Retrieval of ambient aerosol properties from In-situ data

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With direct collaboration from the HSRL-2 and the LARGE teams

Objective and Significance

- The objective of the in-situ aerosol retrieval algorithm (ISARA) is to derive ambient aerosol optical and microphysical products from a suite of direct (i.e., in-situ) measurements
- In particular, the algorithm produces ambient aerosol size and complex refractive index, for both fine-mode and coarse-mode aerosol size distributions
- The ISARA aerosol products can be directly compared to polarimeter aerosol products and to high spectral resolution lidar aerosol measurements
- The ISARA aerosol products can also provide insight into aerosol behavior in complicated atmospheric environments and near clouds

PACE-PAX In-situ Measurements Summary

- Dry aerosol particle size resolved number concentrations
  - DMT UHSAS (optical particle diameters from 0.06 to 1.1 µm)
  - TSI-3321 APS (aerodynamic particle diameters from 1.1 to ~5 µm)
- Dry aerosol particle scattering coefficient at 450, 550, 700 nm wavelengths
  - TSI Scatter Nephelometer
- Dry aerosol particle absorption coefficient at 467, 530 and 660 nm wavelengths
  - PSAP
- Hygroscopic growth function
  - TSI Scatter Nephelometer
- Ambient relative humidity (RH)
  - DLH

<u>Additional Analyses and Measurements</u>

- Inlet efficiency and cut-off based on LARGE/CIRPAS analysis
- Real refractive index (RRI) constrained by LI-Nephelometer

Methods Summary

Step 1 (novel): Alignment of UHSAS and APS (Salibas et al., 20019)

Step 2: Retrieve dry complex refractive index (CRI)

- Spectral dry scattering and absorption coefficients
- Dry aerosol size resolved number concentration (n)

Step 3: Retrieve hygroscopicity (κ)

- Hygroscopic growth function (f(RH))

Step 4: Calculate ambient aerosol properties (Schlosser et al, 2025)

Step 5: Collocation (Schlosser et al., 2024)

Note: All aerosol properties calculated with the Modeled optical properties of ensembles of aerosol particles (MOPSMAP) package

- Schlosser et al., Closing the gap between in-situ and remotely sensed aerosol particle properties, TBD, Atmos. Meas. Tech.
- Schlosser, J. S., and Coauthors, 2024: Maximizing the Volume of Collocated Data from Two Coordinated Suborbital Platforms. J. Atmos. Oceanic Technol., 41, 189–201, https://doi.org/10.1175/JTECH-D-23-0001.1.
- Gasteiger, J. and Wiegner, M.: MOPSMAP v1.0: a versatile tool for the modeling of aerosol optical properties, Geosci. Model Dev., 11, 2739–2762, https://doi.org/10.5194/gmd-11-2739-2018, 2018.
- G. Saliba, et al., Factors driving the seasonal and hourly variability of sea-spray aerosol number in the North Atlantic, Proc. Natl. Acad. Sci. U.S.A. 116 (41) 20309-20314, https://doi.org/10.1073/pnas.1907574116 (2019).

Algorithm

Step 1: Alignment of UHSAS and APS.

$$D_{APS}(\rho) = \frac{D_{UHSAS}}{\sqrt{\rho}}$$

$\rho: < 0.9 : 0.1 : 3.0 > \mathrm{g\ cm^{-3}}$

Measured dry size distributions ($n_{\mathrm{UHSAS}}(\mathrm{D_{UHSAS}})$ and $n_{\mathrm{APS}}(\mathrm{D_{APS}})$)

→

Fit $n_{\mathrm{UHSAS}}(\mathrm{D_{UHSAS}})$ a the power function

$$y^{p}(\mathrm{D_{UHSAS}}) \approx \mathrm{C1} \cdot \mathrm{D_{UHSAS}^{-C2}}$$

→

Iteratively compute objective function for range of ρ given the changing number of overlapping bins ($\mathrm{n_{bins}}$) at each step.

