ACSM measurements robustness, quality assurance, and impact of upper size cutoff diameter

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ACMCC Aerosol Chemical Monitor Calibration Center

COLOSSAL Chemical On-Line cOmpoSition and Source Apportionment of fine aerosoL

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Leibniz Association

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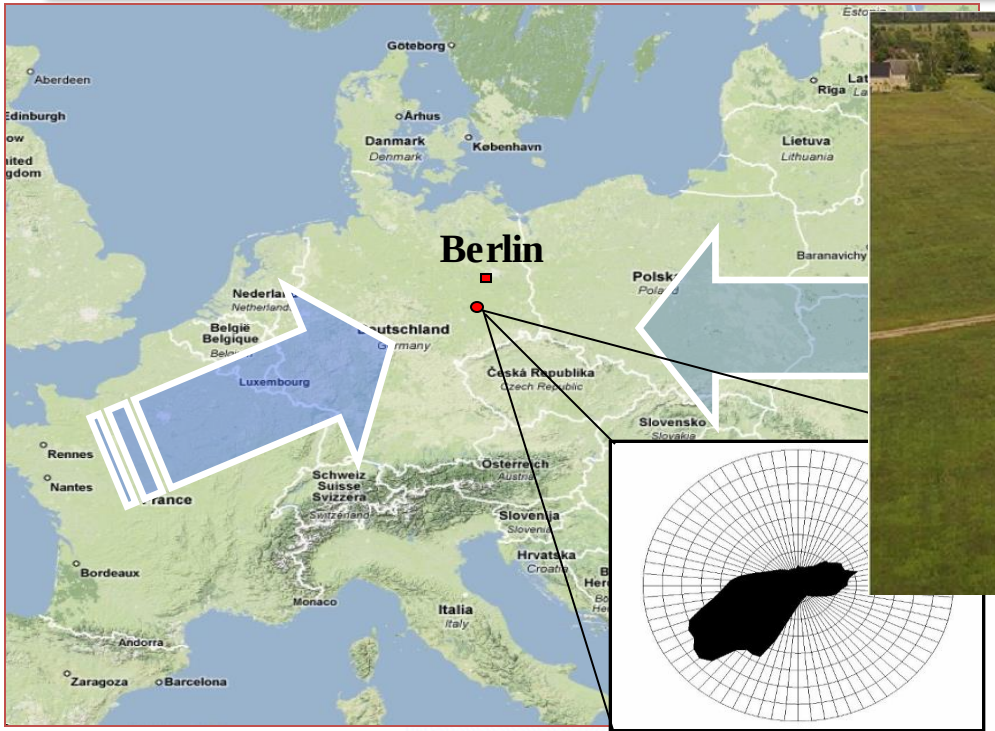
AMS users meeting 20.01.2021

TROPOS Research Station Melpitz (DE44)



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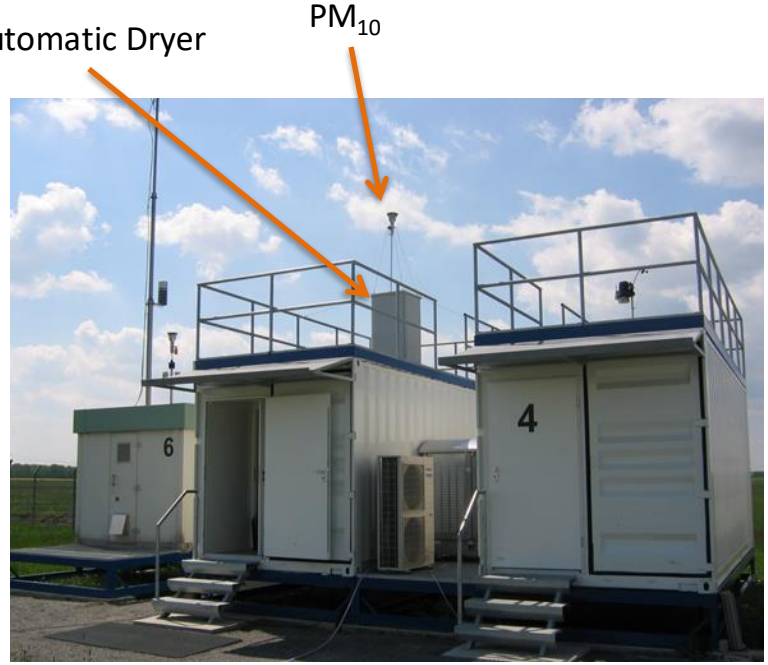

