



Airborne High Spectral Resolution Lidar-2 Measurements of Enhanced Depolarization Associated with Nonspherical Sea Salt

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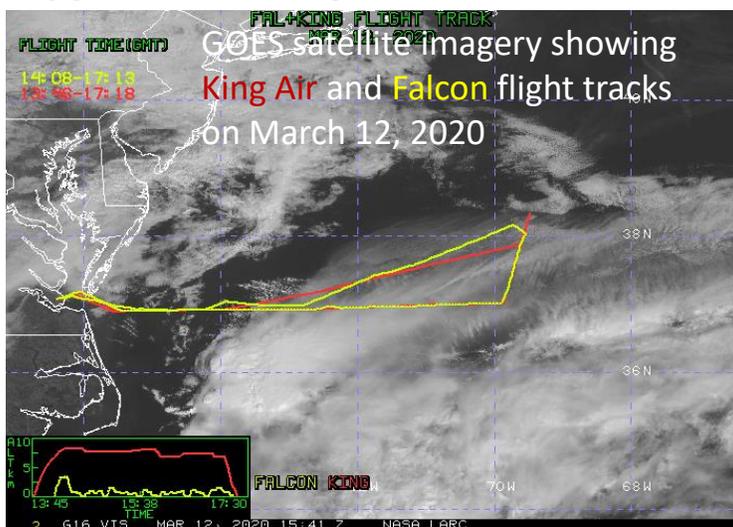
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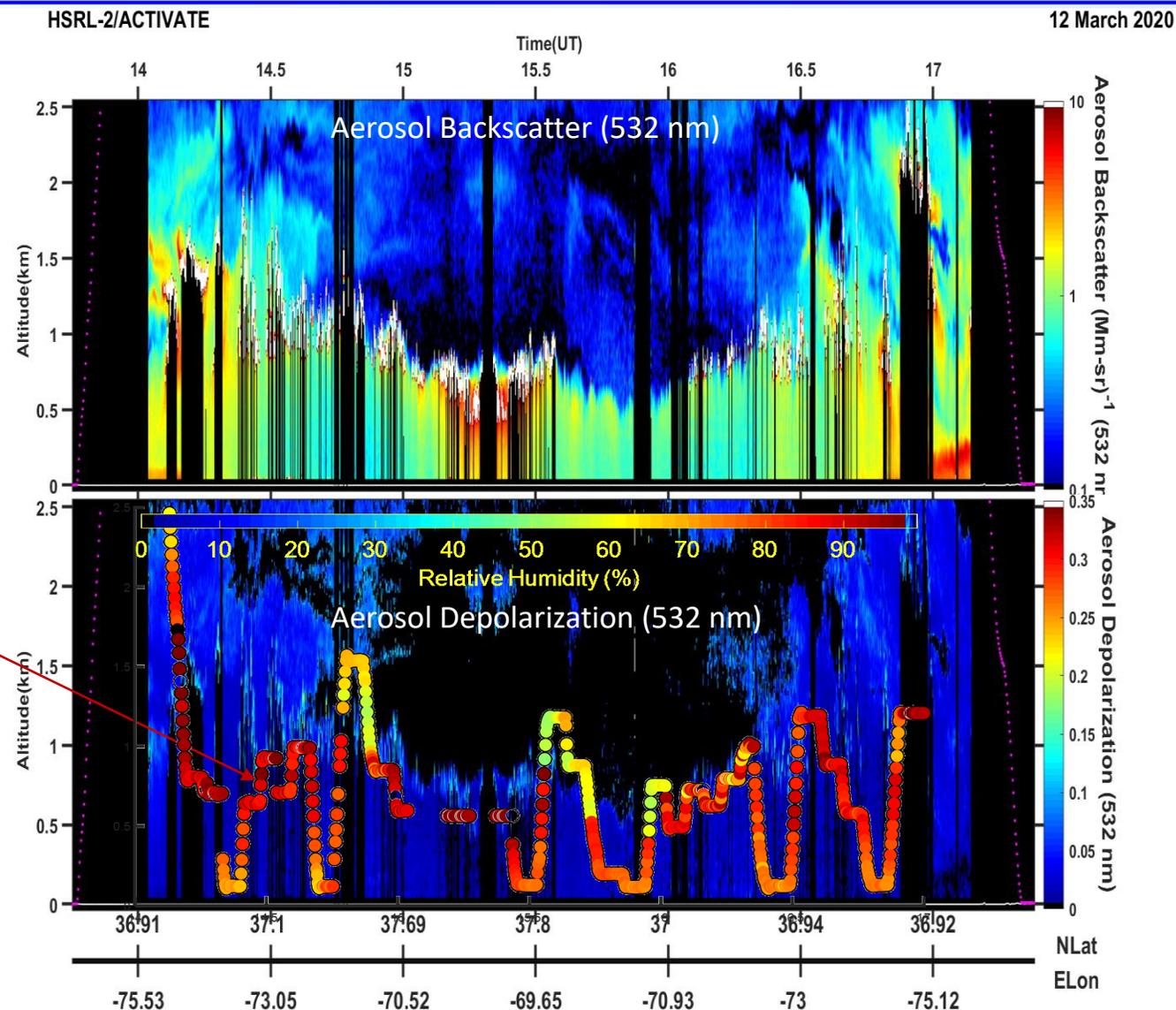
Background – Airborne Measurements of Low Aerosol Depolarization Over the Ocean



- Typically HSRL2 measures low aerosol depolarization over the ocean. This is associated with sea salt aerosols
- Low depolarization is associated with spherical particles
- High depolarization is associated with nonspherical particles
- This example from an ACTIVATE flight on March 12, 2020 shows HSRL2 measured low (<5%) aerosol depolarization in the lowest 1 km
- 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



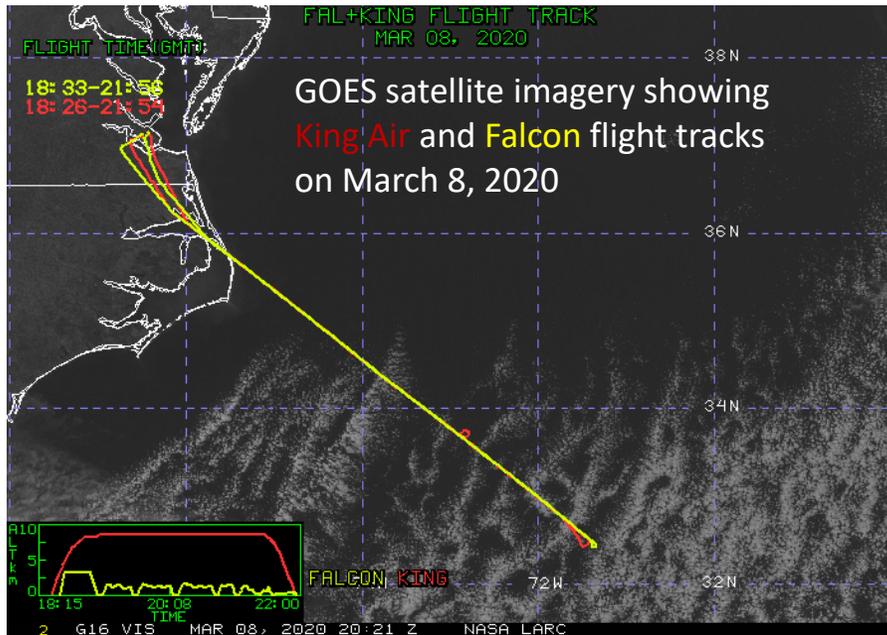
NASA Langley's Satellite Cloud and Radiation Property Retrieval System (SatCORPS)



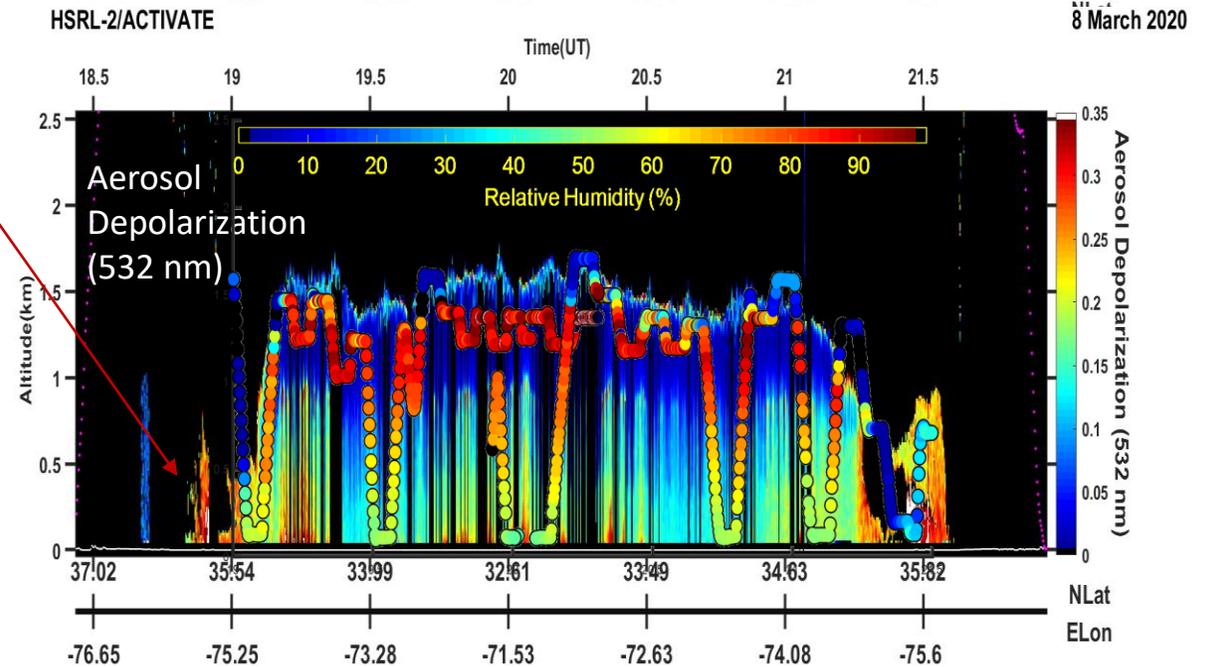
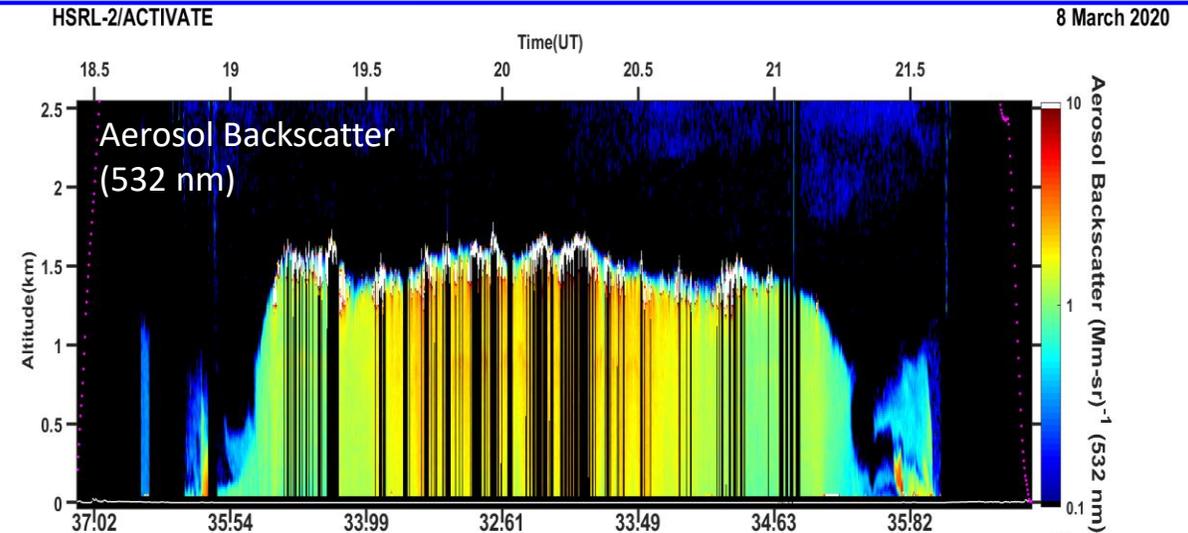
HSRL2 Observations of Enhanced Depolarization over the Ocean



