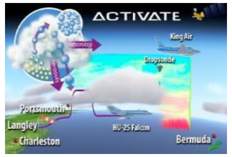

Aerosol Hygroscopic Effects Measured by the Airborne High Spectral Resolution Lidar-2

Richard Ferrare[1], John Hair[1], Chris Hostetler[1], David Harper[1], Shane Seaman[1], Taylor Shingler[1], Ewan Crosbie[2], Edward Winstead[2], Luke Ziemba[1], Michael Shook[1], Lee Thornhill[2], Marta Fenn[3], Marian Clayton[3], Amy Jo Scarino[3], Sharon Burton[1], Anthony Cook[1], Glenn Diskin[1], Rich Moore[1], Claire Robinson[2], Josh DiGangi[1], John Nowak[1], Snorre Stamnes[1], Armin Sorooshian[4], Sue van den Heever[5], Bastiaan van Diedenhoven[6], Brian Cairns[7]

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Motivation

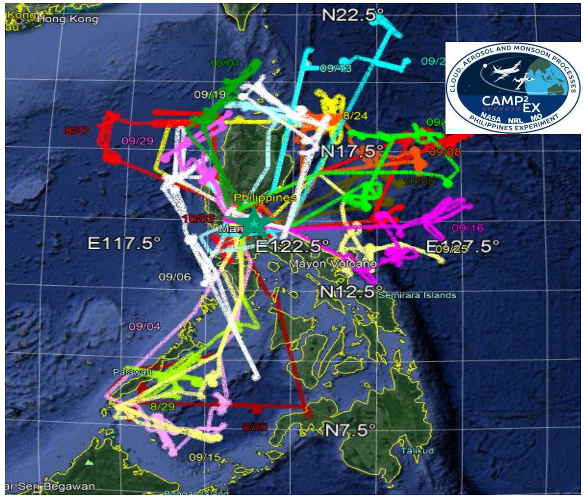
1) Water uptake by aerosols impacts aerosol physical characteristics (size, shape, composition) which in turn affect aerosol optical properties (e.g. scattering, extinction, depolarization)
2) Changes in these aerosol characteristics impact their: 1) radiative effects, 2) ability to act as Cloud Condensation Nuclei and Ice Nuclei, 3) role in aqueous chemistry
3) Large diversity in the magnitude of aerosol humidification in models (e.g. Burgos et al., ACP, 2020)
4) Evidence that some models have too large an increase in aerosol extinction with relative humidity (RH)

- Airborne HSRL measurements often reveal changes of aerosol properties (backscatter, extinction, depolarization) with RH. Can we quantify these changes and relate this variability to aerosol humidification factors derived from in situ measurements and represented in models?

This study uses data from NASA CAMP2Ex and ACTIVATE Missions

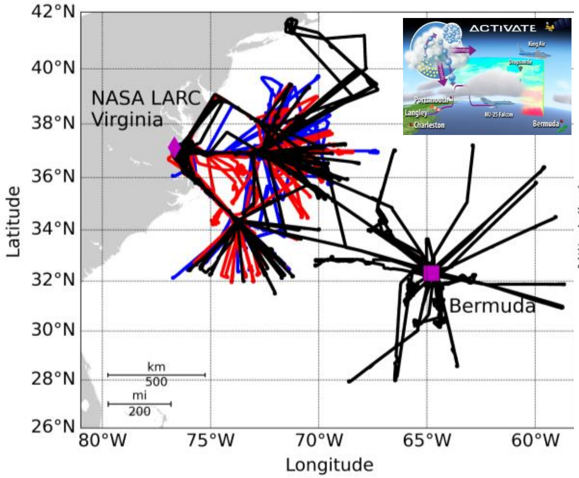
1) NASA CAMP2Ex (Aug-Oct 2019) (Philippines)

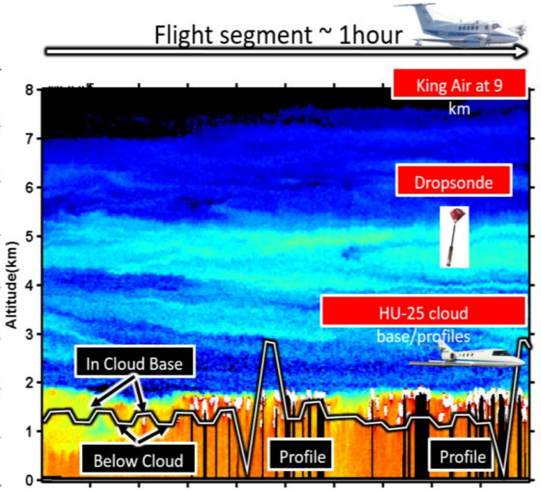
- CAMP2Ex addresses aerosol and cloud microphysics
- NASA LaRC HSRL-2 deployed on P-3B aircraft for nadir viewing measurements
- P-3B, based at Clark Air Base, conducted 19 science flights between Aug. 24 and Oct. 5, 2019
- Dropsondes deployed from P-3B aircraft
- Data available from https://www-air.larc.nasa.gov/cgi-bin/ArcView/camp2ex



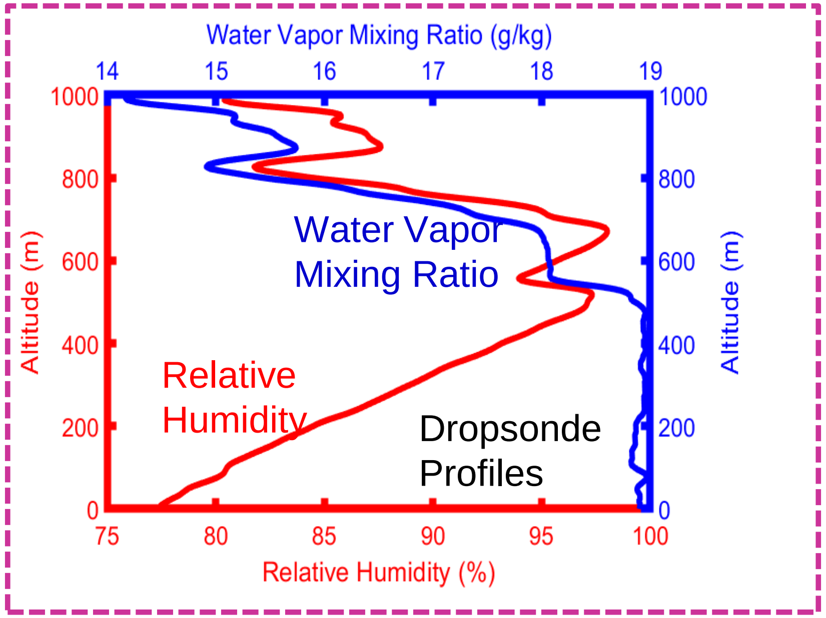
2) NASA EVS-3 ACTIVATE (Feb-Mar, Aug-Sep 2020; Jan-Jun, Dec 2021; Jan-Jun 2022) (western North Atlantic Ocean)

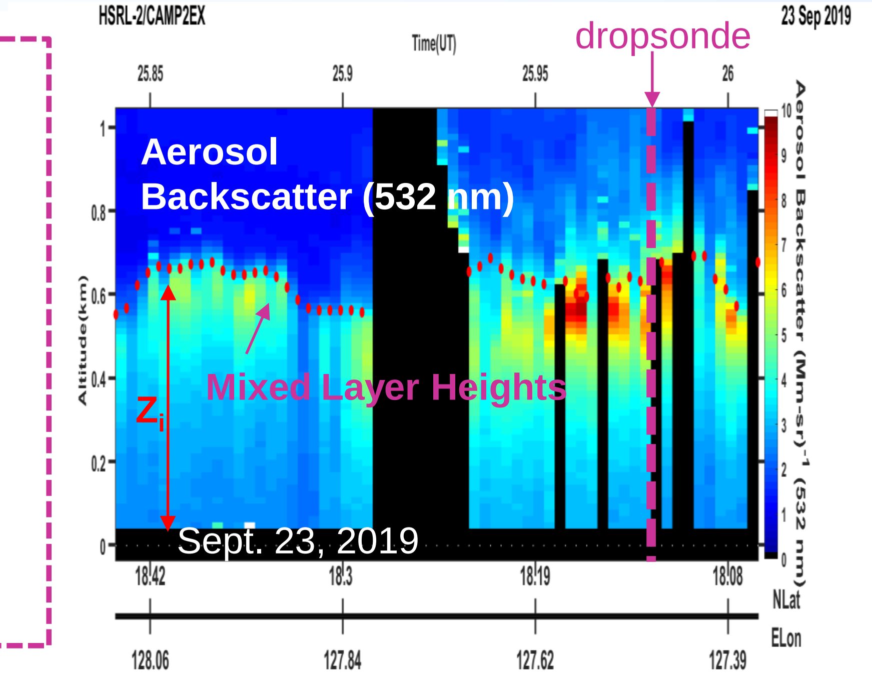
- Focus on marine boundary layer (MBL) clouds off the US Mid-Atlantic Coast
- NASA LaRC HSRL-2 deployed on LaRC King Air aircraft for nadir viewing, Dropsondes deployed from LaRC King Air aircraft
- In situ instruments deployed on NASA LaRC HU-25 Falcon aircraft to simultaneously measure BL clouds and aerosols below King Air
- Data available from https://www-air.larc.nasa.gov/cgi-bin/ArcView/activate.2019, https://www-air.larc.nasa.gov/cgi-bin/ArcView/activate.2021, https://www-air.larc.nasa.gov/cgi-bin/ArcView/activate.2022

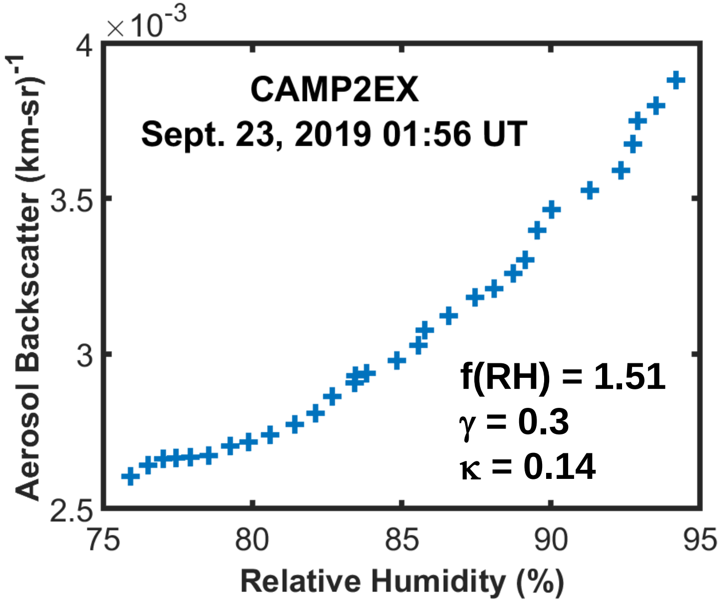




Quantifying the Aerosol Enhancement Factors Associated with the Increase in Relative Humidity (RH) using HSRL-2 and Dropsondes







