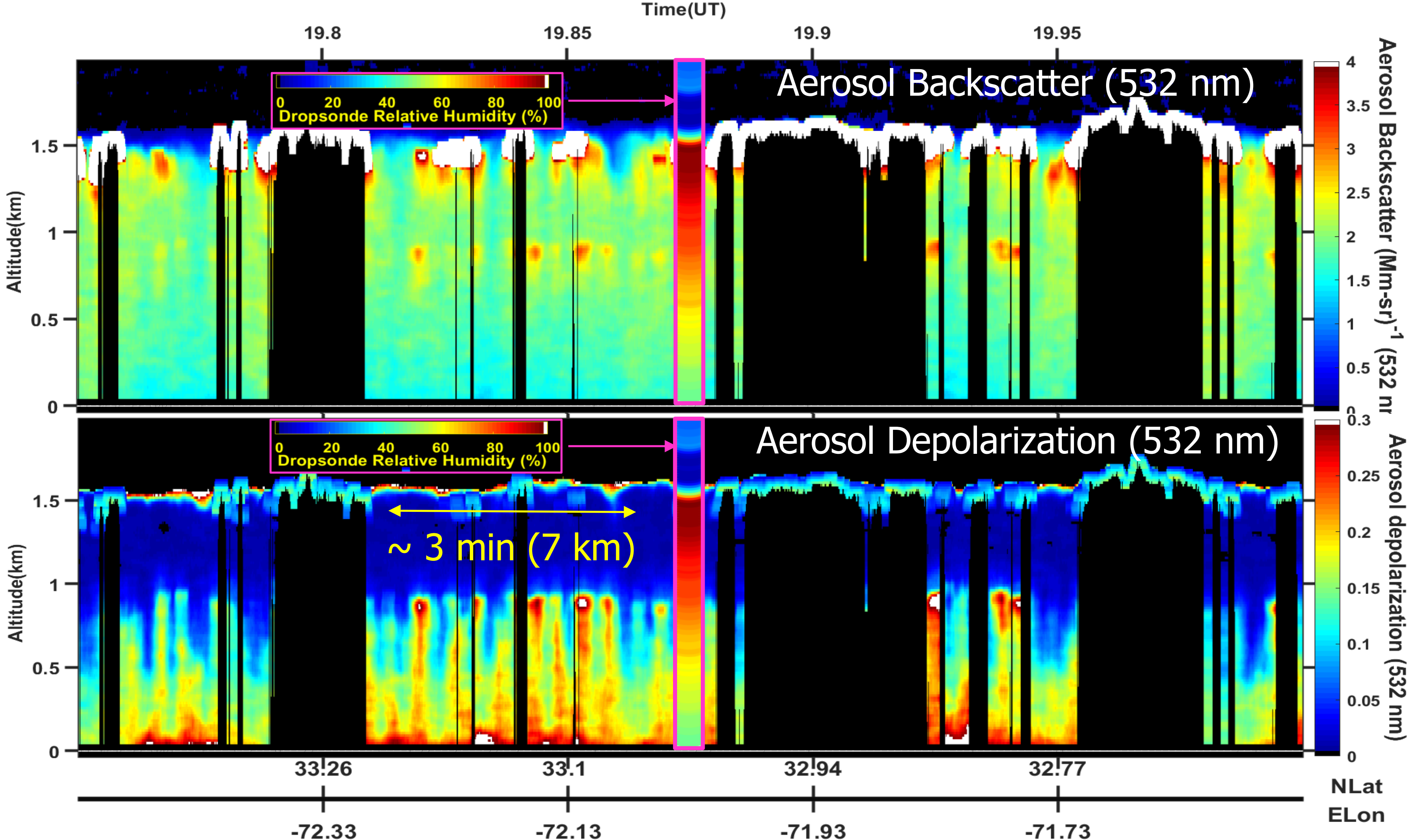


March 8, 2020

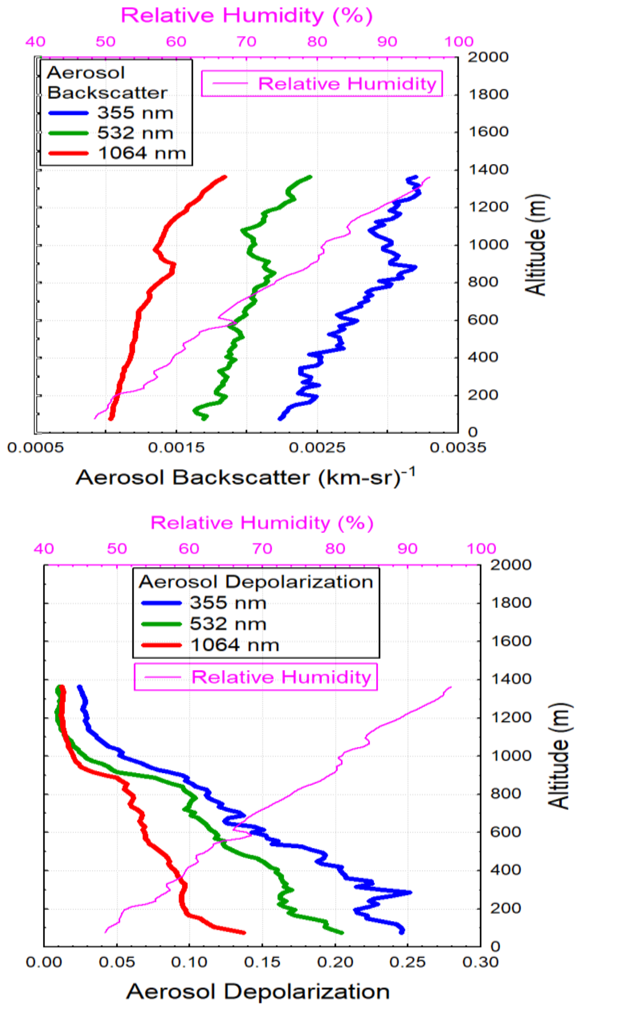


Airborne HSRL-2 data Reveals Spatial and Vertical Variability in Aerosol Depolarization

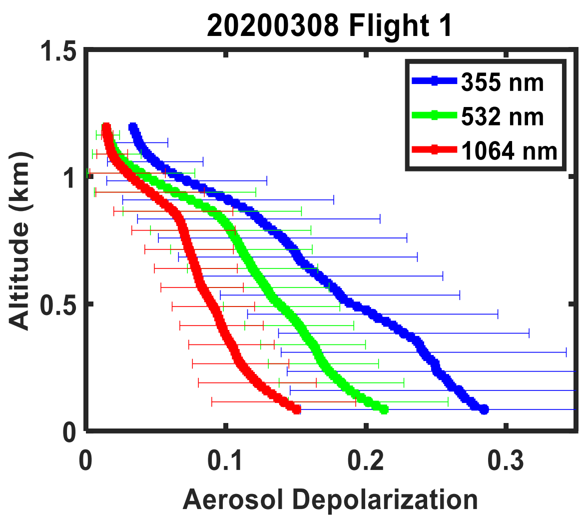
- Small scale (1.5-2 km) "plume-like" variability in the aerosol depolarization
- Abrupt decrease in depolarization as RH increases above about 75%
- Increase in aerosol backscatter near RH~75%

