AOP, AMP, radiation, and clouds

AGES Workshop

September 27-29, 2022

Moderator:
Sunil Baidar (sunil.baidar@noaa.gov)

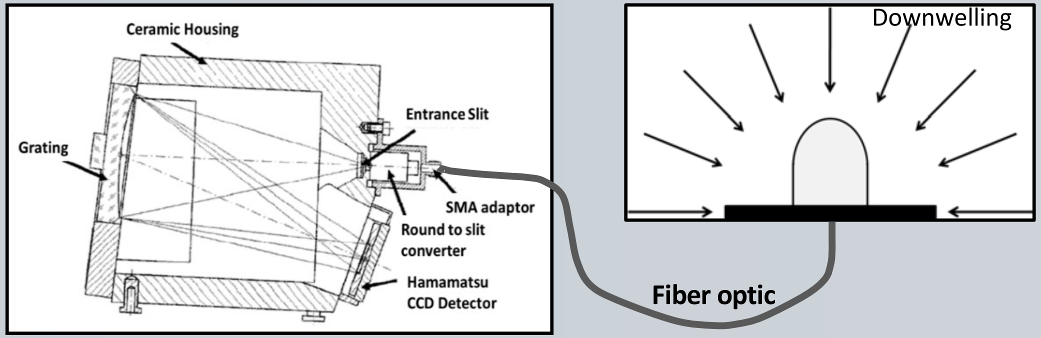
Speaker	Measurement	Mission
Sam Hall	HARP	GOTHAAM
Sarah Woods	PCASP/UHSAS, Cloud probes	GOTHAAM
Rudra Pokhrel	AOP	AEROMMA
Adam Ahern	Li-Neph	AEROMMA
Adam Ahern	AMP	AEROMMA
Shuka Schwartz	SP2	AEROMMA
Hendrik Fuchs	Actinic Flux	AEROMMA

HIAPER Airborne Radiation Package – Actinic Flux
Photolysis frequencies on the NCAR C-130

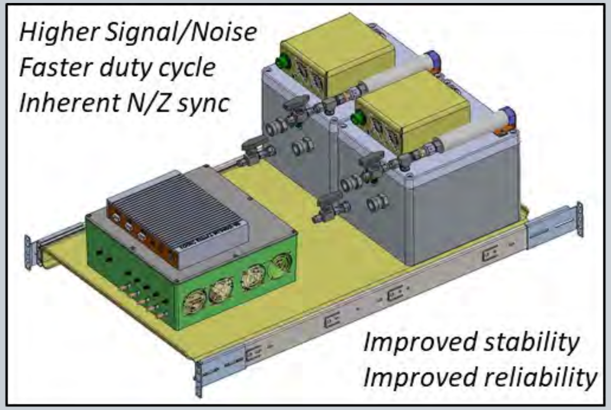
Samuel R. Hall   Kirk Ullmann

Instrumentation and outputs

HARP – Actinic Flux


Spectrally resolved CCD detector

2π steradian optics


Higher Signal/Noise
Faster duty cycle
Inherent N/Z sync

Improved stability
Improved reliability

Time resolution: 1 Hz
Flux precision: ±1-2%

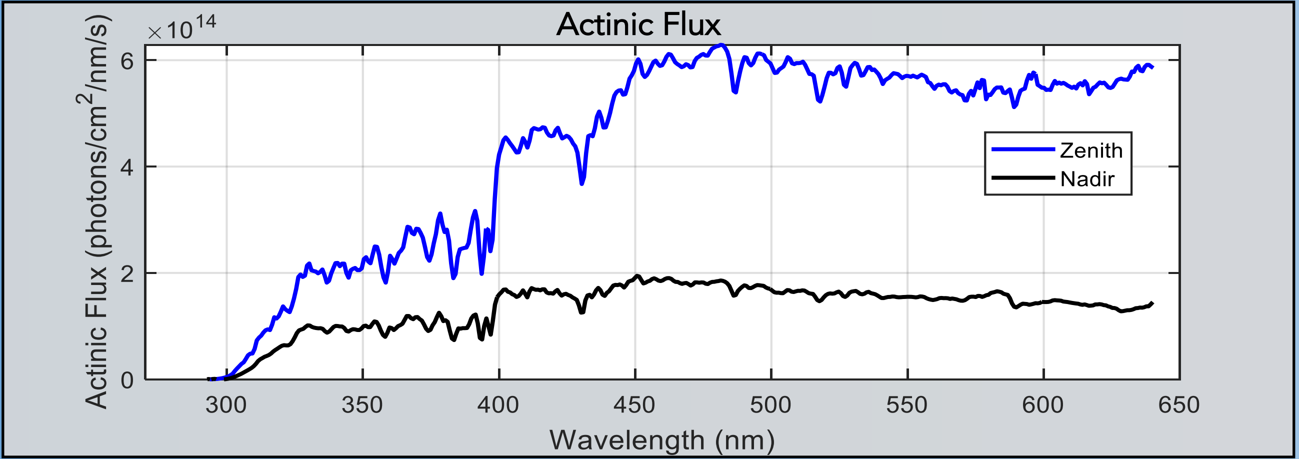
Flux Accuracy:

- UV-B: ±5%
- UV-A/VIS: ±3%

Photolysis Accuracy:

- jNO2: ±12%
- j(O1D): ±20%

- Measurement of spectrally resolved actinic flux (~280-650 nm)
- Measurement of spectrally resolved quasi-actinic flux (~650-1000 nm)
- Calculation of photolysis frequencies

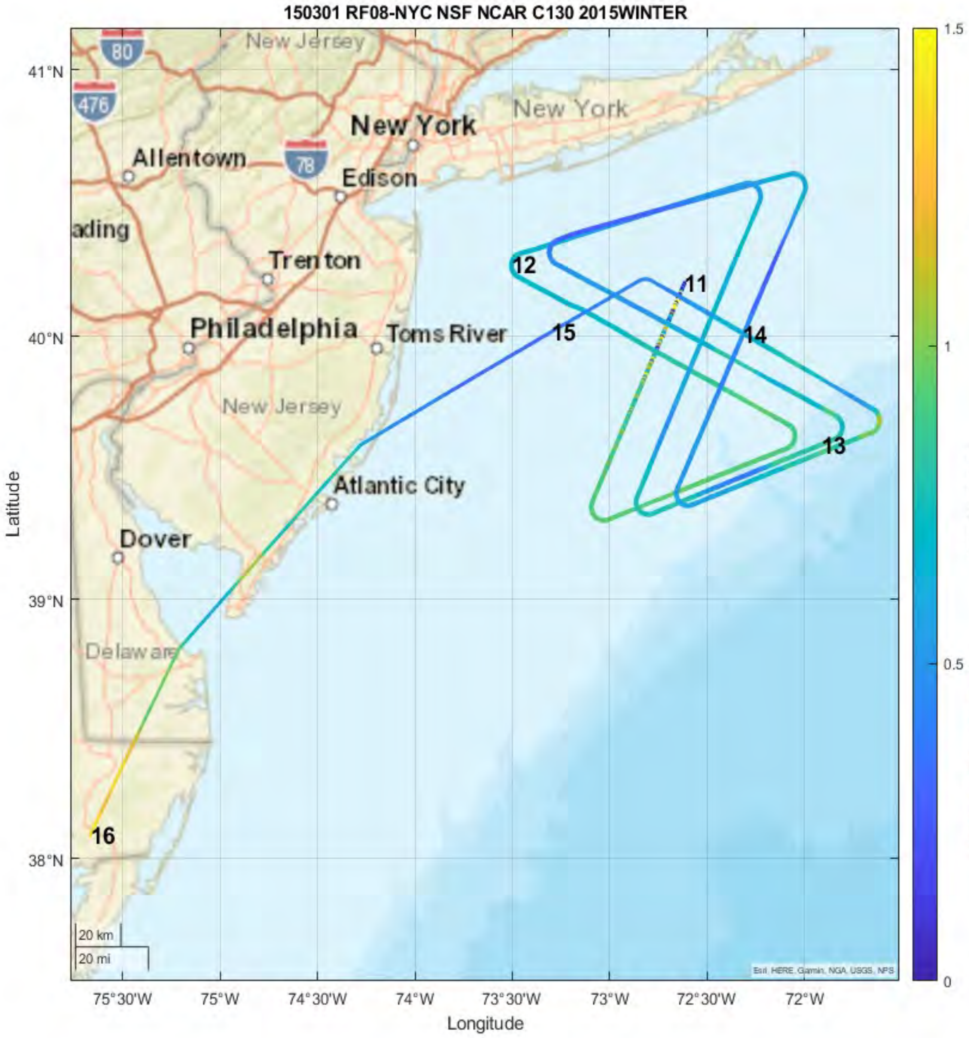


Photolysis Frequencies $= \int F(\lambda)\sigma(\lambda,T,p)\phi(\lambda,T,p)d\lambda$