- As RH increases with height within Mixed Layer, 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 HSRL-2; RH profiles from dropsondes
- Mixed Layer Height (Z_i) derived from HSRL-2 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%)

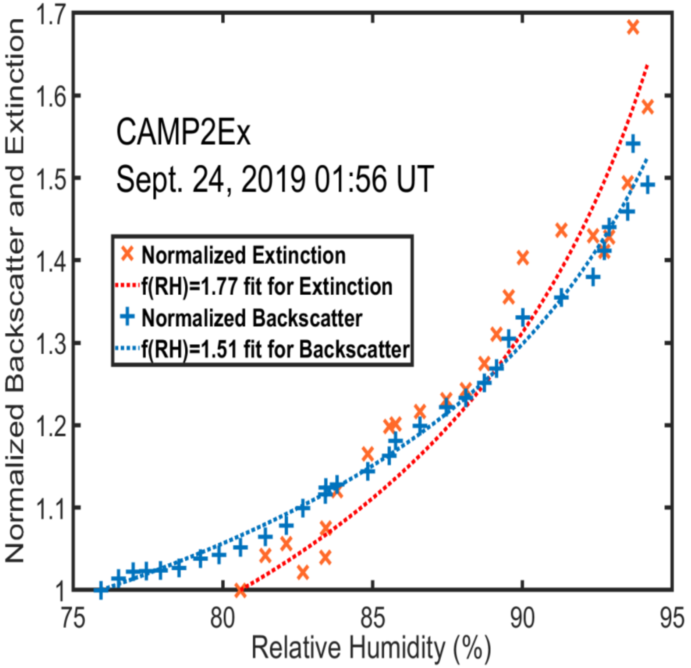
$$f(RH) = \frac{\beta(RH)}{\beta(RH_o)} = \left[\frac{(100-RH_o)}{(100-RH)}\right]^{\gamma}$$

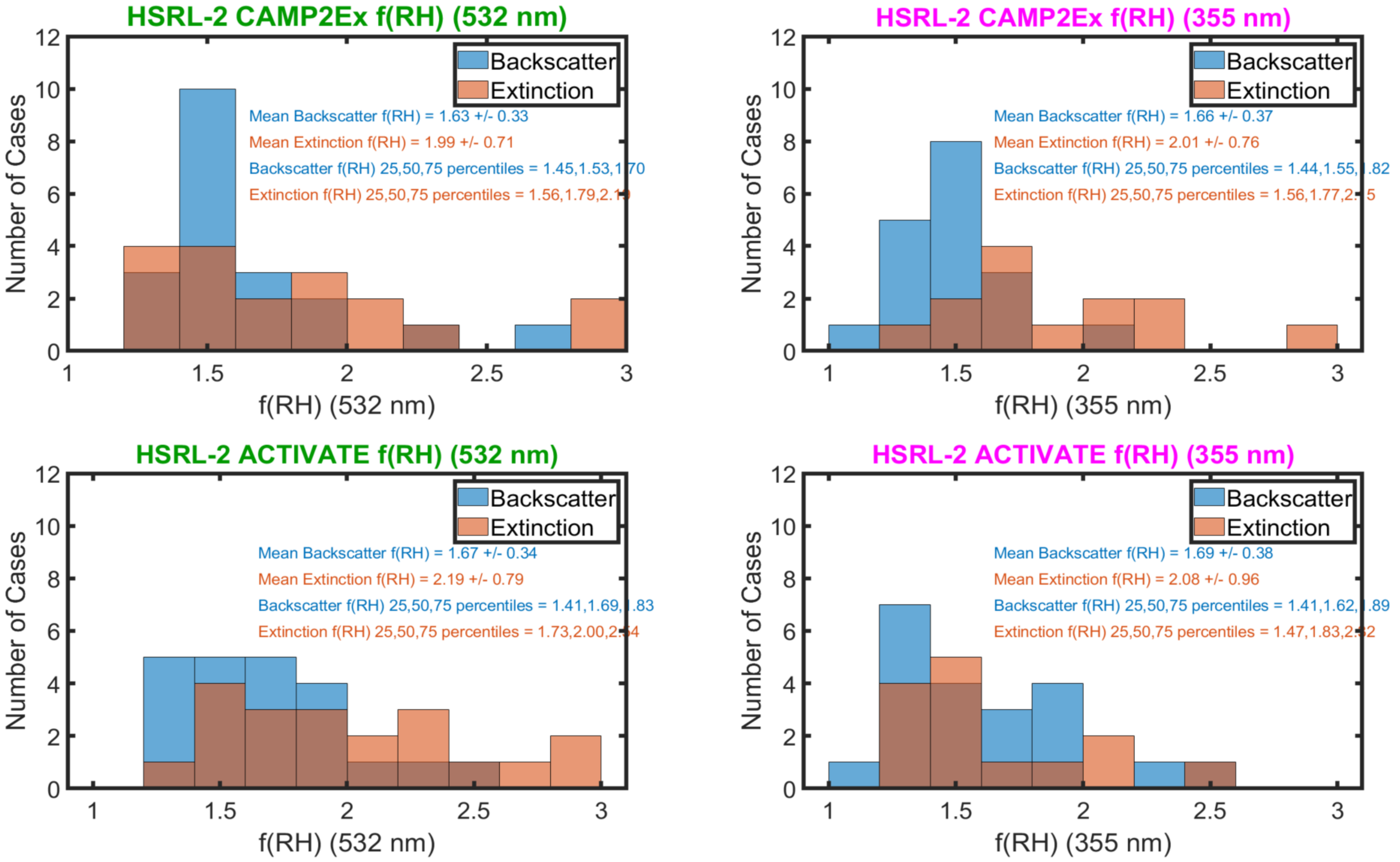
$$\cong 1 + \kappa\left[\frac{RH}{100-RH}\right]$$

$f(RH), \gamma$ computed for aerosol backscatter (β) and aerosol extinction (σ)

Humidification Factors ([f(RH)] for aerosol extinction slightly higher than for aerosol backscatter

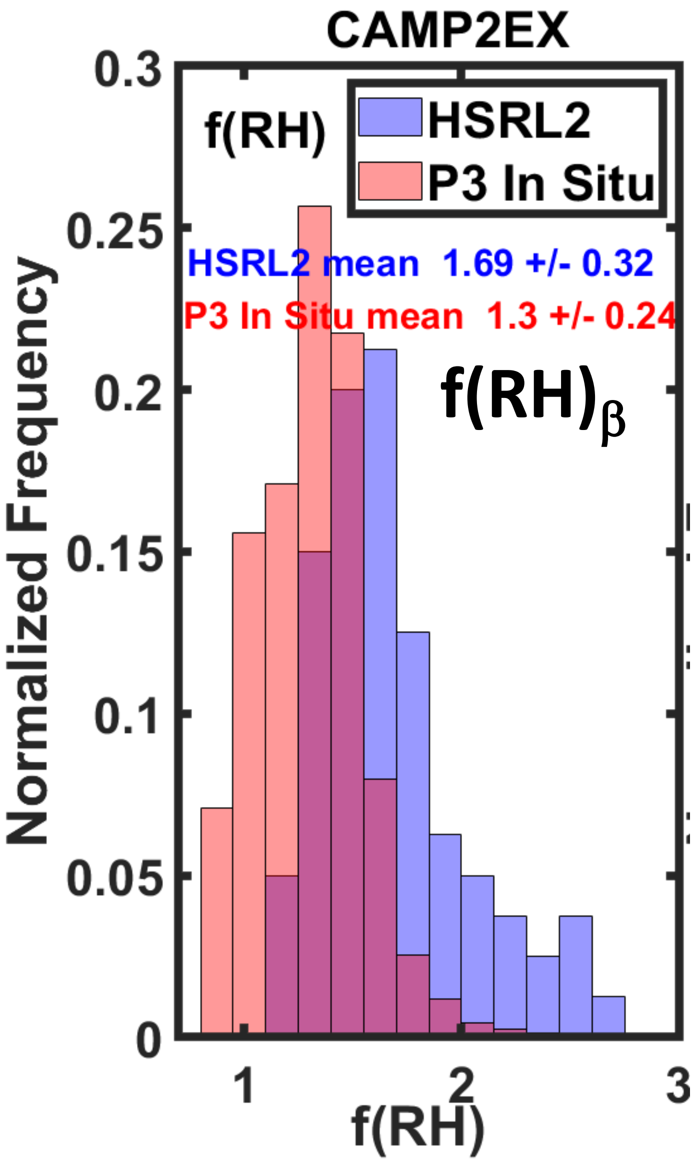
- $f(RH)_\sigma$ for aerosol extinction (~2.1) was slightly higher than $f(RH)_\beta$ for aerosol backscatter (~1.7)
- Consequently, aerosol extinction/backscatter ratio (“lidar ratio”) increases 5-30% with increasing RH, depending on the range of RH
- Average f(RH) appears to be the about the same at 355 nm and 532 nm