- In contrast, during several (10-12) ACTIVATE flights during the winter and summer campaigns, HSRL2 measured elevated (>10-15%) aerosol depolarization in the lowest 1 km. This shows an example from a flight on March 8, 2020.
- Note the lower (<60%) relative humidity (RH) derived from the airborne in situ (Diode Laser Hygrometer-DLH) measurements on the Falcon aircraft



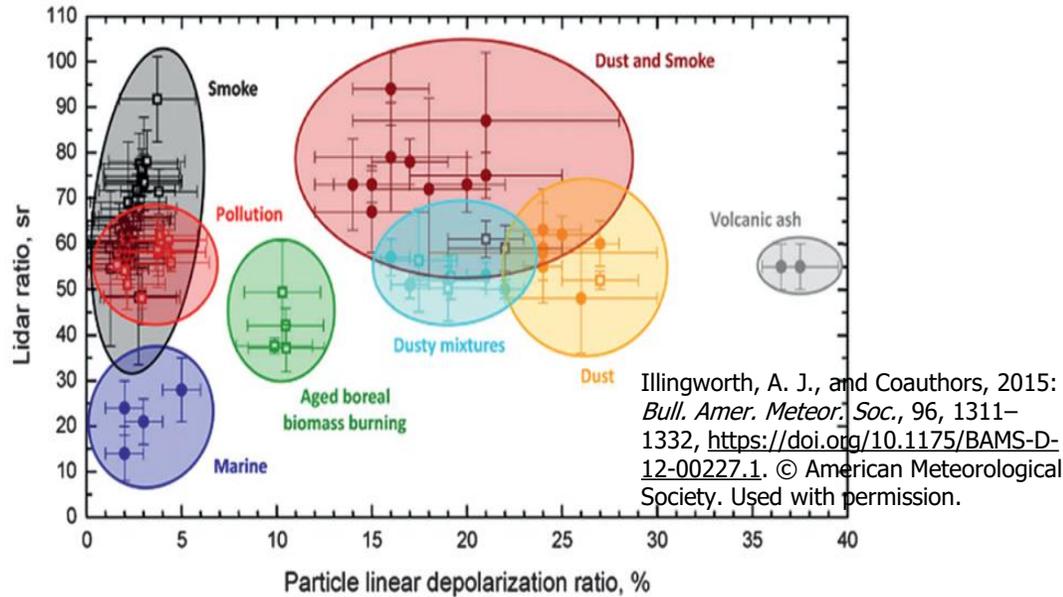
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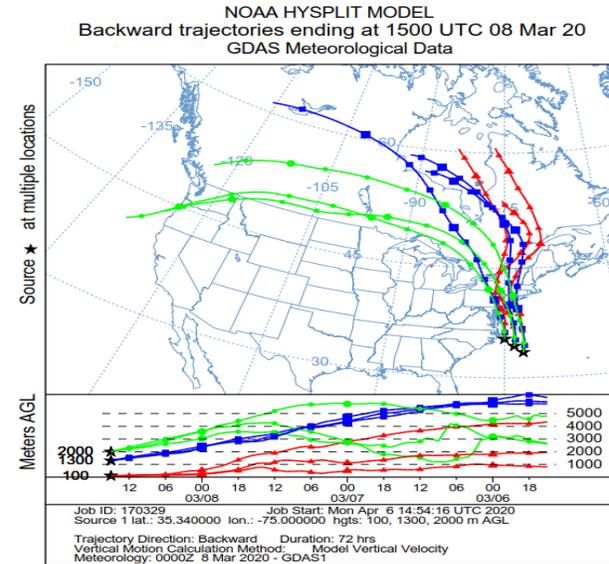
HYSPLIT and FLEXPART Backtrajectories Suggest Enhanced Depolarization is Not Due to Dust



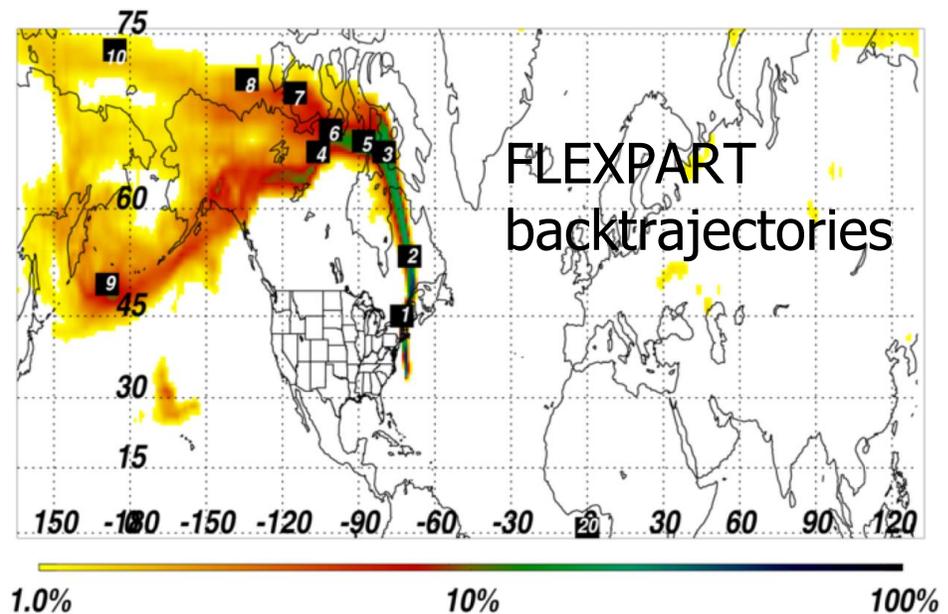
- High aerosol depolarization most often associated with dust and to a lesser extent with smoke and pollen



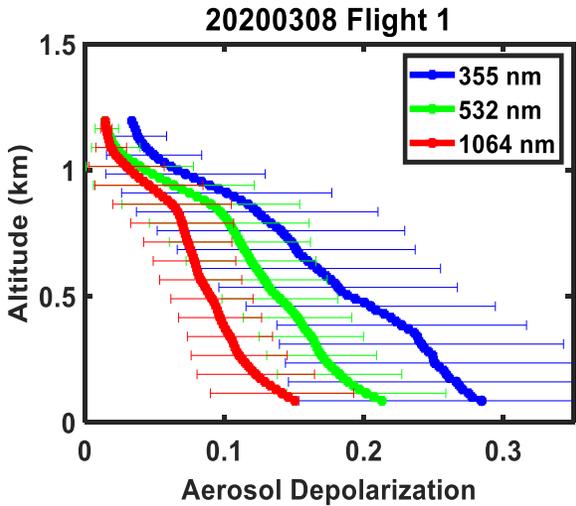
- Backtrajectories from NNW and higher altitudes, and so do not appear consistent with dust transport to the ACTIVATE flight region
- Pollen and air quality reports for March 8 do not indicate high pollen levels



The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (<https://www.ready.noaa.gov>) (see https://www.ready.noaa.gov/HYSPLIT_agreement.php)