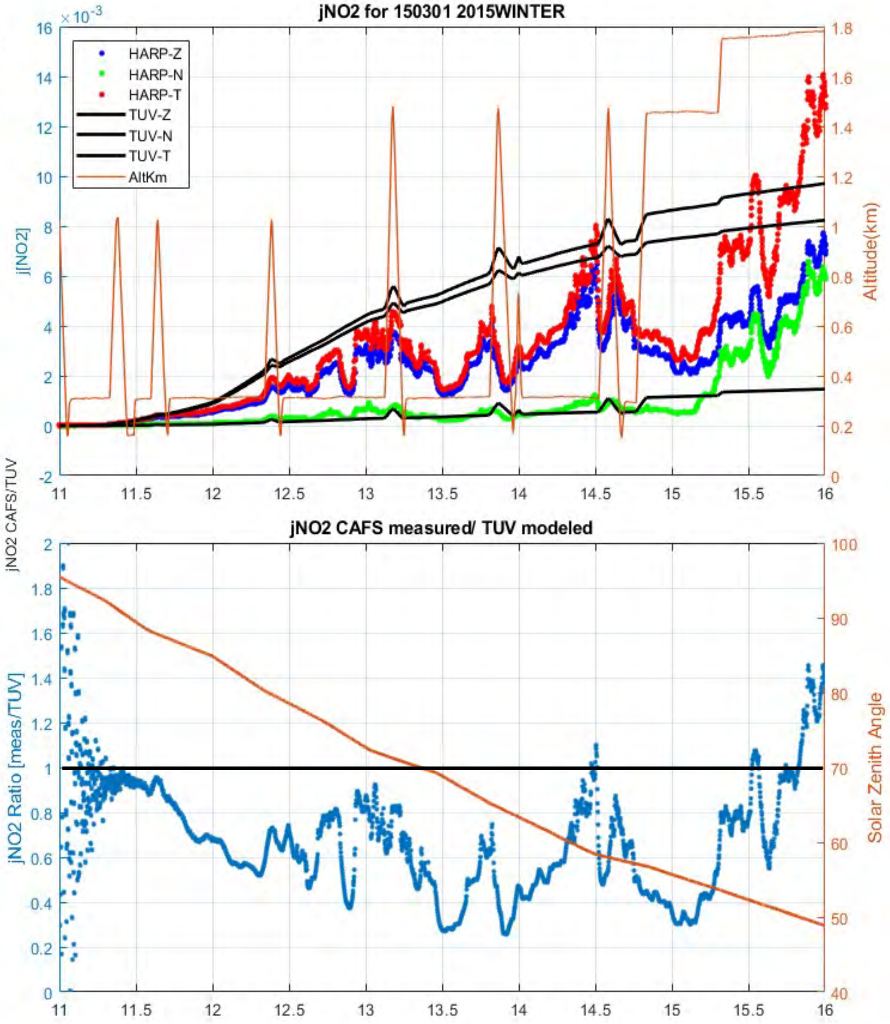
j [O3->O2+O(1D)]
j [NO2->NO+O(3P)]
j [H2O2->2OH]
j [HNO2->OH+NO]
j [HNO3->OH+NO2]
j [CH2O->H+HCO]
j [CH2O->H2+CO]
j [CH3CHO->CH3+HCO]
j [CH3CHO->CH4+CO]
j [C2H5CHO->C2H5+HCO]
j [CHOCHO->products]
j [CHOCHO->HCO+HCO]
j [PAN->products]
j [CH3COCHO->products]

j [CH3COCH3->CH3CO+CH3]
j [CH3OOH->CH3O+OH]
j [CH3ONO2->CH3O+NO2]
j [CH3COCH2CH3->Products]
j [CH3CH2CH2CHO->C3H7+HCO]
j [CH3CH2CH2CHO->C2H4+CH2CHOH]
j [HO2NO2-->HO2+NO2]
j [HO2NO2->OH+NO3]
j [CH3CH2ONO2->Products]
j [Br2->Br+Br]

j [BrO->Br+O]
j [Br2O->products]
j [BrNO3->Br+NO3]
j [BrNO3->BrO+NO2]
j [BrCl->Br+Cl]
j [HOBr->HO+Br]
j [BrONO2->Br+NO3]
j [BrONO2->BrO+NO2]
j [Cl2+hv->Cl+Cl]
j [ClO->Cl+O]
j [ClONO2->Cl+NO3]
j [ClONO2->ClO+NO2]
j [CHBr3->Products]
... plus iodine species



150301 RF08-NYC NSF NCAR C130 2015WINTER



jNO2 for 150301 2015WINTER

jNO2 CAFS measured/ TUV modeled

Photochemical Research

- Chemical evolution
- Tropospheric oxidant chemistry
- Aerosol/cloud impacts on chemistry
- Chemical impacts on clouds/aerosols
- Biomass burning remote sensing

Aerosol, Cloud, & Radiation Measurements for GOTHAAM

Sarah Woods (cloud),
Mike Reeves (aerosol), &
Julie Haggerty (radiometers)
NCAR/EOL/RAF

sfwoods@ucar.edu

Aerosol: PCASP, UHSAS, & CN

- PCASP-100X
  - Passive Cavity Aerosol Spectrometer Probe
  - Not suitable for operation above ~8 km due to its flow through design
  - Splash artifact contamination in cloud

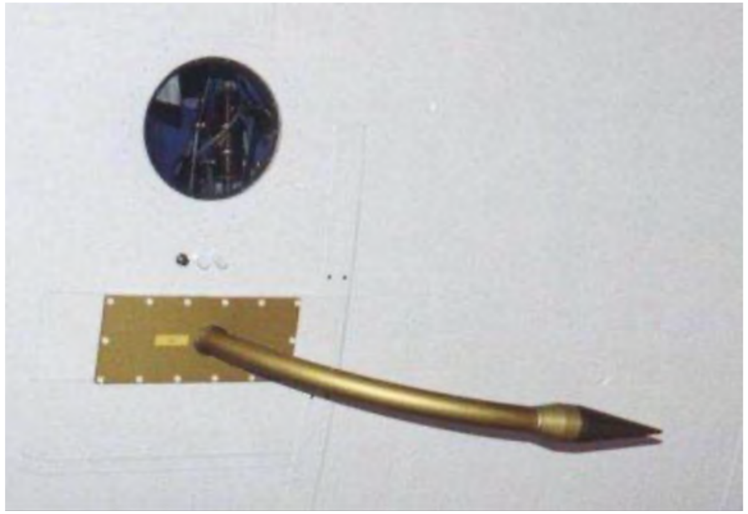
Probe	**Mounting**	**Size**	**Resolution**	**Conc**	**Sampling Rate**
PCASP	Wing	0.1-3 µm	0.02+ µm, Progressive, 30 bins	Spectra	10 Hz; 1 cc/sec
UHSAS	Cabin-SDI	0.055-1 µm	99 bins, logarithmic spacing	Spectra	10 Hz
CN	Cabin-SDI	~0.01-3 µm	n/a	Total	10 Hz