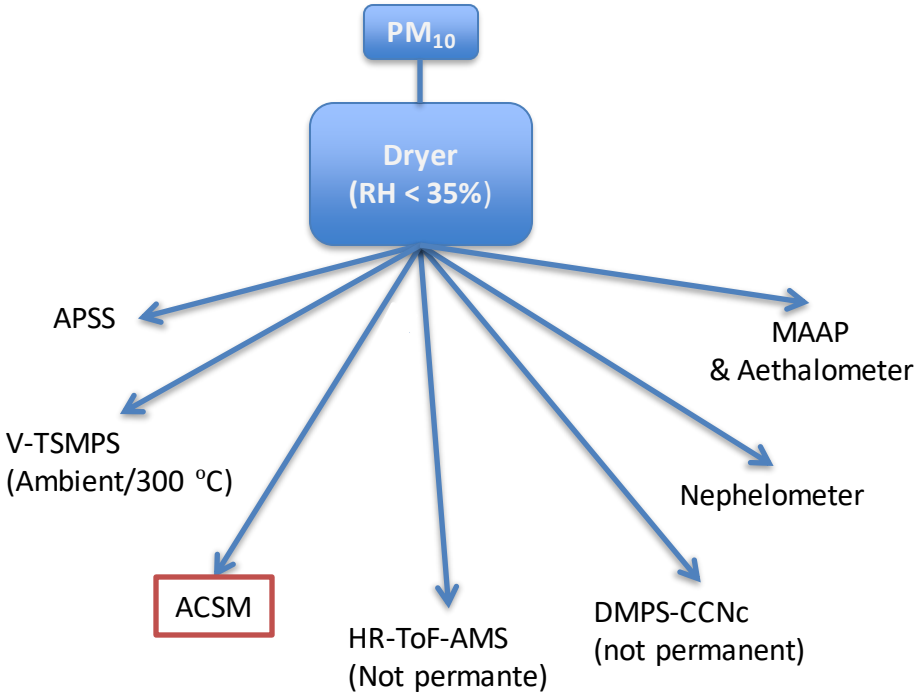

Additional collocated permanent instruments:

- MARGA (PM_{10})
- Digitel PM_1,$PM_{2.5}$, PM_{10}
- Gas tracers (NO_x, O_3, SO_2)
- Ceilometer, solar radiation

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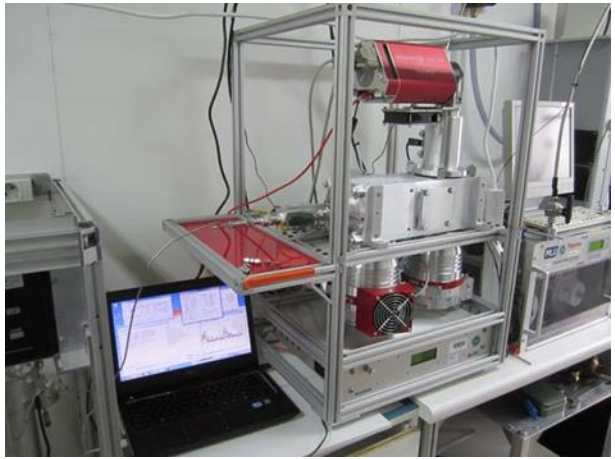
Instrumental comparison



+ MAAP (eBC)

Off-line: High-Vol 24h sampling

- $PM_{2.5}$ daily
- PM_1 every 6d / daily during specific periods

⇒ Total mass, Nitrate, Sulfate, Ammonium, Organics

SMPS:

- Particle number size distribution (PNSD)