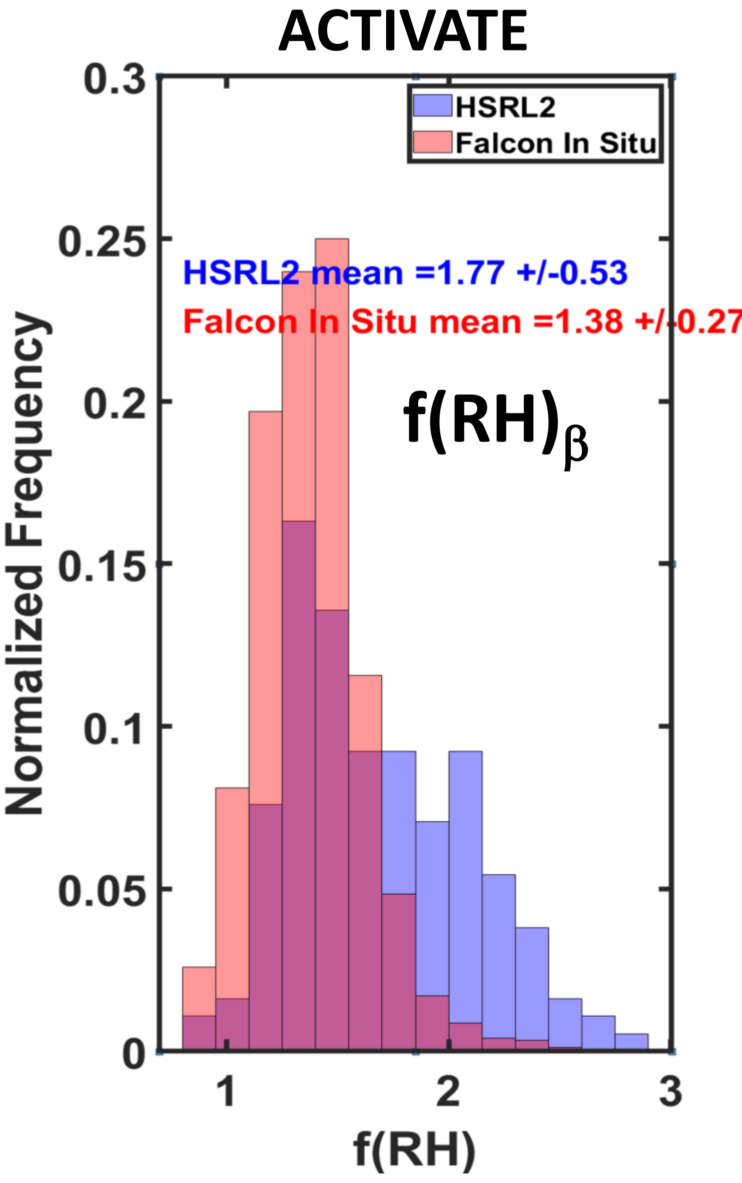




Aerosol Humidification Factors derived from HSRL-2/dropsondes are typically larger than from airborne in situ measurements

- Average f(RH=80%/RH=20%) (532 nm) derived from HSRL-2 and dropsonde data was about 1.7 for backscatter, and 1.8-2 for extinction, during CAMP2Ex and ACTIVATE
- These values were higher than the corresponding values from airborne in situ measurements during both CAMP2Ex and ACTIVATE (1.30-1.39)
- Higher f(RH) values from HSRL-2 & dropsonde are likely because lidar observes both fine and coarse (sea salt) aerosol in contrast to airborne in situ measurements of only fine mode aerosol



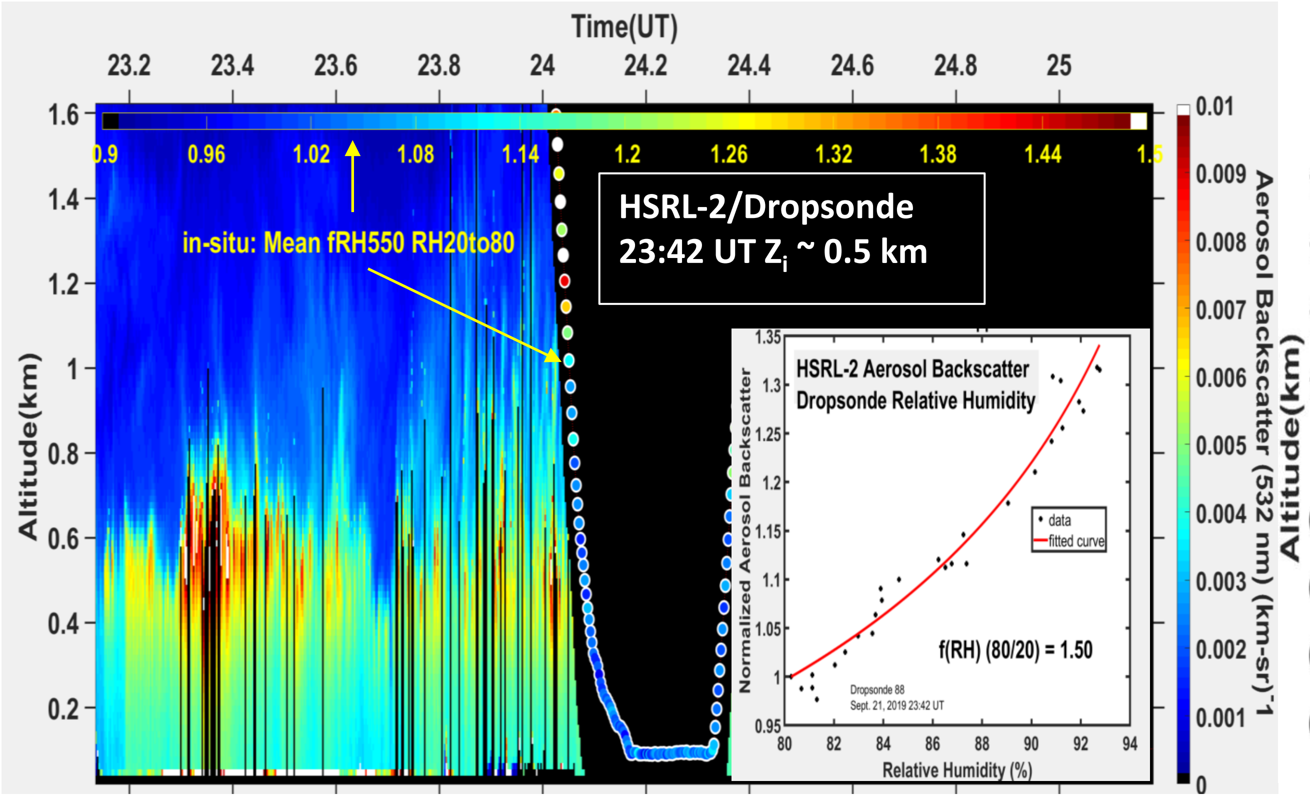


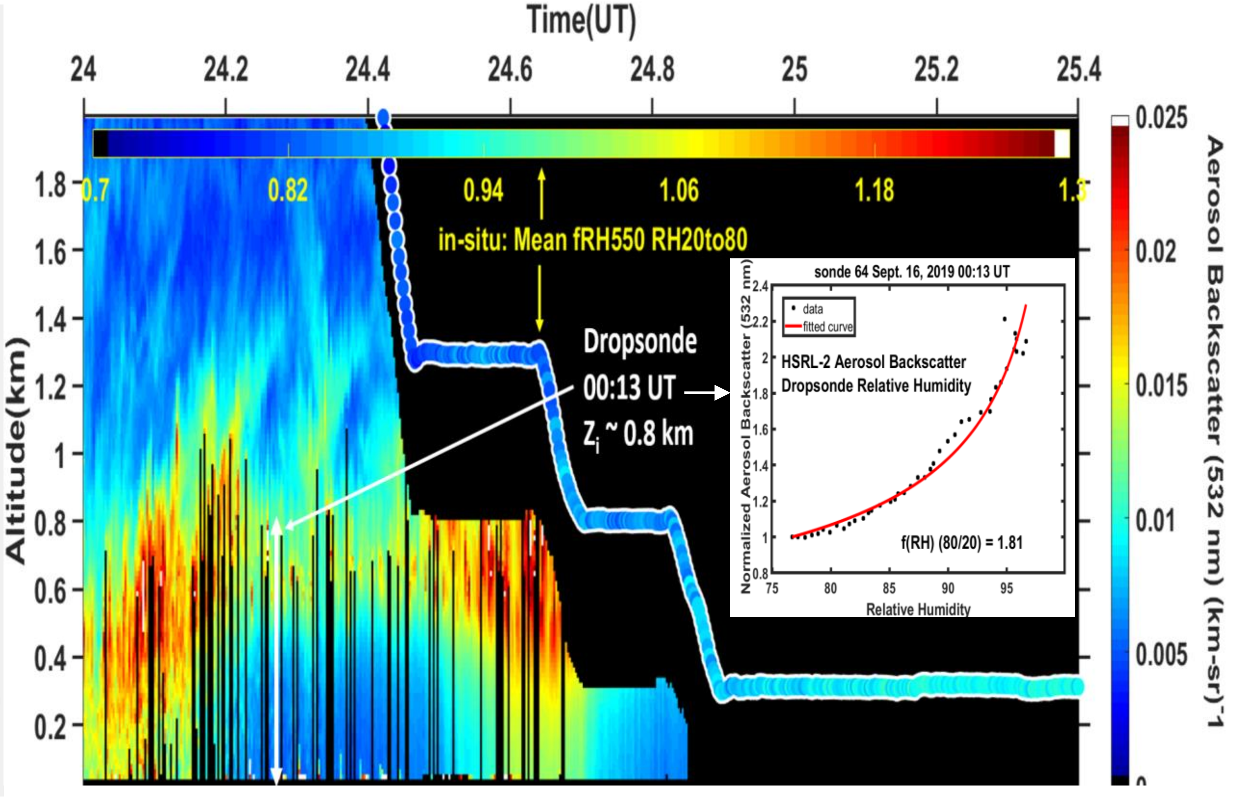
Examples comparing HSRL-2/dropsonde and in situ f(RH)

Higher f(RH) values derived from HSRL-2/dropsonde data are likely because lidar observes both fine and coarse (sea salt) aerosol in contrast to in situ measurements of only fine mode

- Example from CAMP2Ex Sept. 21, 2019 flight
  - In situ f(RH) ~ 1.0-1.1
  - HSRL-2/dropsonde $f(RH)_\beta$ ~ 1.5, $f(RH)_\sigma$ ~ 1.6