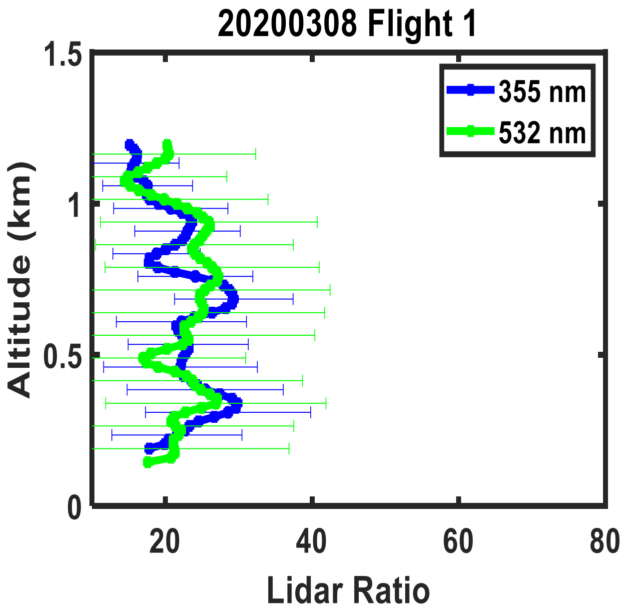


Profiles at dropsonde time (19:52 UT) March 8, 2020

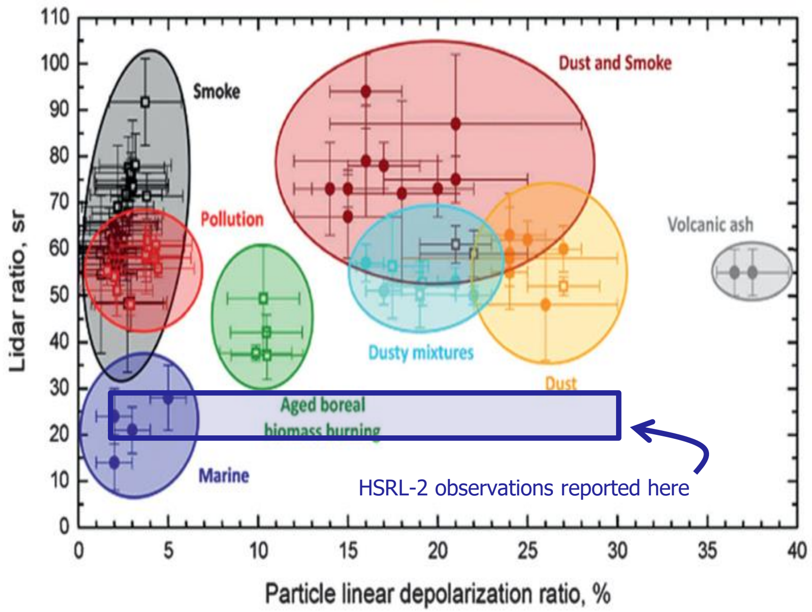


Elevated depolarization appears to be due to sea salt not dust



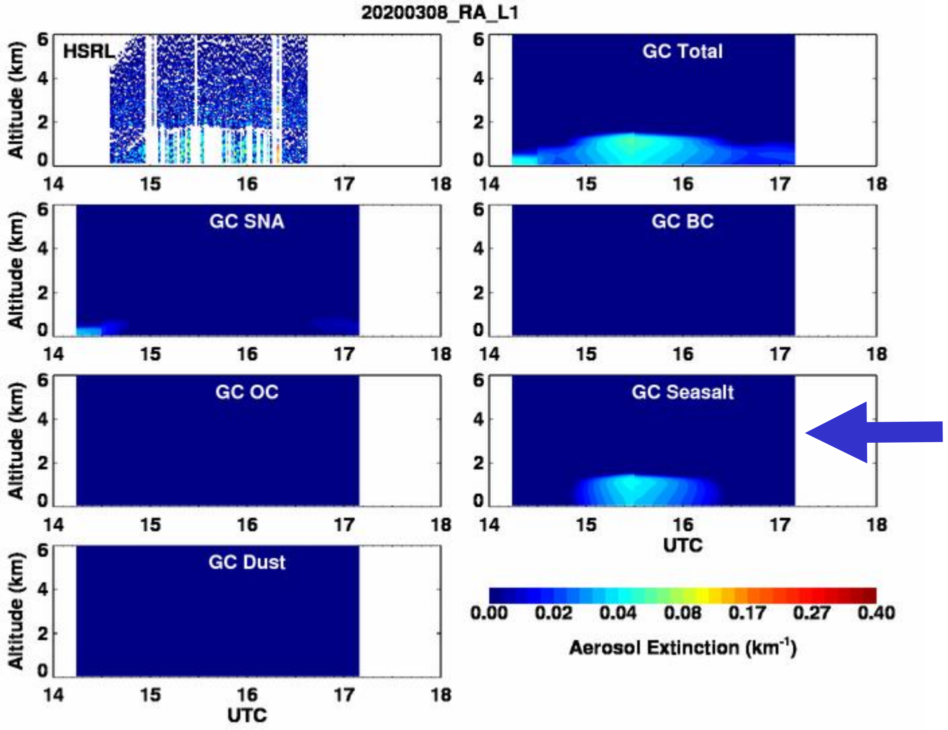


- Wavelength dependence of aerosol depolarization (355-532 nm) is inconsistent with dust
- Aerosol extinction/backscatter ratios (i.e. lidar ratios) were about 20-25 sr and consistent with marine (sea salt) aerosol not dust
- Backtrajectories do not support dust



Illingworth, A. J., and Coauthors, 2015: *Bull. Amer. Meteor. Soc.*, 96, 1311–1332, https://doi.org/10.1175/BAMS-D-12-00227.1. © American Meteorological Society. Used with permission.