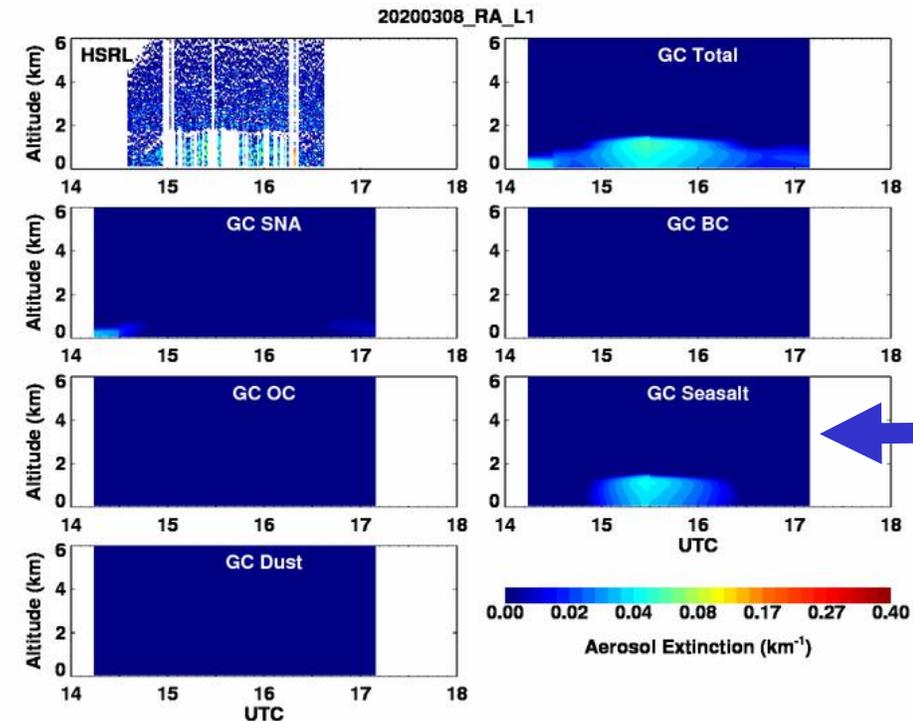
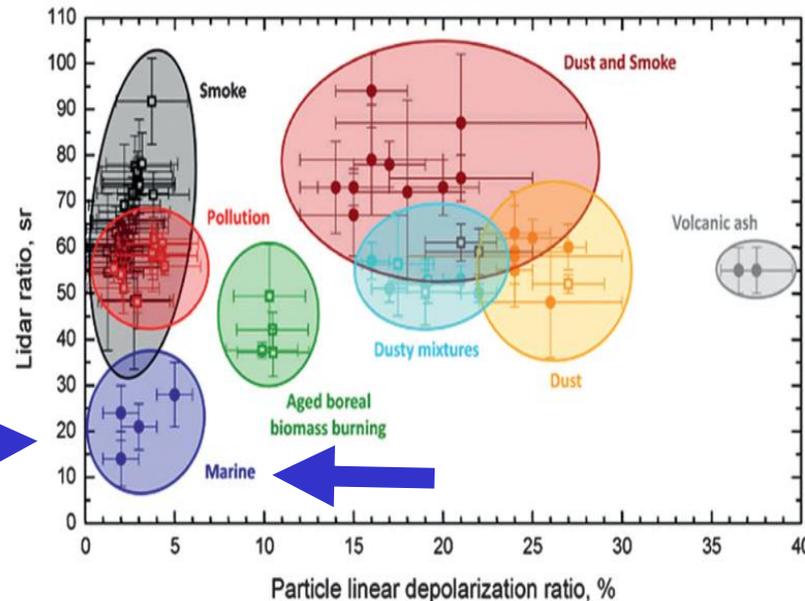
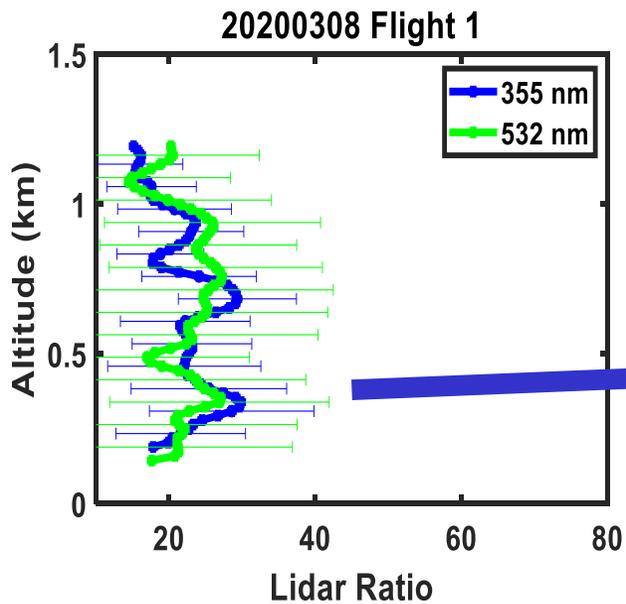


Elevated depolarization appears to be due to sea salt



- Aerosol extinction/backscatter ratios (i.e. lidar ratios) were about 20-25 sr and so appear to be too low for dust
- The low lidar ratios are instead consistent with marine (sea salt) aerosol

➤ 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)

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.

Enhanced aerosol depolarization associated with sea salt has been measured by lidar before



Depolarization from sea salts is known within the science (and lidar) community, but its prevalence has not been noted to the extent and level that we have observed.

- Murayama, T., and Co-Authors (1999), JGR
- Sakai, T. and Co-authors (2000), Atmos Environ
- Haarig, M., and Co-authors (2017): Atmos. Chem. Phys.,

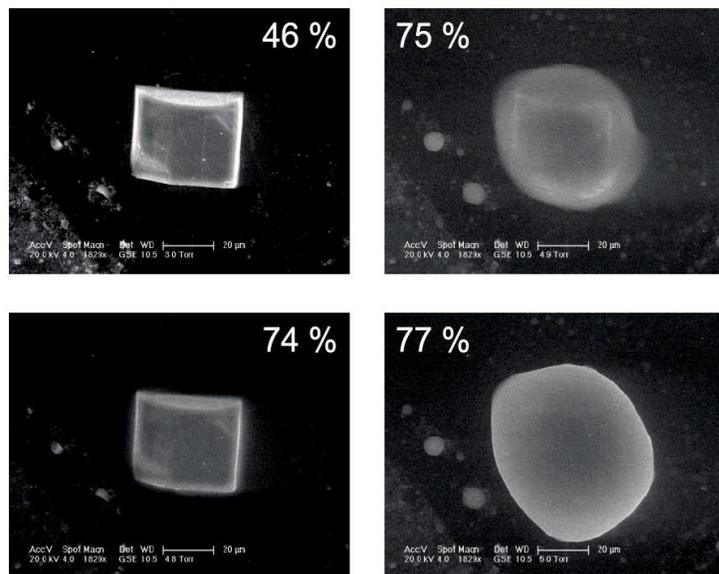


Figure 1. Sodium chloride deliquescence at 75 % RH observed at laboratory conditions (at 4.9 °C) in an environmental scanning electron microscope. The dry cubic particle with sharp edges at RH of 46 % becomes surrounded by a liquid sphere when RH increases to 77 %.

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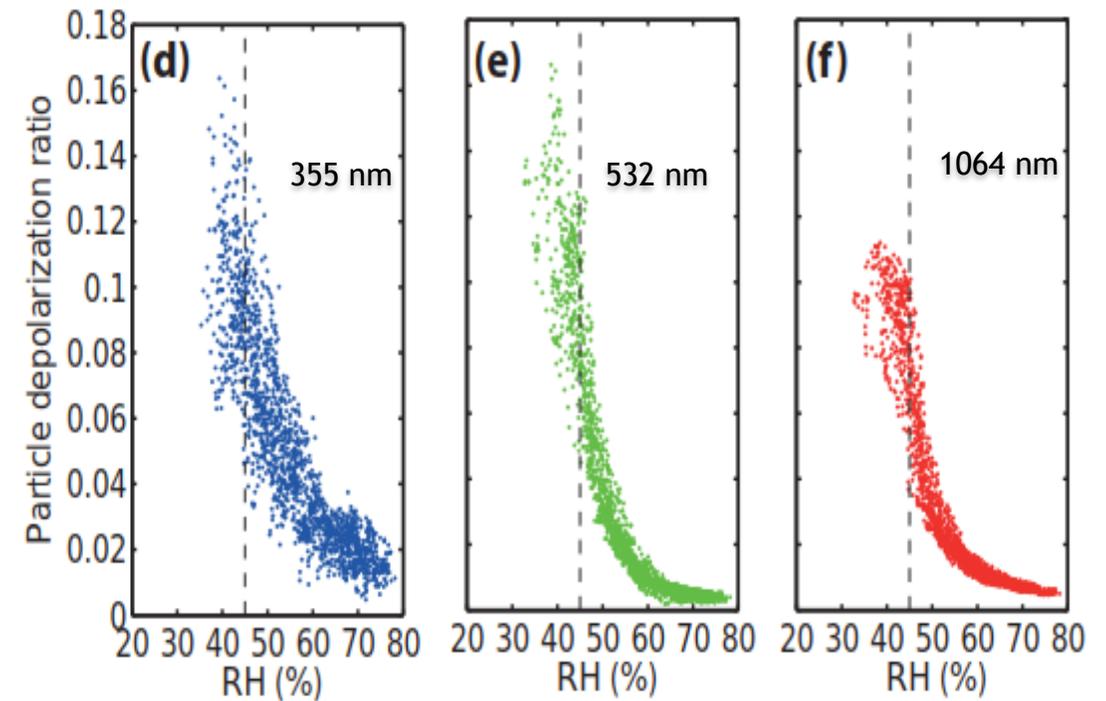
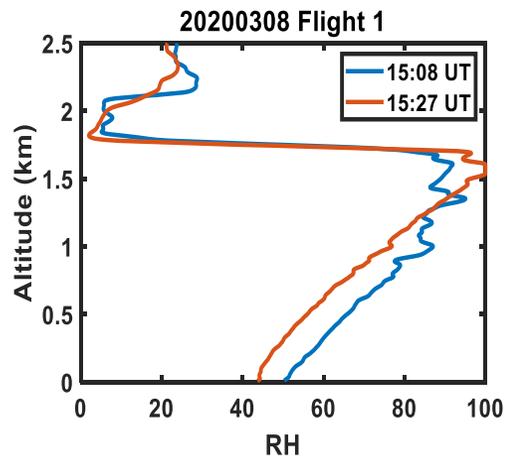
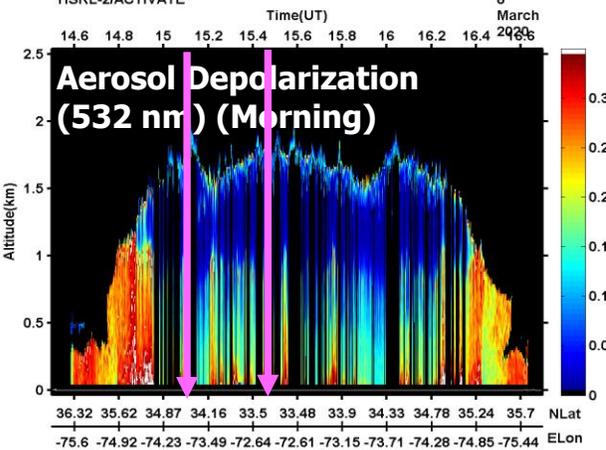
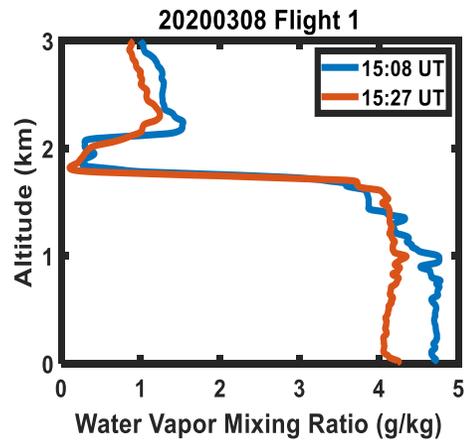
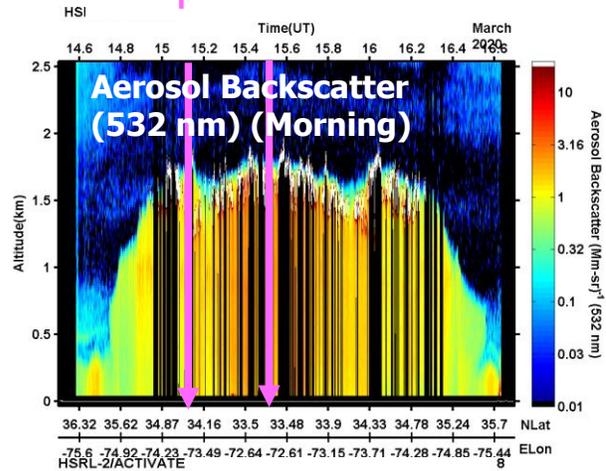


