Evaluating the Consistency of All Submicron Aerosol Mass Measurements (Total and Speciated) from the NASA Atmospheric Tomography Mission (ATom)

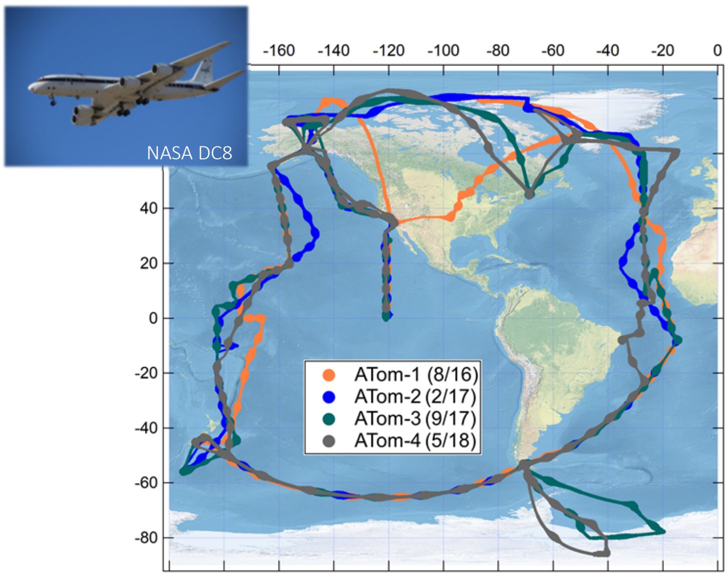
Hongyu Guo[1,2], Pedro Campuzano-Jost[1,2], Benjamin A. Nault[1,2], Douglas Day[1,2], Jason C. Schroder[1,2,*], Christina Williamson[1,3], Agnieszka Kupc[3,4], Charles A. Brock[3], Gregory P. Schill[1,3], Karl D. Froyd[1,3], Daniel M. Murphy[3], Eric Scheuer[5], Jack E. Dibb[5], Joseph Katich[1,3], Jose L. Jimenez[1,2]

*[1] CIRES, Univ. of Colorado Boulder, Boulder, USA; [2] Dept. of Chemistry, Univ. of Colorado Boulder, Boulder, USA; [3] NOAA ESRL, USA; [4] Faculty of Physics, Univ. of Vienna, Austria; [5] EOS, Univ. of New Hampshire, Durham, USA; * Now at Air Pollution Control Division, Colorado Dept. of Public Health and Environment, Denver, USA*

EGU virtual Meeting
May 5th, 2020

The NASA ATom mission:
Profiling the remote atmosphere from 0-13 km (~600x)

University of Colorado Boulder



Motivation of this study:

- Evaluate the consistency of publicly available aerosol measurements suite for ATom:

  https://daac.ornl.gov/ATom

  since this data will be widely used to evaluate and constrain global models.
- Is our current understanding of the uncertainties of the Aerodyne Mass Spectrometer (AMS) consistent with ATom performance?

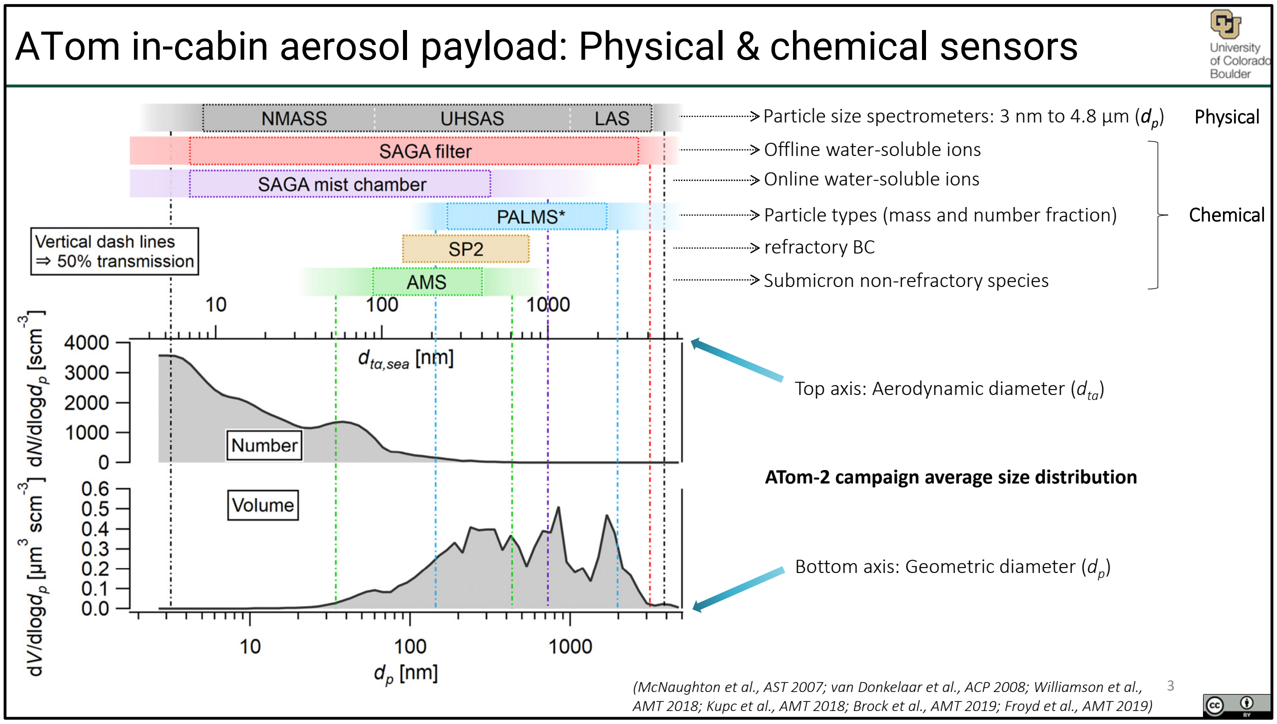
See EGU2020-6155 (AS3.4; May 6th) for science results!