➢ GEOS-Chem Model Identifies Sea Salt as Major Contributor to Aerosol Extinction During March 8 Flights



GEOS-Chem model aerosol extinction (550 nm) curtains along the track of the UC-12 King Air during flights on March 8, 2020. (SNA = sulfate + nitrate + ammonium, OC = organic carbon, BC = black carbon)

Phase transition between spherical sea salt drops and crystalline sea salt responsible for variability of particulate depolarization with RH

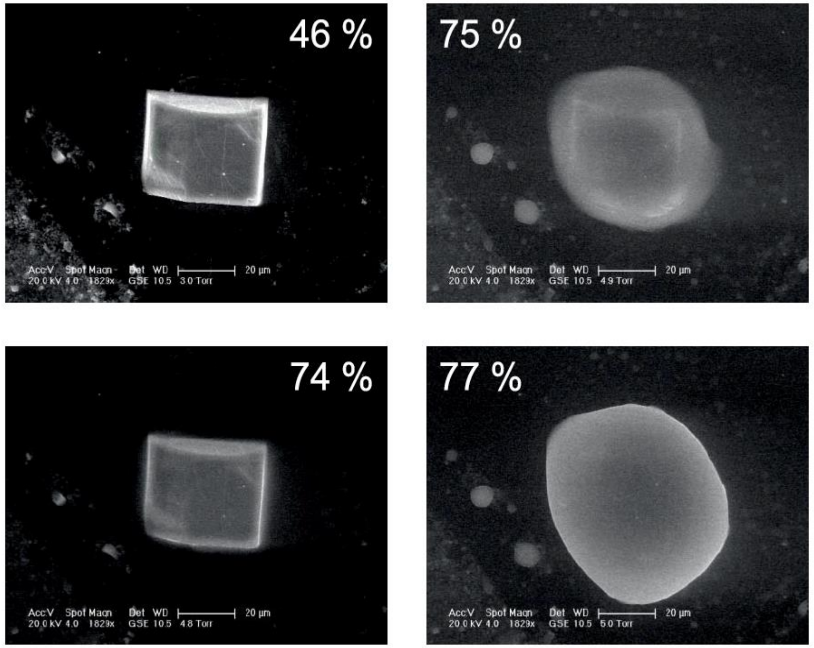


Figure 1. Sodium chloride deliquescence at 75 % RH observed at laboratory conditions (at 4.9 °C) in an environmental scanning electron microscope. The dry cubic particle with sharp edges at RH of 46 % becomes surrounded by a liquid sphere when RH increases to 77 %.