$$O(\rho) = \mathrm{n_{bins}^{-1}} \sum_{\mathrm{i=1}}^{\mathrm{n_{bins}}} \log^{2} \frac{y^{p}(\mathrm{D_{UHSAS}})}{n_{\mathrm{APS}}(\mathrm{D_{APS}})}$$

→

$$\bar{\boldsymbol{\rho}} = \rho \text{ where } O(\rho) = \min(O(\rho))$$

Note: 1100 nm stitching point

Algorithm

Step 2: Retrieve dry complex refractive index (CRI)

$$\mathrm{CRI} = \mathrm{RRI} + \mathrm{IRI} * i$$

RRI: 1.55
IRI: <0.00,0.001:0.001:0.08>

Measured dry scattering and absorption coefficient (C_{dry})

Measured dry size distribution → Compute scattering and absorption coefficient (C_{calc}) →

$$\zeta_{\mathrm{sca}} = \left|\frac{C_{\mathrm{calc}} - C_{\mathrm{dry}}}{C_{\mathrm{dry}}}\right|_{450,550,700}^{\mathrm{sca}} \times 100\%$$

$$\zeta_{\mathrm{abs}} = \left|C_{\mathrm{calc}} - C_{\mathrm{dry}}\right|_{470,532,660}^{\mathrm{abs}}$$

$\overline{\mathrm{IRI}}$ = mean(**IRI**), For all **IRI** where $\zeta_{\mathrm{sca}} < 20\%$ and $\zeta_{\mathrm{abs}} < 1\ \mathrm{Mm}^{-1}$ in all three wavelengths

Algorithm

Step 3: Retrieve hygroscopicity (κ)

$$\left(\frac{D_{\mathrm{amb}}}{D_{\mathrm{dry}}}\right)^3 = g^3 = 1 + \kappa * \frac{\mathrm{RH}}{100 - \mathrm{RH}}$$

κ: <0.01:0.01:1.40>
CRI

Humidified scattering coefficient ($C_{\mathrm{wet}} = C_{\mathrm{dry}} * f(\mathrm{RH})$)

Measured dry size distribution
Ambient RH

Compute scattering coefficient (C_{calc})

$$\zeta_{\mathrm{sca}} = \left|\frac{C_{\mathrm{calc}} - C_{\mathrm{wet}}}{C_{\mathrm{wet}}}\right|_{550}^{\mathrm{sca}} \times 100\%$$

$\bar{\kappa} = \kappa$, for smallest κ where $\zeta_{\mathrm{sca}} < 1\%$

Ambient ISARA products

Inputs

- $n_{\mathrm{UHSAS}}(\mathrm{D}_{\mathrm{UHSAS}})$
- $n_{\mathrm{APS}}(\mathrm{D}_{\mathrm{APS}})$
- Ambient RH