The paper to be submitted to AMT soon.

2

Speaker's notes for EGU audience:

- The title of paper to be submitted: **Evaluating the Consistency of Submicron Aerosol Volume Derived from Sizing vs. Composition measurements during the Atmospheric Tomography Mission (ATom)**



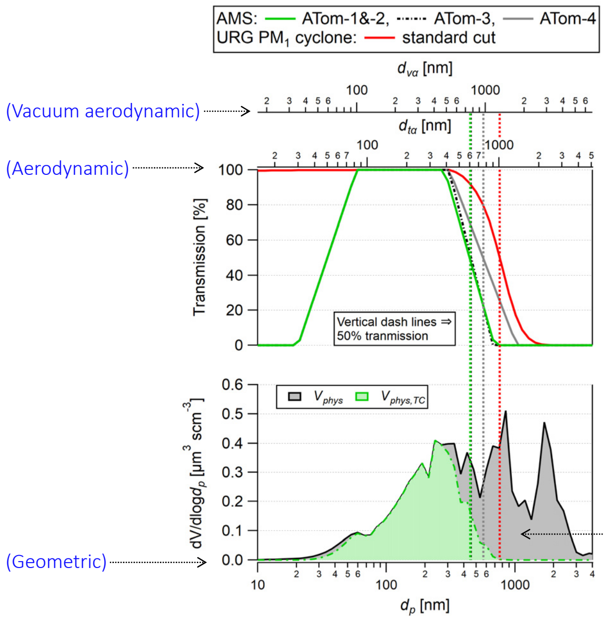
Speaker's notes for EGU audience:

- Each instrument has its own observable particle size range, making the direct intercomparisons not feasible in some cases.
- Thus, instrument inlet transmission needs to be characterized for meaningful comparisons.
- Based on this plot, AMS size range is roughly comparable to SAGA MC.
- $d_{ta,sea}$ refers to the aerodynamic diameter at sea level pressure (1013 hpa), since d_{ta} is pressure dependent.
- NMASS, UHSAS, and LAS comprise the NOAA AMP package.

The list of abbreviations:
NMASS: Nucleation-Mode Aerosol Size Spectrometer
UHSAS: Ultra-High Sensitivity Aerosol Spectrometer
LAS: Laser Aerosol Spectrometer
AMP: Aerosol Microphysical Properties
AMS: Aerosol Mass Spectrometer
SP2: Single Particle Soot Photometer (measured BC)
SAGA filter: Soluble Acidic Gases and Aerosol (SAGA) filter
SAGA MC: SAGA Mist chamber coupled with ion chromatograph
PALSM: Particle Analysis by Laser Mass Spectrometer

What is PM$_1$ and the fraction that AMS observes?

University of Colorado Boulder



Q: What is the cutoff size of AMS?

Compared to a standard PM$_1$ (a URG cyclone operated at ground and ambient condition), the CU AMS is ~ **PM$_{0.75}$** in ATom-2 and **PM$_{0.9}$** in ATom-4 (effect of particle liquid water considered → less difference than shown in the figure).

- AMS transmission differ between instruments and may change with lens alignment or transport.
- Field calibration of the AMS transmission is critical for accurate quantification and intercomparisons.