Figure 1. Sodium chloride deliquescence at 75 % RH observed at laboratory conditions (at 4.9 ◦C) in an environmental scanning electron microscope. The dry cubic particle with sharp edges at RH of 46 % becomes surrounded by a liquid sphere when RH increases to 77 %. (from Haarig, M., and Co-authors: Dry versus wet marine particle optical properties: RH dependence of depolarization ratio, backscatter, and extinction from multiwavelength lidar measurements during SALTRACE, Atmos. Chem. Phys., 17, 14199–14217, https://doi.org/10.5194/acp-17-14199-2017, 2017. (https://www.atmospheric-chemistry-and-physics.net/policies/licence_and_copyright.html, https://www.atmospheric-chemistry-and-physics.net/policies/licence_and_copyright.html, http://creativecommons.org/licenses/by/4.0/)

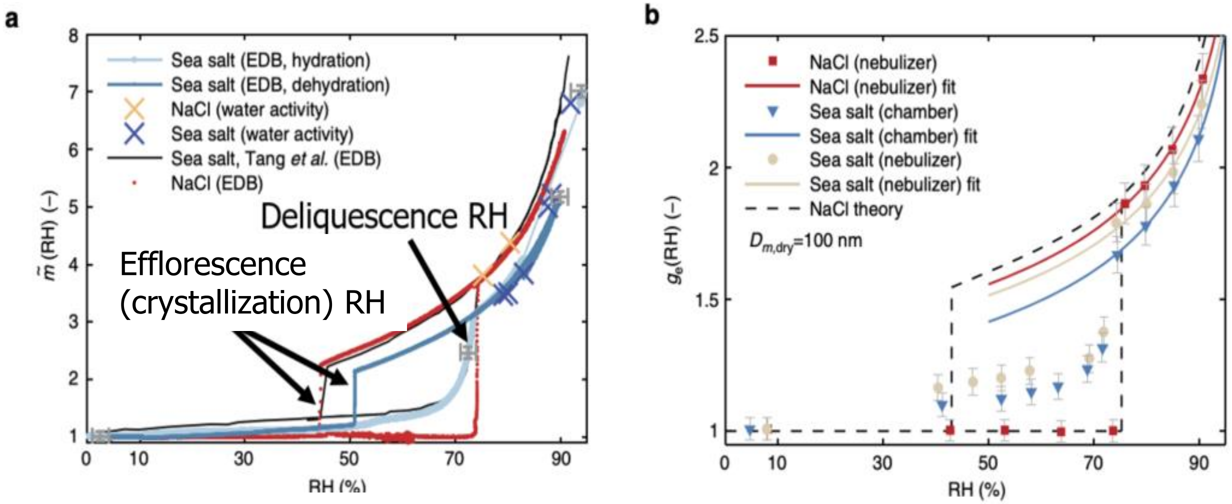


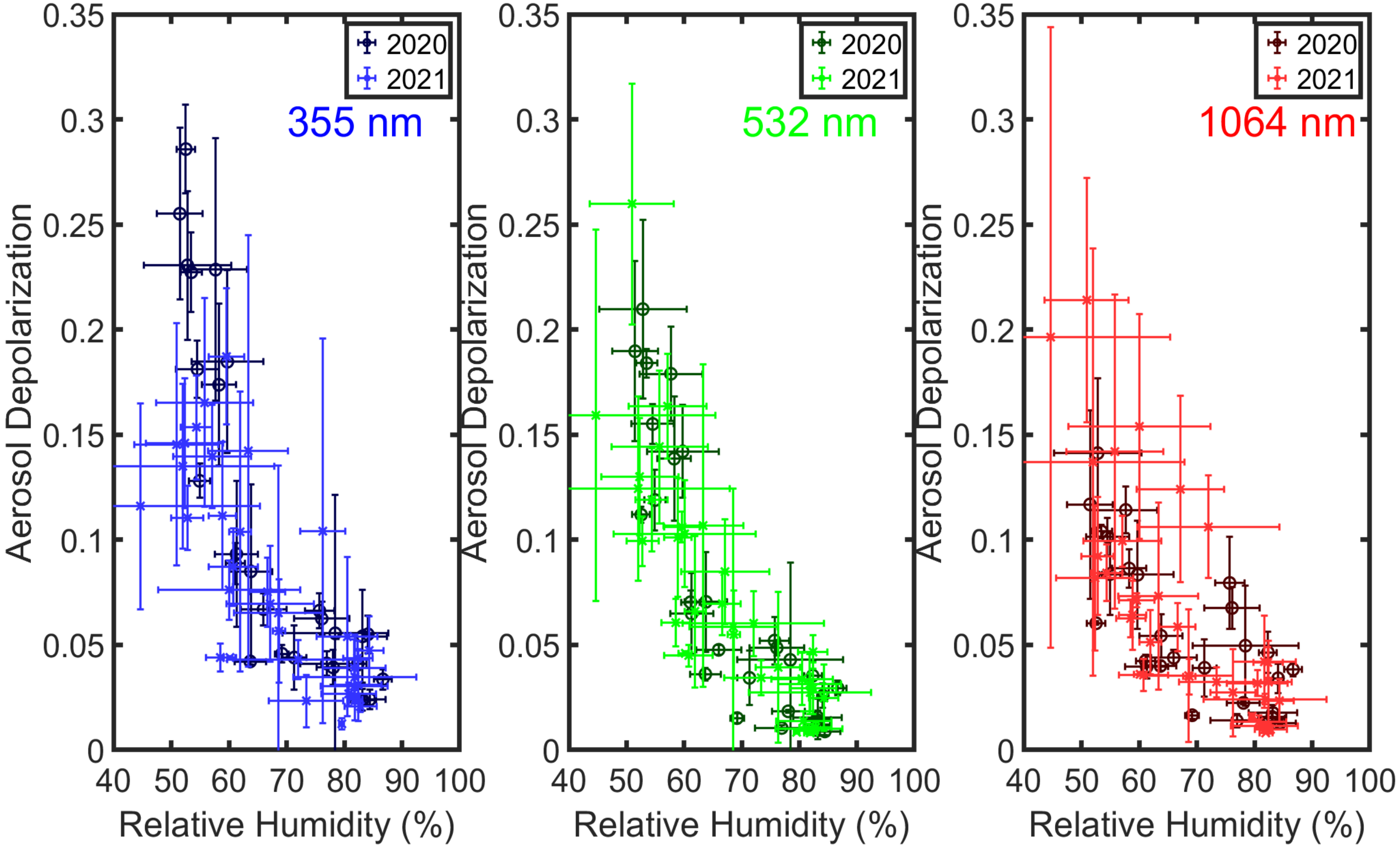
Figure 2 | Hygroscopicity measurements of inorganic sea salt and NaCl particles. (a) Mass growth factor versus RH for artificial sea salt and NaCl. (b) Hygroscopic growth factor versus RH at D(dry)=100 nm. (from Zieger, P., Väisänen, O., Corbin, J. *et al.* Revising the hygroscopicity of inorganic sea salt particles. *Nat Commun* **8,** 15883 (2017). See https://doi.org/10.1038/ncomms15883, https://www.nature.com/articles/ncomms15883#rightslink, https://creativecommons.org/licenses/by/4.0/)

- **Efflorescence (crystallization) for sea salt is at RH~51%**
- **Inorganic sea salt has multiple crystallization points due to complex composition**
- **Deliquescence occurs around RH~75%**
- **Hygroscopic growth during hydration for sea salt continually changes with RH (not a step function)**
- **Both shape modes can be present for 50≤RH≤70% due to hysteresis effect**

HSRL-2 Measurements During ACTIVATE Show Elevated Aerosol Depolarization when RH is below about 60%

- Highest aerosol depolarization is associated with RH below about 60%
- During 2020 and 2021, there were 63 days that had coincident HSRL2 and dropsonde data
  - 20 of these 63 days had observations of high depolarization (>10% at 532 nm) associated with low (<60%) RH
  - 12 (8) days occurred in winter (spring/summer)
- Depolarization decreases with increasing wavelength
- HSRL-2 measurements similar to previous lidar observations of nonspherical sea salt (Haarig et al., 2017)

- Average depolarization in lowest 20% of Mixed Layer coincident with dropsondes during 2020 and 2021
- Each point represents average of the HSRL-2 profiles at the dropsonde times for that day (63 days)