Figure 12. Correlation of the particle linear depolarization ratio (d–f) with RH for the three wavelengths 355, 532, and 1064 nm. (from Haarig, M., and Co-authors: Dry versus wet marine particle optical properties: RH dependence of depolarization ratio, backscatter, and extinction from multiwavelength 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/>)

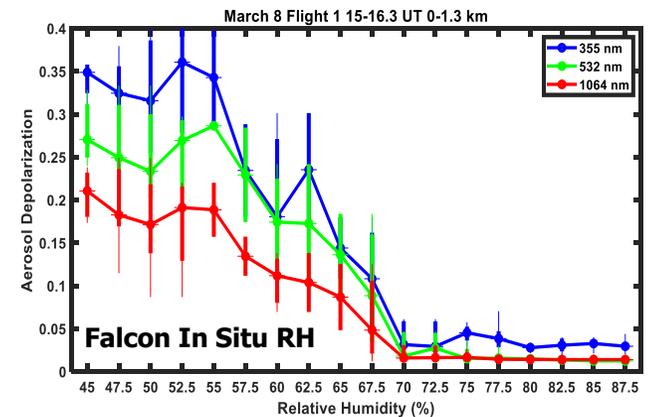
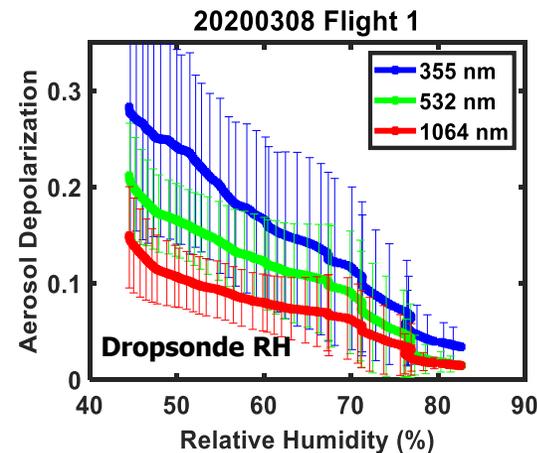
Dropsonde data show well mixed marine BL

- Dropsonde data show increase of RH with altitude within Boundary Layer (BL)
- Constant mixing ratio and nearly constant aerosol backscatter suggest well mixed BL such that vertical variability in particle intensive properties is due to changes in RH, not due to change in particle type

Purple arrows show when dropsondes launched



- Variability in aerosol depolarization appears well correlated with changes in Relative Humidity (RH) as measured by dropsondes and in situ measurements on the Falcon
- High aerosol depolarization is associated with low RH and low depolarization is associated with high RH



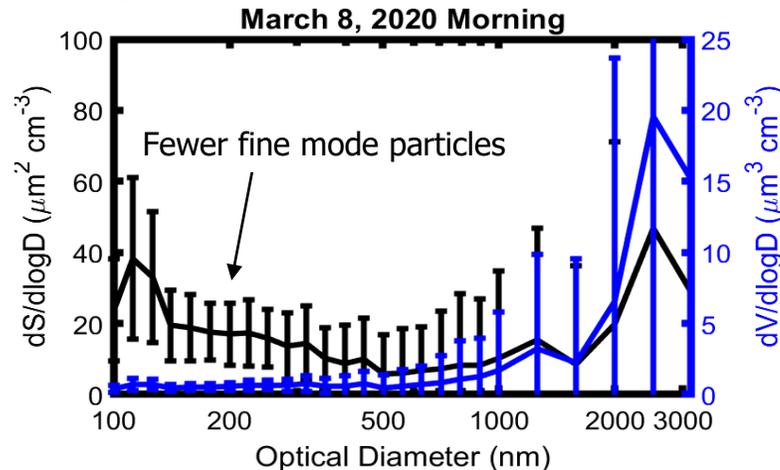


Airborne In situ Aerosol Size Distribution Measurements Show Fewer Fine Mode Particles When Enhanced Depolarization was Observed

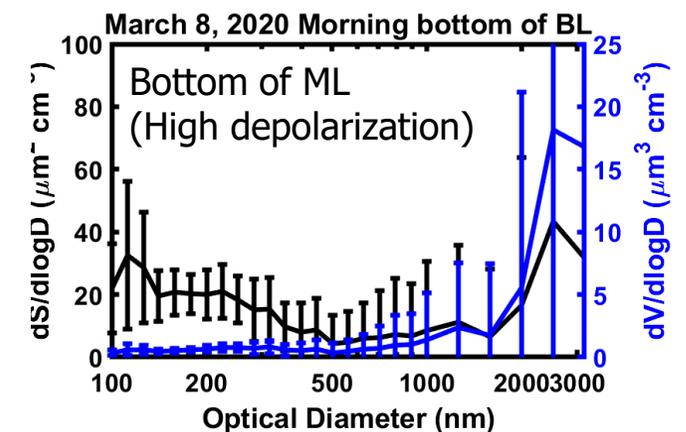
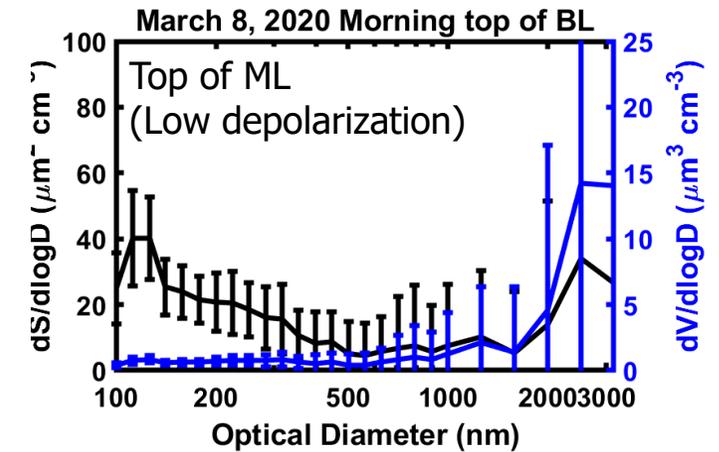
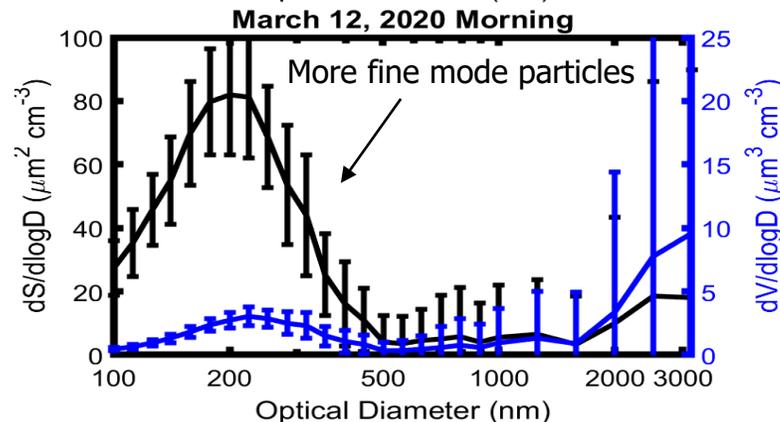
- During March 8 flights (enhanced depolarization) airborne in situ aerosol size distribution measurements show a relative lack of fine mode particles.
- In contrast, during flights on March 12 (low depolarization), airborne in situ aerosol size distribution measurements show considerably more fine mode particles.