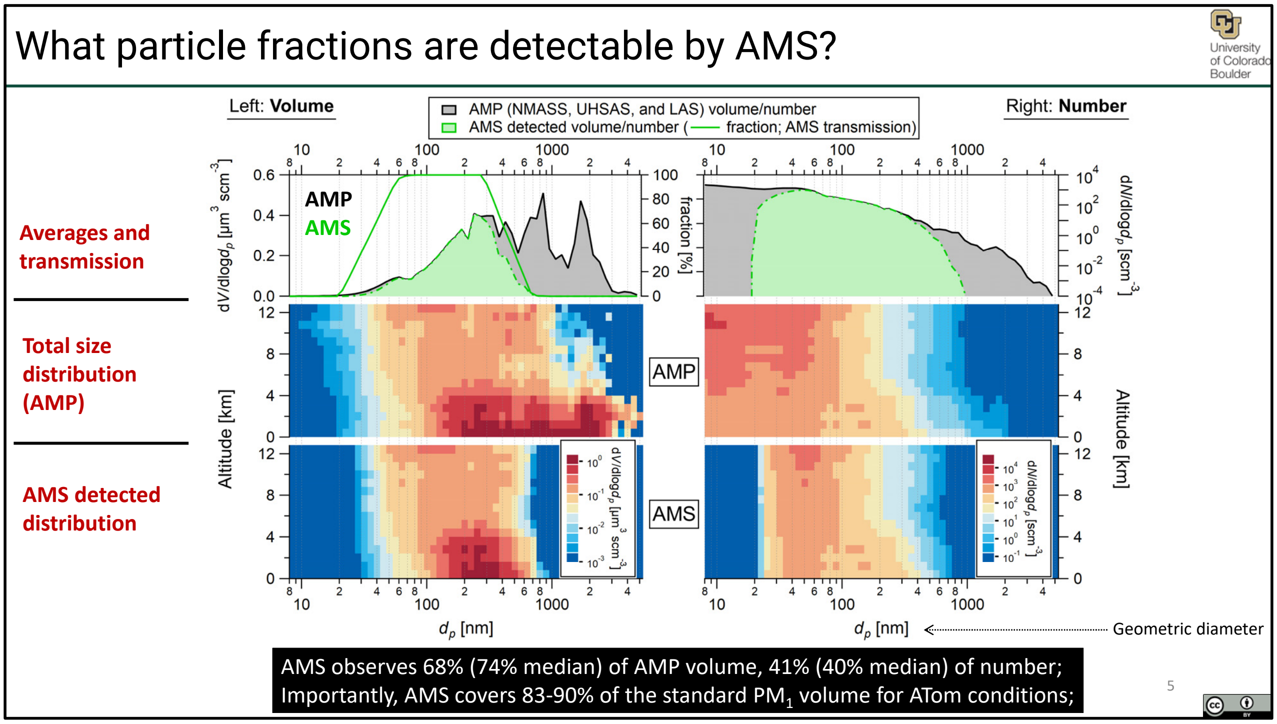
ATom-2 campaign average volume size distribution (the fraction observed by AMS)

4

Speaker's notes for EGU audience:

- Both the AMS and the NOAA AMP size distributions measure dry particles while the ground URG PM$_1$ cyclones are normally operated under ambient humidity. To account for the difference, the URG transmission is applied to the estimated ambient particle size before losing liquid water content.
- Regarding the submicron mass quantification using AMS, the large particle transmission (upper side) is far more relevant for consistent comparisons than the small particle transmission (lower side), as shown by the bottom panel.
- V_{phys}: AMP total volume; $V_{phys,TC}$: AMP total volume corrected with AMS transmission;

What particle fractions are detectable by AMS?

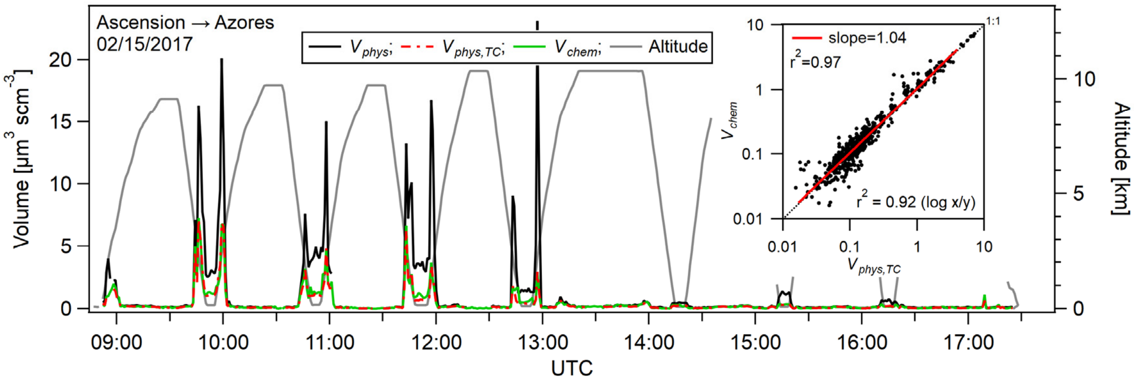


Speaker's notes for EGU audience:

- The right panels show the number of particles in the observable size range for AMS and do not necessarily indicate that AMS detects all these particles. Instead, the chemical speciated info quantified by AMS represents the bulk properties of these particles.
- The size ranges quantified by other instruments are also discussed in the paper. These plots and numbers are important when evaluating global models with the ATom aerosol payload, because different particle size ranges are defined and used by global models.