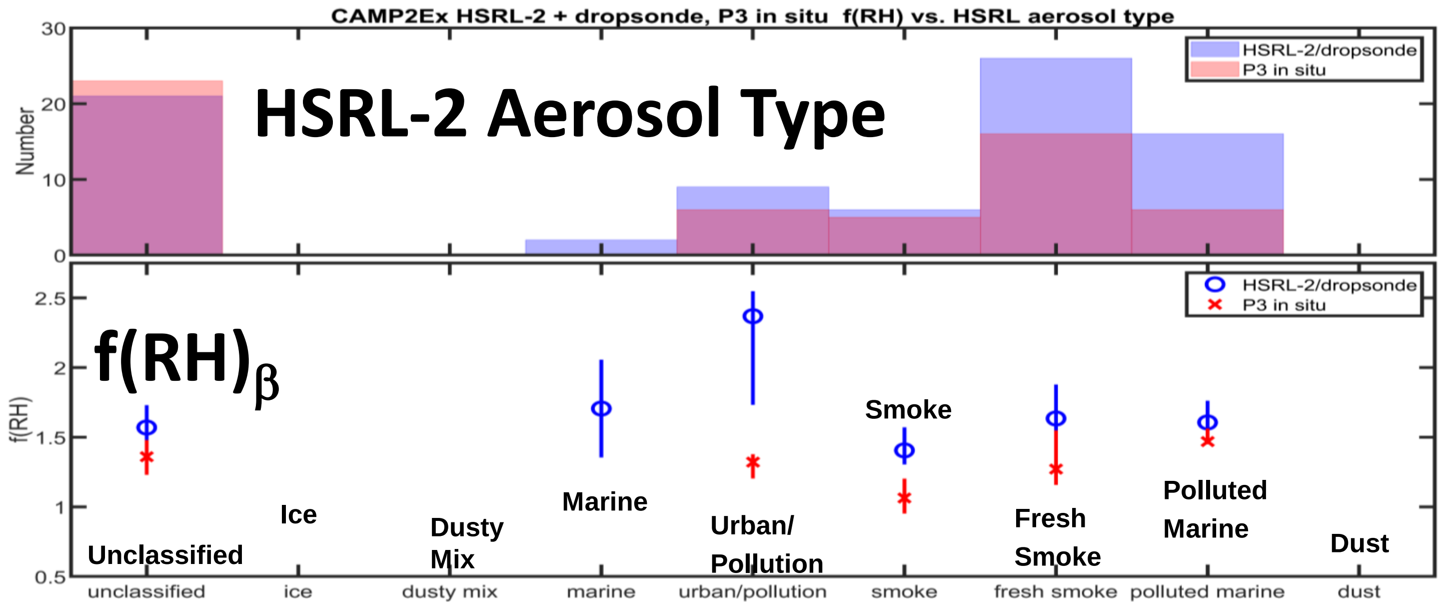
- Example from CAMP2Ex Sept. 15, 2019 flight
  - AOT (~0.8-1.0) dominated by smoke
  - In situ f(RH) ~ 0.82-1.0
  - HSRL-2/dropsonde f(RH) 1.3-1.8

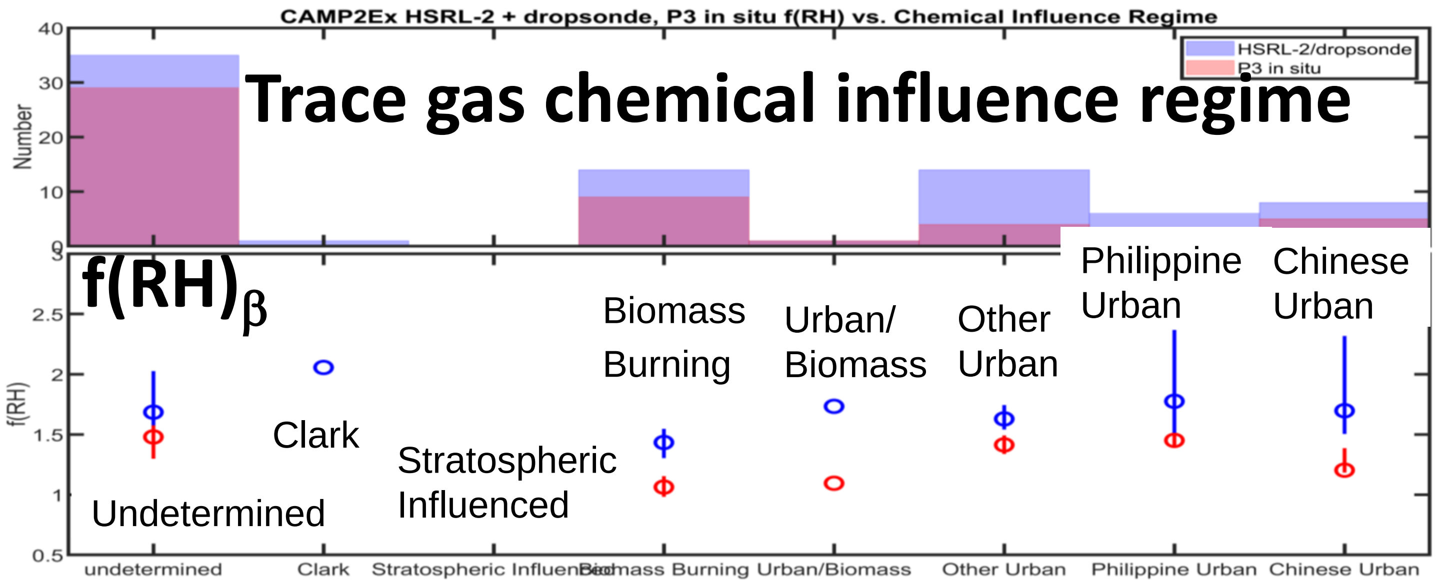




HSRL-2/dropsonde humidification [f(RH)] factors related to HSRL-2 aerosol type and CAMP2EX chemical influence flag

- HSRL-2 measurements of aerosol intensive variables used to infer aerosol type (Burton et al., 2012)
- Trace Gas CO, CH_4, and O_3 and DLH H_2O (Diskin group) measurements used to separate chemical regimes
- During CAMP2Ex, f(RH) values derived from HSRL-2/dropsonde data were somewhat higher for urban/pollution and lower for biomass burning
- HSRL-2/dropsonde f(RH) values higher than in situ for all aerosol types and appear most consistent with previous values for marine & urban aerosol





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

f(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	1.36 ± 0.27
Marine	Urban	Background	Free Trop.

HSRL-2/dropsonde humidification [f(RH)] factors related to HSRL-2 aerosol type during ACTIVATE

- HSRL-2 measurements of aerosol intensive variables used to infer aerosol type (Burton et al., 2012)
- During ACTIVATE, f(RH) values derived from HSRL-2/dropsonde data were somewhat higher for urban/pollution/smoke and lower for marine
- HSRL-2/dropsonde f(RH) values higher than in situ for nearly all aerosol types
- HSRL-2/dropsonde f(RH) appear most consistent with previous values for marine & urban aerosol

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

f(RH=80%)	1.08 ± 0.13	0.99 ± 0.06	1.41 ± 0.13
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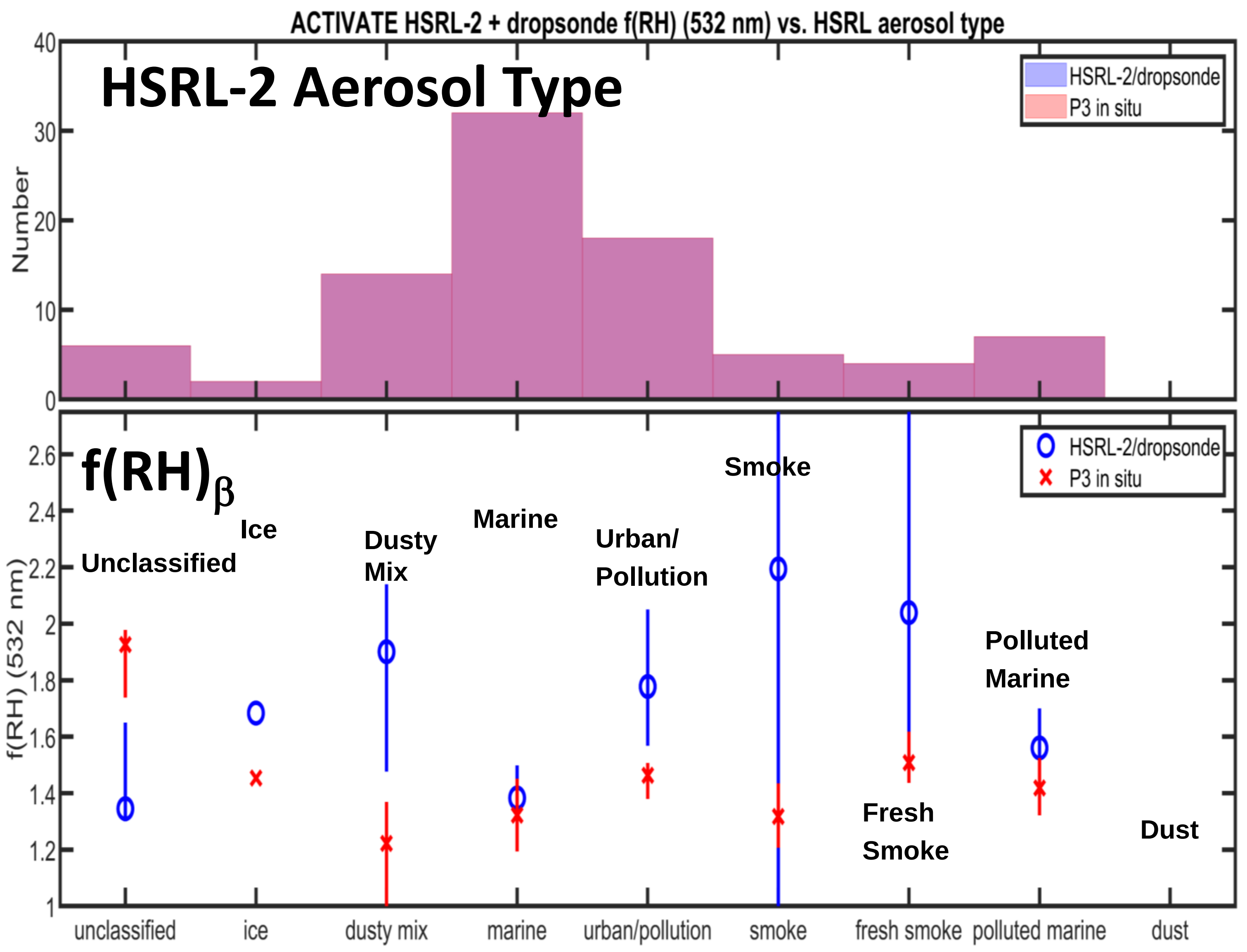
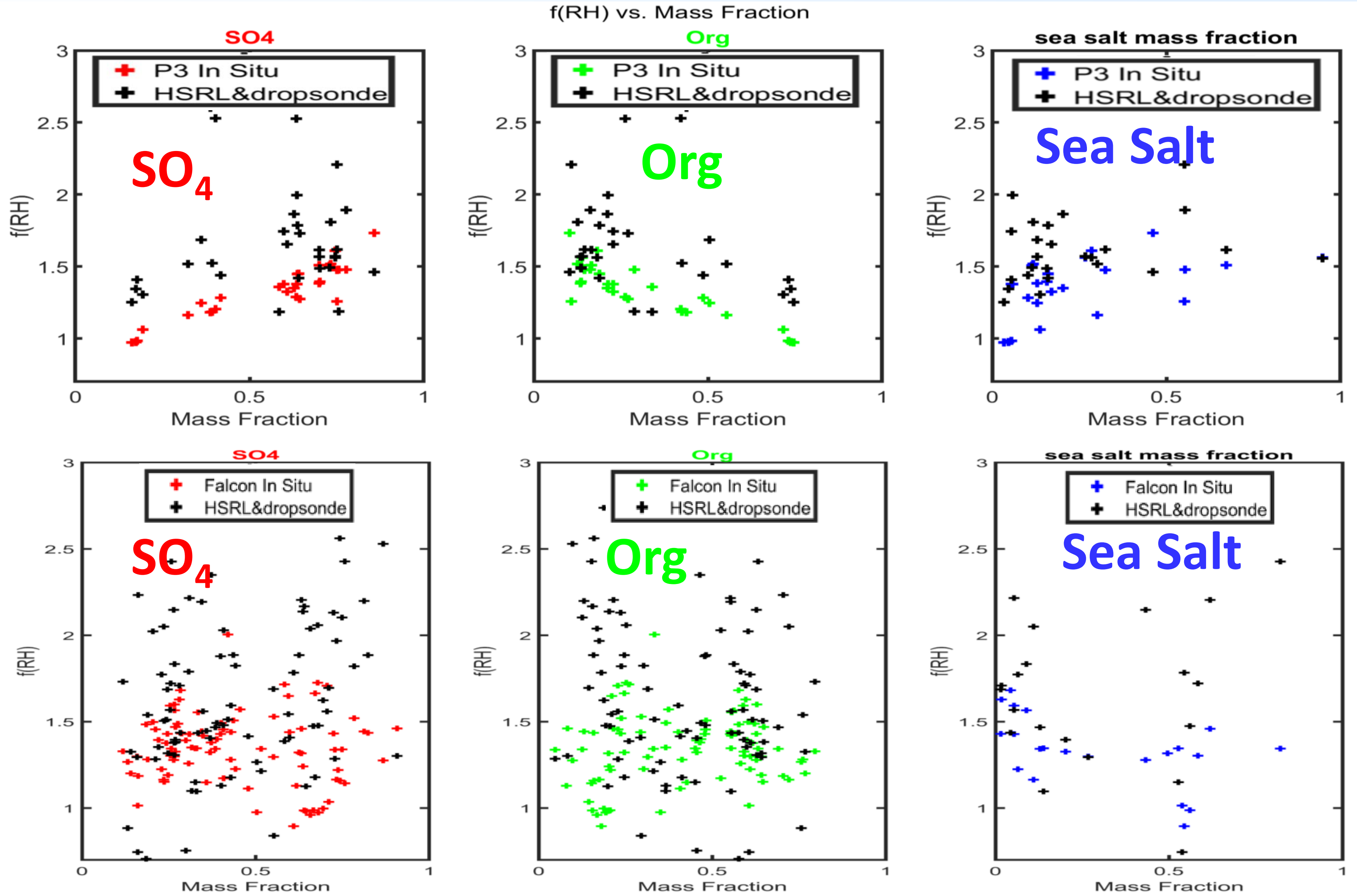