- During March 8 flights, airborne in situ aerosol size distributions were very similar at the bottom (where high depolarization was measured) and at the top (where low depolarization was measured) of the mixed layer
- This suggests particle shape, not size, varied with RH

Flight with Elevated Aerosol Depolarization



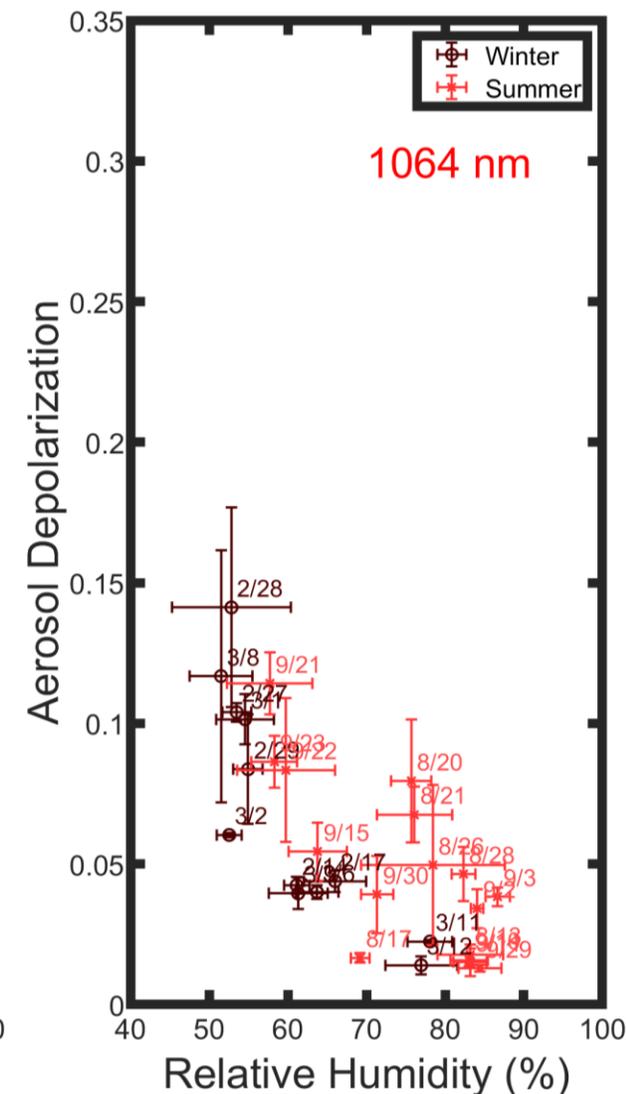
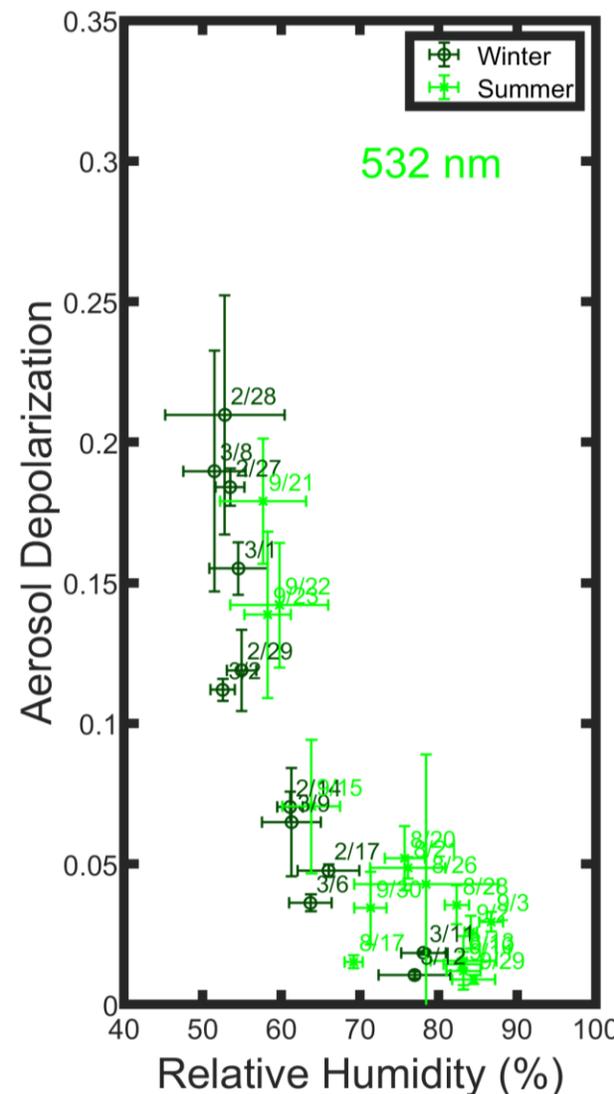
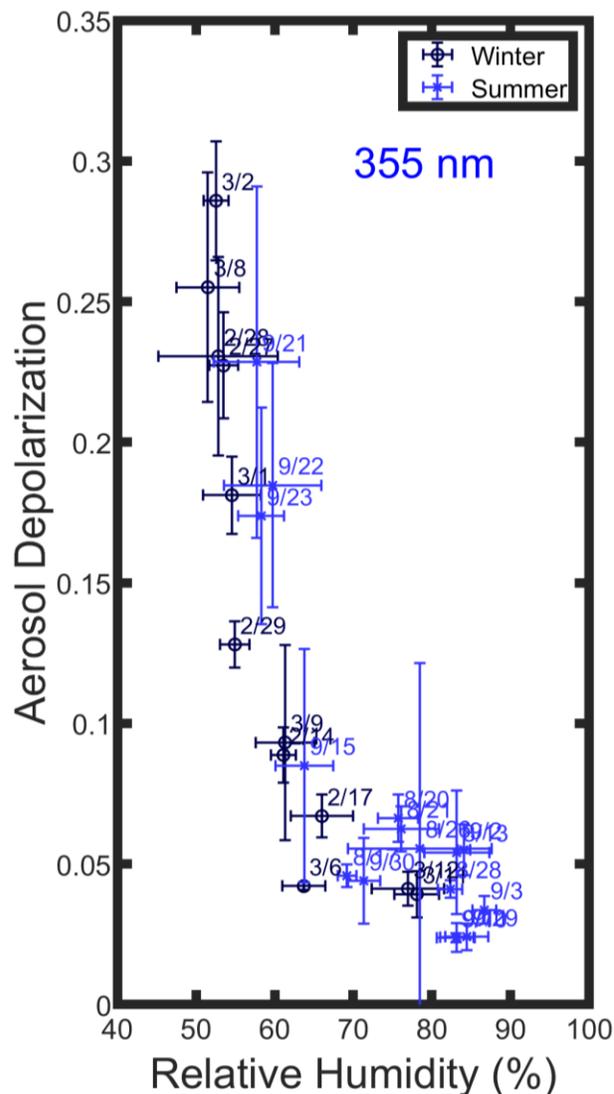
Flight without Elevated Aerosol Depolarization



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



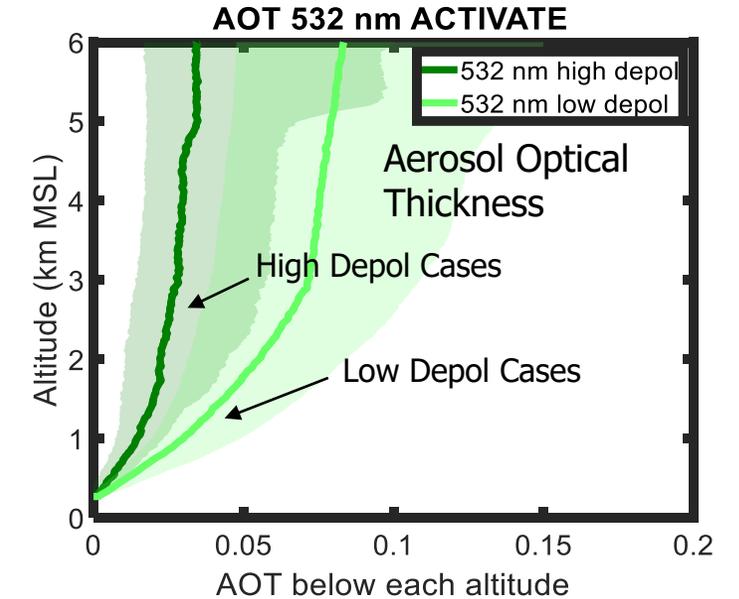
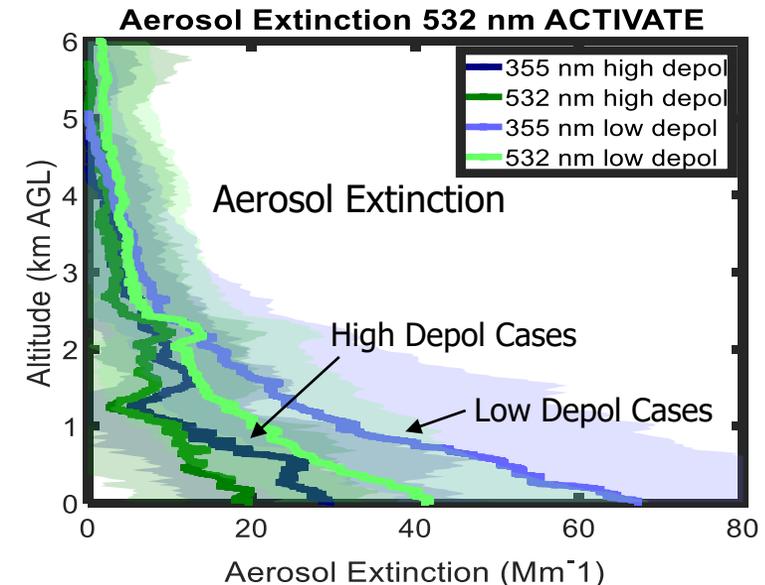
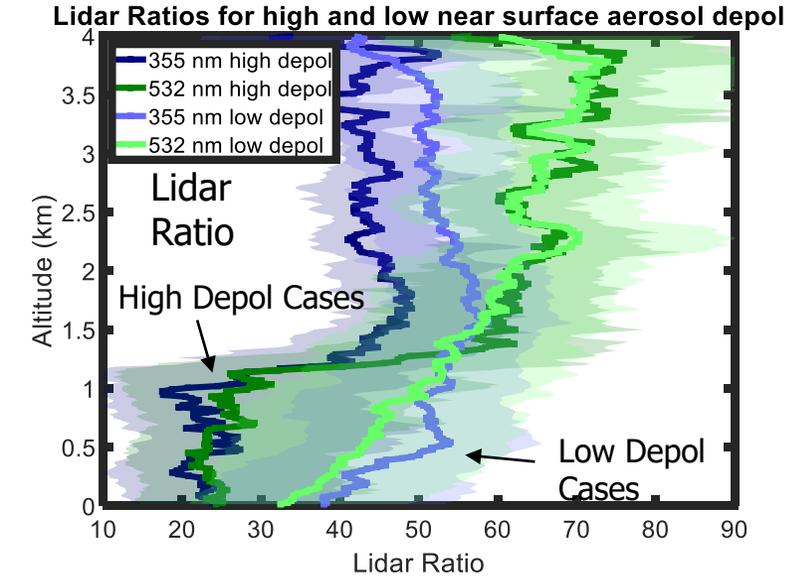
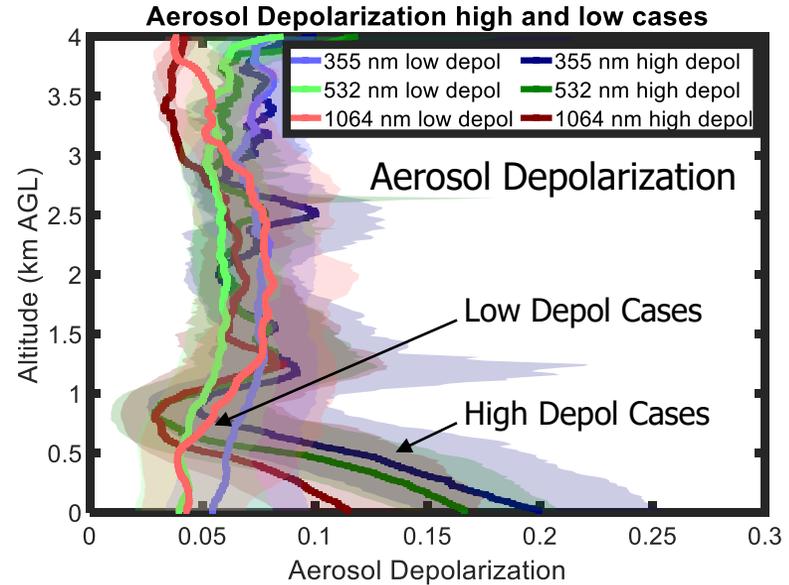
- Average depolarization in lowest 20% of Mixed Layer coincident with dropsondes
- Each point represents average for that date
- High aerosol depolarization is associated with RH below about 60%
- Depolarization decreases with increasing wavelength
- Elevated depolarization during both winter and summer