A good transmission curve allows meaningful comparisons

University of Colorado Boulder



$$V_{chem} = V_{AMS} + V_{BC} = \underbrace{\left(\frac{OA}{\rho_{OA}} + \frac{SO_4 + NO_3 + NH_4}{1.75} + \frac{Chl}{1.52}\right)}_{\text{AMS non-refractory}} + \underbrace{\frac{Sea\ salt}{1.45}}_{\text{AMS refractory}} + \frac{SP2\ BC}{1.77}$$

AMS sea salt: Method of Ovadnevaite et al., JGR 2012, and calibrated in lab

Sea salt density from Froyd et al., AMT 2019

Applying AMS transmission curve →
Good agreement between AMP ($V_{phys,TC}$) and AMS+BC (V_{chem}) volumes

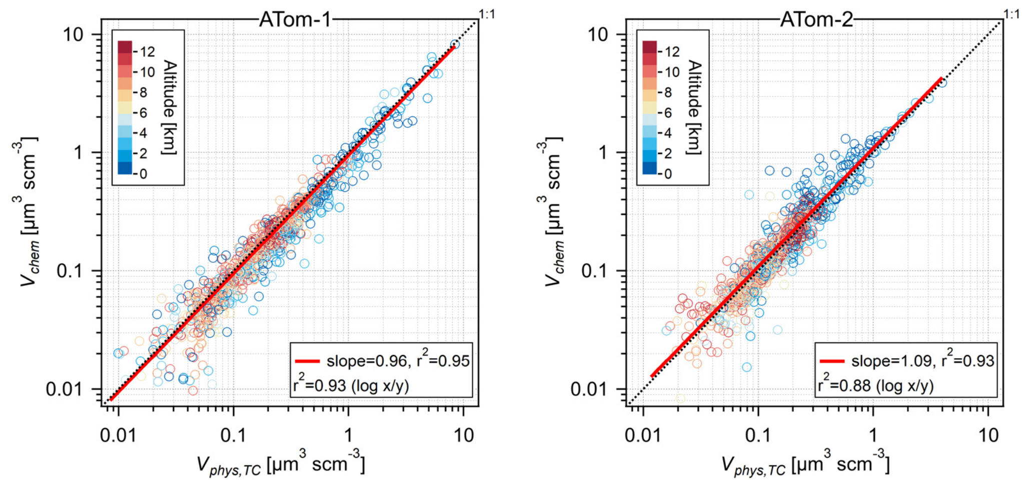
6

Speaker's notes for EGU audience:

- In the last two slides, we show how much AMP volume is captured by AMS. Here, we show an example from a research flight in ATom-2 for volume closure to evaluate the AMS transmission.
- The OA density (ρ_{OA}) is estimated with the AMS measured O/C and H/C atomic ratios of OA using the parameterization of Kuwata et al. (2012).
- Two correlations coefficients (r^2) are listed: one at linear scale (commonly used) and the other at logarithmic scale, which emphasizes the scatter at low concentrations.
- V_{phys}: AMP total volume; $V_{phys,TC}$: AMP total volume corrected with AMS transmission;

Volume comparison: AMS+BC volume and particle sizers agree well

University of Colorado Boulder



Consistent agreement for ATom deployments!

7

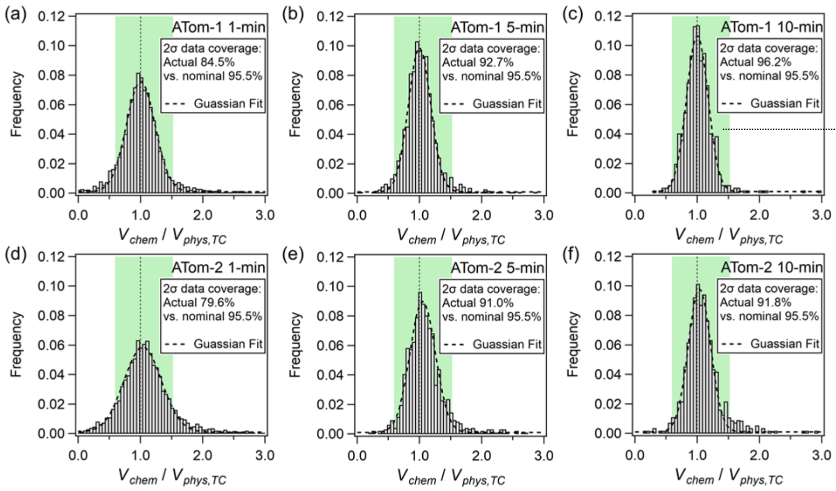
ATom-3 and -4 are similar and not shown here.

Speaker's notes for EGU audience:

- Two correlations coefficients (r^2) are listed: one at linear scale (commonly used) and the other at logarithmic scale, which emphasizes the scatter at low concentrations.
- $V_{phys,TC}$: AMP total volume corrected with AMS transmission; V_{chem}: AMS calculated bulk volume plus BC volume.

Measurements are consistent within the stated uncertainties

University of Colorado Boulder



Green-tinted shade: 2σ combined uncertainty ranges from the two volumes

Evaluating the AMS uncertainty through the volume closure.

The reported 2σ accuracy (Sulfate: 35%, OA: 38%) are reasonable.