HSRL-2+dropsonde f(RH) values show little correlation with in situ measurements of fine mode mass fraction

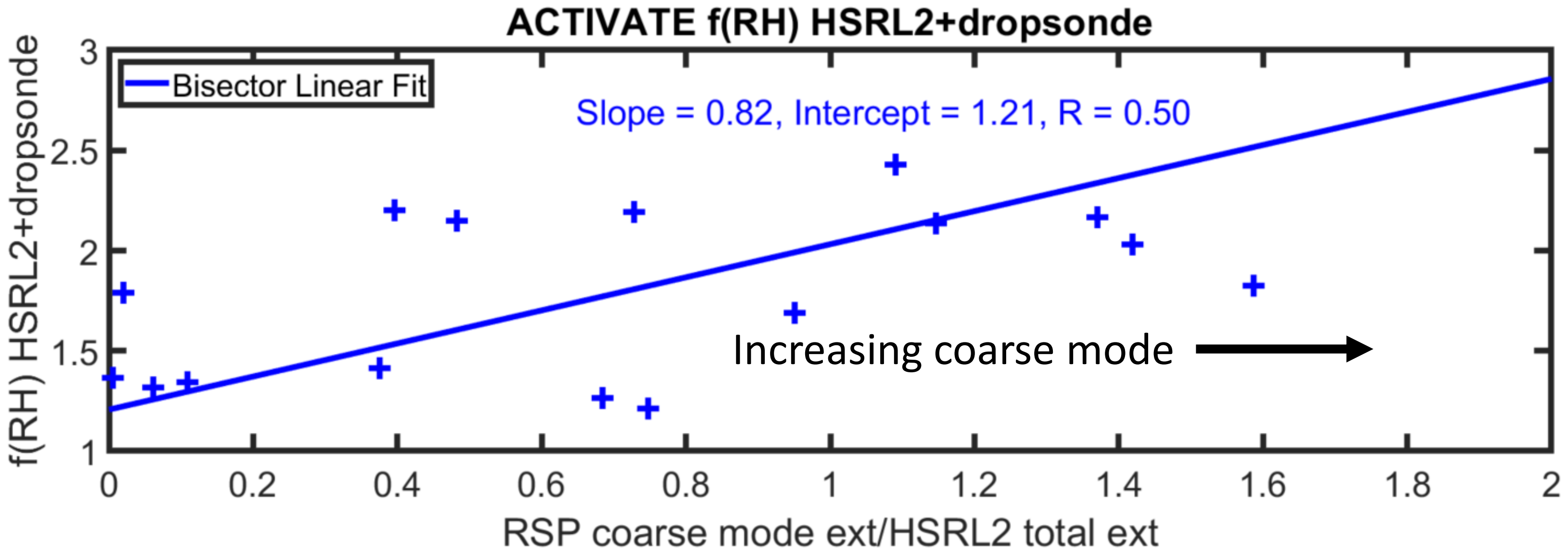
CAMP2Ex

ACTIVATE

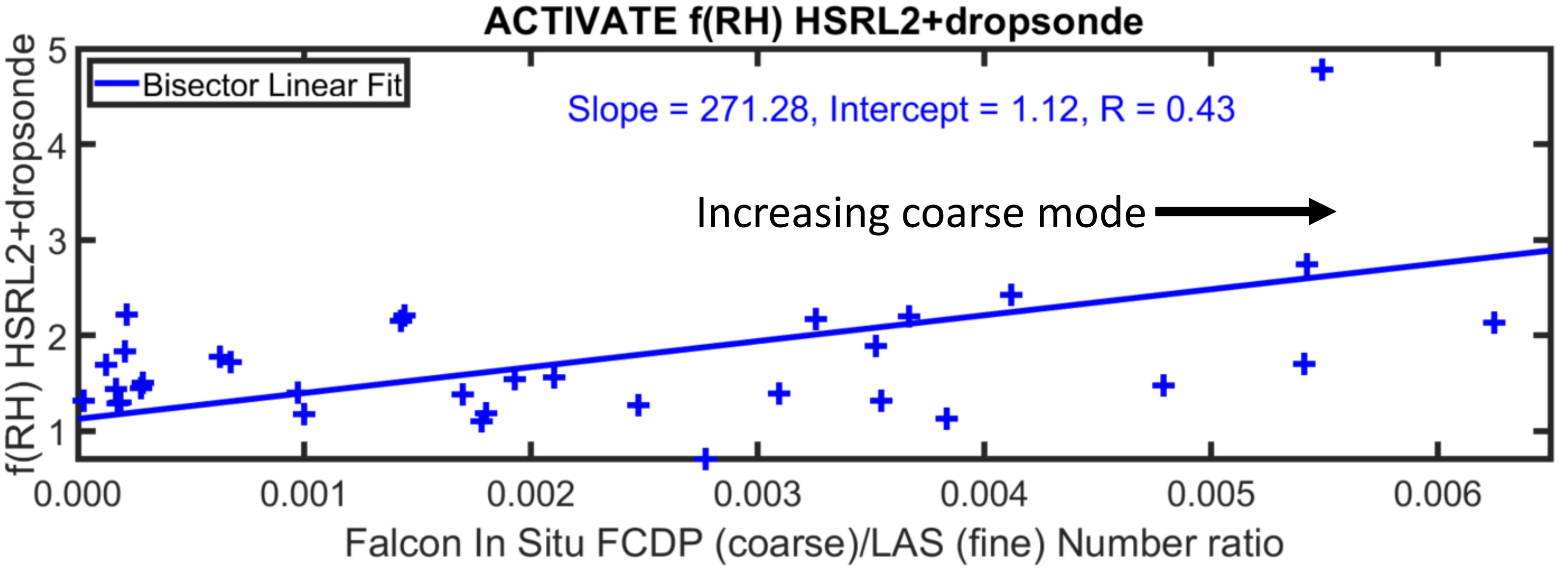


HSRL-2+dropsonde retrievals of f(RH) increase with increasing coarse mode aerosol fraction – f(RH) vs. coarse mode fraction