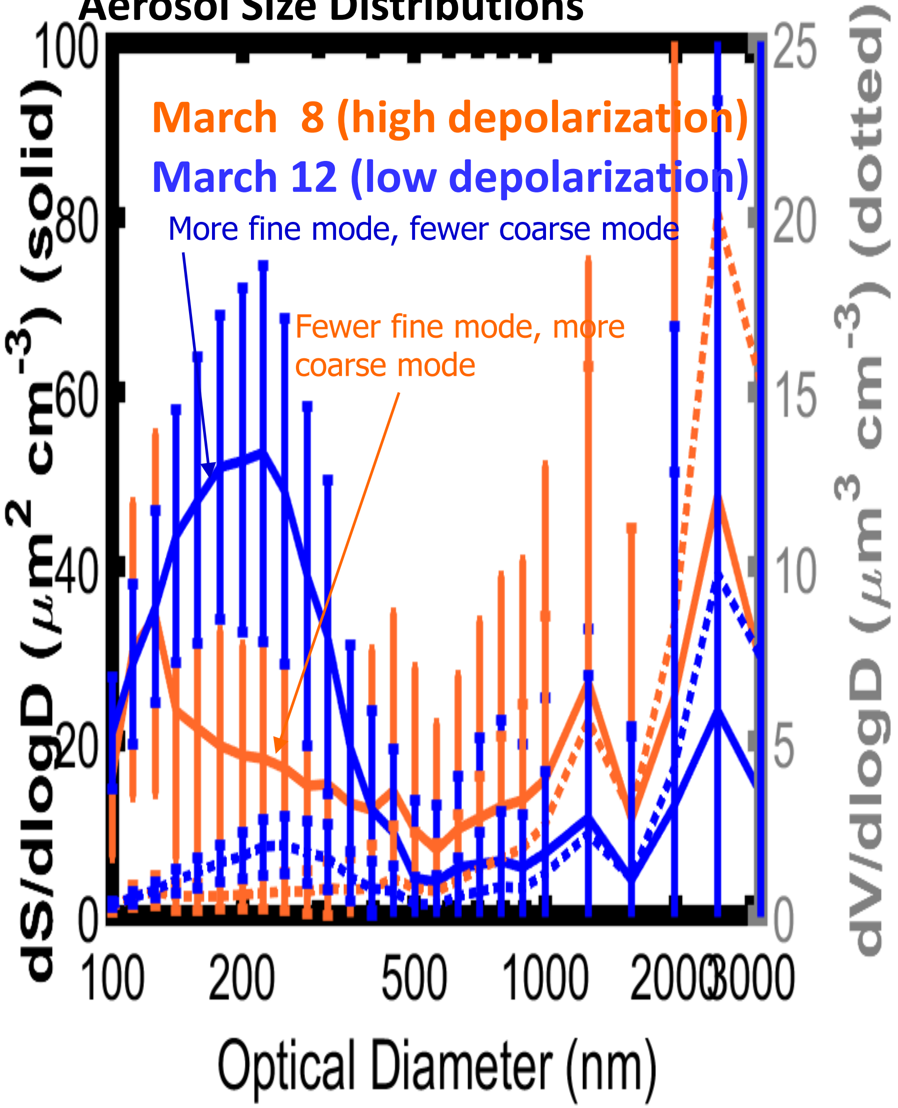


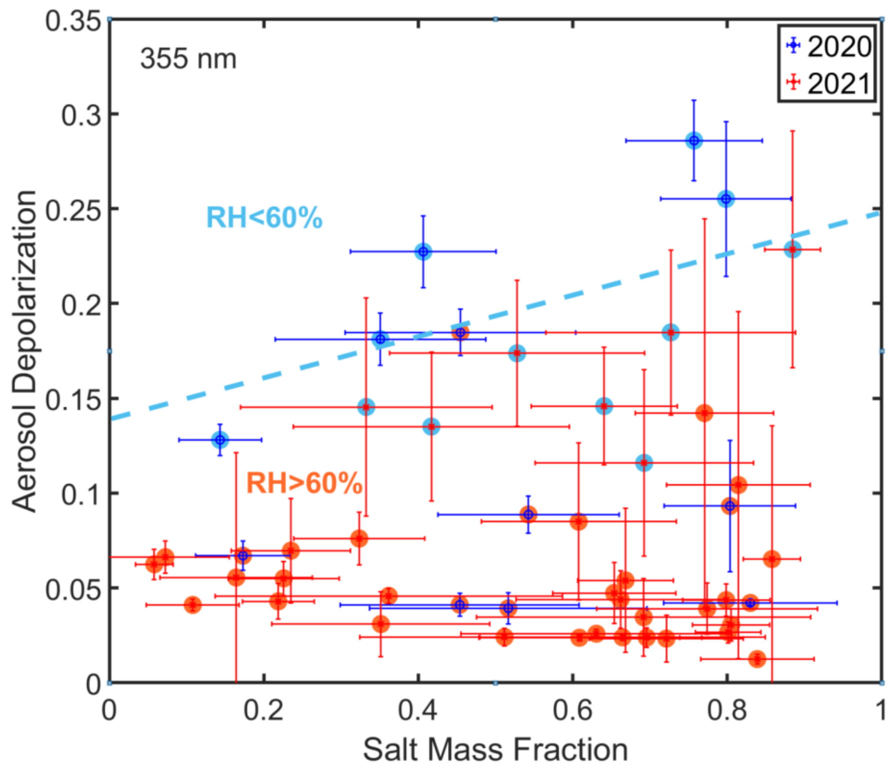
Airborne In situ Aerosol Size Distribution and Salt Mass Fraction Measurements Coincident with HSRL-2 Depolarization Measurements