1-min → 5-min → 10-min

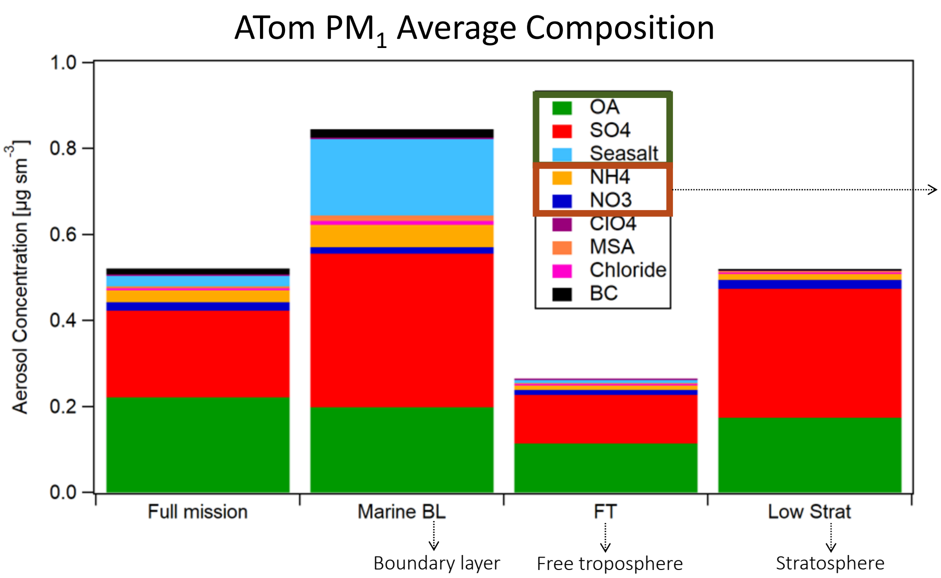
Longer averaging time interval smooths out random noise (prominent in ATom due to the clean remote air)

8

Speaker's notes for EGU audience:

- ATom-1 and -2 sampled remote particles (clean) with low concentrations, 0.50 and 0.38 μg m^{-3}, making the 1 min data suffering from random noises. Longer averaging time can deemphasize these precision errors, especially for a dataset like ATom with few sharp plumes. Thus we plot the volume ratio at three time scales, 1 min, 5 min, and 10 min. It shows a clear improvement in the spread of the ratio as the averaging time scale increases, with the 10 min data being consistent with the reported uncertainties. This supports the good quality and consistency of the ATom aerosol dataset, and it also supports the reported AMS uncertainties.
- $V_{phys,TC}$: AMP total volume corrected with AMS transmission; V_{chem}: AMS plus BC volume.

OA, sulfate & sea salt are the major components of PM_1 in ATom



See EGU2020-11366 (AS2.14; May 8th)
For related presentation on global survey of aerosol acidity

9

Speaker's notes for EGU audience:

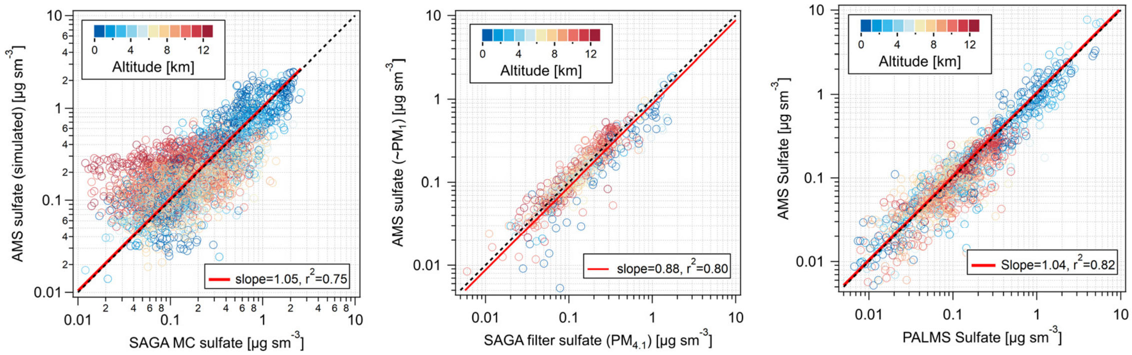
- With a good confidence on the volume closure, in the next, we compare the three main submicron aerosol components: organic aerosol (OA), sulfate, and sea salt.
- Sea salt is an important submicron aerosol component when sampling marine boundary layer in ATom.

Sulfate: Good agreement vs water-soluble Ions and Single Particle MS

University of Colorado Boulder

(Simulated based on the tailing effect of SAGA MC; see next slide)

ATom-1: AMS vs. SAGA MC | AMS vs. SAGA Filters | ATom-1: AMS vs. PALMS (PM_1)



Resolution: 75 sec | Resolution: 5-20 min | Resolution: 3 min

From PALMS and AMS Data: organosulfate is very low (~1%)

SAGA MC, SAGA Filters, PALMS, and AMS sulfate agree well.

