AMS Quantification across 14 time zones: Physical and chemical closure during the NASA ATom mission

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AMS Users Meeting (virtual)
Jan 20th, 2021

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

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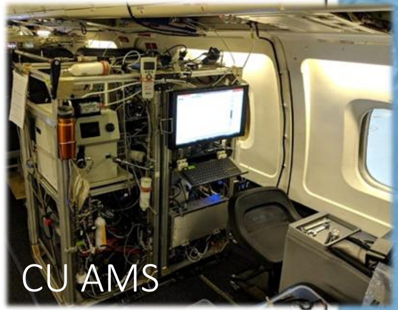

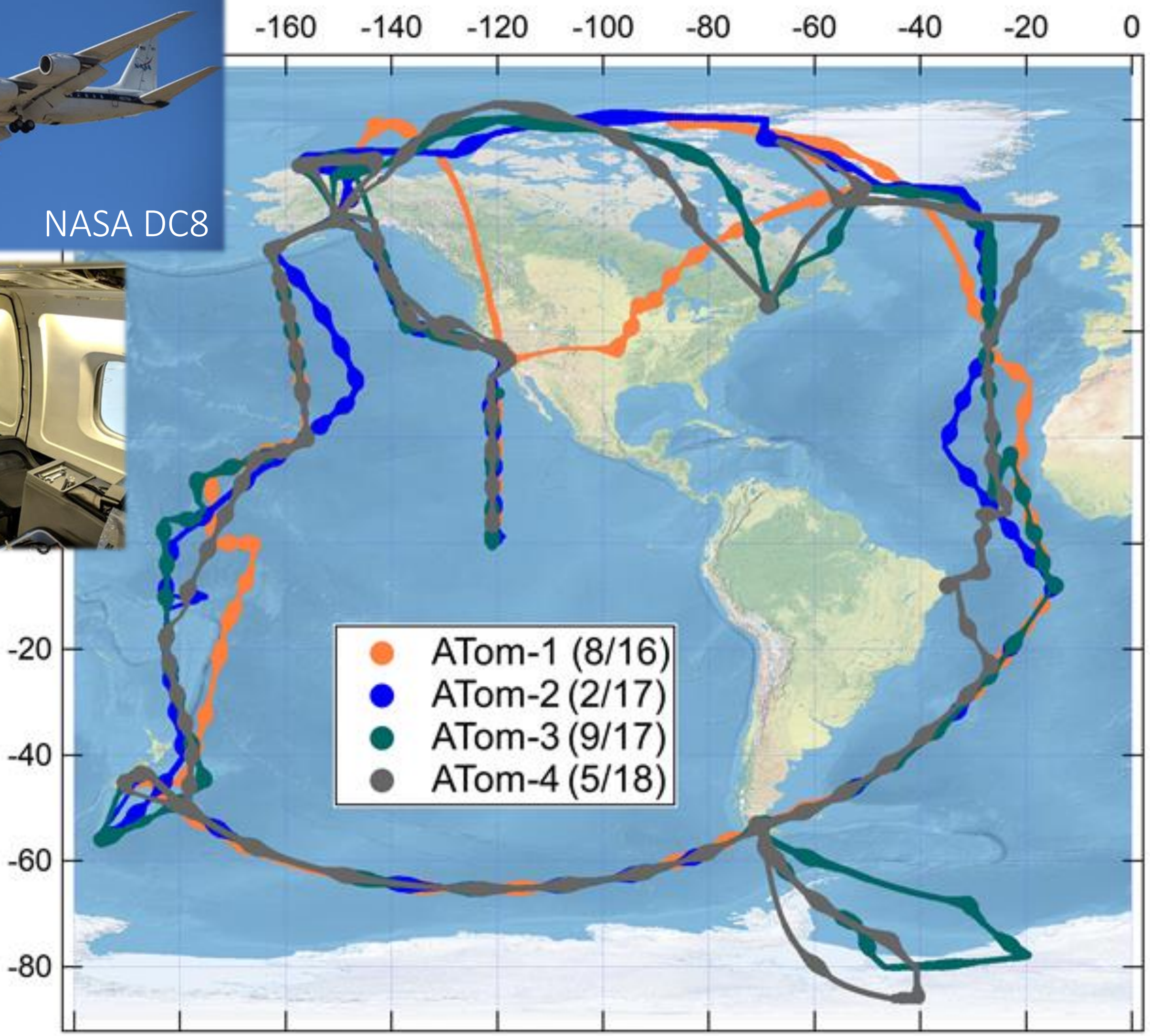

Motivation of this study:

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

  https://daac.ornl.gov/ATom

  This data will be widely used to evaluate and constrain global models.
- Is the current understanding of the uncertainties of the Aerodyne Mass Spectrometer (AMS) consistent with ATom performance?
- Paper on AMTD (https://www.atmos-meas-tech-discuss.net/amt-2020-224/)

ATom in-cabin aerosol payload: Physical & chemical sensors

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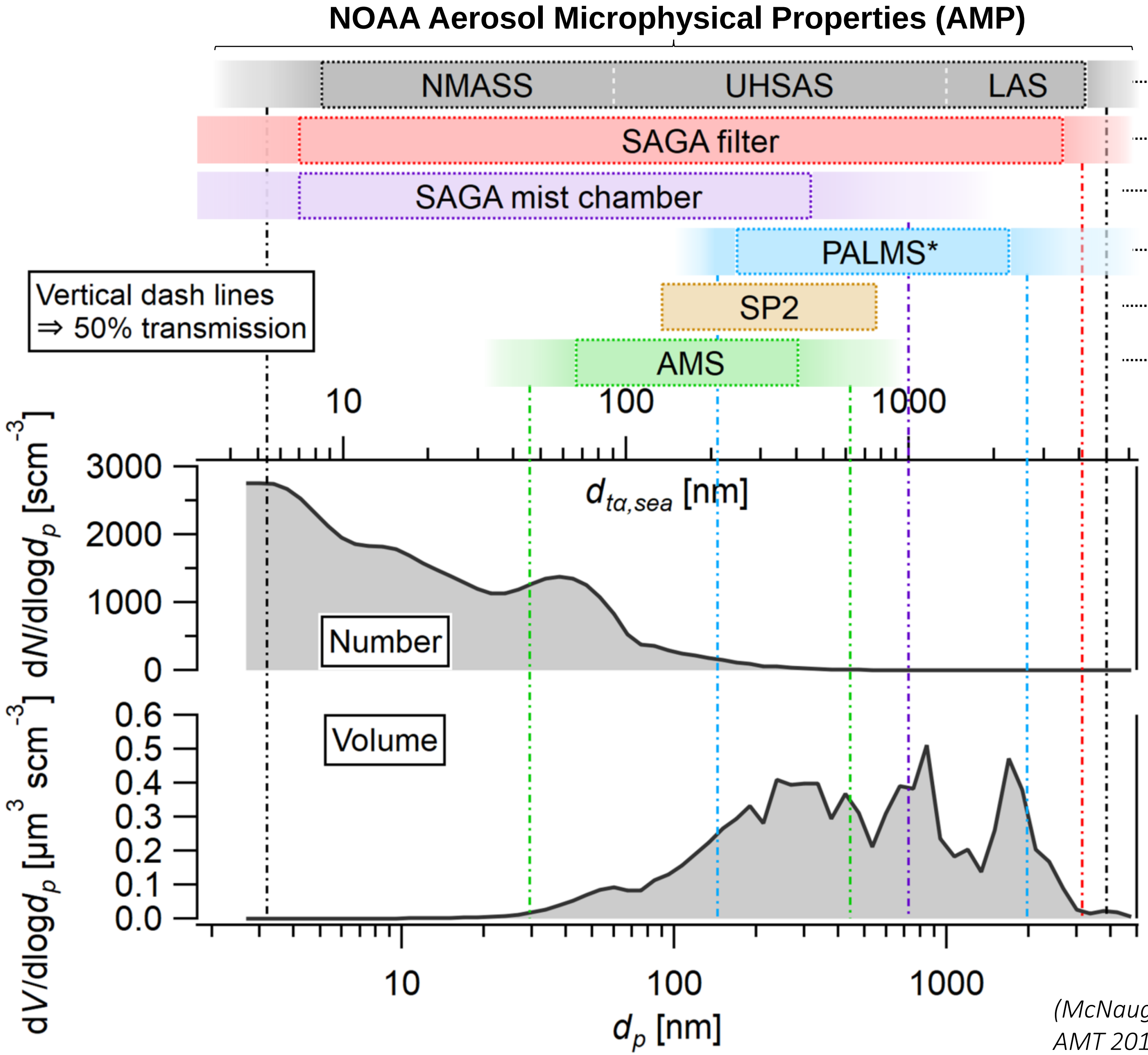