Retrieved

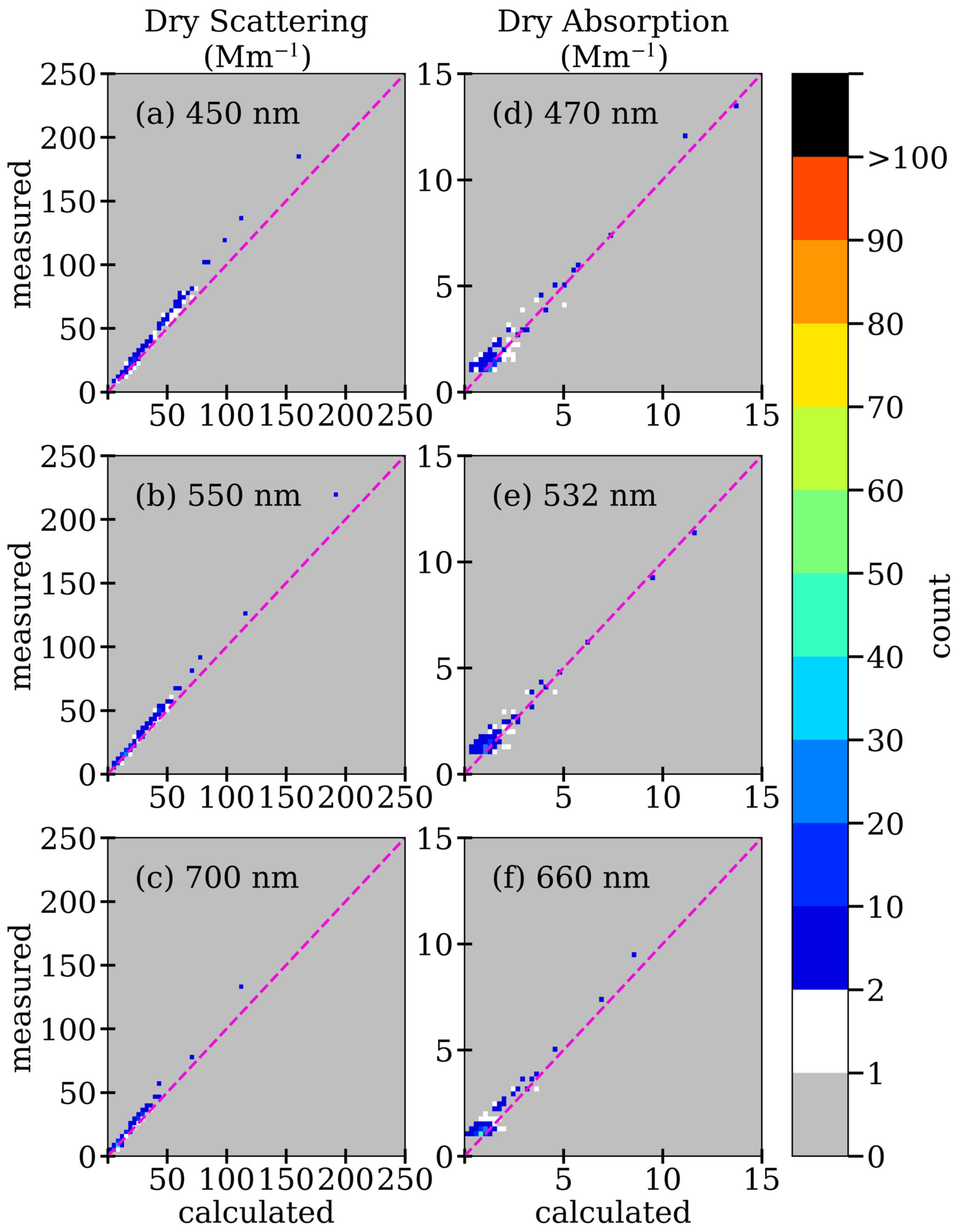
ρ, κ, and CRI

Ambient aerosol properties:

- Number, surface and volume concentrations
- Spectral extinction coefficient and cross section
- Spectral single scattering albedo

Internal Closure (Dry)

- All results from PACE-PAX
- Scattering coefficients agree to within 2.5%
- Absorption coefficients agree to within 5%



	Scattering			Absorption		
Wavelength (nm)	450	550	700	470	532	660
NMAD (%)	2.1	1.8	2.4	2.3	2.8	4.3

Internal Closure (Dry)