Coarse mode extinction estimated from RSP retrievals

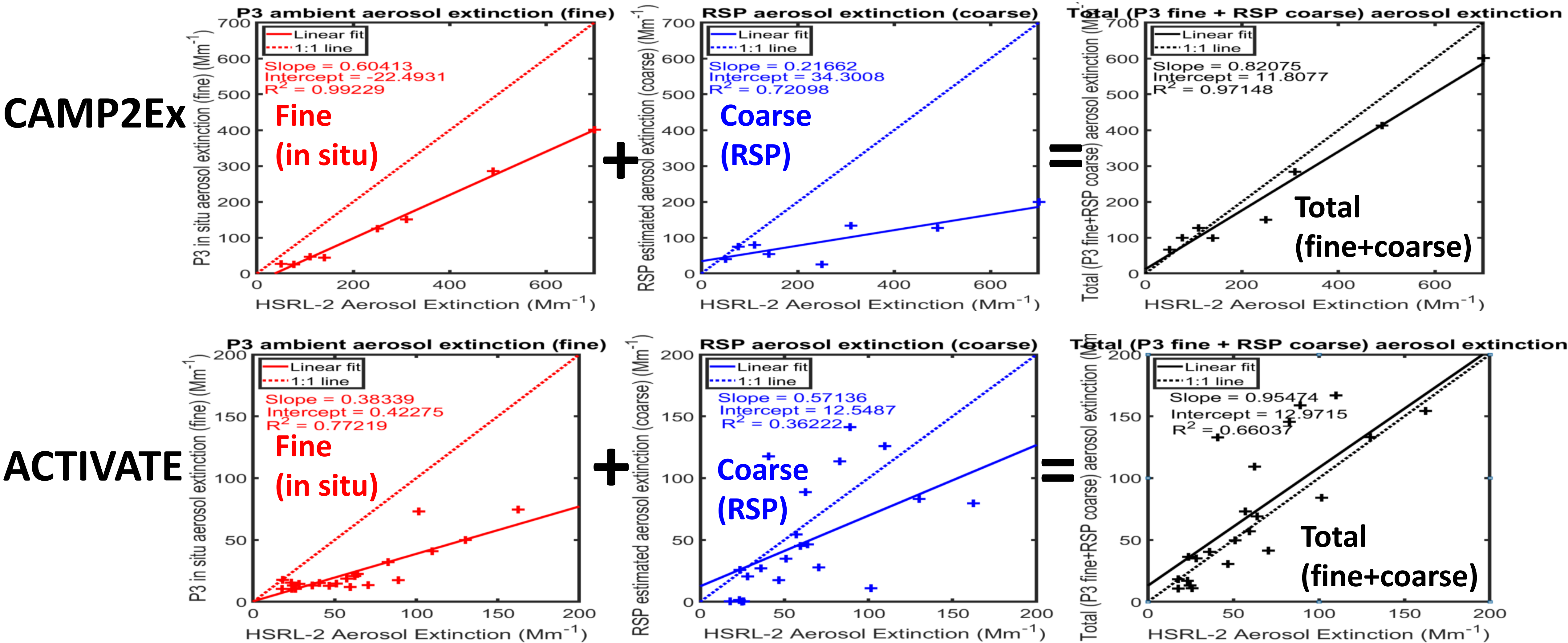


Coarse mode fraction from in situ (FCDP number/ LAS fine mode number)



Aerosol Extinction Comparisons

HSRL-2 aerosol extinction compares best with sum of fine (in situ) and coarse (RSP) aerosol extinction

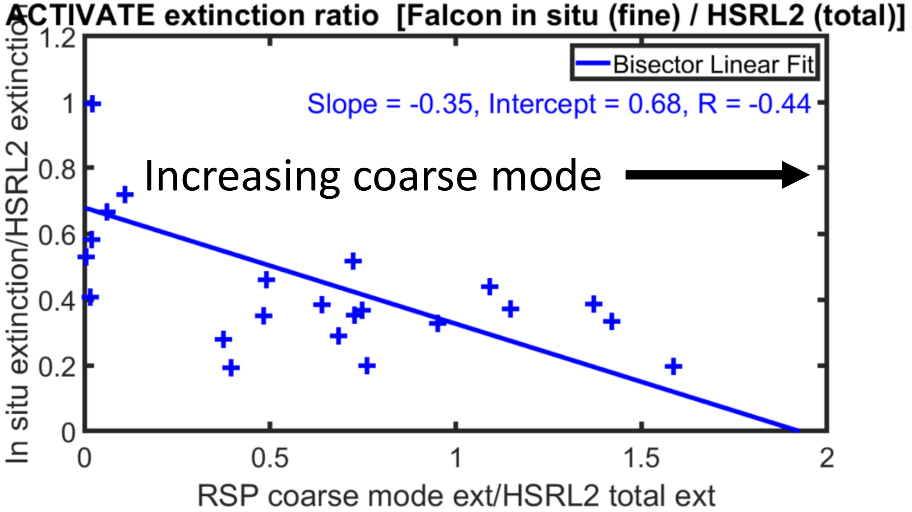


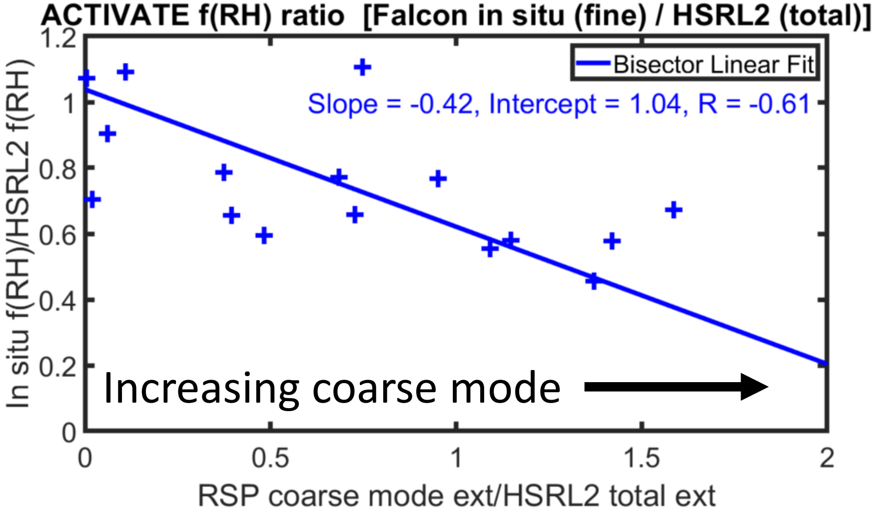
Ratios of In Situ/HSRL-2 Aerosol Extinction and In Siu/HSRL f(RH) decrease with increasing coarse mode aerosol fraction

Extinction Ratio

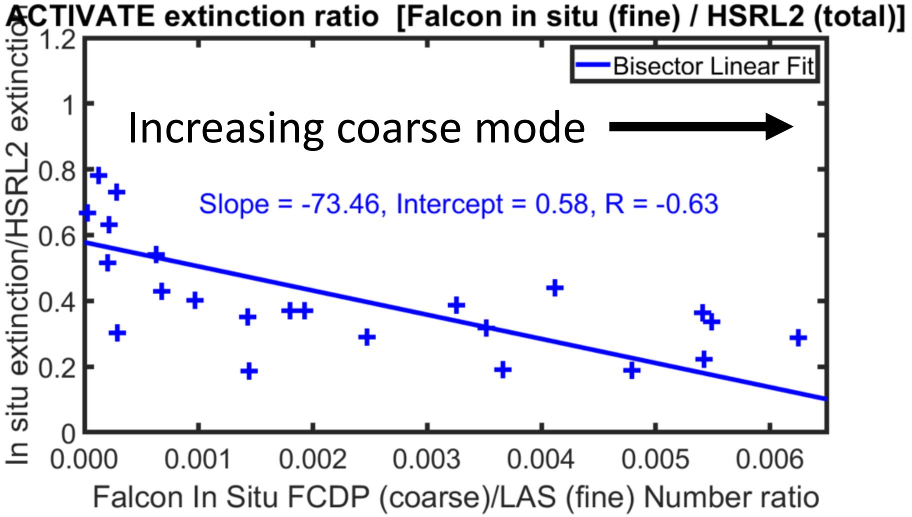
f(RH) Ratio

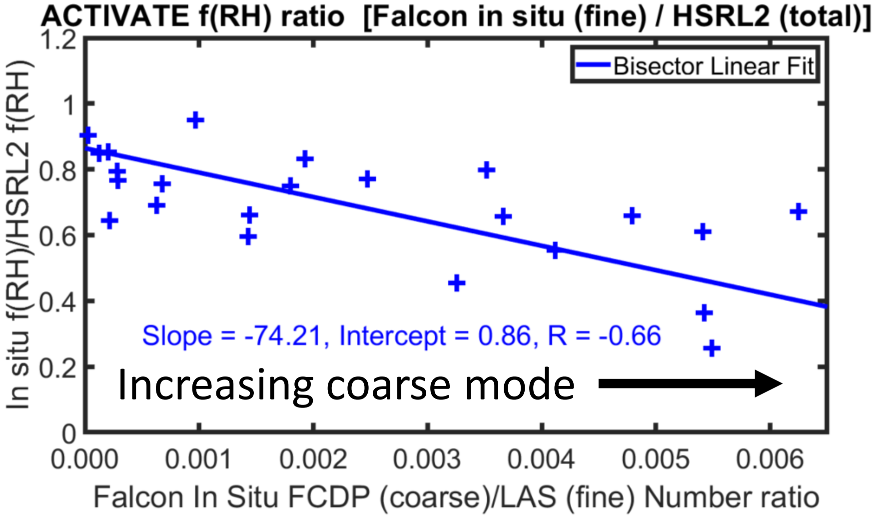
Coarse mode extinction estimated from RSP retrievals





Coarse mode fraction from in situ (FCDP number/ LAS fine mode number)