For SAGA MC: the tails and the correction made for them → extra noise

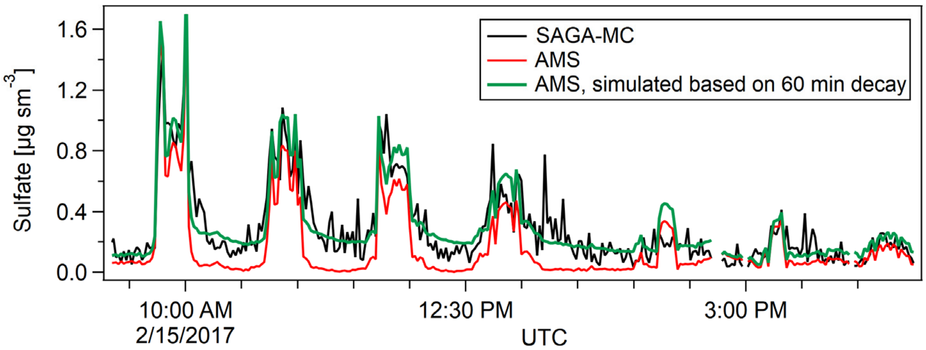
10

Speaker’s notes for EGU audience:

- Sulfate is virtually nonvolatile in troposphere (e.g., no evaporation bias as ammonium nitrate when collected via filters) and a ubiquitous fine particle component, thus commonly used for instrument intercomparisons.
- Excluding the dust and rich supermicron particle events, AMS sulfate (PM_1) agrees well with the SAGA filter sulfate ($PM_{4.1}$)
- Since the PALMS composition data is derived by mapping particle composition/type to the NOAA size distributions, the AMS transmission is applied to the PALMS mass product to allow an apple-to-apple comparison with AMS.

Sulfate: AMS vs. SAGA Mist Chamber (IC-based)

University of Colorado Boulder



SAGA-MC vs AMS
Slope = 1.10, r^2 = 0.73

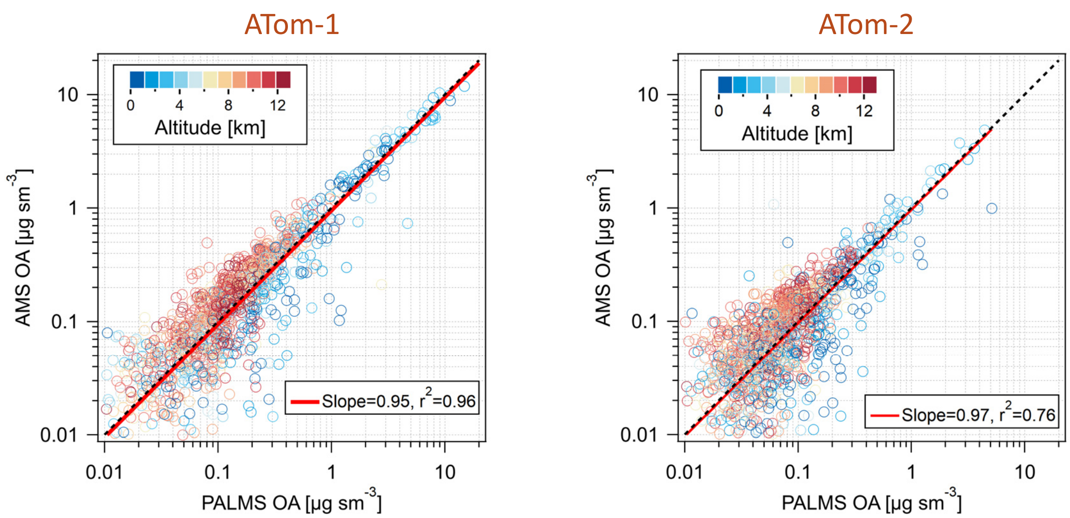
Slope = 1.00, r^2 = 0.81

- SAGA Mist Chamber has strong tailing due to the liquid sampling and analysis (carryover between samples)
- ATom sampling strategy (constantly up and down) requires adding fairly heavy tails to the AMS SO_4, to simulate the tailing of the SAGA-MC, leading to better agreement.

11

Speaker's notes for EGU audience:
- The simulation of the tailing in SAGA MC is supported by the good agreement between AMS and SAGA filter.

OA: Good agreement between AMS and PALMS



PALMS OA: completely independent calibration/determination of OA vs. AMS OA

PALMS data *(Froyd et al., AMT, 2019)*

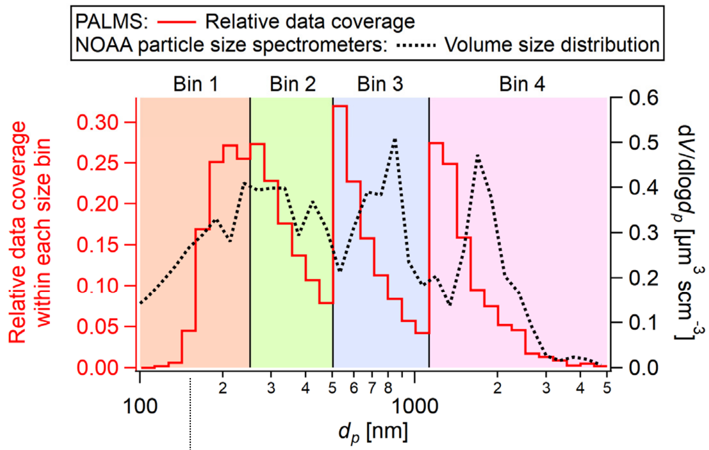
12

Speaker's notes:

- Since the PALMS composition data is mapped to the NOAA size distributions, the AMS transmission is applied to the PALMS mass product to allow an apple-to-apple comparison with AMS.
- The agreement between the two completely different methods of quantifying OA is convincing and exciting since OA represents a large family of organic molecules with different physiochemical properties. This makes the OA mass measurement inherently more uncertain than sulfate for instance.