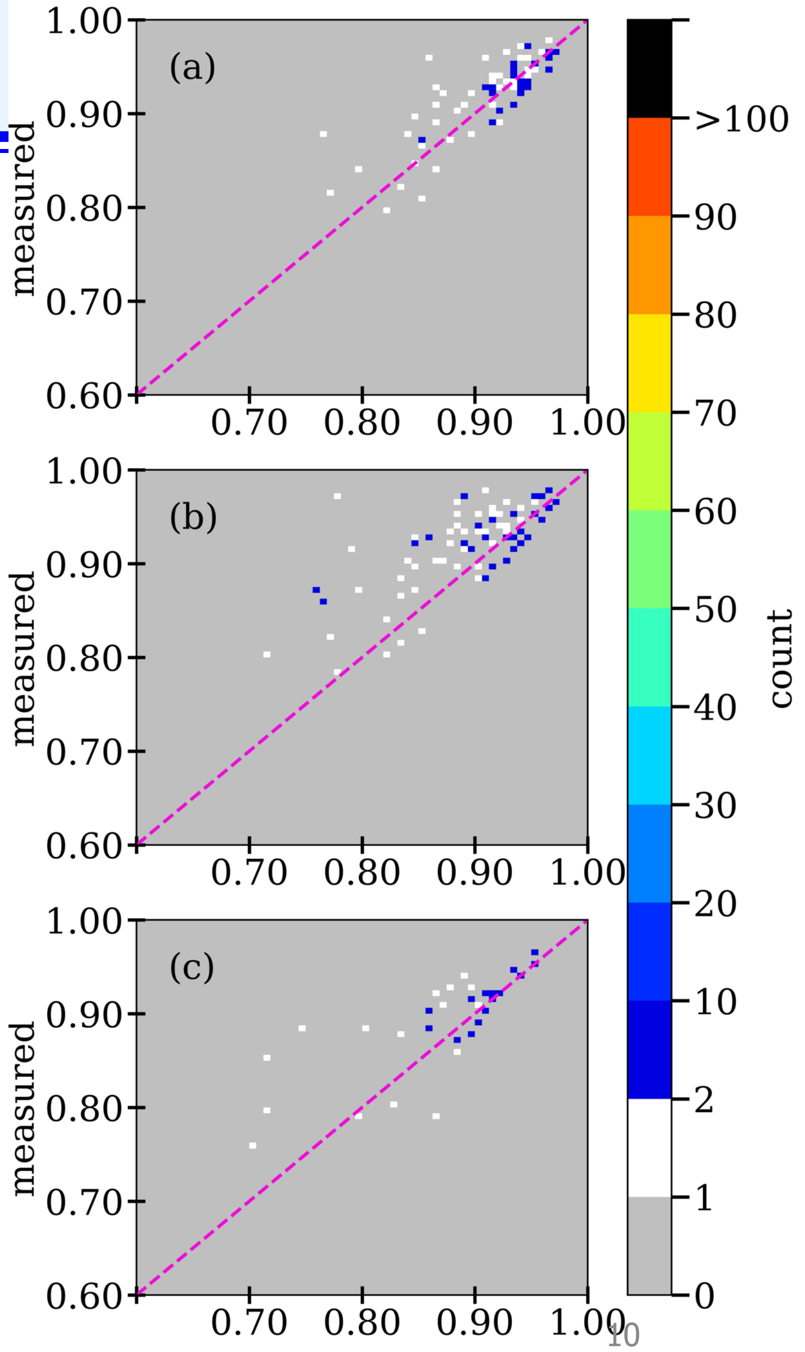
- Single scattering albedo (SSA) to within 15%.

Normalized-range mean absolute deviation (NMAD)

$$\sum_{j=1}^{n} \frac{|Y_j - X_j|}{n} \cdot \frac{100\%}{\max(X) - \min(X)}$$

where Y_j is set of calculated data, X_j is set of measurement data, and n is the total number of points.