- Particle size spectrometers: 3 nm to 4.8 µm (d_p) — **Physical**
- Offline water-soluble ions
- Online water-soluble ions
- Particle types (mass and number fraction)
- refractory BC
- Submicron non-refractory species

Chemical

Top axis: Aerodynamic diameter (d_{ta})

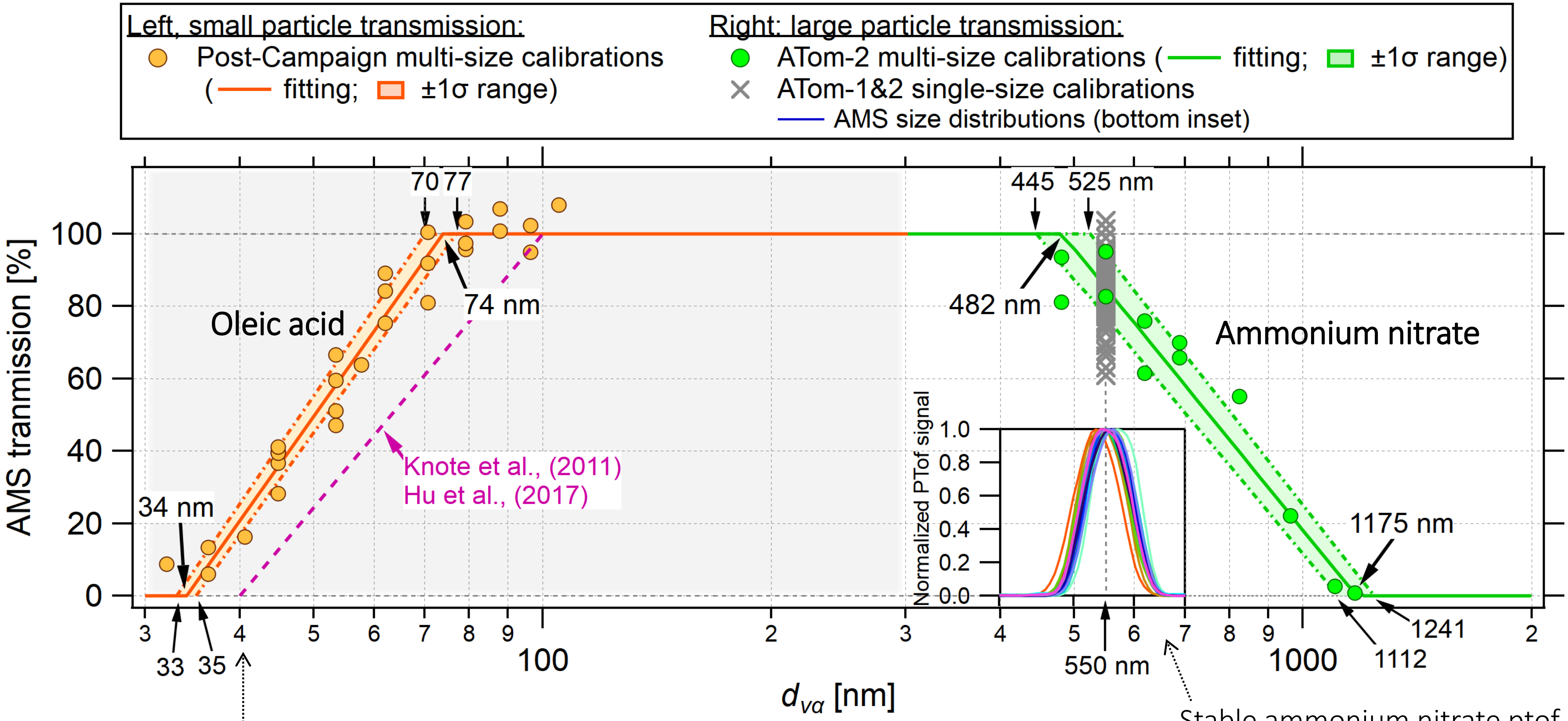
ATom-2 campaign average size distribution (AMP)

Bottom axis: Geometric (optical) diameter (d_p)

(McNaughton et al., AST 2007; van Donkelaar et al., ACP 2008; Williamson et al., AMT 2018; Kupc et al., AMT 2018; Brock et al., AMT 2019; Froyd et al., AMT 2019)

Calibration of the AMS inlet transmission (ATom-1&2)

Derive AMS transmission by comparing the number/mass observed by AMS to CPC for monodisperse particles



See Dongwook's prez (Jan 21st) on how to generate small and monodisperse particles

Vacuum Aerodynamic diameter

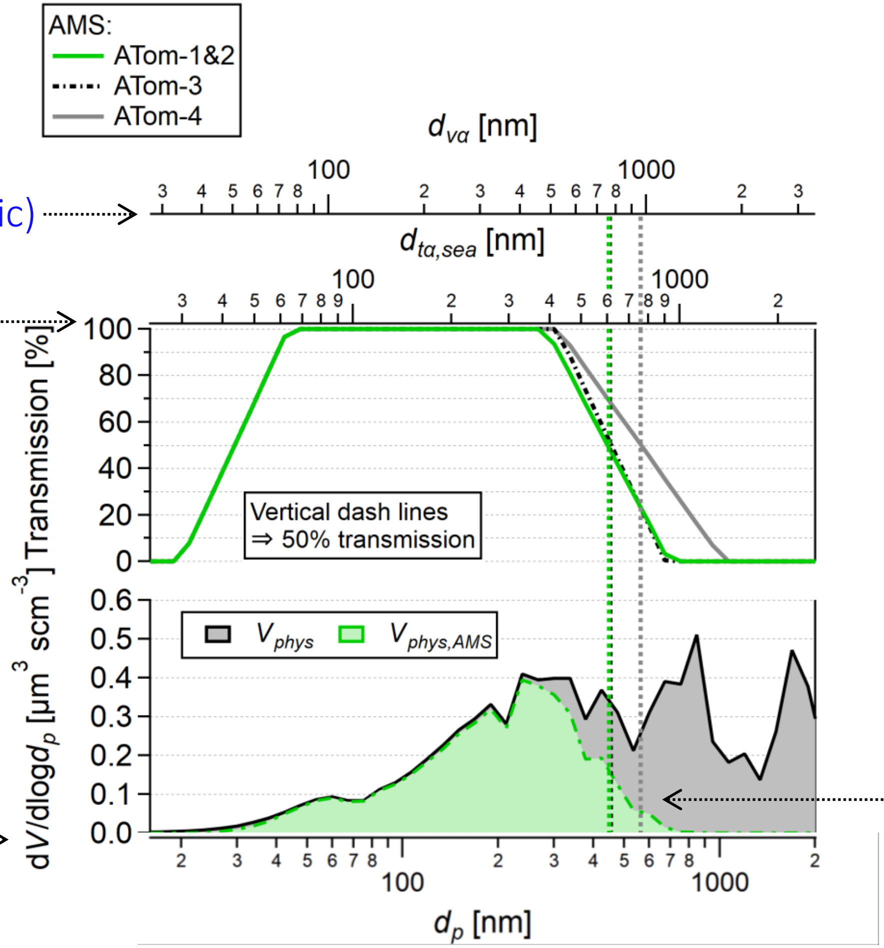
Stable ammonium nitrate ptof throughout the campaign

What is PM_1 and the fraction that AMS observes?