PALMS composition coverage

University of Colorado Boulder



- Uneven data coverage within a bin.
- For the smallest bin, weighted towards > 140 nm particles; if homogeneous composition in this bin, no bias; otherwise, bias exists.
- No data coverage below 100 nm.

Next, we calculate the small particle number fraction in Bin 1 ($\frac{N_{100-150nm}}{N_{100-240nm}}$) to investigate the potential bias.

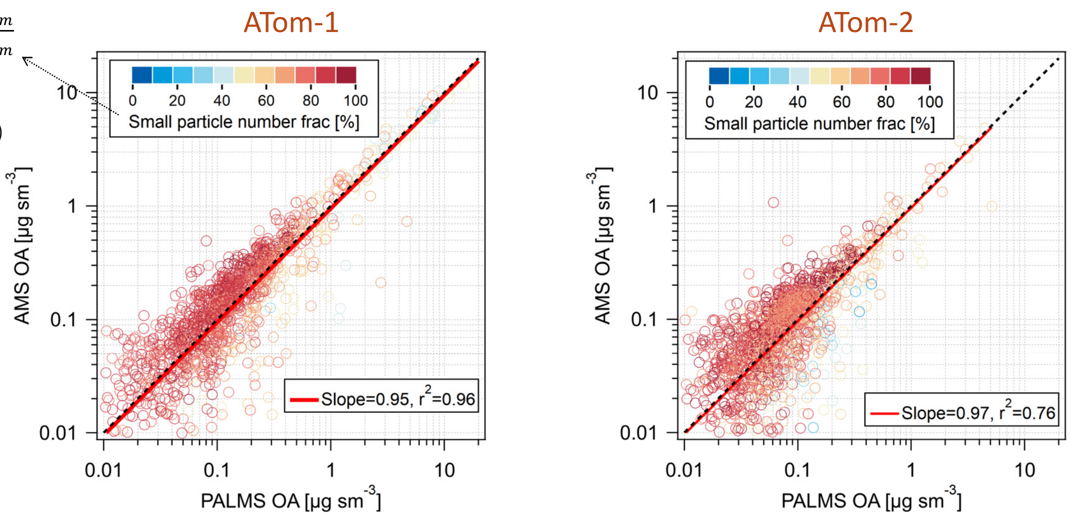
(Froyd et al., AMT, 2019)

Looking at the effect of small particles for AMS vs PALMS

University of Colorado Boulder

$\frac{N_{100-150nm}}{N_{100-240nm}}$

For Bin 1 (see the last slide)



Extrapolating the composition from larger particles, which sometimes has more SO_4 and less OA

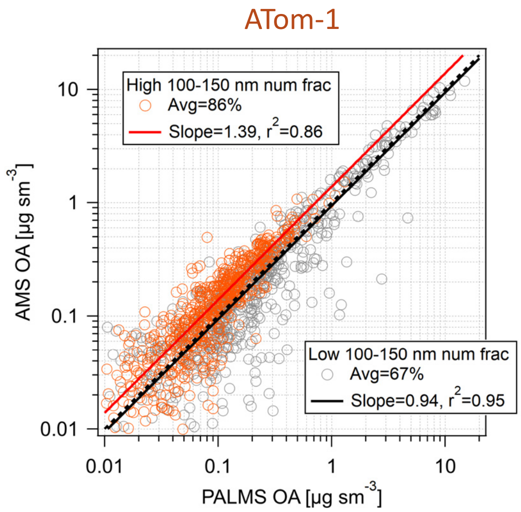
➢ PALMS will be biased when 100-150 nm particle composition ≠ 150-250 nm particles

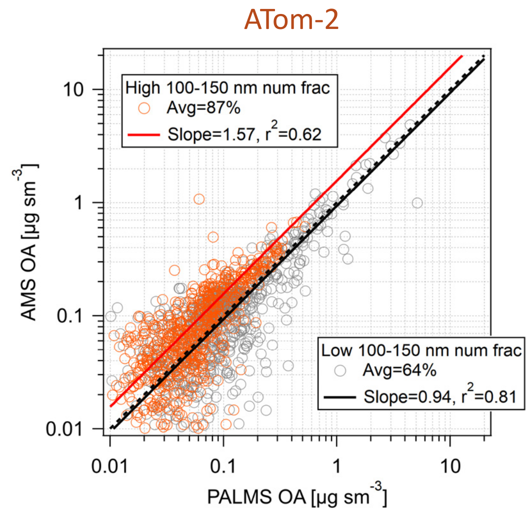
14

Speaker's notes:

- The color gradients suggest the existence of heterogeneous aerosol composition within Bin 1 (100-240 nm).