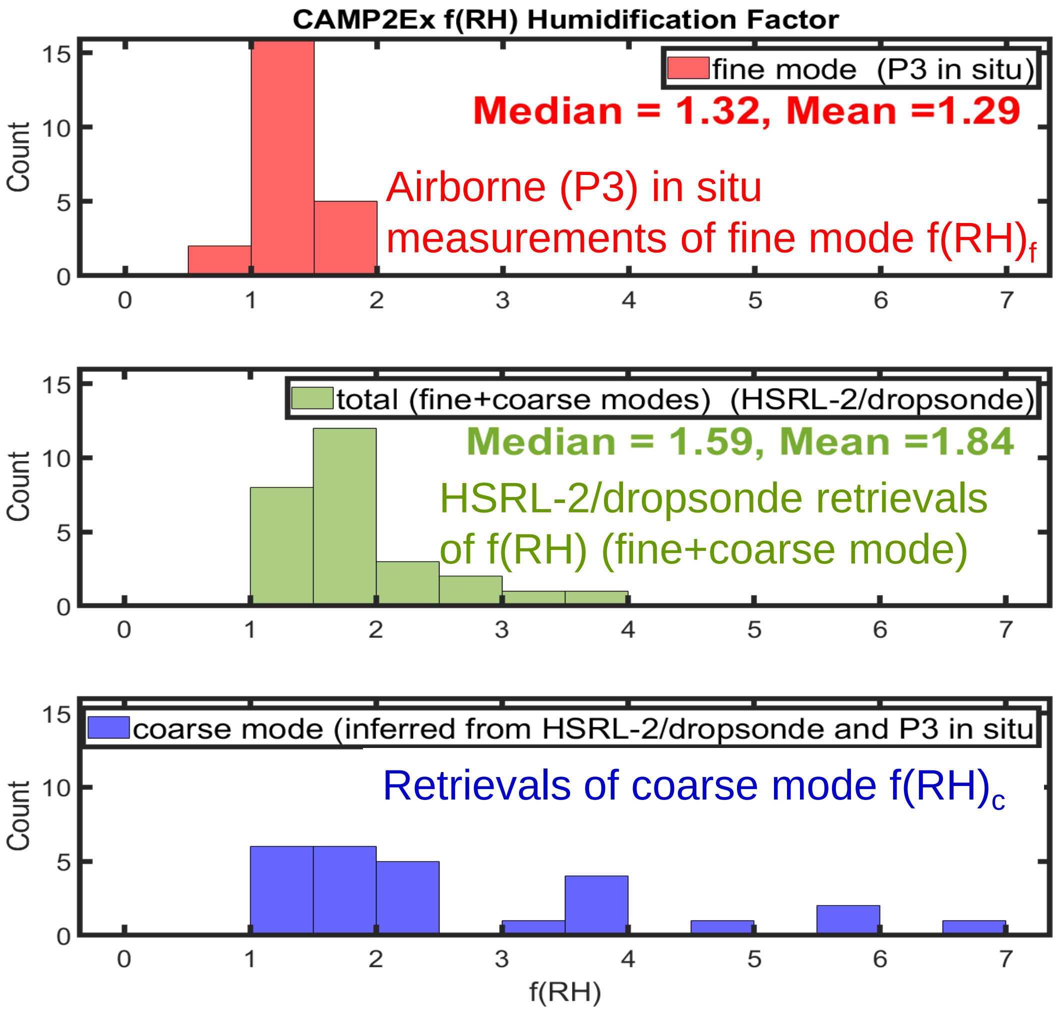


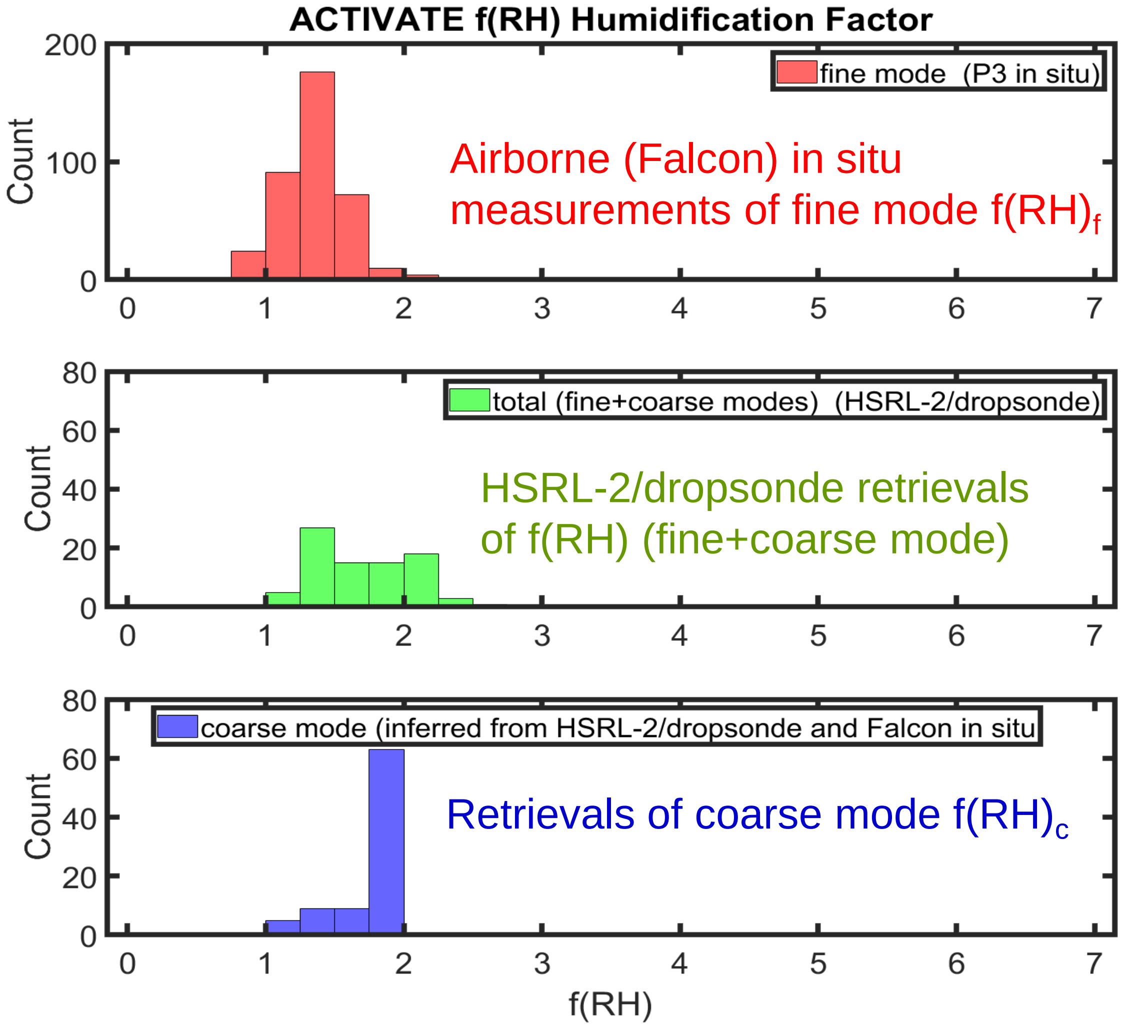
Inferring f(RH) for coarse mode sea salt

- HSRL-2/dropsonde humidification factors are larger than airborne in situ values because lidar observes both fine and coarse (sea salt) aerosol in contrast to in situ measurements of only fine mode. Therefore, we attempted to infer coarse mode humidification factors.
- Assumptions:
  – Coarse mode aerosol is comprised of sea salt and is entirely within PBL
  – In situ measurements of f(RH) correspond to only fine mode
- Coarse mode aerosol extinction is estimated from Research Scanning Polarimeter (RSP) retrievals of coarse mode AOD and HSRL-2 retrievals of PBL height
- Fine mode aerosol extinction is derived from HSRL-2 measurements of total aerosol extinction and estimates of coarse mode aerosol extinction
- Coarse mode $f(RH)_c$ is derived from change in aerosol extinction with RH measured by HSRL-2, in situ measurements of fine mode $f(RH)_f$, and estimates of fine and coarse mode aerosol extinction

Inferring f(RH) for coarse mode sea salt

Mean values of coarse mode $f(RH)_c$ are around 2 and are consistent with values from literature (e.g. Titos et al. 2016, Atm. Environ.)





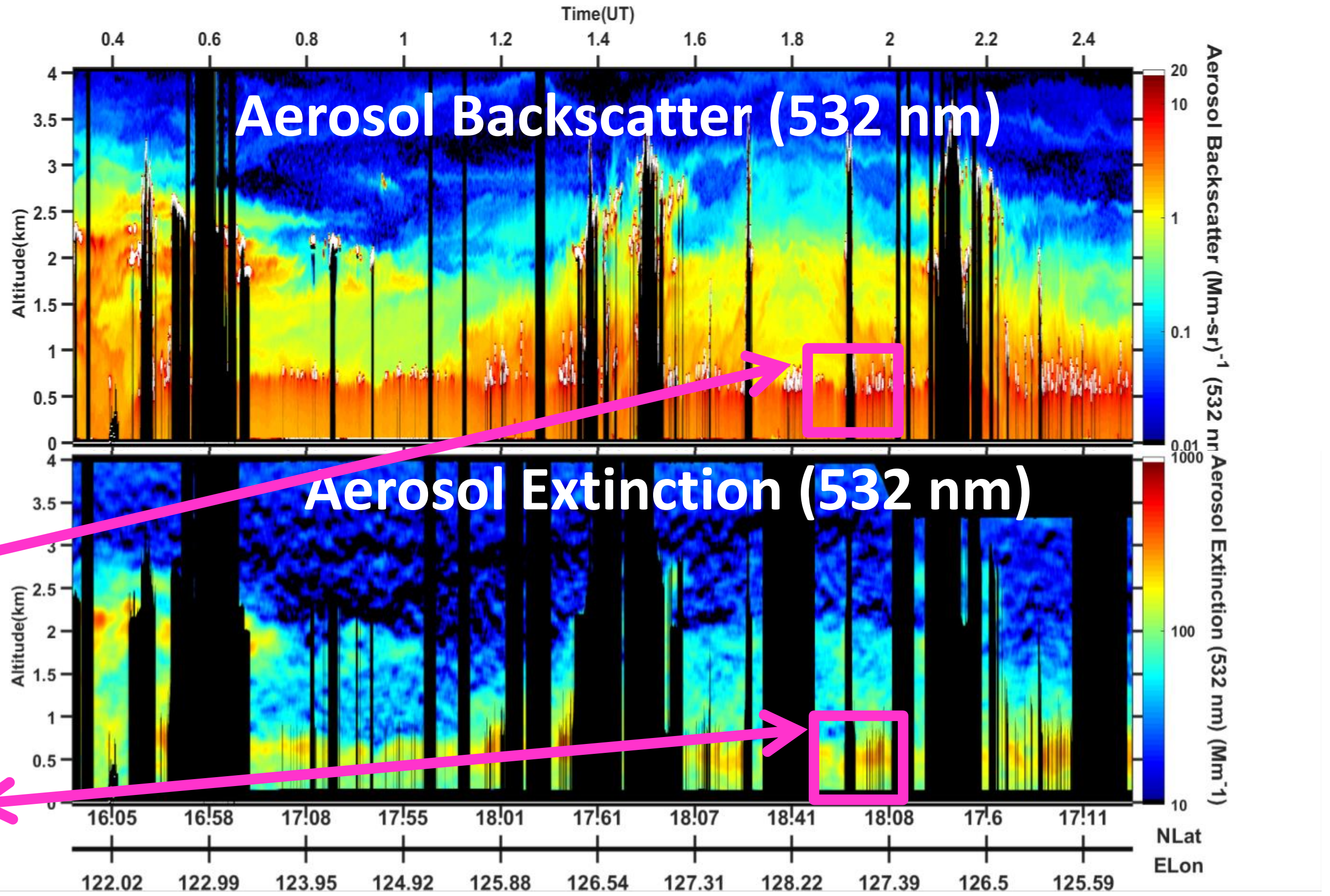
Summary

- Aerosol humidification factors [e.g. f(RH)] for aerosols within well mixed PBL are derived using HSRL-2 measurements of aerosol backscatter and dropsonde measurements of RH during NASA CAMP2EX and ACTIVATE missions.
- Average $f(RH=80\%/RH=20\%)_{\beta}$ (532 nm) was about 1.7, $f(RH)_{\sigma}$~2, during CAMP2Ex & ACTIVATE.
- These values were higher than values from airborne in situ measurements (1.30-1.39). Higher f(RH) values derived from HSRL-2 & dropsonde data are likely because lidar observes both fine and coarse (sea salt) aerosol in contrast to in situ measurements of only fine mode aerosol.
- Higher f(RH) values were derived for urban aerosols and lower values for biomass burning.
- Higher values of f(RH) for aerosol extinction than for aerosol backscatter indicates lidar ratio increases with RH.
- Estimates of coarse mode (sea salt) f(RH) are around 2-3 based on HSRL-2, in situ f(RH), RSP measurements and retrievals. These values are consistent with previous measurements.
- In situ and RSP retrievals indicate that the ratios of in situ/HSRL-2 extinction and in situ/HSRL2 f(RH) decrease with increasing coarse mode aerosols