(Vacuum aerodynamic)

(Aerodynamic)

(Geometric, also optical)



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-1&2 and **$PM_{0.9}$** in ATom-4 (effect of particle liquid water considered).

- 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)

See DeCarlo et al., AST 2004 for the diameter conversions, which is density dependent

What is PM_1 and the fraction that AMS observes?

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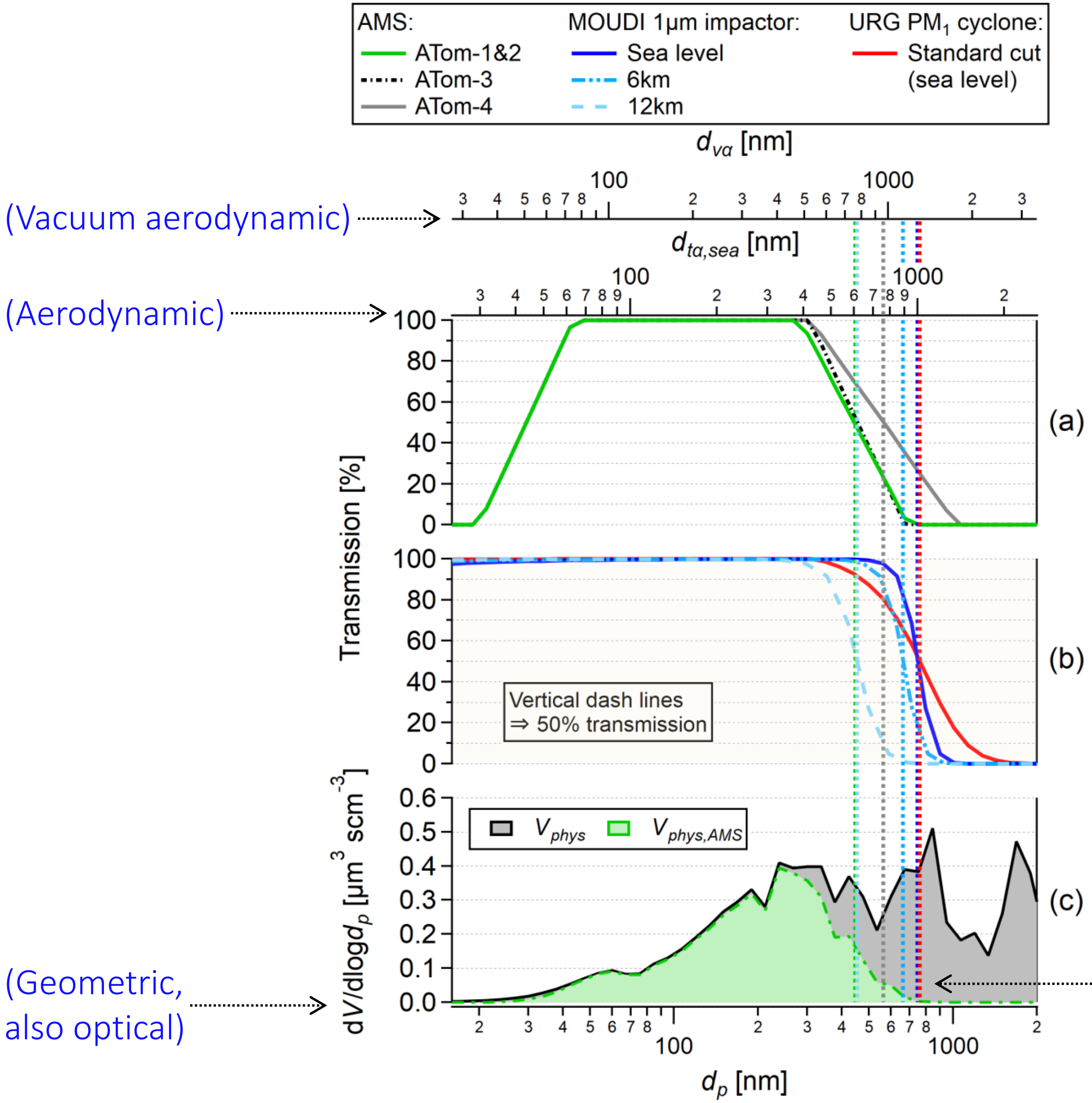

- An advantage of using a pressure-controlled inlet (PCI) has a constant transmission up to ~9 km, while the aerodynamic cutoff of MOUDI keeps decreasing with altitude (SAGA MC is similar).

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

What is PM_1 and the fraction that AMS observes?

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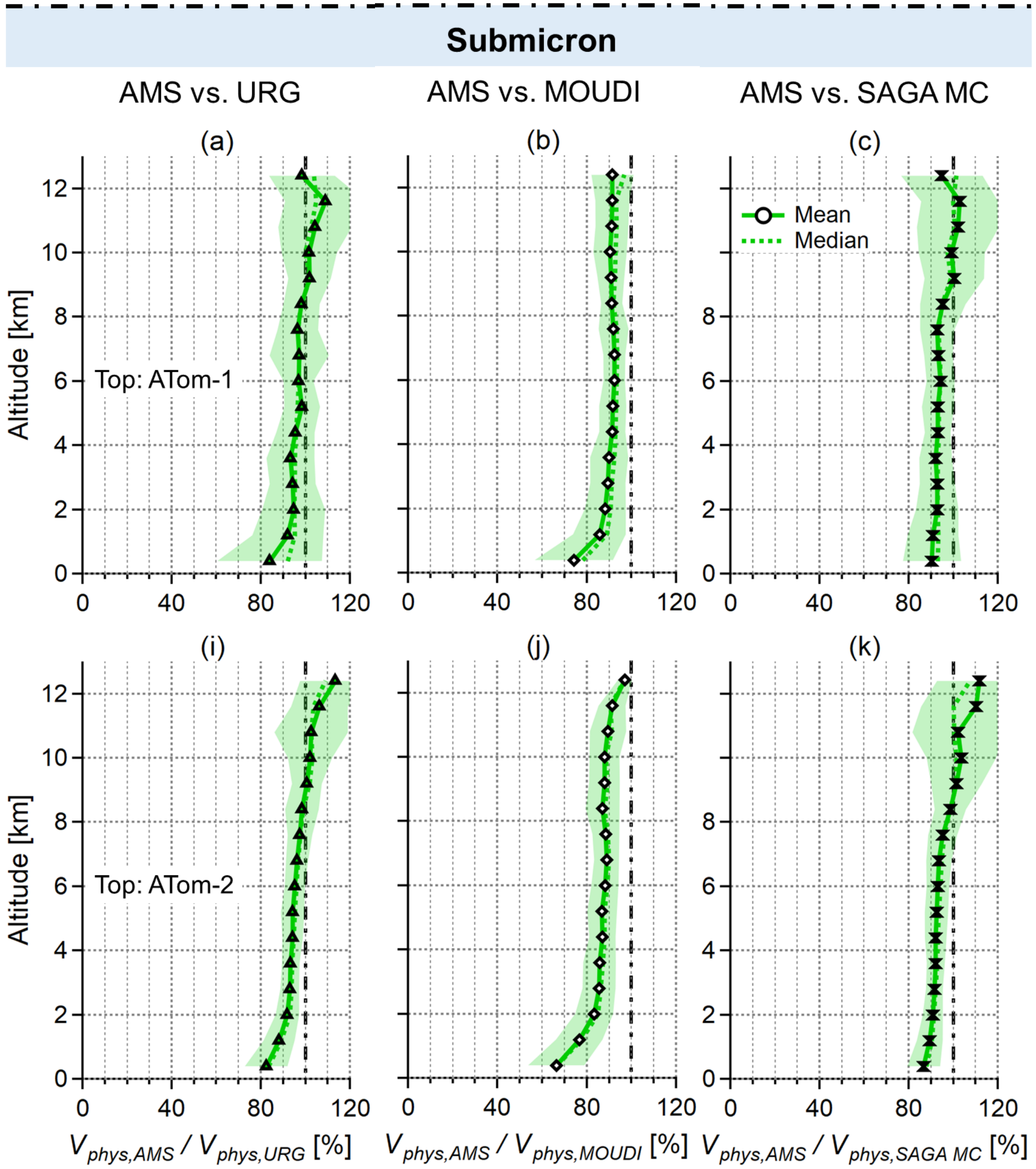