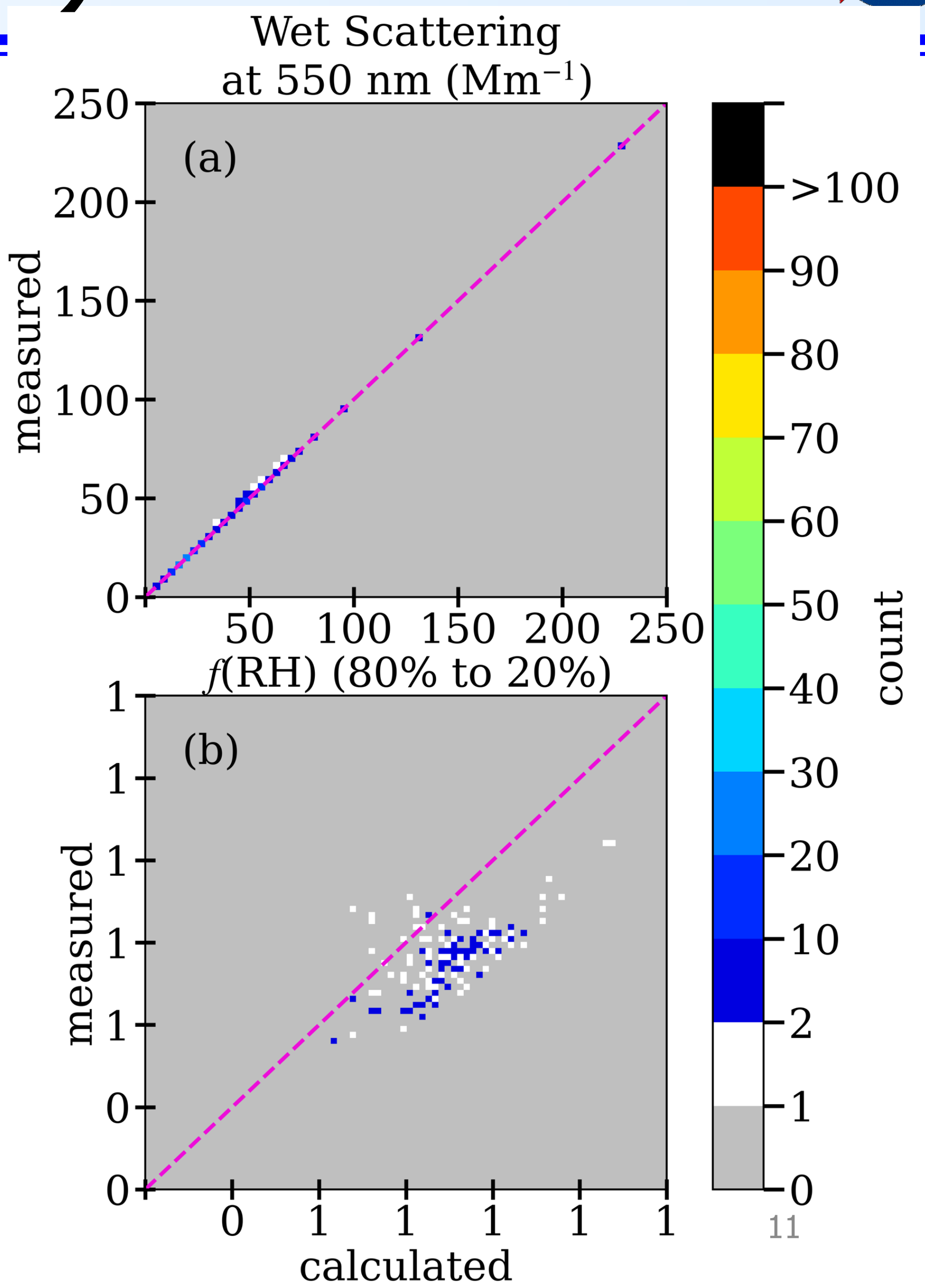
	SSA		
Wavelength (nm)	450	550	700
NMAD (%)	9.2	13.1	10.8



Internal Closure (Humidified)

