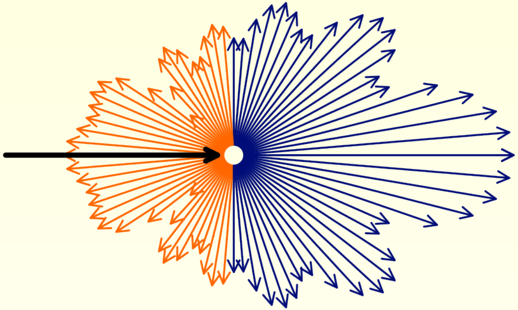
Synthesis of Aerosol Physical, Chemical, and Radiative Properties from Various Sources: Consistency and Closure



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Allison McComiskey
Elisabeth Andrews
Gary Hodges
Don Collins
Thomas Watson

May 23, 2018

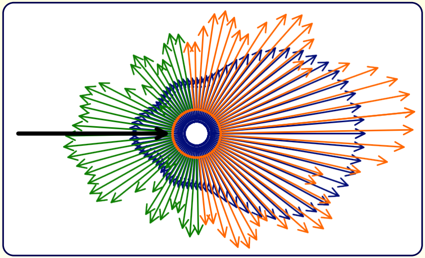
introduction

- Closure study of aerosol properties: scattering coefficient (σ), hemispheric backscattering fraction (g), hygroscopicity (*fRH*)

$\Rightarrow$ assess the consistency and understand benefits and limitation of different techniques

$\Rightarrow$ σ, g, *fRH* needed to understand aerosol radiative forcing

- data-products are from in-situ measurements at DOE ARM Southern Great Plains (SGP) site
- time frame: the year 2012



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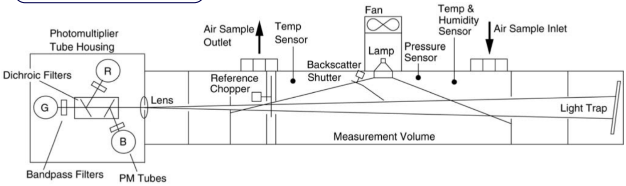
introduction – nephelometer

Nephelometer schematic



scattering coefficient – σ

- measures light that is scattered by aerosols $\Rightarrow$ scattering coefficient
- 3 channels, red, green, blue $\rightarrow$ only green (550 nm) considered here

hemispheric backscattering fraction – $g = \sigma_{\text{back}} / \sigma_{\text{total}}$

- backscattering is measured by blocking forward fraction

hygroscopicity – $f\text{RH} = \sigma_{\text{wet}} / \sigma_{\text{dry}}$

- two nephelometers in series $\rightarrow$ 1st measures σ_{dry} ($RH < 40\%$), second σ_{wet} ($RH \lessapprox 80\%$)

introduction – nephelometer

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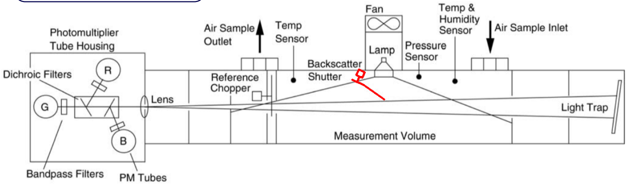

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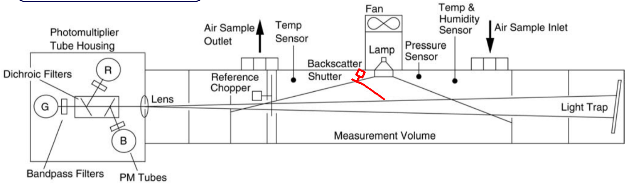

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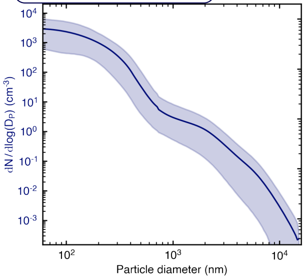
introduction – size distribution

Size/scattering distribution



size distributions

- particles with $d < 750$ nm $\leftrightarrow$ scanning mobility particles sizer (SMPS)
- particles with $d > 500$ nm $\leftrightarrow$ aerodynamic particle sizer (APS)

scattering coefficient – σ

- derived using Mie theory
- $\sigma(d, \lambda, n)$ with $\lambda = 550$ nm and $n = 1.5$

hemispheric backscattering frac. $g = \sigma_{\text{back}} / \sigma_{\text{total}}$