⇒ Total volume / Mass

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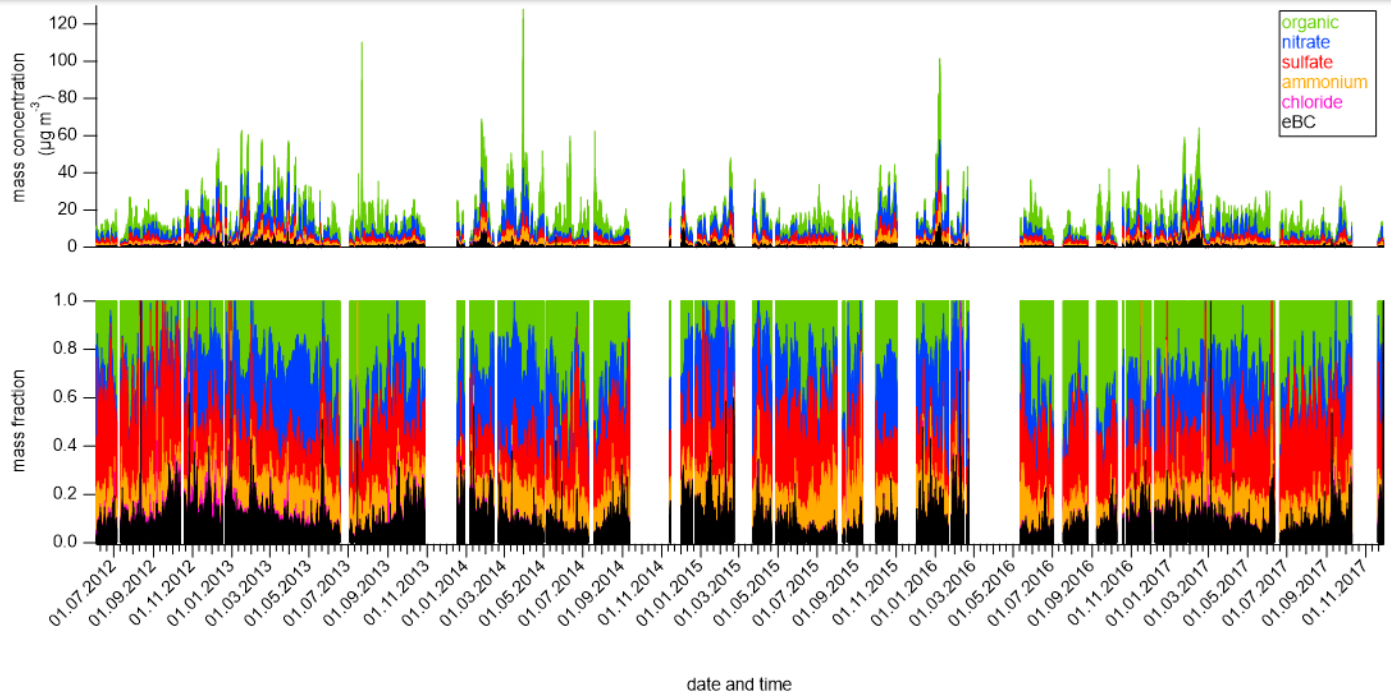
Overview of Melpitz ACSM aerosol chemical composition



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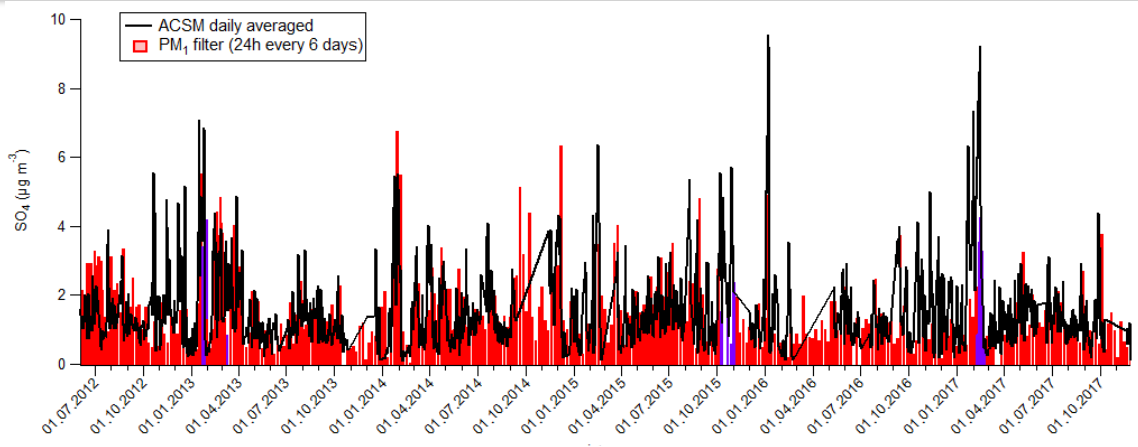
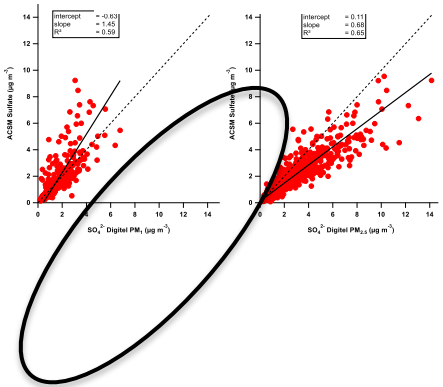
ACSM vs offline: Sulfate





⇒ No significant change on the sulfate fragments ratios

⇒ Cutoff effect?