Aerosol Properties (High vs. Low Aerosol Depolarization)

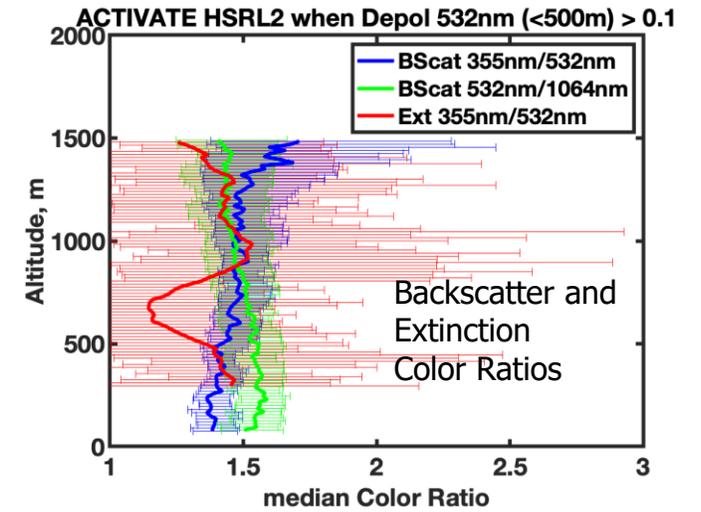
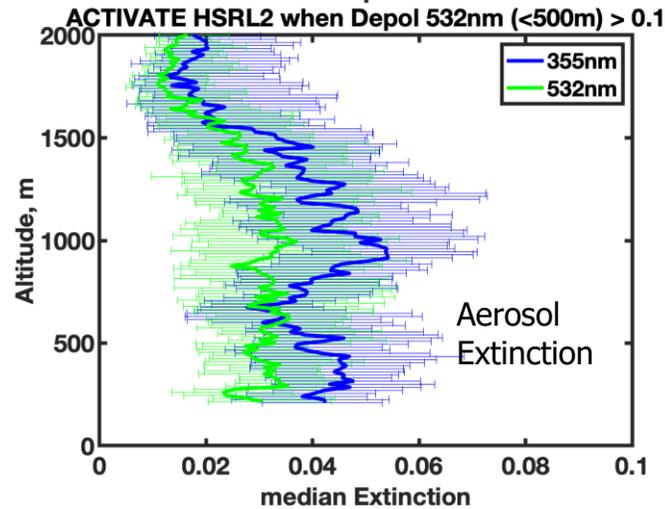
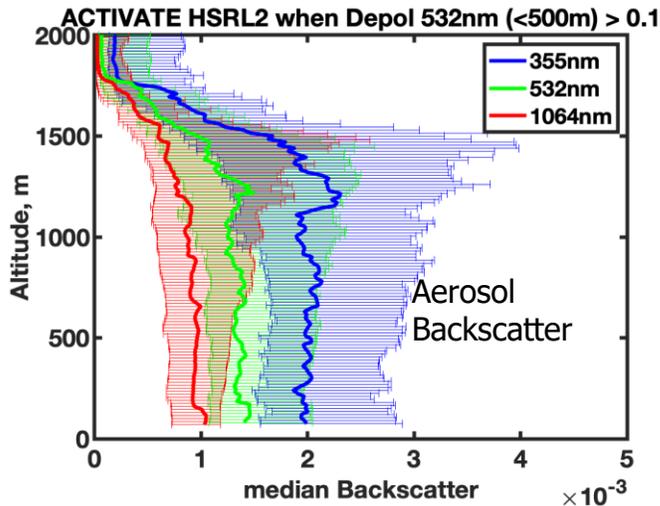
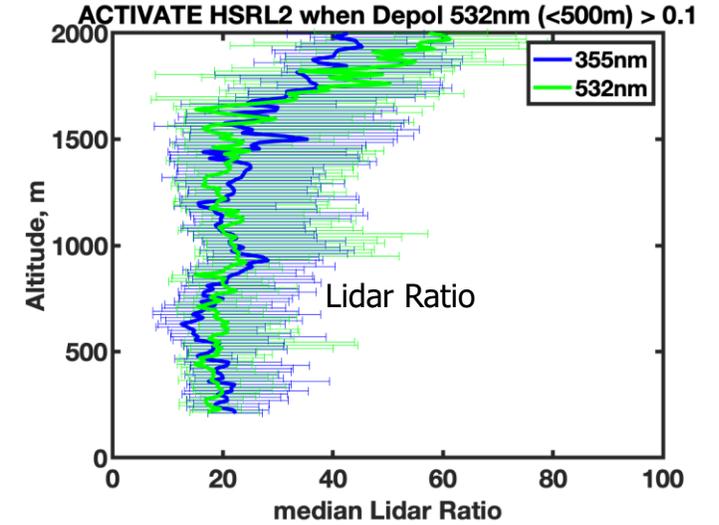
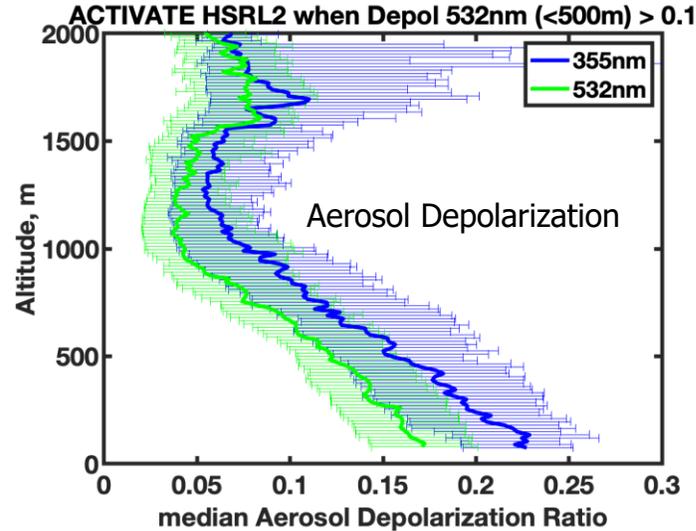
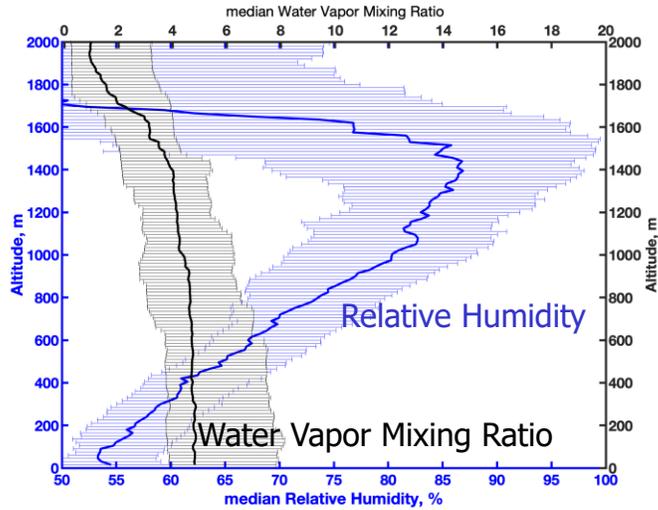


- HSRL2 data from 2020 divided into high and low near-surface depolarization cases
- High depolarization observed in about 18% of the HSRL2 profiles and during 23 of 33 flights
- Median lidar ratios at 355 and 532 nm were around 20-25 sr during these high depol (nonspherical sea salt) cases
- Higher lidar ratios observed when other aerosol types (e.g. smoke) within BL
- On average, lidar ratio increased significantly with altitude
- Median AOT (0-7 km) for high depol cases was about 0.04 (532 nm)
- Median AOT (0-7 km) for low depol cases was about 0.09 (532 nm)
- AOT contributed by the nonspherical sea salt particles was small (0.03-0.04) (532 nm)



Median Profiles for Dropsonde Cases of High Near-Surface Depolarization

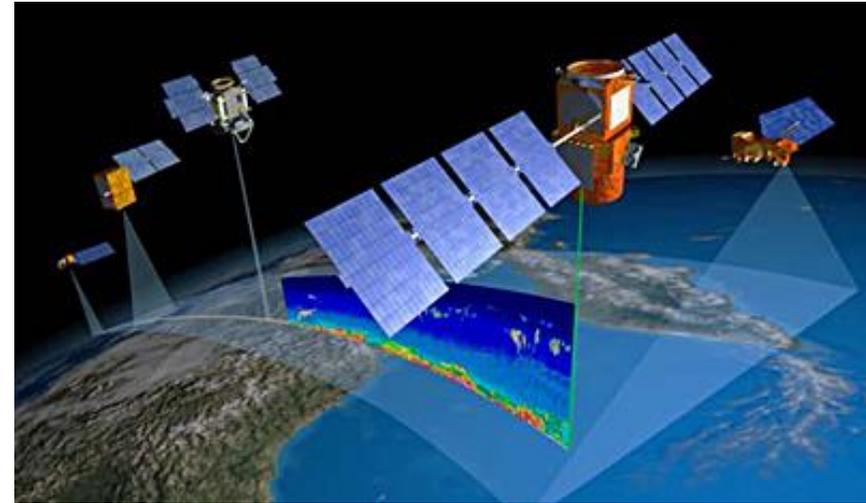
- High near-surface depolarization coincident with 17 of 126 dropsondes during 2020
- Aerosol depolarization decreased as RH increased; little change in other parameters