- Mie provides phase function $\mathcal{P}$

$$\sigma_{back} = \sigma_{total} \cdot \int_{\pi/2}^{3\pi/2} \sin(\theta)\mathcal{P}(\theta) \cdot d\theta$$

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- tandem differential mobility analyzer (TDMA)
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$\Rightarrow$ *fRH* from dry and wet size distribution using Mie

CIRES

introduction – size distribution

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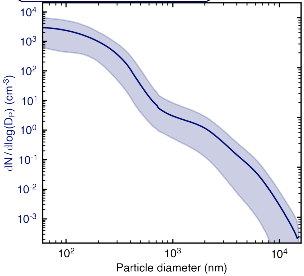

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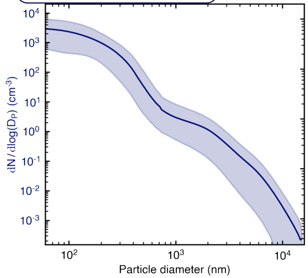
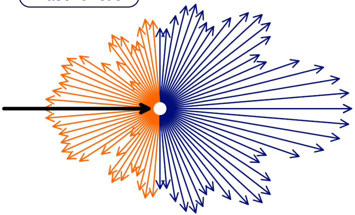


Phase function



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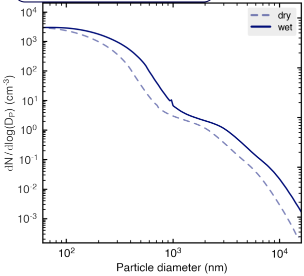

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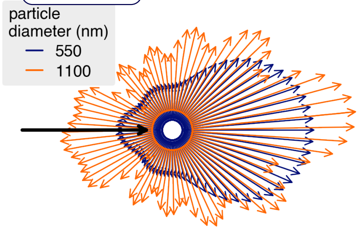

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introduction – chemical composition

chemical composition

- Aerosol Chemical Speciation Monitor (ACSM)

→ mass of NO_3, SO_4, NH_4, Cl and Organic fraction

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⇒ growth factor gRH

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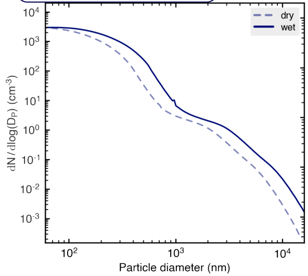
introduction – chemical composition

Size/scattering distribution



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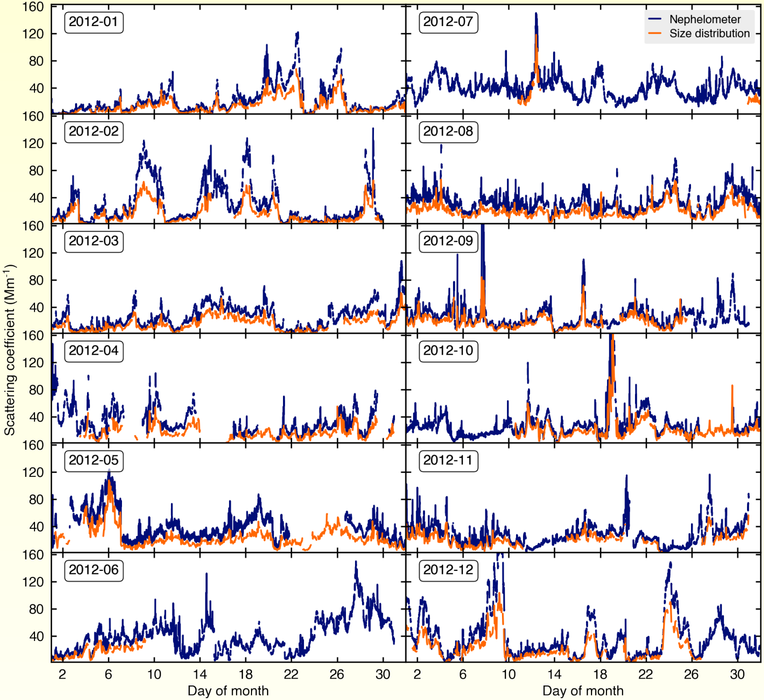
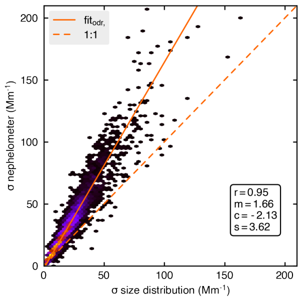