Looking at the effect of small particles for AMS vs PALMS





Extrapolating the composition from larger particles, which sometimes has more SO_4 and less OA

➢ PALMS will be biased when 100-150 nm particle composition ≠ 150-250 nm particles

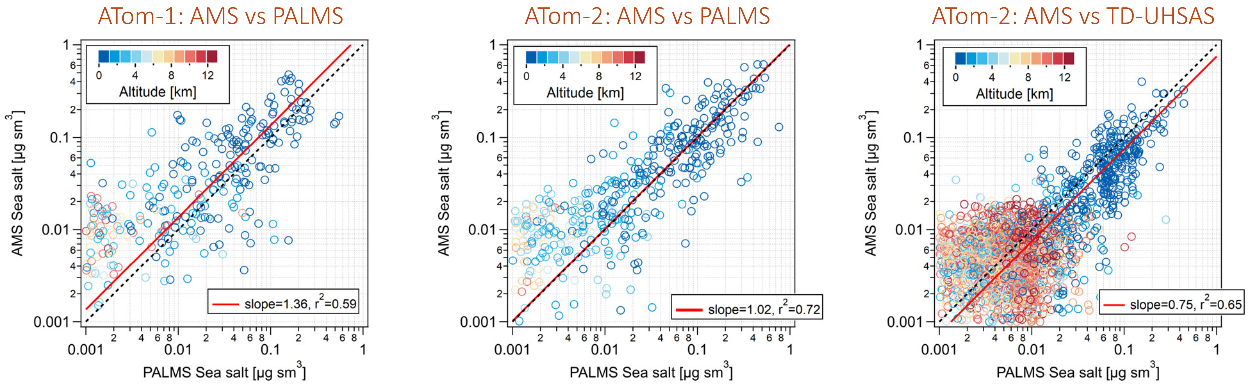
15

Speaker's notes:

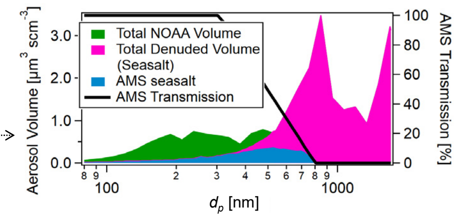
- Fitting for high vs low 100-150 nm number fraction data shows clearly the potential bias when composition is not homogeneous within a PALMS assigned particle bin.
- Exclusion of those higher 100-150 nm number fraction points doesn't affect linear regression slope and r^2 for ATom-1 and slight change in ATom-2.

Submicron sea salt: Good agreement of AMS, PALMS and TD-UHSAS

University of Colorado Boulder



- Good agreement
- High noise:
  - due to the enhanced steepness combined from AMS transmission and sea salt size distribution



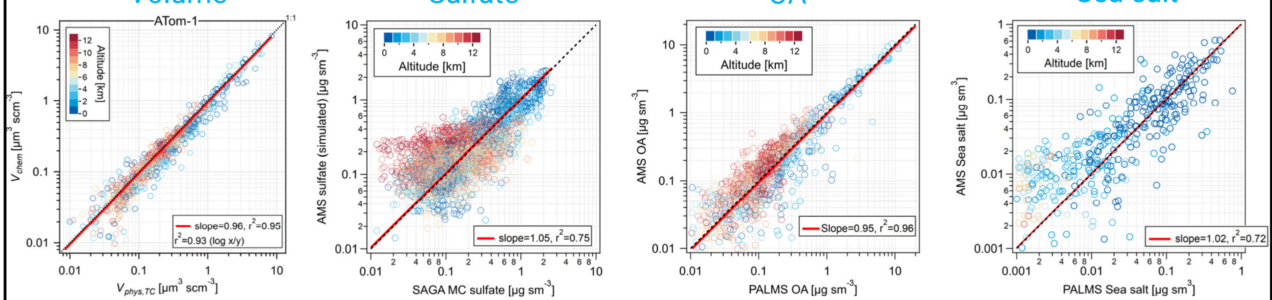
AMS seasalt: Method of Ovadnevaite et al., JGR 2012, calibrated in lab

Speaker's notes for EGU audience:

- TD-UHSAS: thermally-denuded (300 °C) Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)
- Other than the sea salt agreement between AMS and PALMS, we also compare the AMS sea salt volume to TD-UHSAS volume when there were low BC concentrations. Thus, sea salt is expected to be the main TD-UHSAS volume.

Summary

University of Colorado Boulder



i. Physical and chemical measurements of submicron aerosols for the ATom mission are consistent within uncertainties.

ii. For the AMS, reported uncertainties (2σ accuracy: Sulfate: 35%, OA: 38%, Sea salt: 50%) seem consistent with the comparisons.

iii. Size transmissions and instrument idiosyncrasies need to be considered.

Contact info for personal communication:
Hongyu Guo: hongyu.guo@Colorado.edu; Pedro Campuzano-Jost: Pedro.CampuzanoJost-1@colorado.edu; Jose-Luis Jimenez: jose.jimenez@colorado.edu