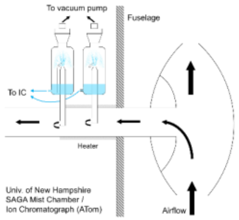

URG: standard cut URG 1µm cyclone (widely used on the ground, size-selecting <u>ambient condition</u> particles);

MOUDI: micro-orifice uniform deposit impactor (1µm stage), size-selecting <u>dry condition</u> particles in the cabin of airplane;

SAGA MC: Soluble Acidic Gases and Aerosol Mist Chamber, onboard DC8 size-selecting <u>ambient condition</u> particles;

- AMS covers **95±14%** of the standard PM_1 volume (URG standard-cut cyclone) for ATom-1&2 conditions (more in ATom-4);
- For ATom, AMS is directly comparable to SAGA MC (**97±14%**).

V_{phys}: the integrated volume of AMP from 2.7 nm to 4.8 µm
$V_{phys,AMS}$: the V_{phys} applied with AMS inlet transmission

What particle fractions are detectable by AMS for the full AMP size range?

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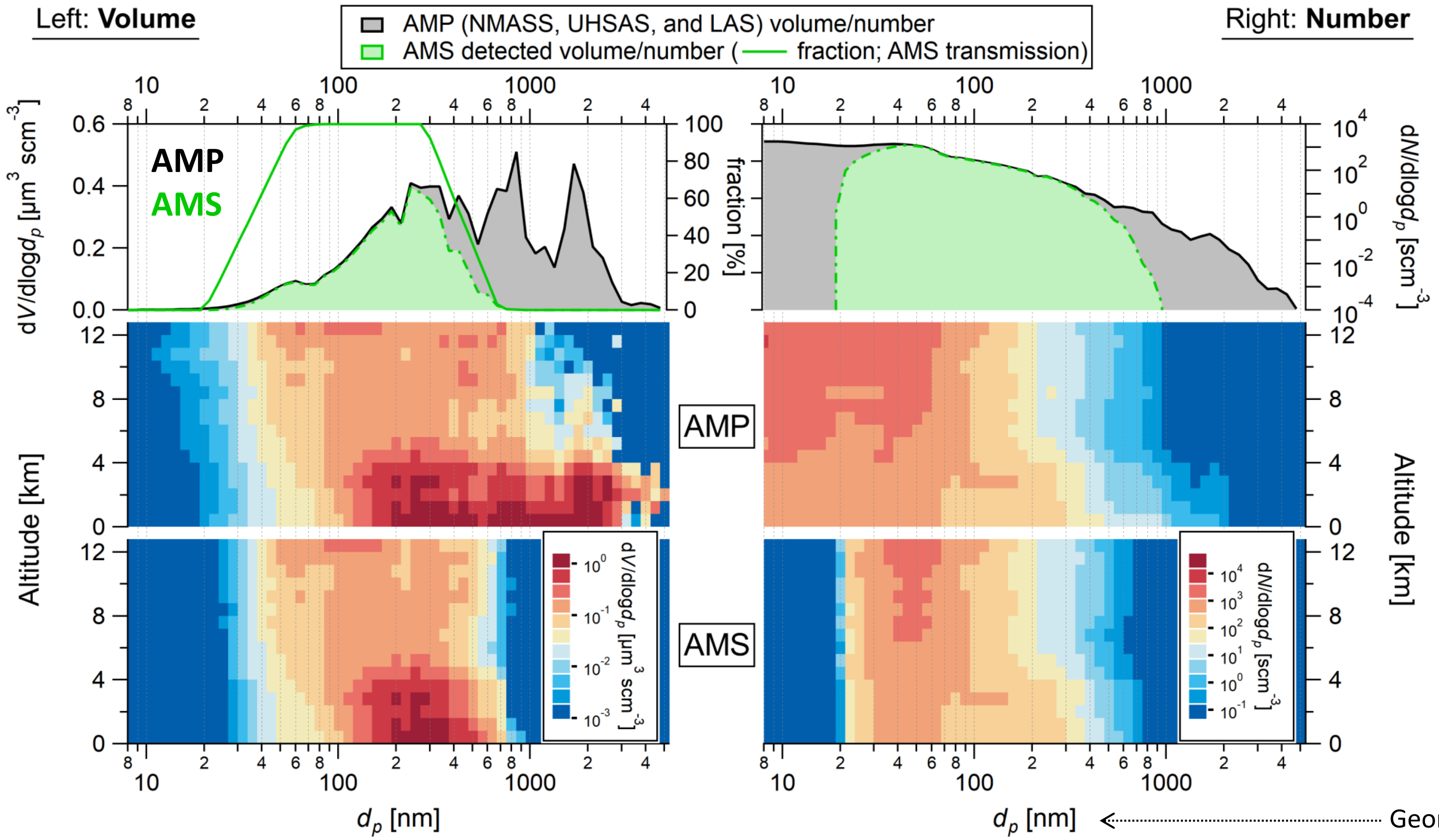
Averages and transmission

Total size distribution (AMP)

AMS detected distribution



AMS observes 68% (74% median) of AMP volume, 41% (41% median) of number;

What particle fractions are detectable by the companions?