2012-01
2012-02
2012-03
2012-04
2012-05
2012-06
2012-07
2012-08
2012-09
2012-10
2012-11
2012-12
Nephelometer
Size distribution
Scattering coefficient (Mm-1)
Day of month
160
120
80
40
2
6
10
14
18
22
26
30

scattering coefficient – closure



correlation

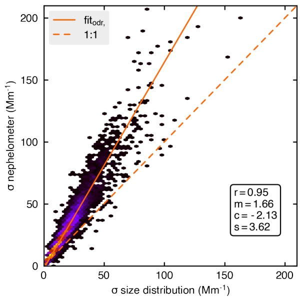
- high correlation and linear relationship
- σ(nephelometer) $>$ σ(size distribution)

uncertainty (85% confidence)

nephelometer $\pm$10% $\Leftarrow$ truncation, particle loss

size distribution $\pm$ 42 %

scattering coefficient – closure



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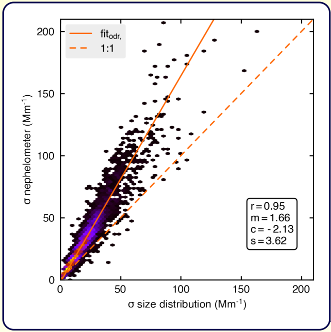

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- diameter 28%
  - APS 11%
    - shape 7%
    - density 11%
    - instrument 2%
  - SMPS 25%
    - shape 22%
    - instrument 12%
- counting efficiency 30%
  - APS 12%
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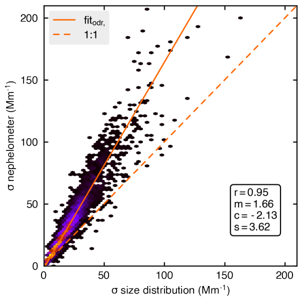
- Mie 13%
  - shape 5%
  - $n_{accu.}$ 10%
  - $n_{coarse.}$ 7%

scattering coefficient – closure



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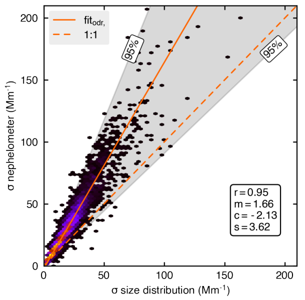

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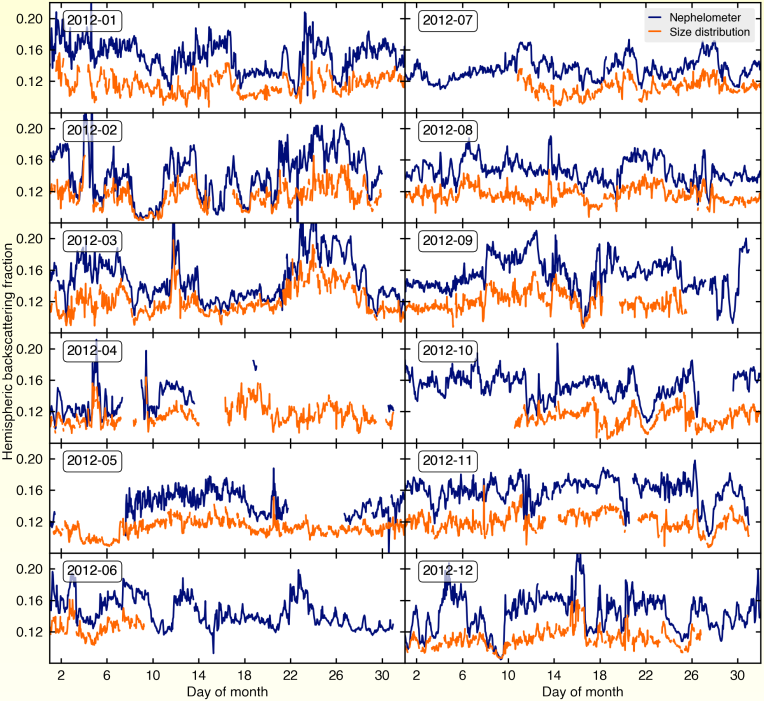
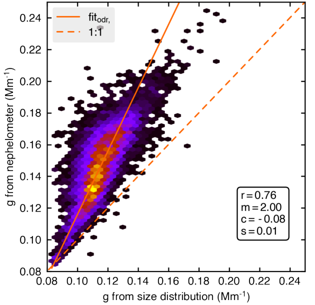
- 44% combined uncertainty
- the 1:1- line is within the 95% confidence interval