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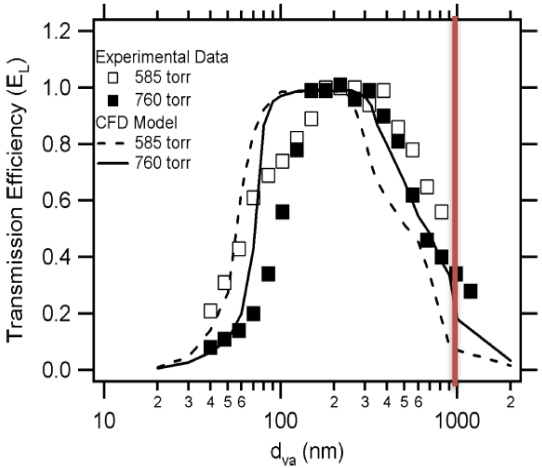
Sulfate: impact of upper size cutoff diameter

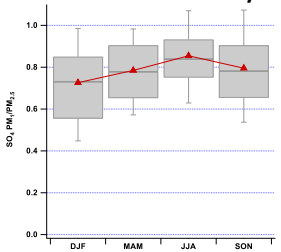
ACSM



Liu, P. S. K., et al., Aerosol Sci. Technol., 41, 2007.

⇒ ACSM has transmission efficiency of 30 - 40 % at 1 µm (d_{va})

Seasonal variability of the PM_1:$PM_{2.5}$ ratio



⇒ More coarse mode sulfate particle in winter compare to summer

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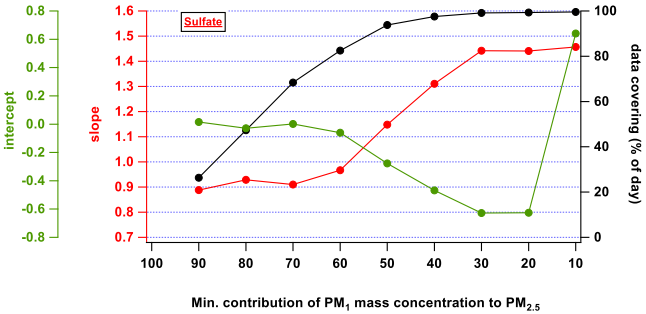
Sulfate: impact of upper size cutoff diameter

Variability of the correlation between ACSM and PM_1 sulfate parallel to the change of the PM_1:$PM_{2.5}$ ratio



⇒ Best correlation parameters when PM_1:$PM_{2.5}$ > 60 %

⇒ Clear impact of the individual size cutoff of the instruments

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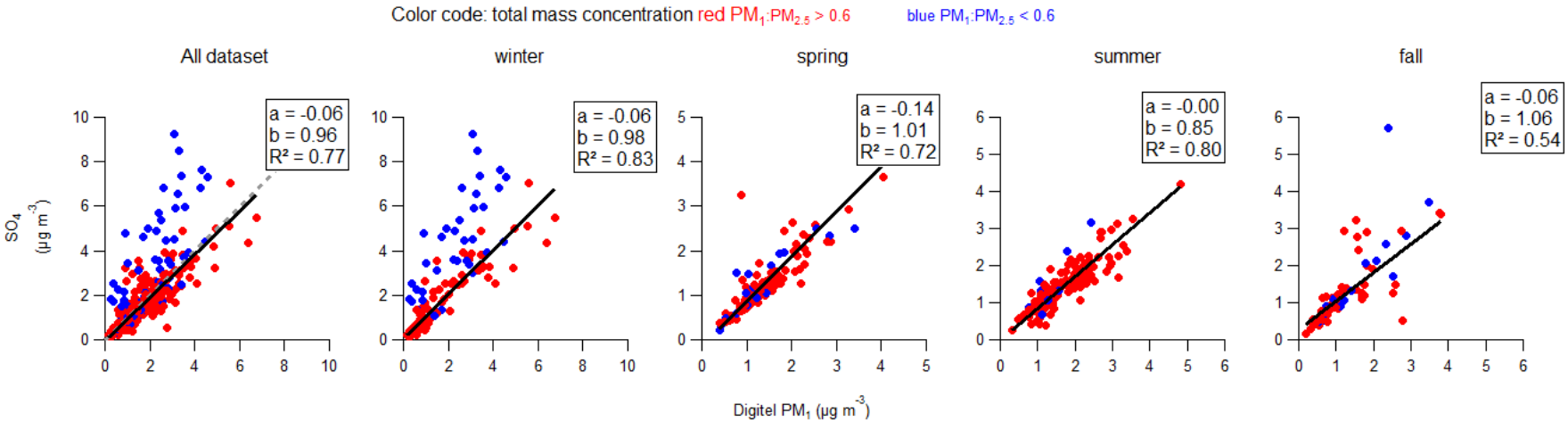
ACSM vs off-line: Sulfate