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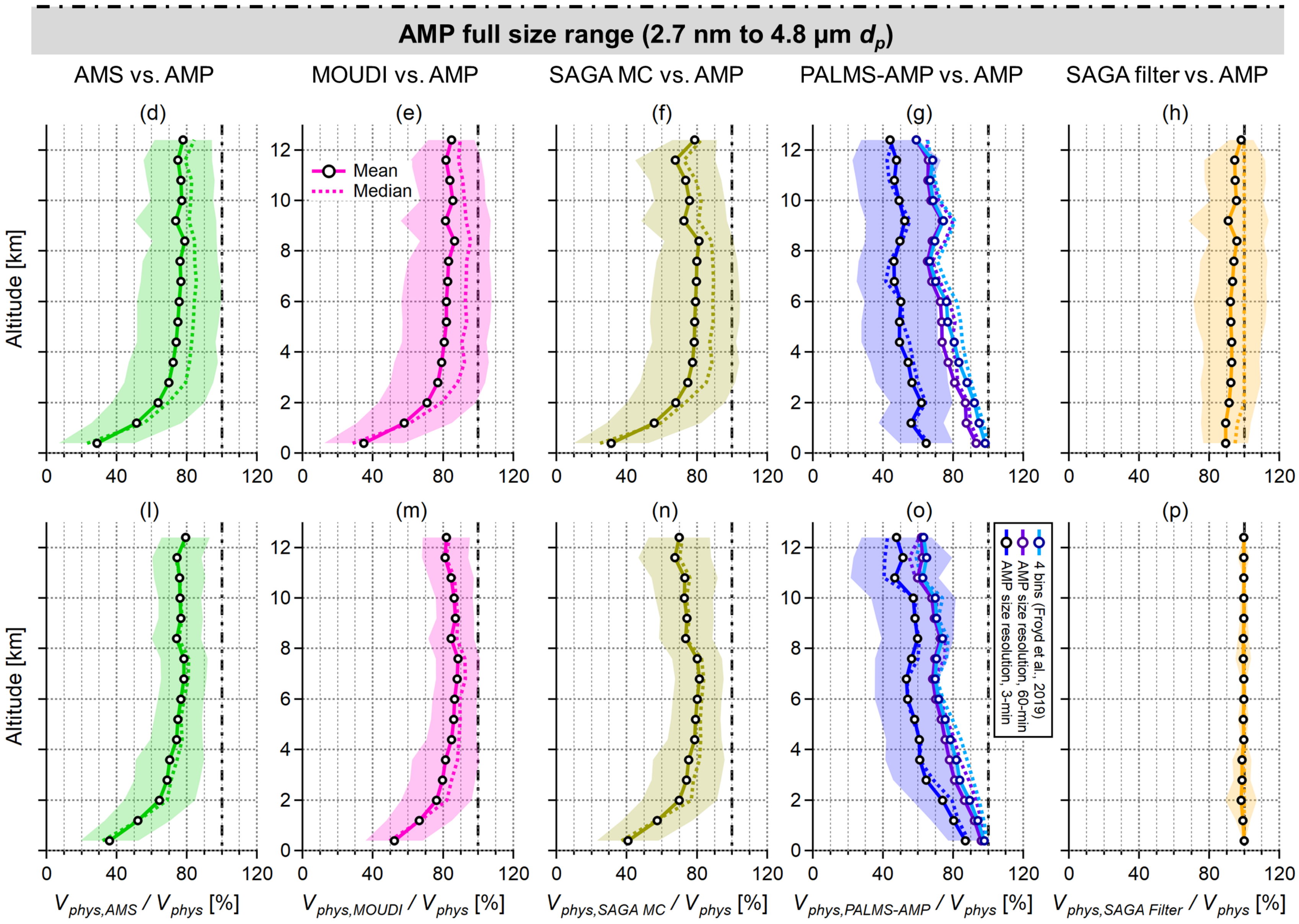
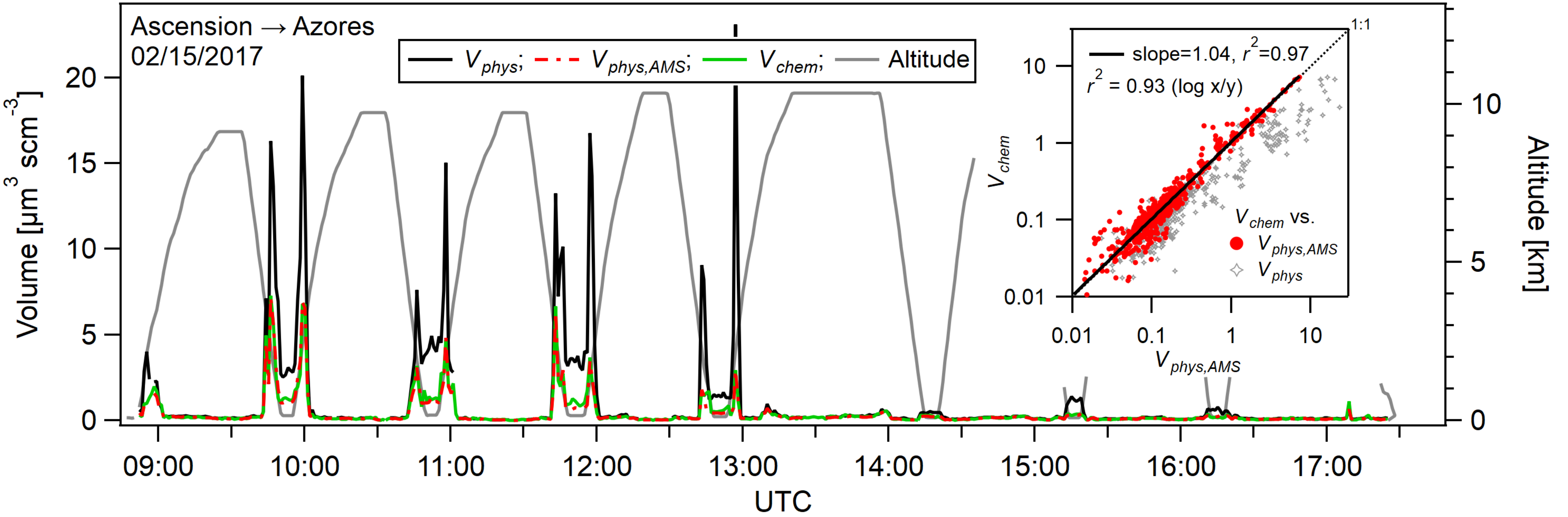


- SAGA filter collects almost the same as the AMP;
- The vertical trends of PALMS-AMP are different to others (since it doesn't report <100 nm, and particles are smaller at higher elevation), complimentary to the other collocated aerosol instruments;

A good transmission curve allows meaningful comparisons



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

AMS non-refractory — AMS refractory

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 ($V_{phys,AMS}$) and AMS+BC (V_{chem}) volumes