to improve bias better knowledge of sub-micron particle shapes and the counting efficiency of the SMPS is needed



Hemispheric backscattering fraction
Day of month
Nephelometer
Size distribution
2012-01
2012-02
2012-03
2012-04
2012-05
2012-06
2012-07
2012-08
2012-09
2012-10
2012-11
2012-12
0.20
0.16
0.12
2
6
10
14
18
22
26
30

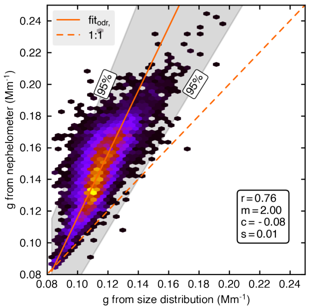
hemispheric backscattering fraction – closure



- moderate correlation and large bias
- uncertainty (20%) can not fully explain bias
- correlation improves when data with weak scattering signal is removed max r when $\sigma > 20$ Mm^{-1}

→ more then 50% of data excluded

hemispheric backscattering fraction – closure



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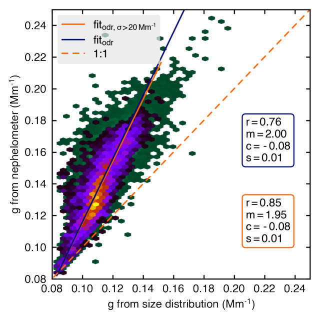