⇒ Overestimation days were systematically correlated to day with low PM_1:$PM_{2.5}$ ratio and high sulfate concentration

⇒ Cannot completely exclude a small contribution of organo-sulfate to the ACSM sulfate signal (Farmer et al., 2010)

⇒ ACSM strongly correlates with off-line PM_1 sulfate without any pronounced seasonal effect

Least Orthogonal linear fit y = a + bx

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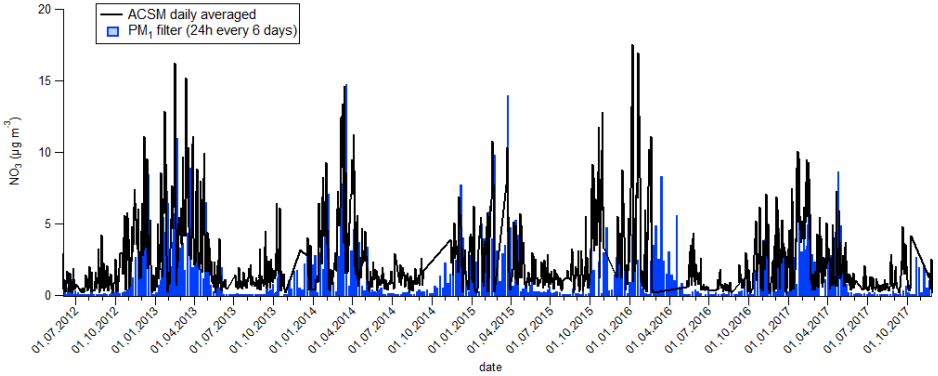
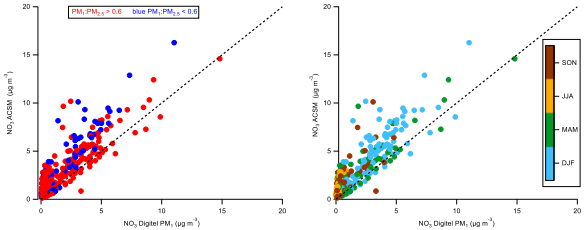
ACSM vs off-line: Nitrate





⇒ Strong seasonal variability

⇒ Small impact of the PM_1:$PM_{2.5}$ ratio

⇒ Other sources of artefact

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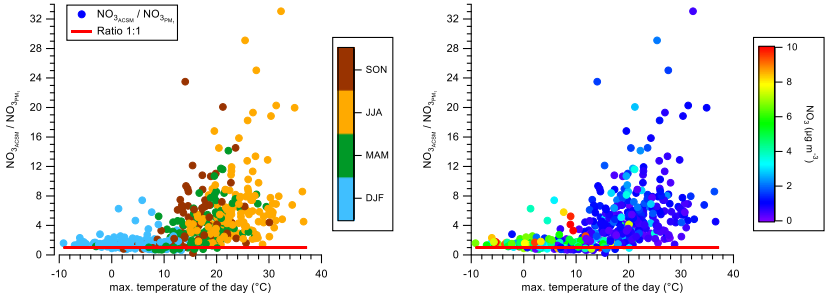
ACSM vs off-line: Nitrate

1- temperature effect on filter samples:

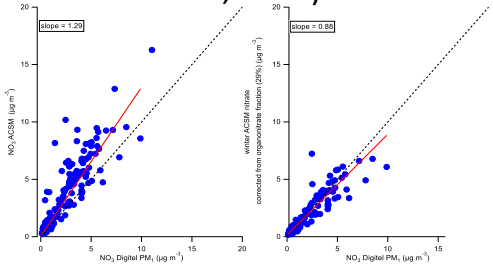
Evaporation of ammonium nitrate on quartz filter for temperature > 20 ºC (Schaap et al., 2004)



⇒ Confirm the lost of ammonium nitrate on filter samples

2- Organo-nitrate compounds:

Organo-nitrate compounds contribute to 29 % of the AMS nitrate signal during the Feb-March 2009 campaign (Kiendler-Scharr et al., 2016)



⇒ Real impact of organo-nitrate to the ACSM nitrate signal but difficult to quantify due to the unit mass resolution of the mass spectrometer

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ACSM vs off-line: organic aerosol

Filters: organic carbon (OC) using EUSAAR-2 protocol

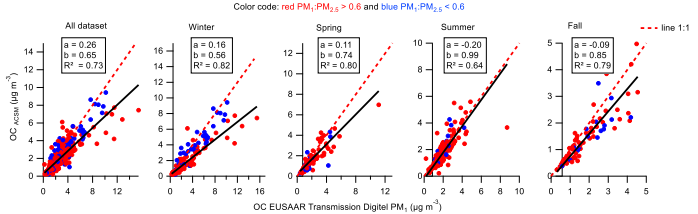
ACSM : organic mass (OM)

$$f_{44} = [CO_2{}^+]/[Organic]$$

$$O{:}C = 0.079 + 4.31 \times f_{44}$$

$$OM{:}OC = 1.29 \times O{:}C + 1.17$$

Aiken et al., 2008, Canagaratna et al., 2015



⇒ limitations: no correction from possible interference due to Pieber effect

⇒ More systematic comparison at various locations are needed

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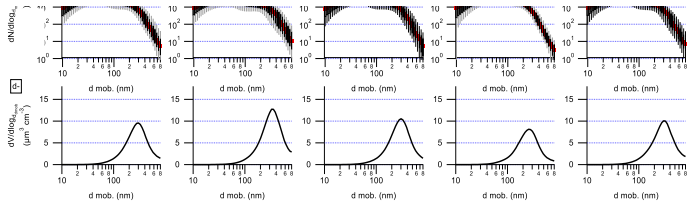
ACSM and SMPS mass closure

- Time dependent density based on the ACSM + MAAP chemical composition
- Performed mass closure in both Volume and Mass => no difference

$$density = \frac{[Total_{ACSM} + eBC]}{\frac{[NO_3^-]+[SO_4^{2-}]+[NH_4^-]}{1.75} + \frac{[Cl^-]}{1.52} + \frac{[Org]}{1.2} + \frac{[eBC]}{1.77}}$$

Adapted from Salcedo et al., 2006



⇒ Small size cutoff effect in cold seasons

⇒ PNSD can be used for near-real time quality control of the ACSM => ACTRIS CAMS 21a

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Conclusion

- ACSM vs. offline $PM_{1:}$ The influence of the individual instrument cutoff **becomes non-negligible as soon as the PM_1:$PM_{2.5}$ < 60%**.

- ACSM vs. offline $PM_{1:}$ It appears to be a **crucial parameter to ensure the ACSM sulfate validation** as well as to support the ACSM's sulfate calibration

- ACSM vs. offline $PM_{1:}$ Nitrate suffers from **strong sampling artifacts**: temperature for offline sampler and organo-nitrate for ACSM

  ⇒ **More investigations on the quantification of organo-nitrate by ACSM are required**

- ACSM vs. offline $PM_{1:}$ **Promising comparison between OC_{ACSM} and OC_{PM1}.** OC is the only regulated organic aerosol-related variable commonly monitored with air quality networks, while ACSM provides directly OM at high time resolution

  ⇒ **More systematic comparison at various locations are needed to confirm the correlation with offline OC**

- Mass balance with PNSD certainly represents the best way for in-situ quality control as well as tracking a possible drift on the ACSM performance

  ⇒ **near-real time mass closure should be considered in the near future as a standard way for in situ quality control of measurements (implementation within ACTRIS CAMS 21a in few months)**

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Multi-year ACSM measurements at the central European research station Melpitz (Germany) – Part 1: Instrument robustness, quality assurance, and impact of upper size cutoff diameter

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Thanks for your attention