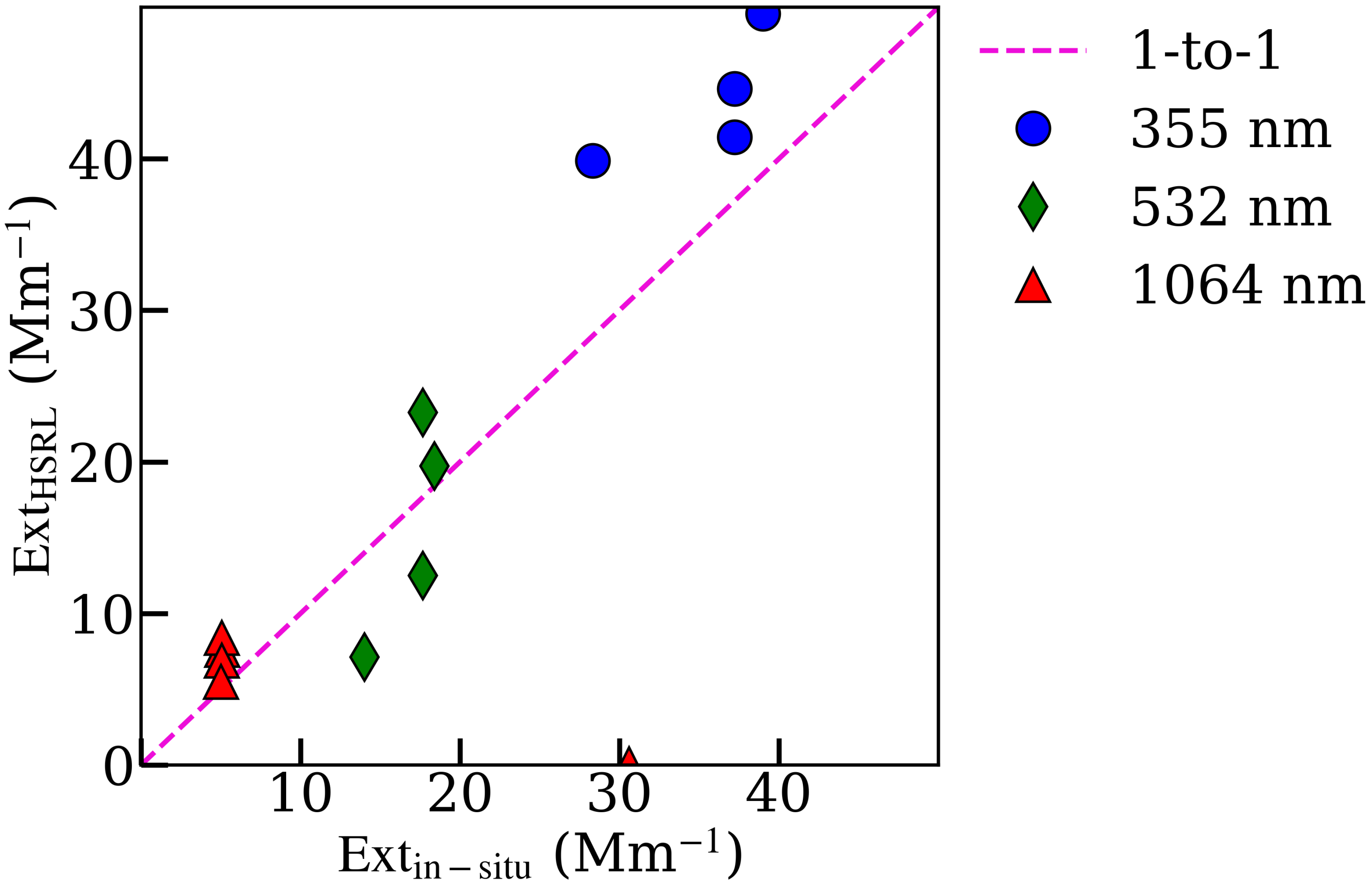
- Humidified (wet) scattering at 550 nm agrees to within 0.5%
- f(RH) agrees to within 7%