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

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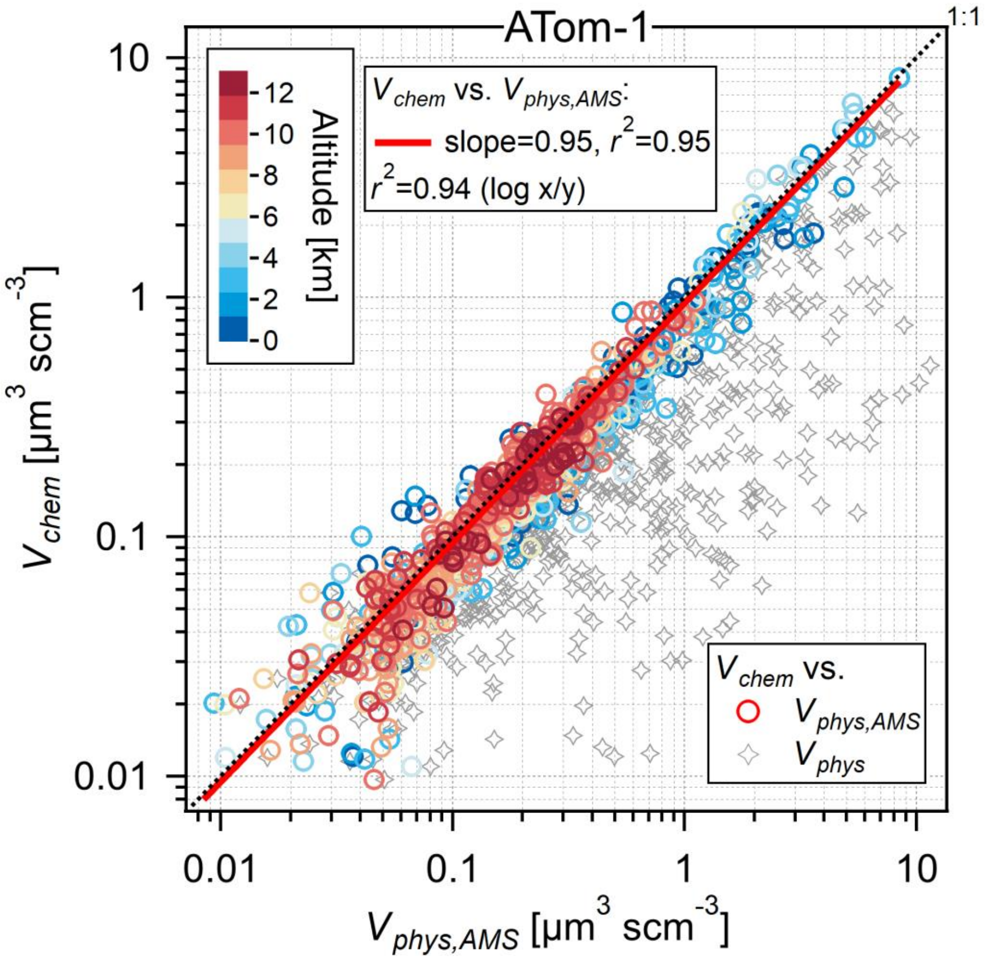
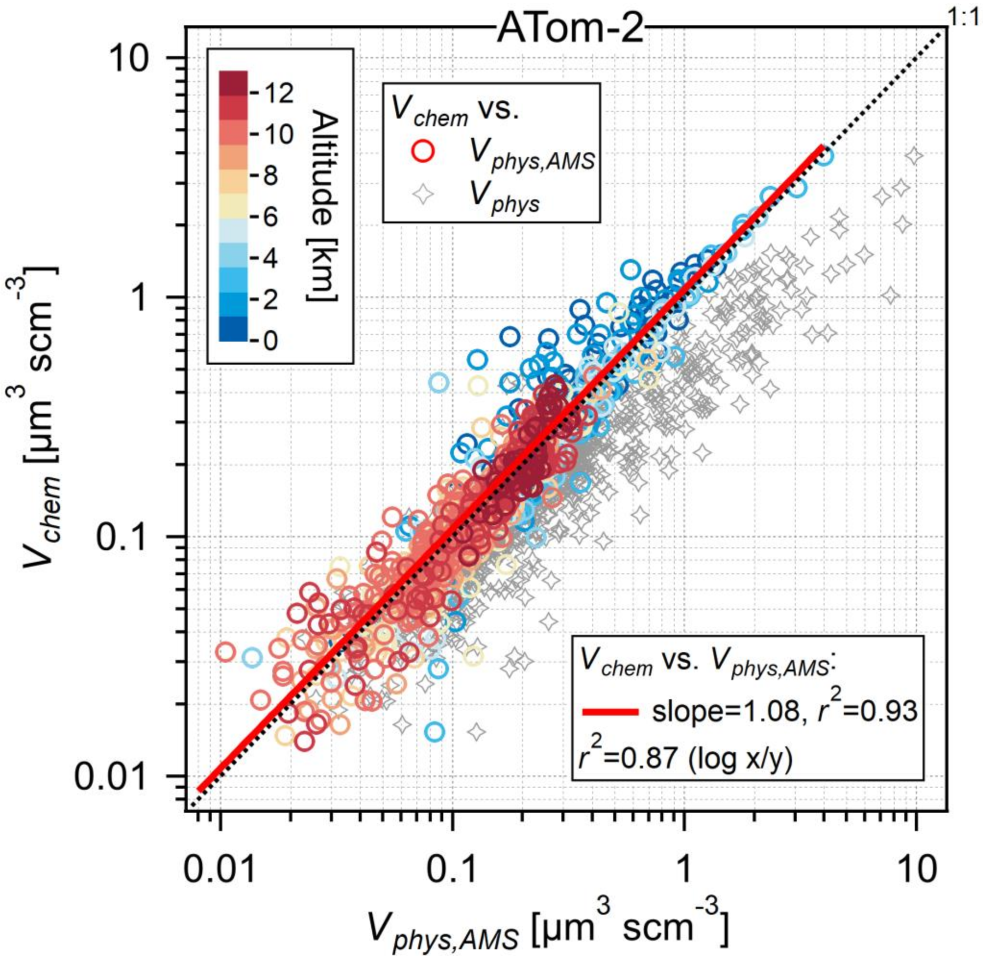