- UHSAS-G
  - Ultra-High Sensitivity aerosol Spectrometer
  - Ground-based UHSAS, modified for flight

- CN
  - Condensation Nucleus Counter
  - butanol CPC (TSI-3760A)
  - Low counting efficiency at pressures < 250 mb
  - Processing converts measured conc to equivalent ambient (not standard) conditions
  - Uncertainty <10% up to concentrations around 10,000 cm^{-3}

Aerosol: SDI

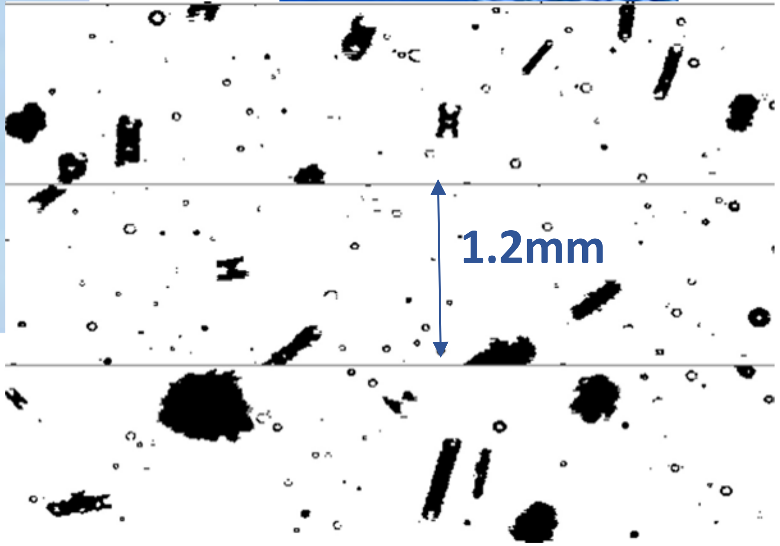
- SDI (Solid Diffuser Inlet)
  - Isokinetic inlet with controlled flow
  - Cone chosen depending on flow needs, shrouded or unshrouded. Available diameters:
    - 4.4 mm (5.4 shrouded)
    - 5.9 mm (6.35 shrouded)
    - 5.4 mm (7.75 shrouded)
  - High flow - up to 500 lpm
  - The cone half-angle is only 4.4 deg to prevent internal flow separation and re-circulation
  - Sample manifold in C-130 cabin to feed multiple instruments:
    - UHSAS-G
    - CN
    - TRAC (Purdue)
    - A-AToFMS (UMich)
    - HR-ToF-AMS (CSU)



Cloud: CDP & F2D-S

- CDP (Cloud Droplet Probe)
  - Good for liquid, mixed, and ice clouds
  - Some sizing uncertainty in ice (assumes spherical particles)
  - Also "sees" wetted aerosols and haze

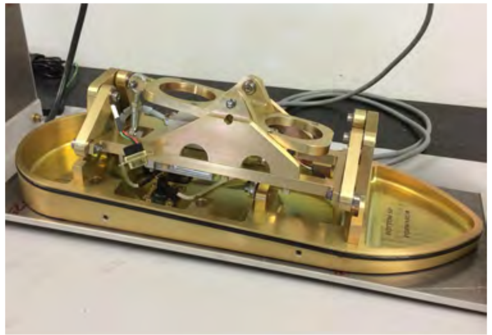
- F2D-S (Fast 2D-Stereo Optical Array Probe)
  - Good for liquid, mixed, and ice clouds
  - Particle imagery (shape, extinction, area, mass)
  - H & V are redundant measurements



Probe	Mounting	Size	Resolution	Conc	Area	Mass
CDP	Wing	2-50 µm	2 µm	Spectra, Measured	Spectra, Assume circular	Spectra, Assume spherical
F2D-S	Wing	10 µm – 1+mm	10+ µm	Spectra, Measured	Spectra, Measured	Spectra, Estimated from Cross-sectional area

Radiometers

Measurement	Sensor	Manufacturer/ Model	Spectral Range (µm)	Scientific Application
Irradiance (up/down)	Broadband radiometer (visible)	Kipp&Zonen CMP22	0.2-3.6	Radiation balance; cloudy/clear determination
Irradiance (up/down)	Broadband radiometer (IR)	Kipp&Zonen CGR4	4.5-42	Radiation balance; cloudy/clear determination
Radiometric surface/sky temperature (up/down)	Radiation pyrometer	Wintronics-Heimann KT19.85	9.6-11.5	Surface, cloud base/top, sky temperature



Aerosol, Cloud, & Radiometer Data Applications

- Aerosol layers, plumes
- Cloud flag context for other measurements
- Aerosol & Cloud Microphysical processes
    - Number conc., extinction, LWC/IWC, R_{eff}, D_{max}
    - Conc, Area, Mass size distributions
    - Aerosol-cloud interaction
        - Polluted vs unpolluted
        - Aerosol composition and CCN at CB have been shown to affect cloud microphysics

NOAA Aerosol Optical Properties Suite

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NOAA CHEMICAL SCIENCES LABORATORY

CIRES

NOAA AOP Suite

Measurements:

Cavity-Ringdown Spectrometer (Langridge et al., 2011)
- Extinction, dry: 405, 532, and 660 nm
- Extinction, high RH (75% and 85%): 660 nm
- 5% accuracy, 0.1 Mm^{-1} detection limit (1s average)
- Sampling rate: 1Hz

Photoacoustic Absorption Spectrometer (Lack et al., 2012)
- Absorption, dry: 405, 532, and 660 nm
- Absorption, thermally denuded: 405, 660 nm
- 25% accuracy, 0.5 Mm^{-1} detection limit (10s average)
- sample rate: 1Hz

TSI Nephelometer(Bodhaine et al., 1991)
- Scattering, dry: 450, 550, and 700 nm
- 10% accuracy, 0.3 Mm^{-1} detection limit (30s average)
- sample rate: 1Hz

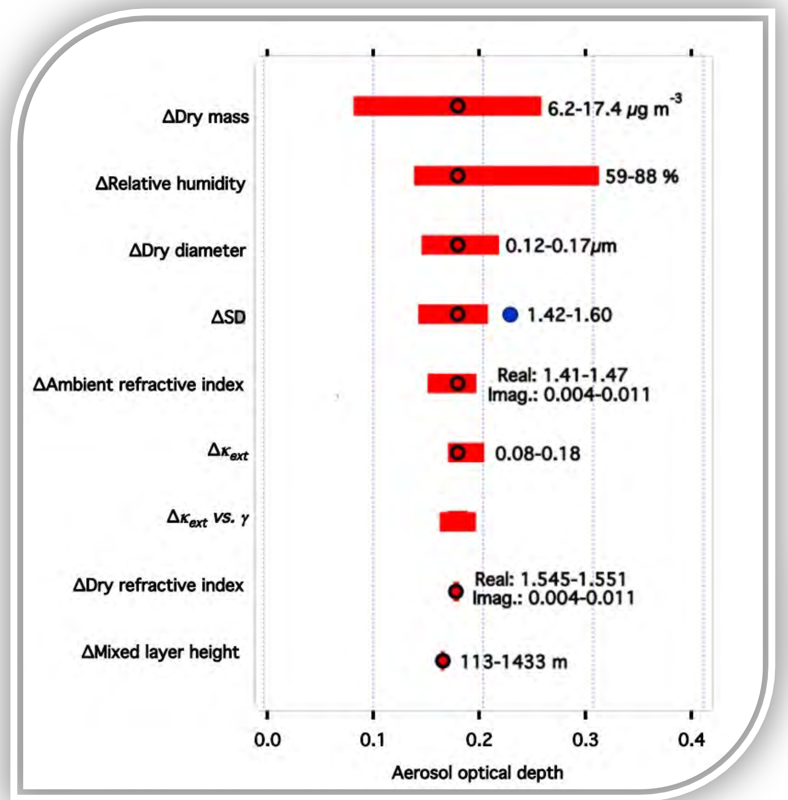
Particle Soot Absorption Photometer (Bond et al., 1999)
- Absorption, dry: 467, 530, and 660 nm
- 25% accuracy, 0.5 Mm^{-1} detection limit (10s average)
- sample rate: 0.1 Hz

NOAA PAS , CRDS and PSAP

TSI-Neph

Science Goals

Sensitivity of AOD and radiative effect to aerosol microphysics (i.e., SSA, size distribution, f(RH)).