The Classification of These Aerosols is Very Relevant to the CALIOP Operational Aerosol Retrievals



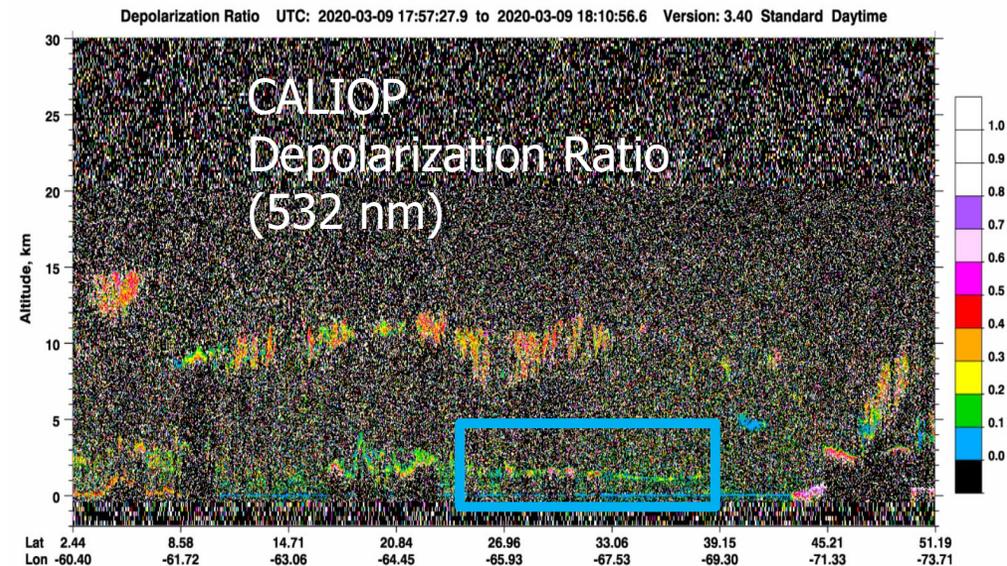
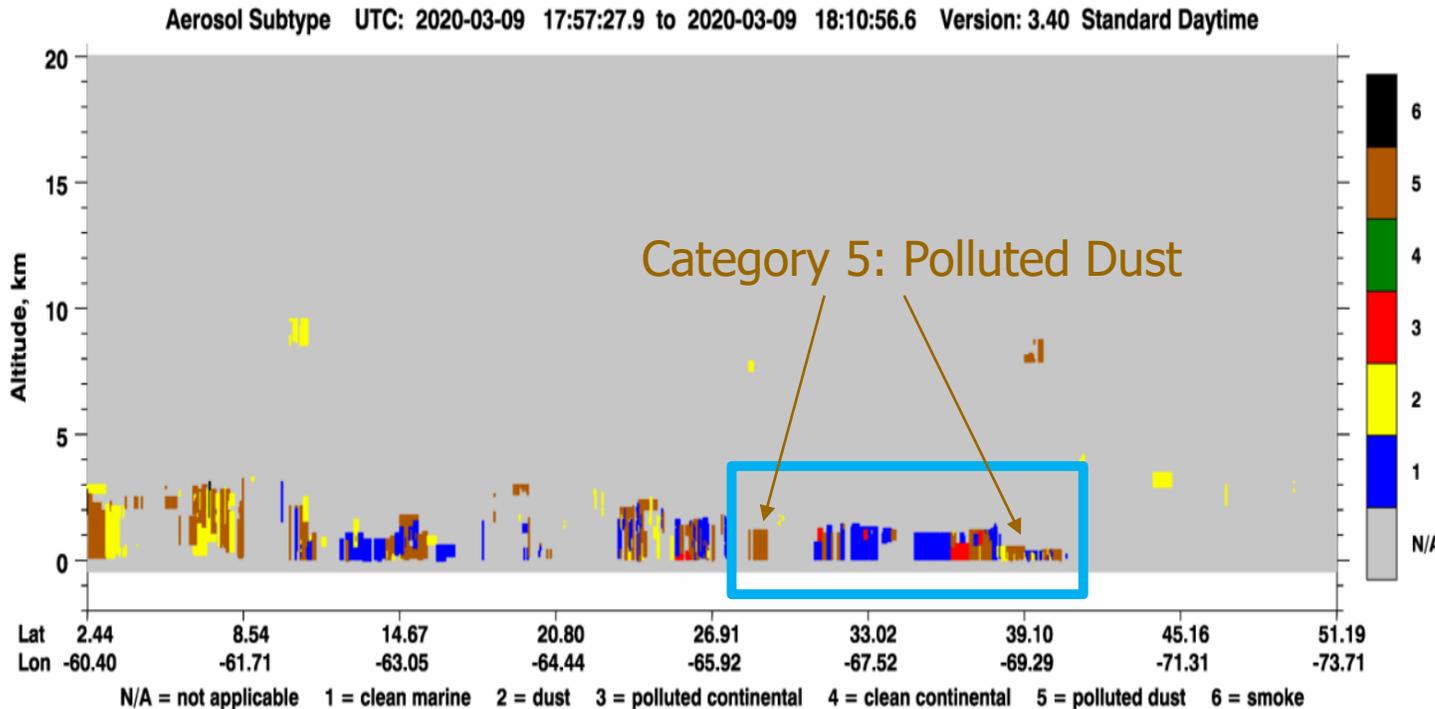
- Difference in sea salt vs. dust can have major impact on CALIOP (on CALIPSO) operational aerosol retrievals
- If CALIOP detects elevated depolarization, the aerosol is classified as dusty marine, polluted dust, or desert dust depending on altitude and location
- This classification specifies the aerosol 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)



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

CALIOP Measurements of Enhanced Depolarization March 9, 2020

- On March 9, a day after the HSRL-2 observations of enhanced depolarization in lower marine BL, CALIOP measurements show enhanced depolarization in this same region off the east coast of USA
- Based on this elevated depolarization, CALIOP aerosol typing algorithm classified some of the aerosol in this marine BL as polluted dust
- Modeled RH profiles show relative dry ($RH < 60\%$) conditions near the surface consistent with the ACTIVATE measurements on the previous day



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



- The occurrences of elevated depolarization associated with sea salt observed in both winter and late summer 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, every single one included some aerosol that the CALIOP algorithm classified as 'dusty marine'.
- Were these misclassified as 'dusty marine' instead of sea salt?
 - 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
- The large majority of these CAO cases also suggest that the CALIOP operational algorithm misclassified depolarizing sea salt as 'dusty marine' aerosol

Summary and Key Findings

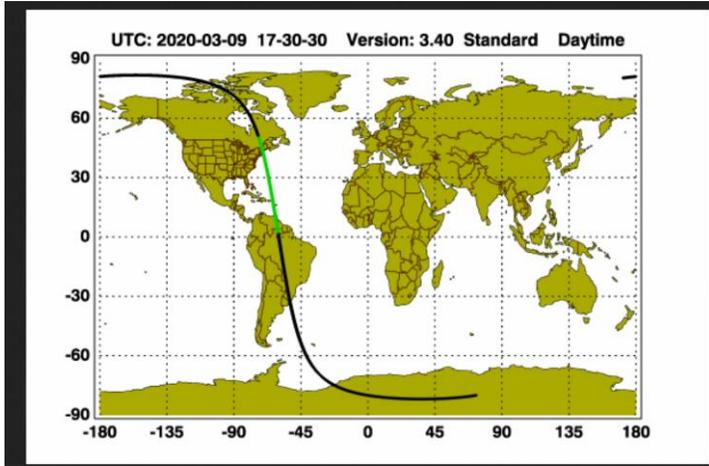


- Airborne NASA Langley Research Center (LaRC) High Spectral Resolution Lidar-2 (HSRL-2) measurements acquired during several (10-12) flights during both the winter and summer 2020 NASA EVS-3 ACTIVATE campaigns revealed enhanced (>15-20%) particulate linear depolarization associated with aerosols within the marine boundary layer.
- These aerosols do not appear to be associated with dust or pollen.
- The strong correlation of elevated depolarization with low (<60%) relative humidity (RH), and the HSRL-2 measurements of low (20-25 sr at 532 nm) aerosol extinction/backscatter (i.e. lidar) ratio, strongly suggest that these elevated depolarization measurements are associated with crystalline sea salt, consistent with previous lidar observations of enhanced depolarization associated with sea salt.
- These cases tended to appear during Cold Air Outbreaks (CAO).
- Examination of CALIOP measurements during several CAO episodes reveal that CALIOP also measured enhanced depolarization during such cases and that these typically also had low (<60%) RH.
- The CALIOP operational aerosol algorithms tended to classify these aerosols as dust, polluted dust, or dusty marine, rather than marine aerosols. Such misclassification leads to overestimates in the assumed lidar ratio and in the resulting retrievals of aerosol optical depth and aerosol extinction.
- Future work will examine the prevalence and location of such observations in the CALIOP data record.