	Extinction	f(RH)
NMAD (%)	0.2	21



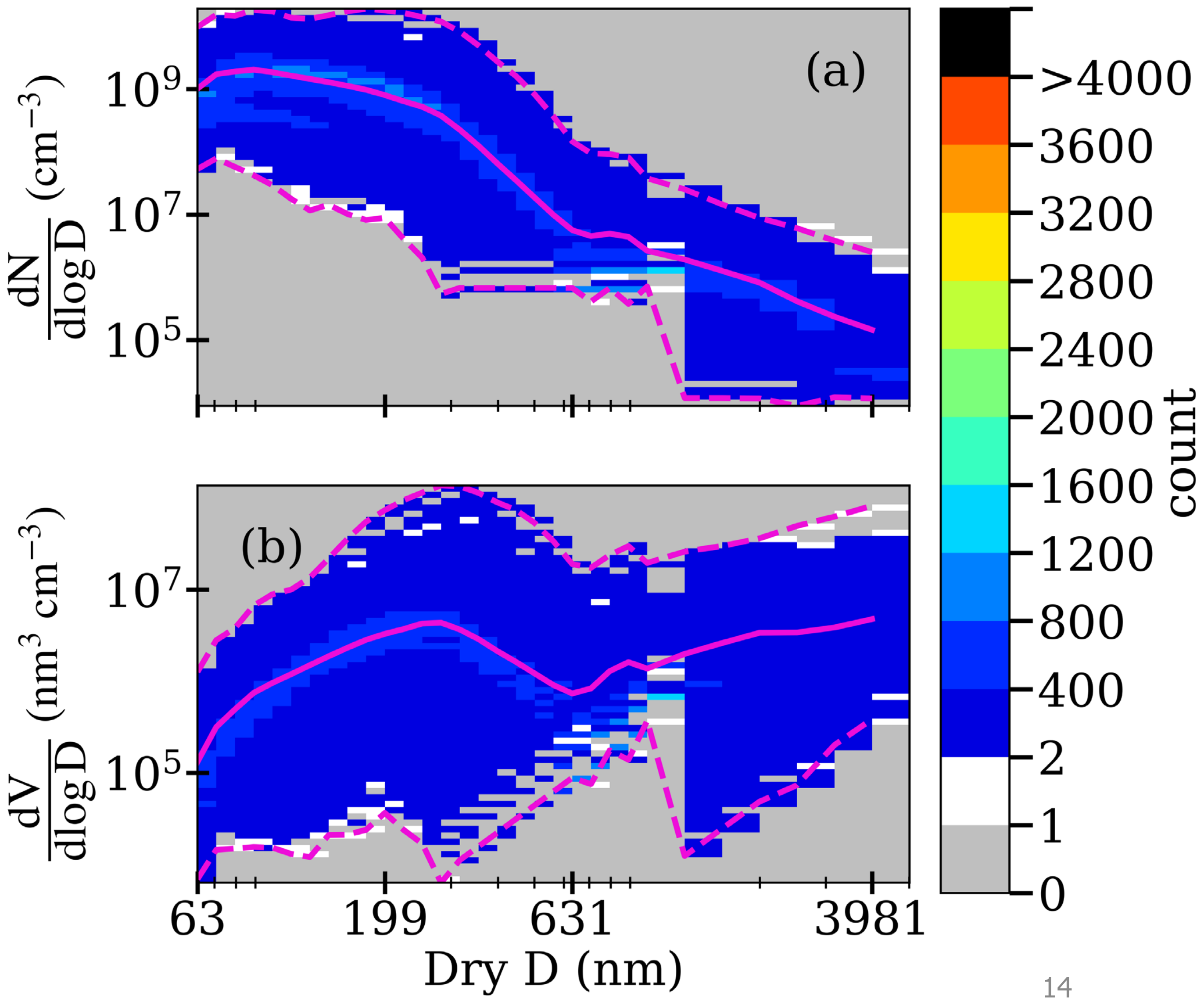
External Closure

- HSRL-2 derived vs. in-situ derived spectral ambient aerosol extinction
- 532nm (NMAD: 26%, Count: 5, p-value: 0.01)