(Brock et al., ACP 2016)

Work with modeling and satellite groups to validate aerosol products (AOD, Extinction, AE). Study the impact of source and morphology on validation.

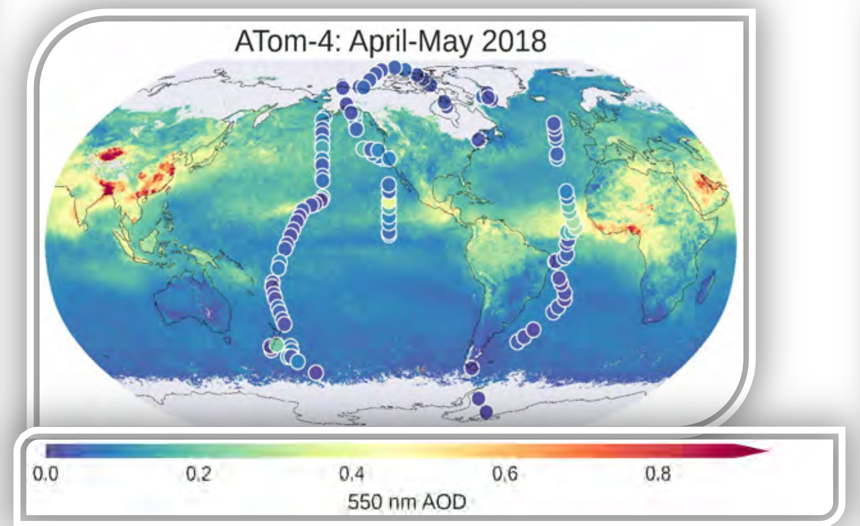
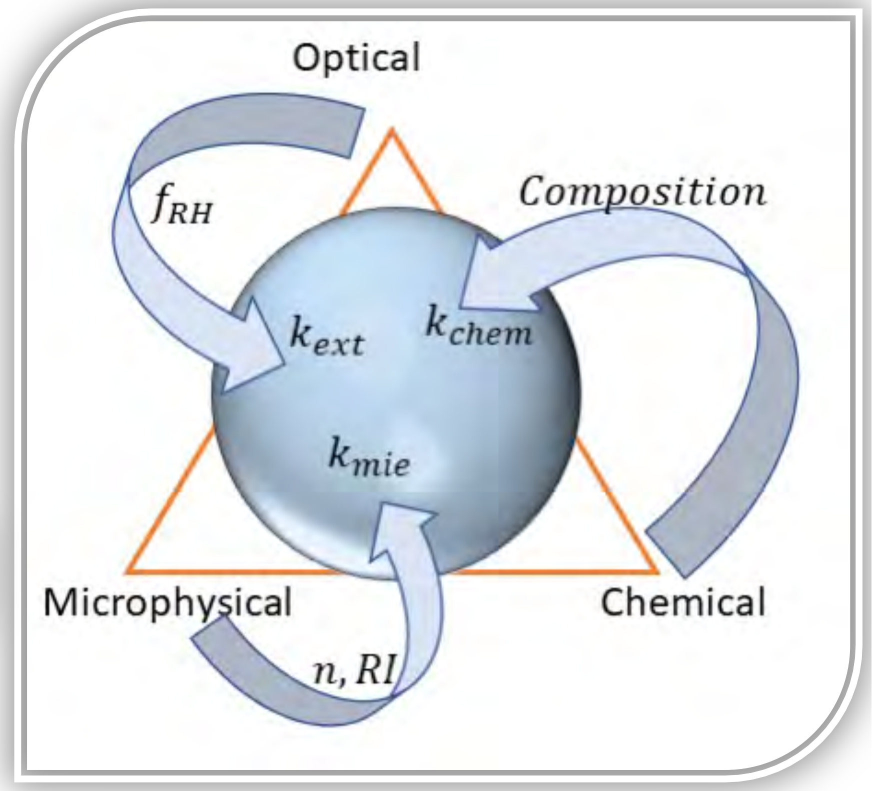


Figure courtesy of Siyuan Wang

Intercompare the aerosol hygroscopicity estimated based on optical, microphysical, and chemical approaches.



NOAA Nephelometers

Laser Imaging Nephelometer (LiNeph)
Integrating Nephelometer (IntNeph)

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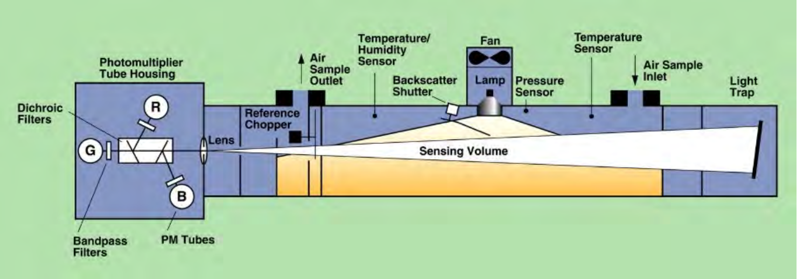
Measuring directionality of aerosol light scattering

Measurements:

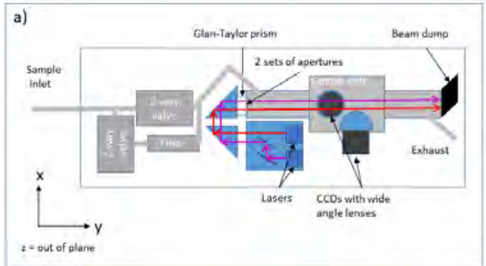
Integrating Nephelometer (Anderson et al., 1996)

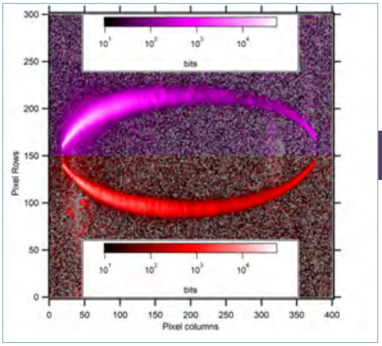
- Dry $PM_{2.5}$ Scattering: 450, 550, and 700 nm
- Hemispheric backscatter fraction *(b)*
- ~5% accuracy, 4 Mm^{-1} detection limit
- sample interval: 2s

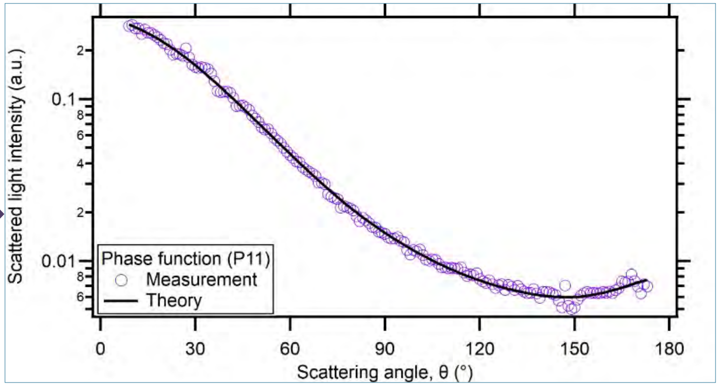


Laser Imaging Nephelometer (Ahern et al., 2022)