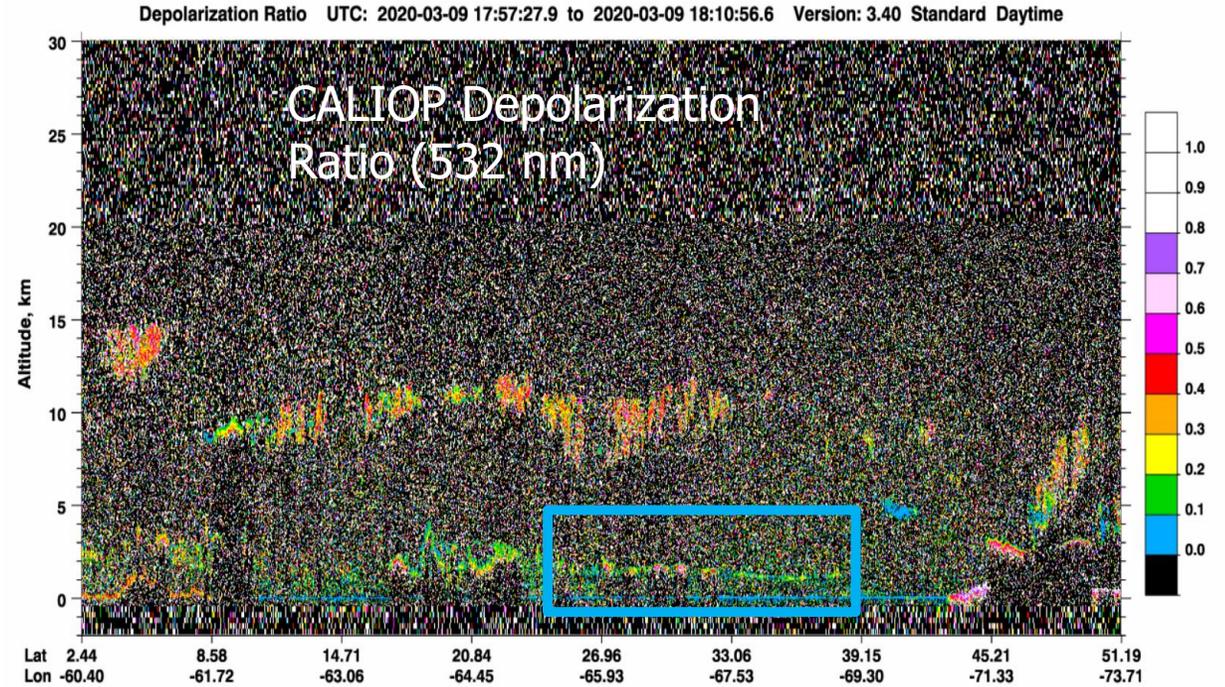
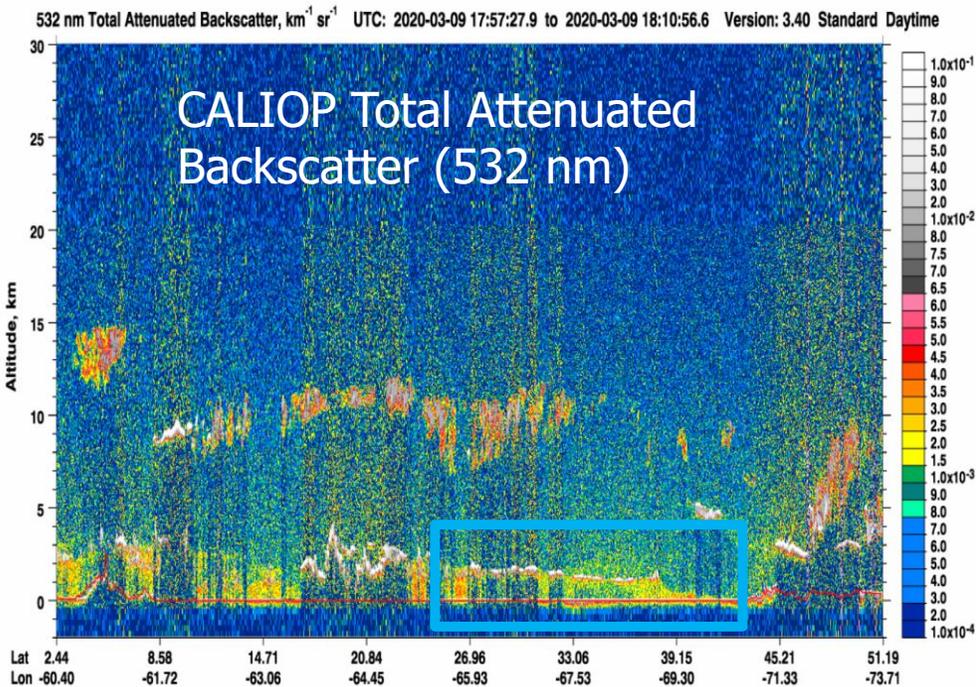


Backup

CALIOP Measurements of Enhanced Depolarization - March 9, 2020



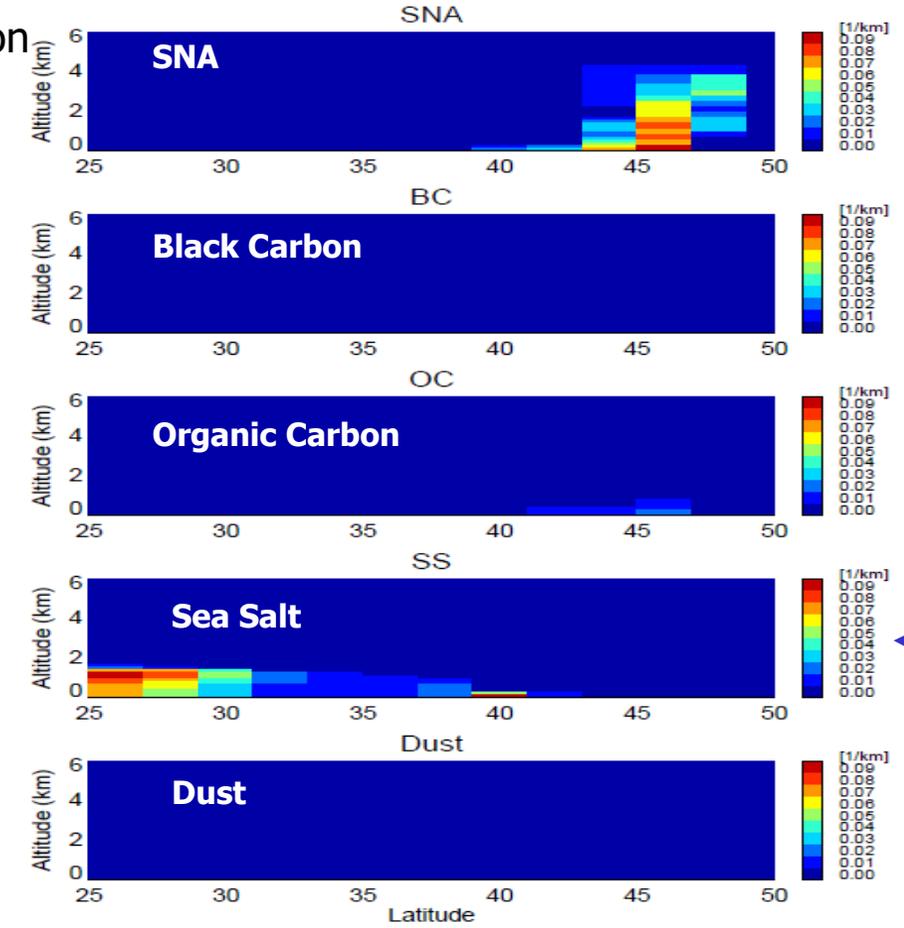
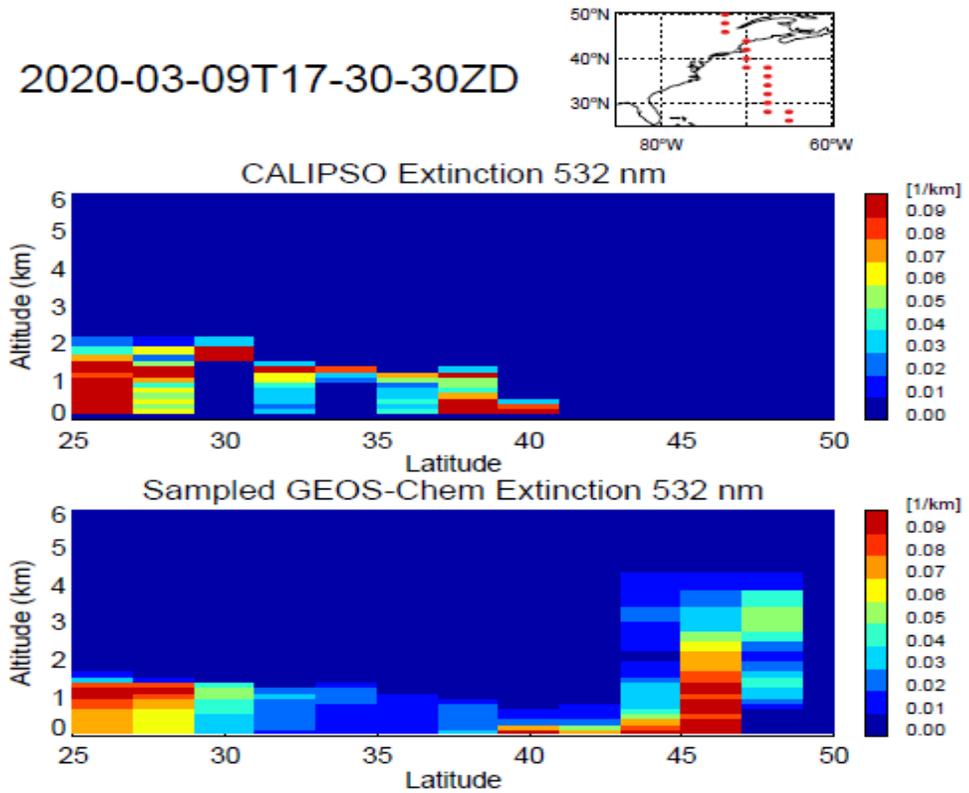
On March 9, a day after the HSRL-2 observations of enhanced depolarization in lower marine BL, CALIOP measurements show enhanced depolarization in this same region off the east coast of USA



GEOS-Chem Model Identifies Sea Salt as Major Contributor to Aerosol Extinction During March 9 CALIOP Overpass



GEOS-Chem model also suggests that the source of elevated depolarization along this CALIOP track is sea salt rather than polluted dust



Comparisons of GEOS-Chem aerosol extinction (550nm) with CALIPSO measurements (532 nm) over the ACTIVATE analysis domain (17:30UTC, Mar.9, 2020). Model output are sampled at 1:30pm LT. (SNA = sulfate + nitrate + ammonium, OC = organic carbon, BC = black carbon)

Variability with aerosol properties with RH



- High near-surface depolarization coincident with 17 of 126 dropsondes during 2020
- Aerosol depolarization decreased as RH increased; little change in other parameters