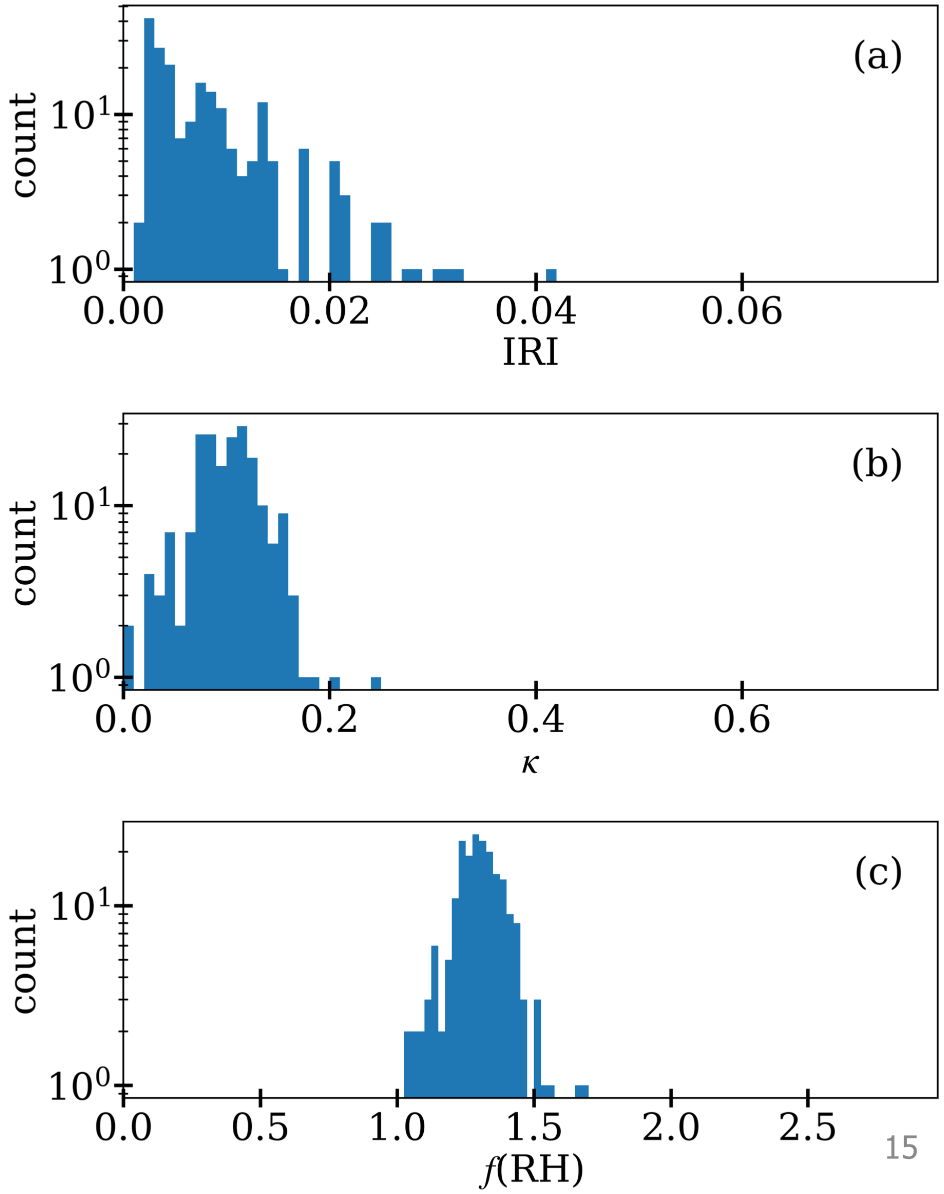



Questions?

- There is a small coarse mode present in low number concentrations but even for PACE-PAX, this is not common.



- Retrieved IRI and hygroscopicity (κ) follow expected trends.
- Low-absorption and generally low hygroscopicity.



HSRL-2 and RSP AOD Comparison

- HSRL-2 to RSP derived spectral AOD (count = 125).

	AOD	
Wavelength (nm)	355	532
NMAD (%)	20	3