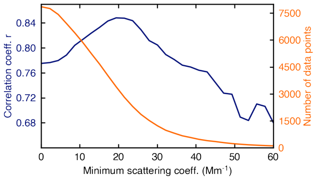

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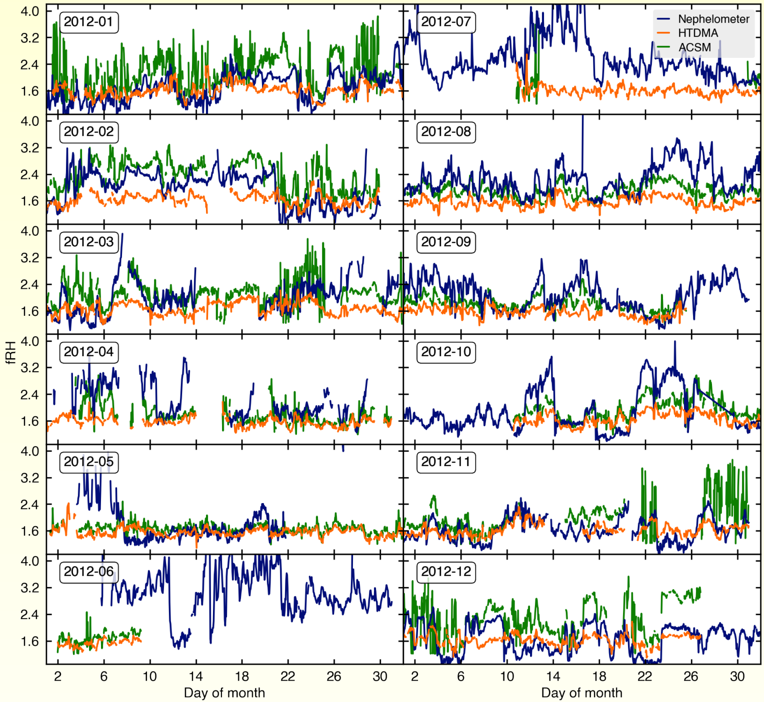
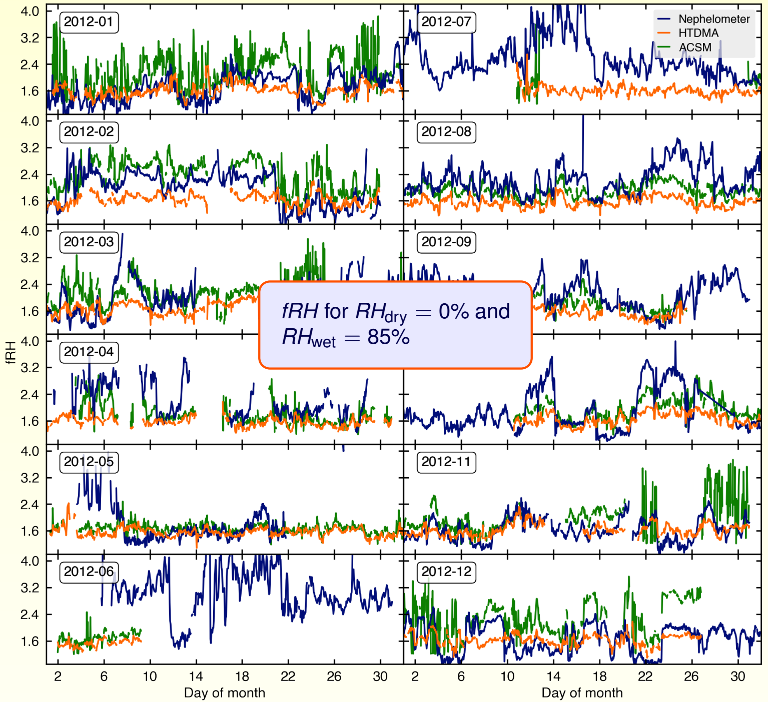
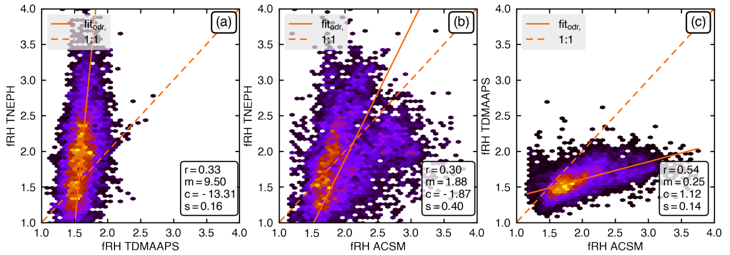


Nephelometer
HTDMA
ACSM
2012-01
2012-02
2012-03
2012-04
2012-05
2012-06
2012-07
2012-08
2012-09
2012-10
2012-11
2012-12
fRH
4.0
3.2
2.4
1.6
2
6
10
14
18
22
26
30
Day of month



2012-01
2012-02
2012-03
2012-04
2012-05
2012-06
2012-07
2012-08
2012-09
2012-11
2012-12
Nephelometer
HTDMA
ACSM
fRH
Day of month
fRH for RHdry = 0% and RHwet = 85%

hygroscopicity – closure



- weak correlation and strongly biased
- correlation improves if data is limited to $RH_{dry} < 20\%$

$\Rightarrow$ is $RH < 40\% \equiv$ dry good assumption?!?

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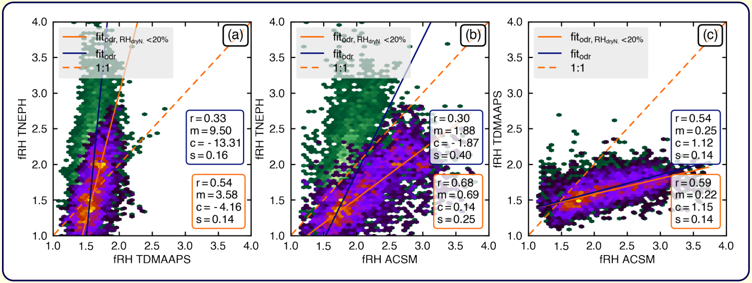