- All measurements at two wavelengths, $PM_{2.5}$ dry: 405 and 660 nm
- Phase function ($\mathbf{P}_{11}$) and degree of linear polarization ($-\mathbf{P}_{12}/\mathbf{P}_{11}$)
- Asymmetry parameter (*g*)
- Hemispheric backscatter fraction (*b*)
- ~5% accuracy, 5 Mm^{-1} detection limit
- sample interval: 2.5 s

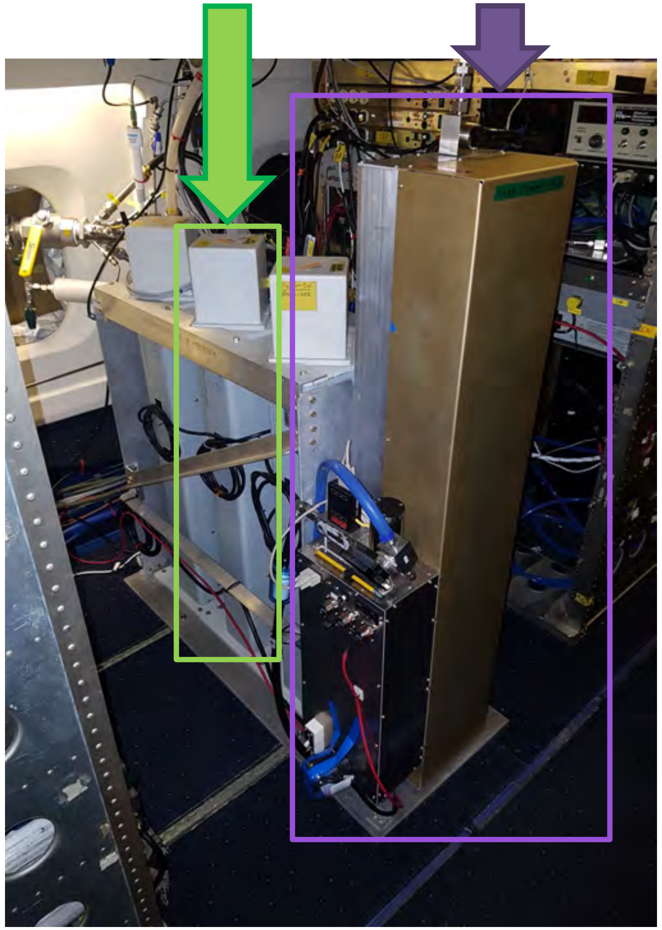






IntNeph (TSI Model 3563)

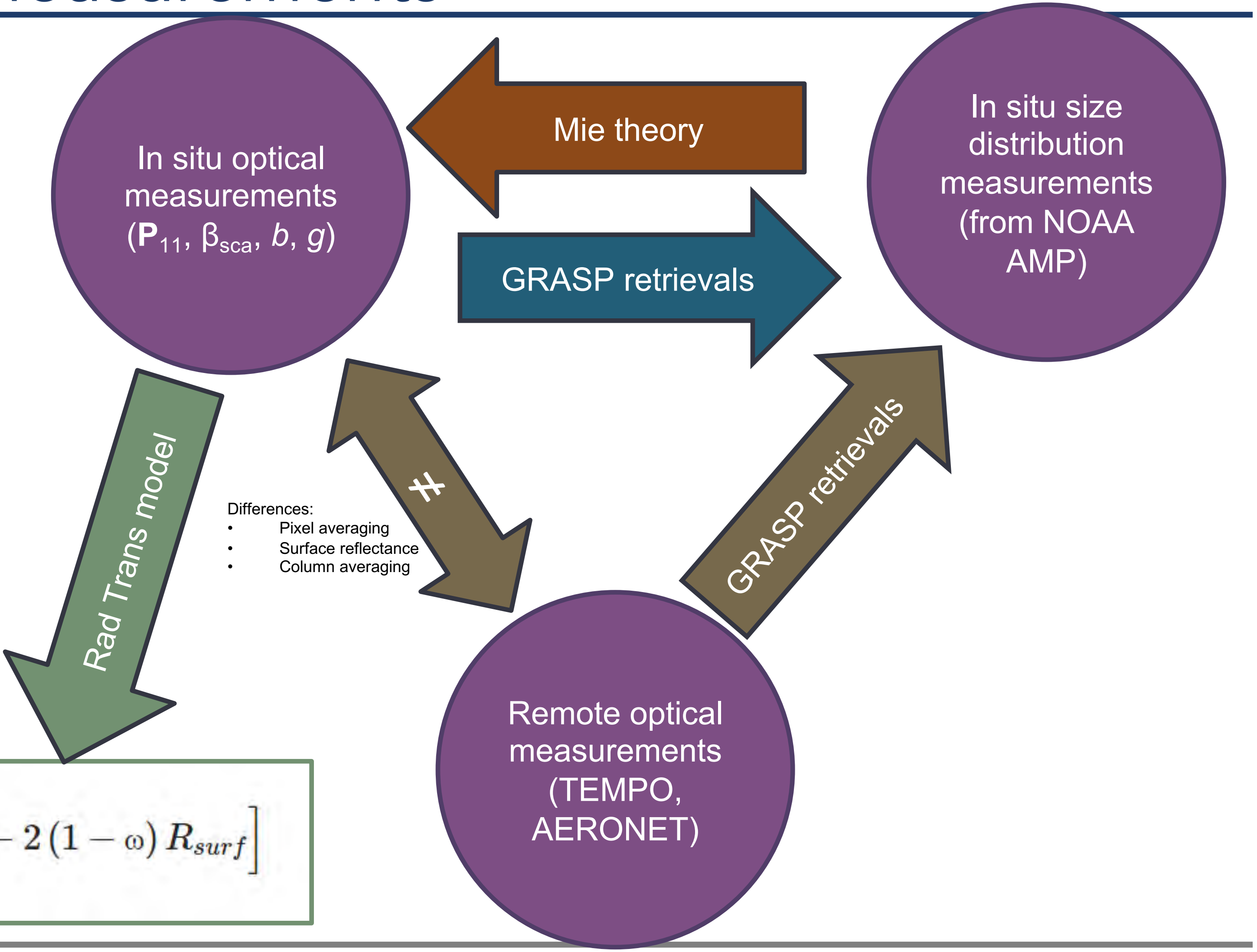
NOAA LiNeph



NOAA Chemical Sciences Laboratory

Comparing models of aerosol scattering with measurements

- Compare in situ optical measurements with predictions based off in situ size distributions.
- Retrieve aerosol microphysical and optical properties using GRASP algorithm using in situ measurements of P_{11}.
- Retrieve aerosol microphysical and optical properties using GRASP algorithm using remote measurements of P_{11}.
- Evaluate direct aerosol radiative forcing based on in situ optical measurements.