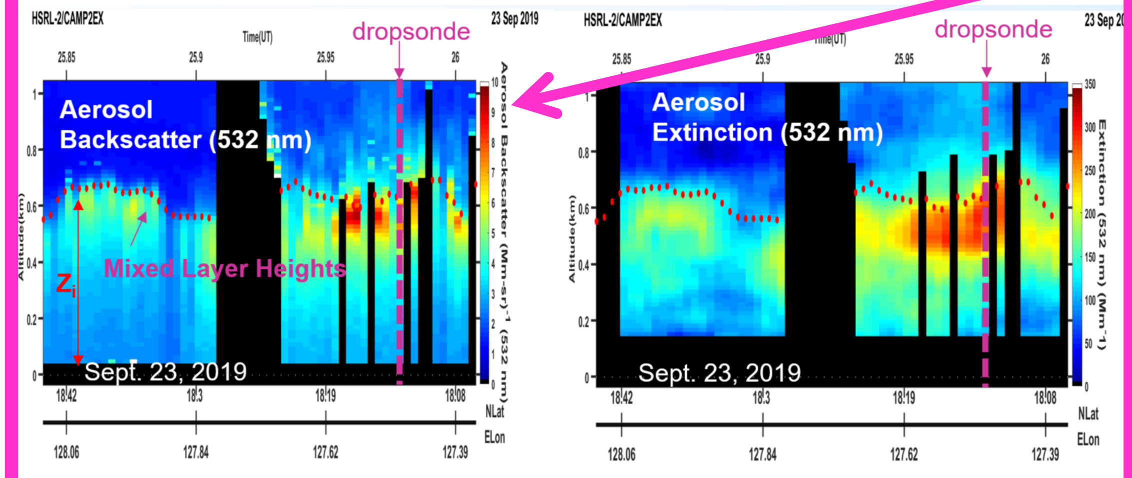
HSRL-2 Products from CAMP2Ex and ACTIVATE

- Aerosol Backscatter and Depolarization Profiles (355, 532, 1064 nm)
- Aerosol Extinction, Lidar Ratio, and AOT Profiles (355 and 532 nm)
- Aerosol Color Ratio Profiles (1064/532, 532/355)
- Aerosol Type
- Mixed Layer Heights
- Aerosol humidification enhancement factors for aerosols within well-mixed PBL are computed using HSRL-2 measurements of aerosol backscatter and dropsonde measurements of RH

HSRL-2 Aerosol Measurements show Variability with Relative Humidity



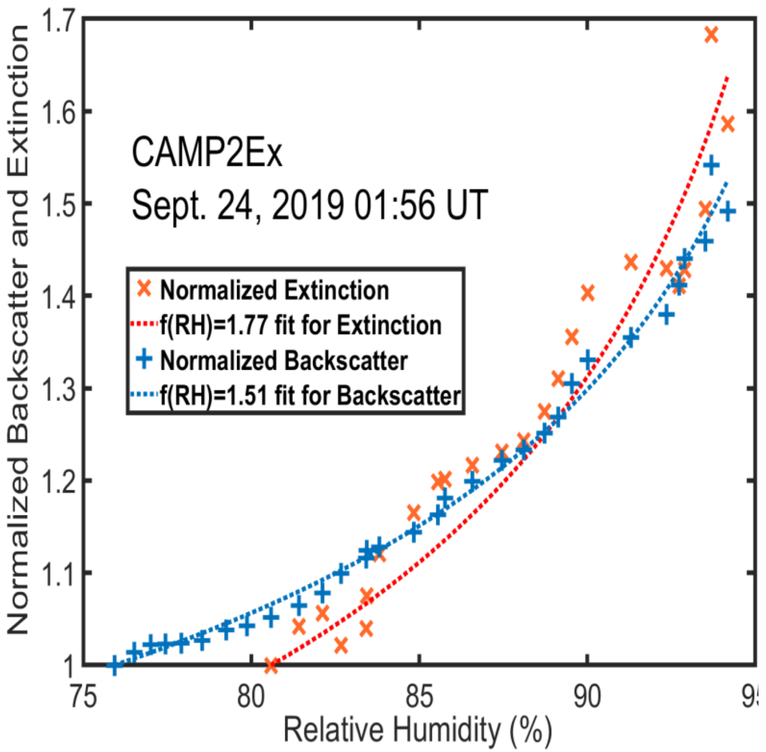
Expanded View of Aerosol Backscatter and Extinction Images

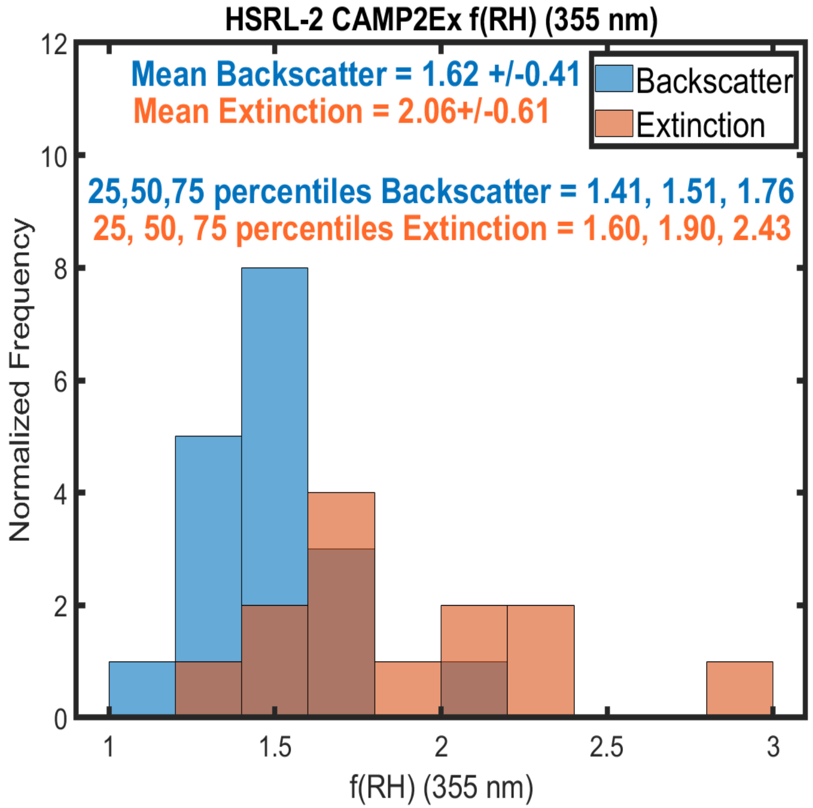


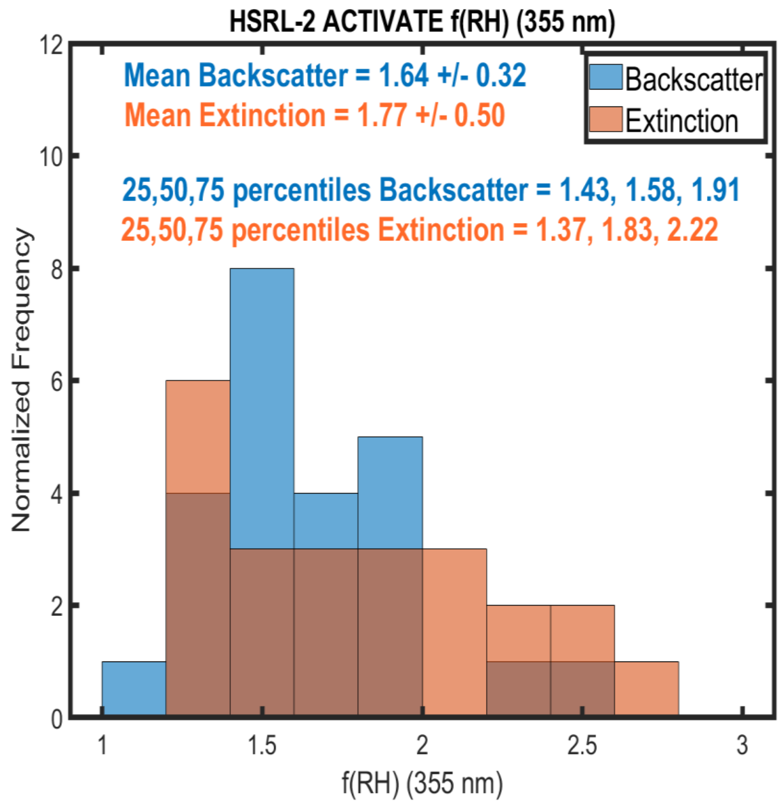
HSRL-2 data from CAMP2Ex at
https://www-air.larc.nasa.gov/cgi-bin/ArcView/camp2ex#HOSTETLER.CHRIS/
HSRL-2 data from ACTIVATE at
https://www-air.larc.nasa.gov/cgi-bin/ArcView/activate.2019#HOSTETLER.CHRIS/

Humidification Factors ([f(RH)] for aerosol extinction slightly higher than for aerosol backscatter

- $f(RH)_\sigma$ for aerosol extinction was slightly higher than $f(RH)_\beta$ for aerosol backscatter
- Consequently, aerosol extinction/backscatter ratio ("lidar ratio") increases 5-30% with increasing RH, depending on the range of RH
- Other aerosol intensive parameters (e.g. depolarization, color ratio) also vary with RH







Gamma vs. Kappa Fit for f(RH)

“Gamma” relationship was found to provide a consistently better fit to the HSRL-2/dropsonde data than the “kappa” fit (e.g. Brock et al., 2016, ACP)

“Gamma” $$f(RH) = \frac{\beta(RH)}{\beta(RH_o)} = \left[\frac{(100-RH_o)}{(100-RH)}\right]^{\gamma}$$

“Kappa” $$f(RH) \cong 1 + \kappa_{bsc}\left[\frac{RH}{100-RH}\right]$$