- 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

In situ LAS Aerosol Surface and Volume Aerosol Size Distributions

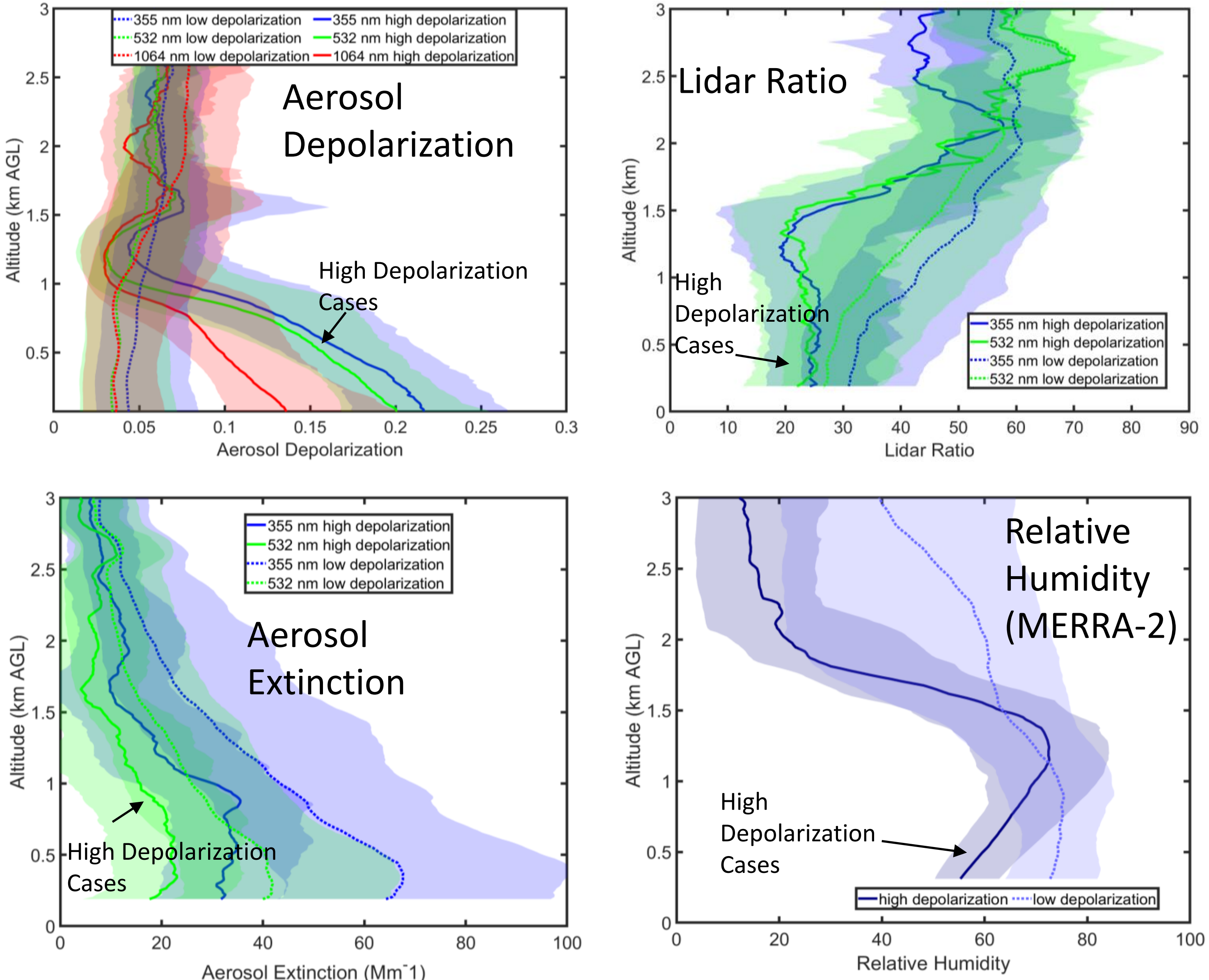




- Elevated depolarization was correlated with salt mass fraction for RH<60%
- Elevated depolarization was not correlated with wind speed regardless of RH

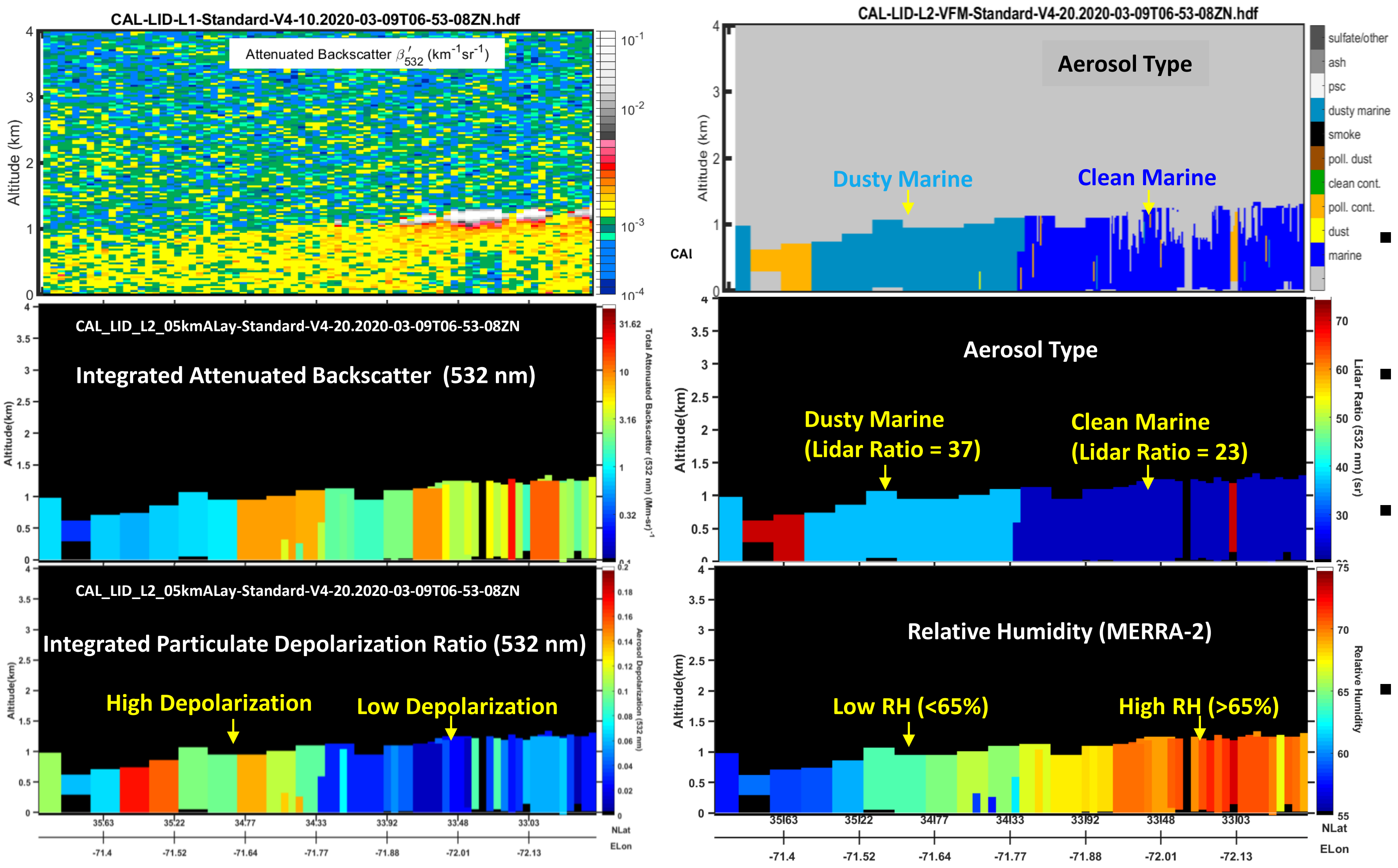
Lower Lidar Ratios, Aerosol Extinction, and AOD associated with the high depolarization, nonspherical sea salt cases