In situ optical measurements ($\mathbf{P}_{11}$, β_{sca}, b, g)

Mie theory

GRASP retrievals

In situ size distribution measurements (from NOAA AMP)

Rad Trans model

≠

Differences:
- Pixel averaging
- Surface reflectance
- Column averaging

GRASP retrievals

Remote optical measurements (TEMPO, AERONET)

$$\frac{\Delta F_{aer}}{\tau} = -\frac{S_0}{2} T_{atm}^2 \left(1 - A_{cld}\right) \left[\bar{\beta}\,\omega\left(1 - R_{surf}\right)^2 - 2\left(1 - \omega\right) R_{surf}\right]$$

NOAA Aerosol Microphysical Properties Suite

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NOAA Aerosol Microphysical Properties Suite

Measurements:

Nucleation-Mode Aerosol Size Spectrometer-NG (NMASS-NG)

- 8-channels of sizing using Kelvin diameter
- Inversion to recover size distribution
- Size distributions 0.003-0.06 µm
- sample interval: 1s

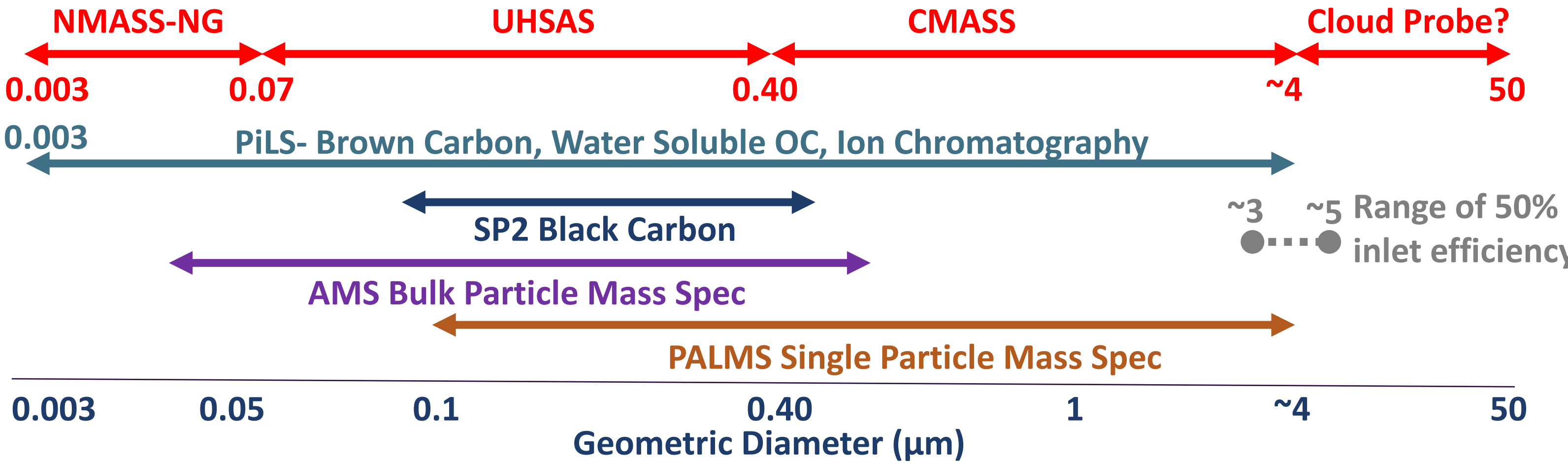
Ultra-High Sensitivity Aerosol Size Spectrometer (UHSAS)

- Optically based size distribution using laser
- Size distributions 0.07-1 µm
- sample interval: 1s

Coarse-Mode Aerosol Size Spectrometer (CMASS)

- Optically based size distribution using broadband LED
- Size distributions 0.4-10 µm (inlet-limited to ~4 µm)
- sample interval: 1s

Underwing Cloud Probes? TBD



NMASS-NG 0.003 0.07
UHSAS 0.07 0.40
CMASS 0.40 ~4
Cloud Probe? ~4 50

0.003 PiLS- Brown Carbon, Water Soluble OC, Ion Chromatography

SP2 Black Carbon

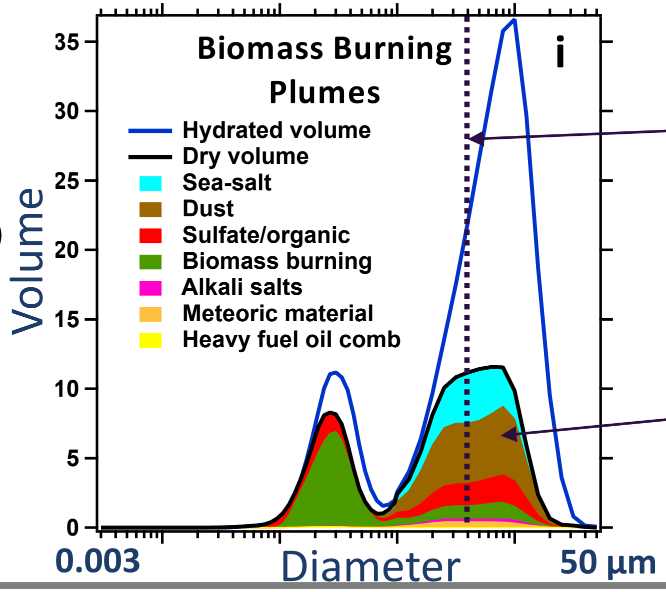
AMS Bulk Particle Mass Spec

PALMS Single Particle Mass Spec

~3 ~5 Range of 50% inlet efficiency

0.003 0.05 0.1 0.40 1 ~4 50

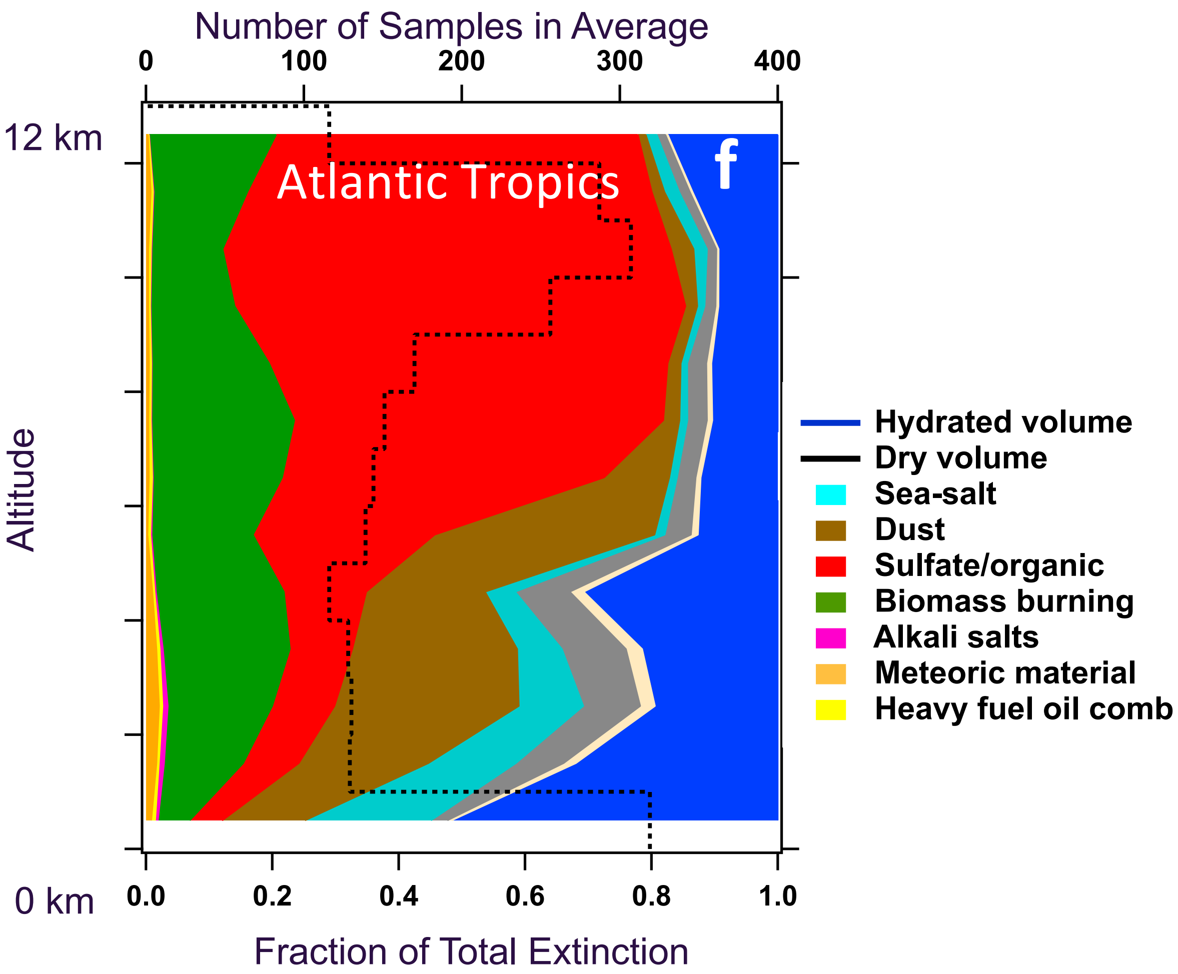
Geometric Diameter (µm)