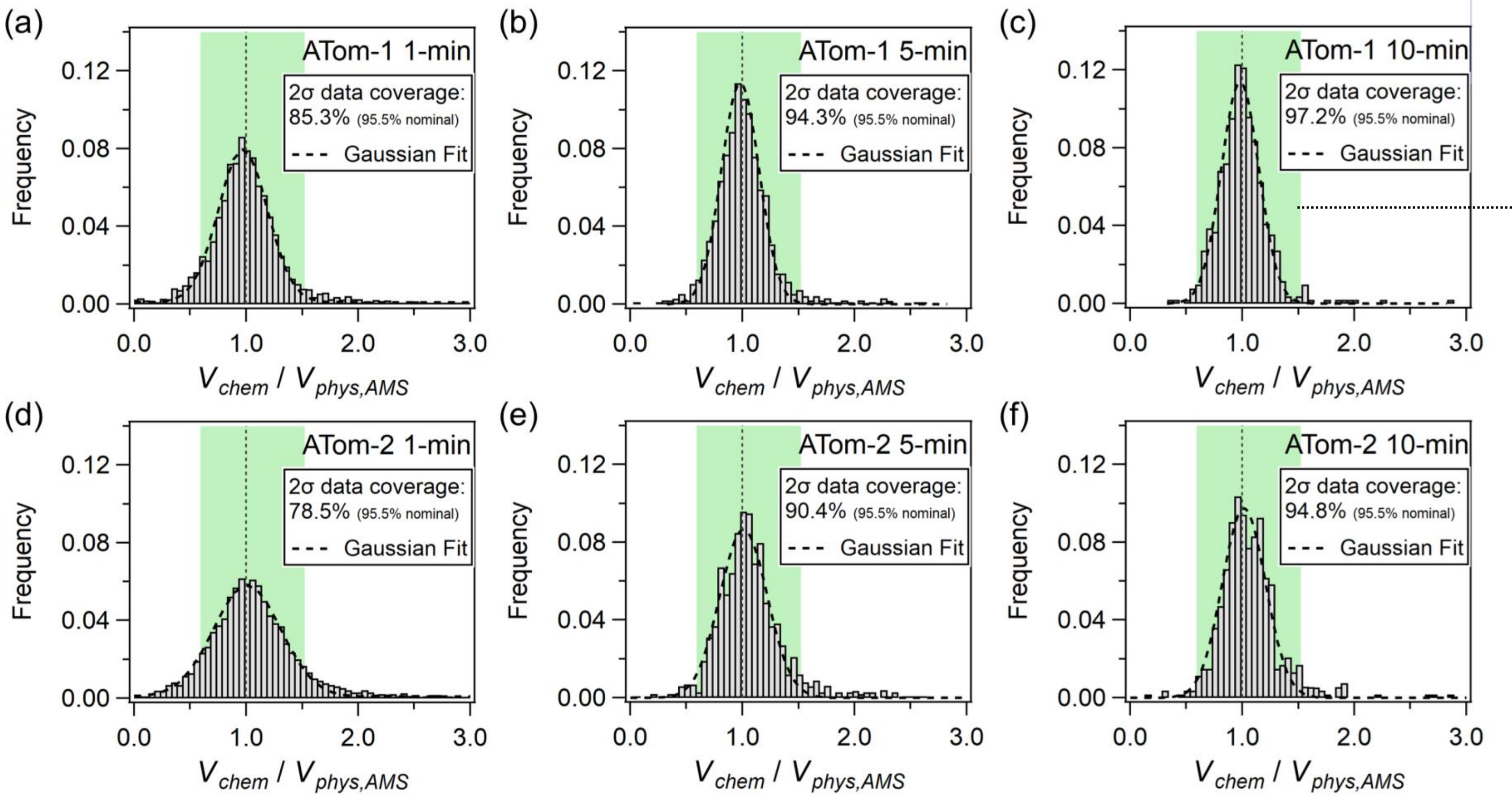


Consistent agreement for ATom deployments

→ No unknown large bias in *CE*, *RIE,* etc. for the remotely aged particles sampled during ATom

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

Measurements are consistent within the stated uncertainties



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 for a well calibrated instrument.

Bahreini et al., JGR 2009

1-min → 5-min → 10-min

Longer averaging time interval smooths out random noise (prominent in ATom due to the clean remote air, ATom-1 0.50 μg m^{-3} and ATom-2 0.38 μg m^{-3})

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

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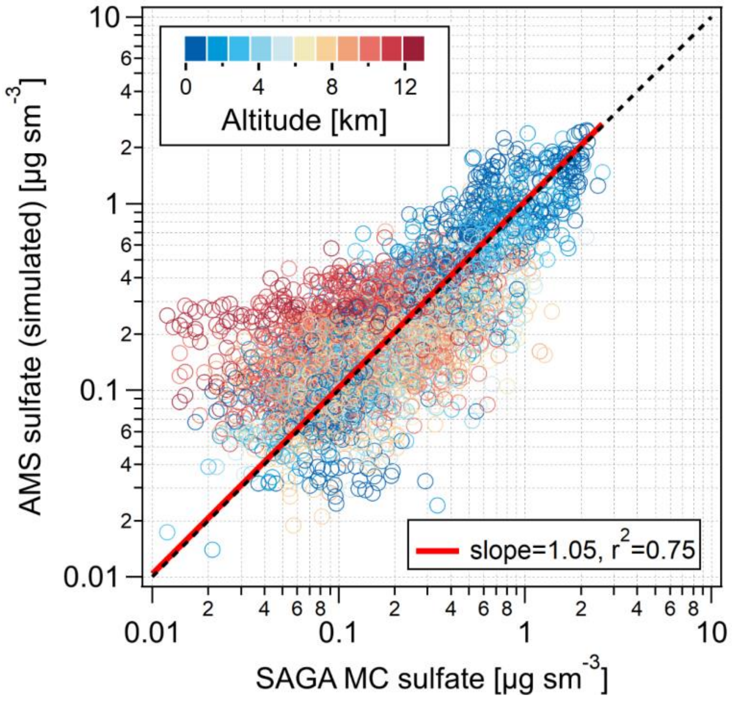
(Simulated based on the tailing effect of SAGA MC; see next slide)

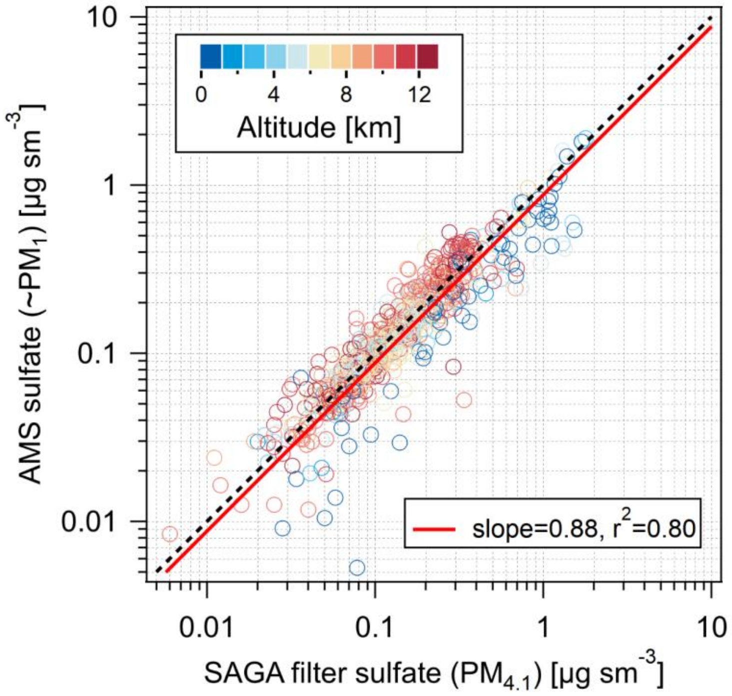
ATom-1: AMS vs. SAGA MC



Resolution: 75 sec

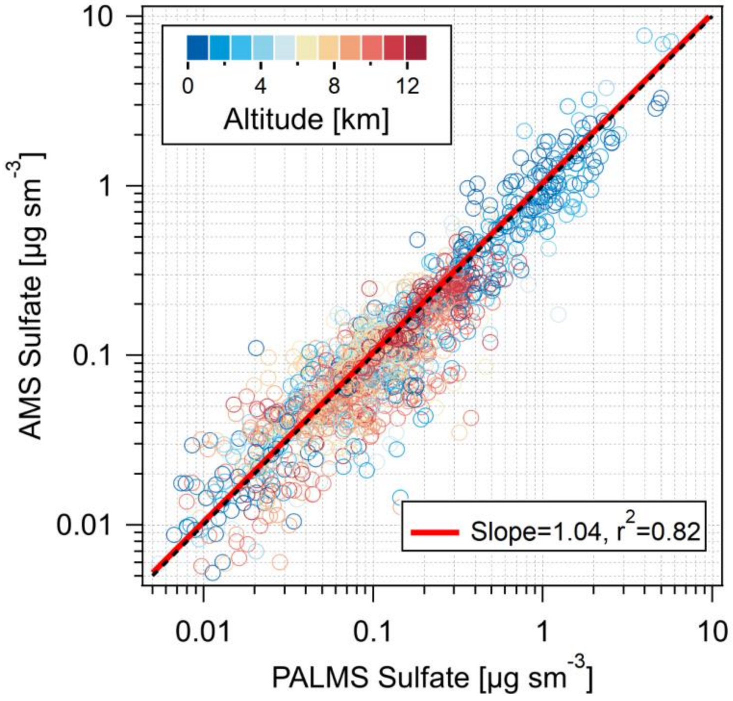
AMS vs. SAGA Filters

(excluding supermicron particle events)



Resolution: 5-20 min

ATom-1: AMS vs. PALMS (PM_1)