- All HSRL-2 data divided into high (solid lines) and low (dotted lines) near-surface depolarization cases
- For high near-surface depolarization cases:
  - Median lidar ratios at 355 and 532 nm were around 20-25 sr, consistent with marine (sea salt) aerosol
  - Median extinction (532 nm) was about 20 Mm^{-1} which is about half that for the low depolarization cases
  - MERRA-2 profiles show low (<60%) near-surface RH
- Higher lidar ratios observed when other aerosol types (e.g. smoke) were present within BL
- AOT (532 nm) contributed by the nonspherical sea salt particles was small (0.03-0.04)



Example of Probable CALIOP Misclassification of Sea Salt as Dusty Marine

March 9, 2020



- As an example, we examine CALIOP nighttime data on March 9, 2020, a day after HSRL-2 observed high aerosol depolarization associated with nonspherical sea salt in the same region
- Elevated aerosol depolarization led to classification of dusty marine (lidar ratio = 37)
- Lower aerosol depolarization led to classification as marine (lidar ratio = 23)
- The aerosol depolarization transition seems to correspond to transition from low to high RH (from MERRA-2 model)
- GEOS-Chem model indicates sea salt as the dominant aerosol type within the BL throughout this region

Frequency of CALIOP Dusty Marine type – Examination of Cold Air outbreaks (CAO) during Jan-Mar 2019

- Elevated depolarization associated with sea salt during ACTIVATE flights tended to occur during Cold Air Outbreaks (CAO)
- To investigate the frequency of CALIOP observations of elevated depolarization associated with these aerosols, CALIOP observations during 15 CAO events in January-March, 2019 were examined
- Of the 15 CAOs visually identified, each included some aerosol that was classified as 'dusty marine'.
- Were these misclassified as 'dusty marine' instead of 'marine'?
  - in 3 cases, air above 2km includes dust (one in Gulf of Mexico)
  - in 12 cases, no aerosol identified above cloud, suggesting little higher level dust transport off of the continent
  - In most of these cases, ERA-5 model indicated drier (RH<70%) air near the surface

➢ Results suggest that the CALIOP operational algorithm sometimes misclassifies depolarizing sea salt as "dusty marine" aerosol

How can such misclassification and associated aerosol retrieval bias be avoided?

- Aerosol depolarization spatial and vertical variability highly correlated with RH variability may provide an indication of nonspherical sea salt
- Spectral depolarization ratio (532-1064 nm) and backscatter color ratio (532-1064 nm) probably won't work reliably since nonspherical sea salt and dust can have similar variabilities
- If the AOD from overlying aerosols is negligible, then column AOD constraints from passive sensors (e.g. MODIS, Polarimeter, GOES) or ocean surface reflection (e.g. ODCOD, SODA) may provide better estimates of lidar ratio
- Models and backtrajectories can provide guidance as to the presence of sea salt vs. dust
- Direct HSRL measurements of the lidar ratio (e.g. ATLID on EarthCARE, Clio on AOS) could distinguish nonspherical sea salt and dust (with sufficient spatial and vertical averaging)

…Coming Attraction…

- HSRL-2 data clearly show aerosol depolarization varies with relative humidity
- How do aerosol backscatter and extinction vary with relative humidity?

➢ **See our Fall AGU presentation "Aerosol Humidification Observed by the Airborne High Spectral Resolution Lidar-2" in Session A073-Models, In situ, and Remote Sensing on Aerosols)**