- Max size if no cloud probes
- Will miss a major fraction of aerosol mass in dust, biomass burning, marine cases

- Map composition onto combined size distribution
- Calculate hygroscopic growth
- Calculate optical properties

Science Goals



- **Build comprehensive microphysical, chemical and optical description of the aerosol**
- **Apportion optical properties like aerosol optical depth (AOD) to different aerosol types**
- **Evaluate satellite retrieval assumptions—particularly useful for new sensors/algorithms on TEMPO**
- **Evaluate HSRL2 classification**
- **Evaluate model emissions and processes**
- **More profiles=better for these goals**
- **Prefer profiles coordinated with AERONET sites and HSRL2 to compare derived AOD with more direct measurements**

NOAA SP2 on NASA DC8 measuring black carbon aerosol

Samantha
Lee
Software

Georgia
Michailoudi
SP2

Behind the camera:
Shuka Schwarz, SP2

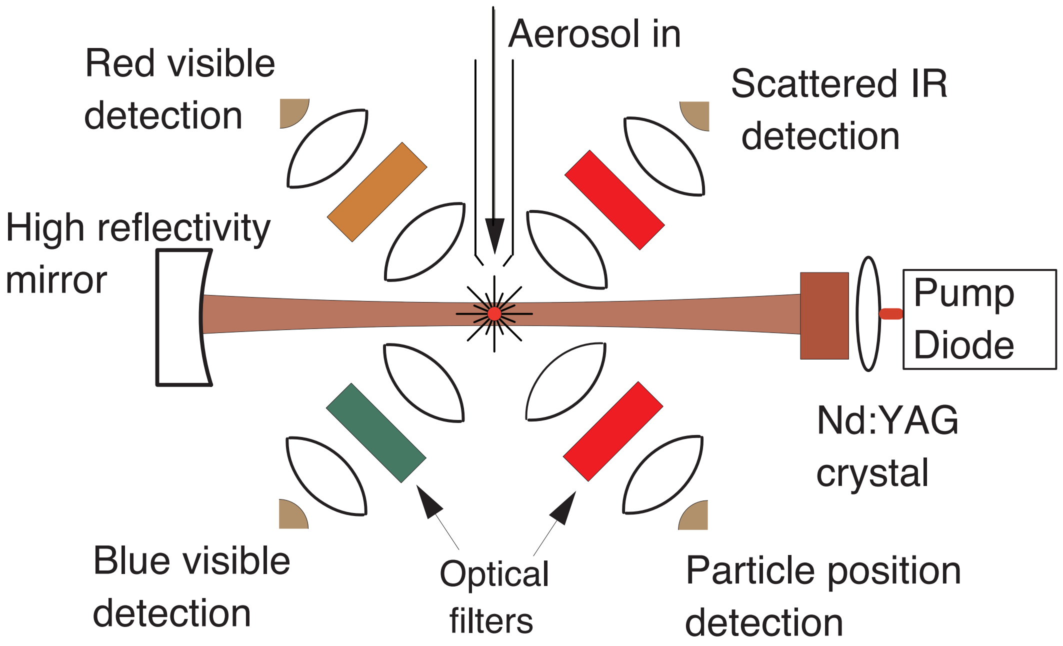
Anne
Perring
(U-Colgate)
+ Student
SP2

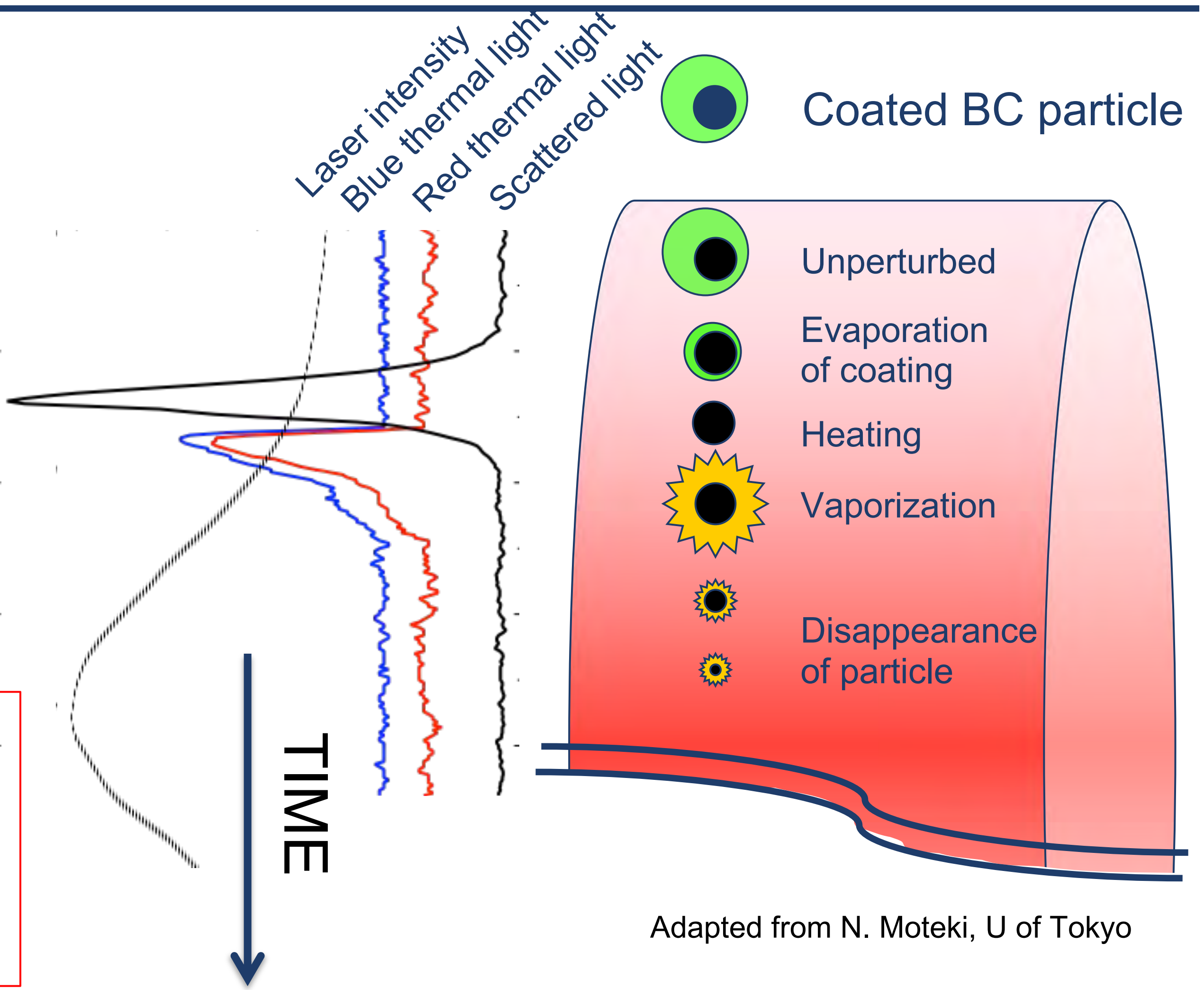
Data Products:

- Black carbon (BC) concentration
- BC microphysical state (size distribution, quantitative mixing state)

Contact: Joshua.p.schwarz@noaa.gov

NOAA SP2 on NASA DC8 measuring BC



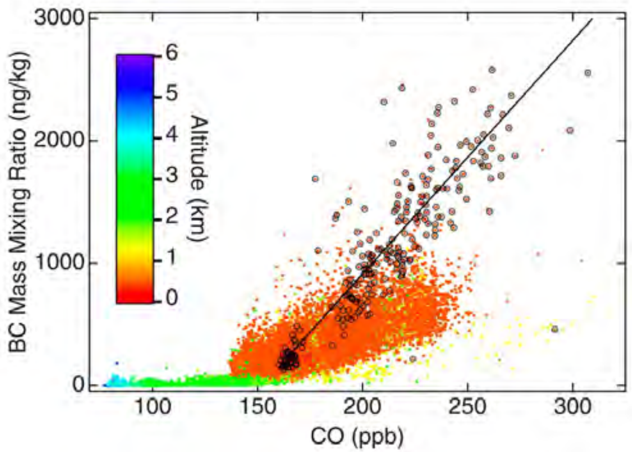


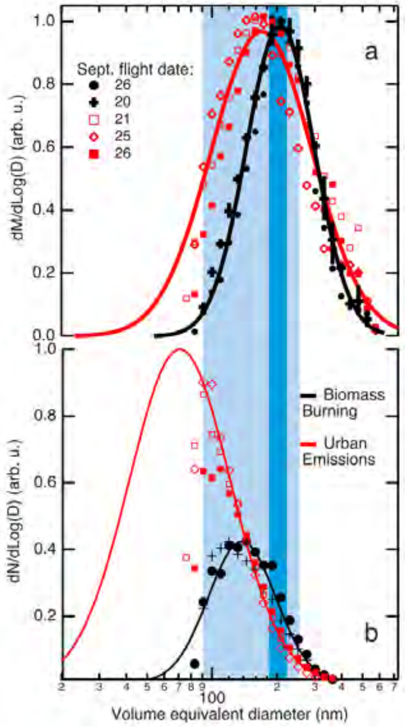
Adapted from N. Moteki, U of Tokyo

- Provides single particle:
- BC mass: **0.15 – 0.6 µm**
- Total particle optical size (BC-containing, limited size range)

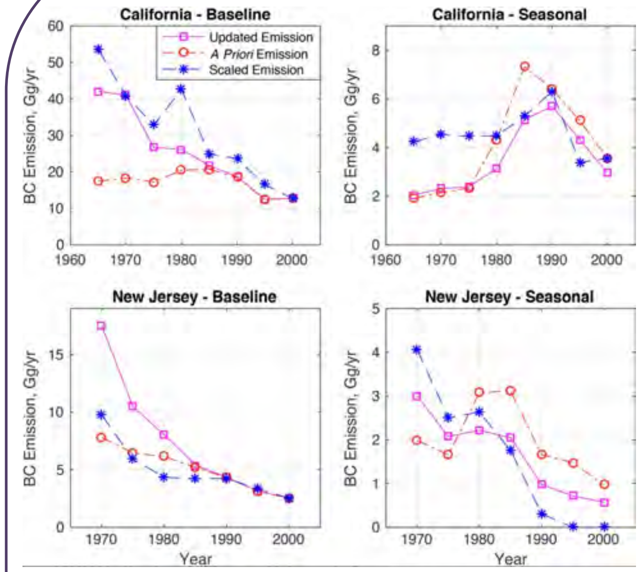
Morphology, mixing state, pressure, et cet. do not affect BC mass measurement.

Science Goals and Foci

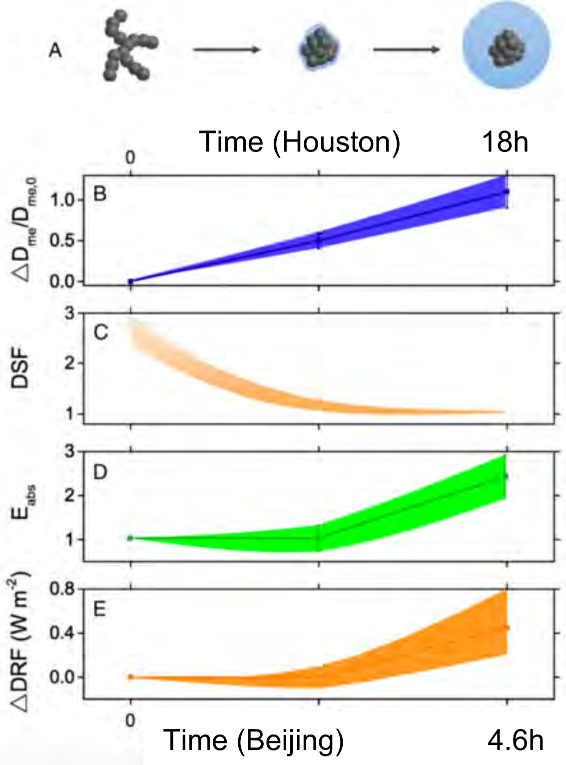




Determine tracer-relationships and aerosol properties that are critical for both AQ and climate impact evaluations



Long-term inventories
e.g.: Sun et al., 2019 *JGR*



Aging processes, e.g. Peng et al, *PNAS* 2016

Determine Urban BC/CO, BC microphysics

Test emissions inventories, model performance, and bulk aerosol process understanding.

AEROMMA Photolysis frequencies: J-CAFS

Birger Bohn, Hendrik Fuchs and Anna Novelli

27.09.2022

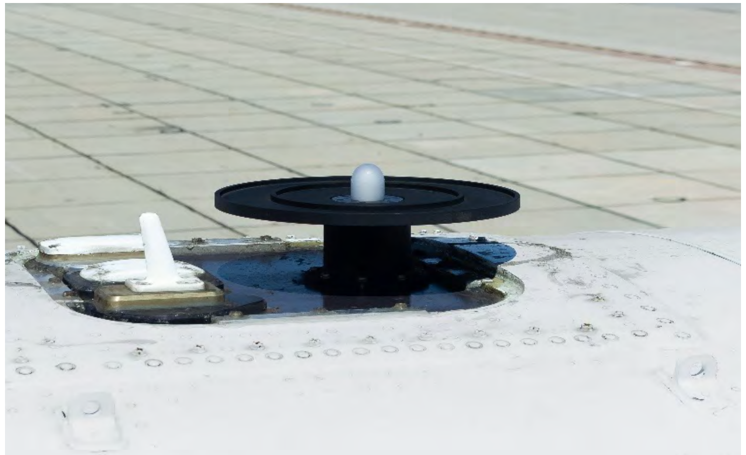
J-CAFS (Jülich - CCD Actinic Flux Spectroradiometer)

- Hemispherical zenith and nadir measurements of downward and upward actinic flux densities → calculation of photolysis frequencies

- Original NASA DC-8 design: CAFS by NCAR (Rick Shetter, Samuel Hall)

- AEROMMA: Optical receivers (photo) connected with two Jülich spectroradiometers → J-CAFS

- Jülich spectroradiometers approved on research aircraft HALO (DLR, Germany)



DC-8 top fuselage (NCAR photo)

J-CAFS

- Key parameters:
  - wavelength range: 280 - 650 nm
  - spectral resolution: ≈ 2 nm
  - time resolution: ≈ 1 s
  - uncertainty: ≈ 10% (radiation measurements)

- Example: HALO flight over East China Sea, north-east of Taiwan

- Strongest variability induced by clouds, similar for photolysis frequencies