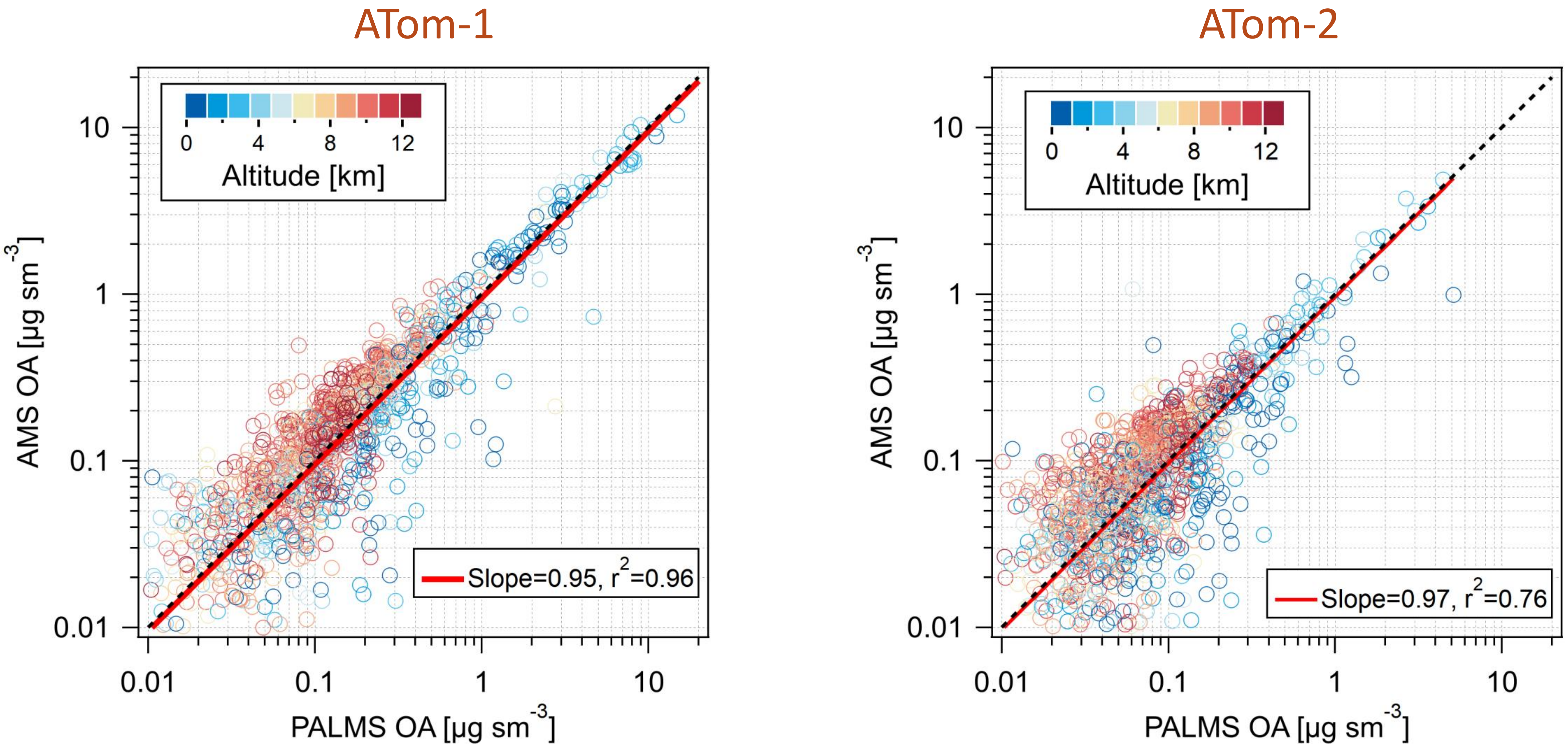
Resolution: 3 min

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

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)*

Summary

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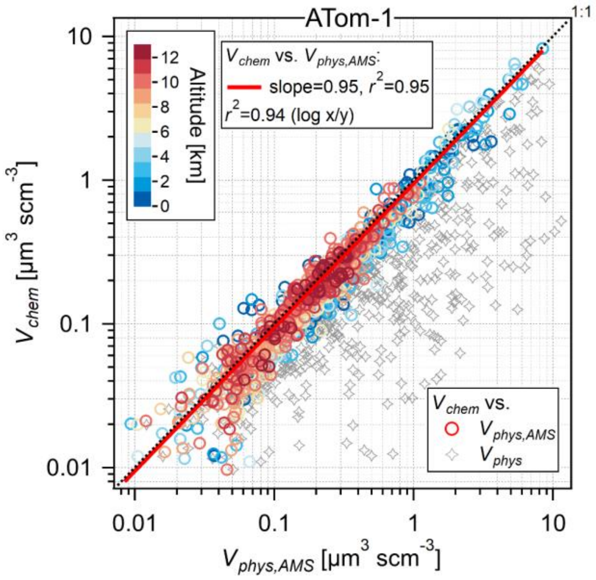
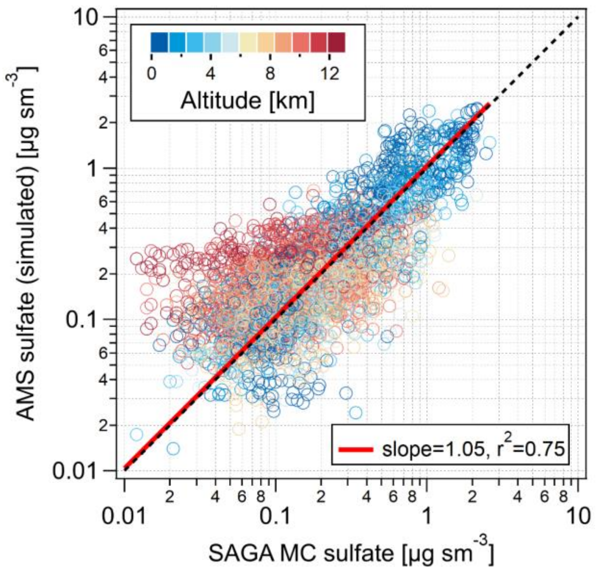
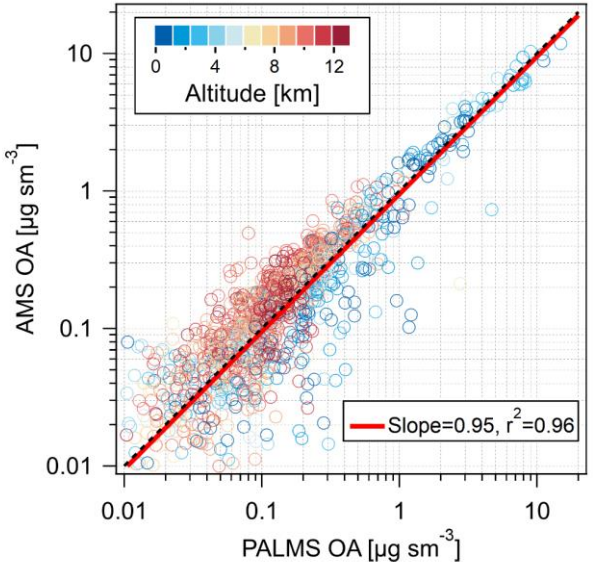
Volume



Sulfate



OA



i. In-field calibration of inlet transmission, especially the upper end, is important for meaningful intercomparisons.

ii. The AMS was an equivalent PM_1 measurement during ATom compared to other submicron size-selections.

iii. Physical and chemical measurements of submicron aerosols are consistent within uncertainties.

iv. The reported AMS uncertainties (2σ accuracy: Sulfate: 35%, OA: 38%) are consistent with the comparisons.

v. Size transmissions (e.g., the pressure dependency for aircraft studies) and instrument idiosyncrasies need to be considered for intercomparisons.

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