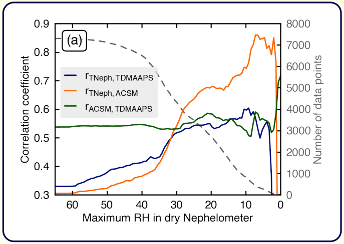

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conclusions

closure of scattering coefficient

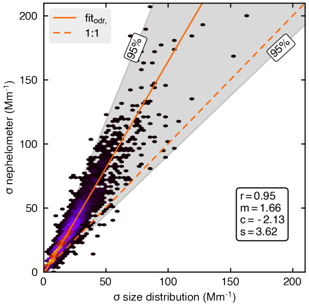
- nephelometer and size distribution products are highly correlated but show significant bias
- large uncertainties likely origin of bias, in particular related to particle shape and counting efficiency of SMPS

closure of hemispheric backscattering fraction

- nephelometer and size distribution products are moderately correlated and show significant bias
- correlation is improved when data is removed where scattering is low

closure of hygroscopicity

- correlation of nephelometer and size distribution products are low to moderate and show very large bias
- correlation and bias greatly improve if “dry” is defined as $RH < 20\%$



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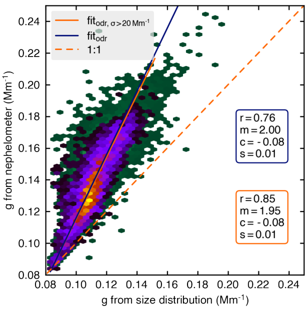

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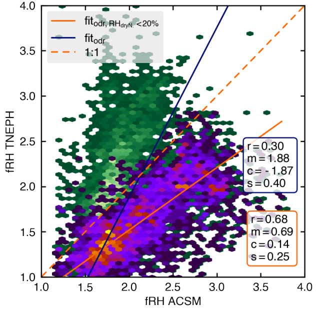

acknowledgment

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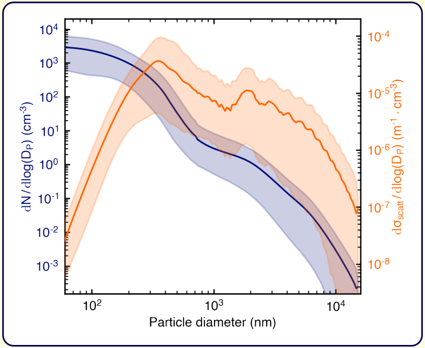
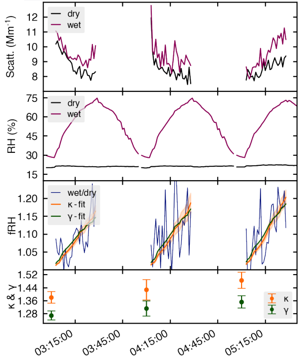
FINE

scattering distribution



hygroscopicity – introduction

nephelometer → *fRH*



$$fRH(\kappa) = \frac{1 + \kappa \frac{RH_{wet}}{100 - RH_{wet}}}{1 + \kappa \frac{RH_{dry}}{100 - RH_{dry}}}$$

nephelometer

two nephelometers in series → 1st measures σ_{dry} ($RH < 40\%$), second σ_{wet} ($RH \lessapprox 80\%$)

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⇒ growth distribution ⇒ *fRH* from dry and wet size distribution using Mie

chemical composition

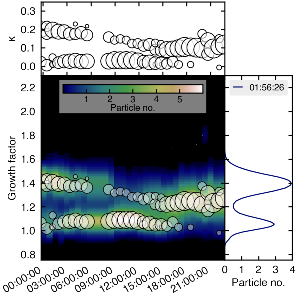
- Aerosol Chemical Speciation Monitor (ACSM) → mass of NO_3, SO_4, NH_4, Cl and Organic fraction

⇒ growth factor *g*RH

⇒ *f*RH from dry and grown size distribution using Mie

hygroscopicity – introduction

TDMA $\rightarrow$ growth distribution



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size distribution

- two SMPS in series *bka* tandem differential mobility analyzer (TDMA)
- 1st runs under dry ($RH = 20\%$) second under wet ($RH = 90\%$) conditions

$\Rightarrow$ growth distribution $\Rightarrow$ *fRH* from dry and wet size distribution using Mie

chemical composition

- Aerosol Chemical Speciation Monitor (ACSM) $\rightarrow$ mass of NO_3, SO_4, NH_4, Cl and Organic fraction

$\Rightarrow$ growth factor *g*RH

$\Rightarrow$ *f*RH from dry and grown size distribution using Mie