Evaluation and Improvement of the Parameterization of Aerosol Hygroscopicity in Global Climate Models Using In-situ Surface Measurements

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Aerosols take up water

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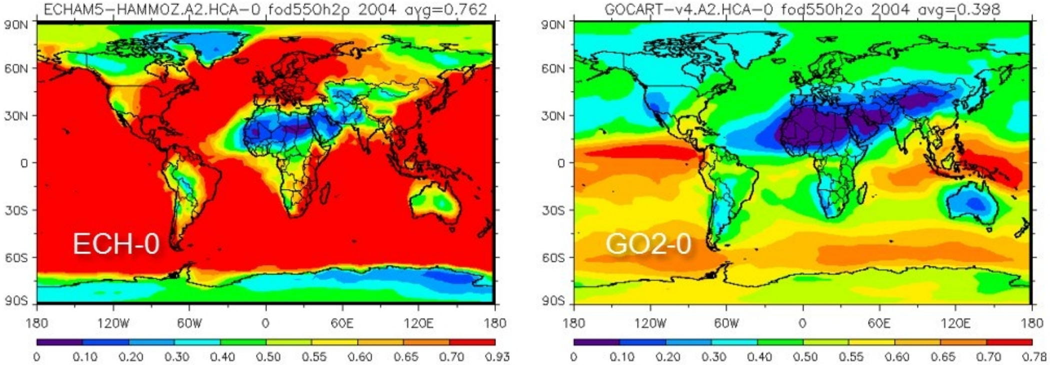
- Hygroscopicity also important for clouds, atmospheric resident times / removal, measurement artefacts, etc.

Example of differences in hygroscopicity in GCM's (AeroCom II for 2004)



Fraction of AOD due to water (ECHAM5 with global annual average of 76 %; GOCART with 40 %)

Figures from Mian Chin (NASA Goddard)

The effect of relative humidity on aerosol light scattering

Scattering enhancement factor

$$f(\mathrm{RH}, \lambda) = \frac{\sigma_{\mathrm{sp}}(\mathrm{RH}, \lambda)}{\sigma_{\mathrm{sp}}(\mathrm{RH}_{\mathrm{dry}}, \lambda)}$$

with λ: wavelength, σ_{sp}: scattering coefficient, RH: relative humidity

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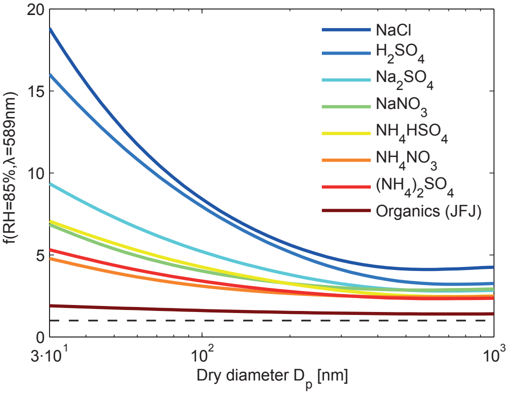

Modelled scattering enhancement vs. dry diameter assuming a single lognormal size distribution

Source: Zieger et al. (2013)

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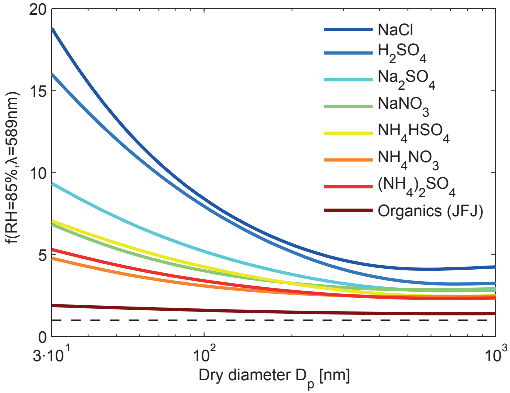
Both size distribution <u>and</u> chemical composition determine $f(\mathrm{RH})$



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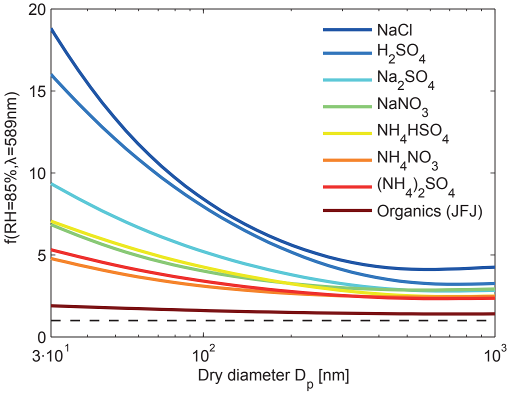
$f(\mathrm{RH})$ can be measured using humidified nephelometer systems



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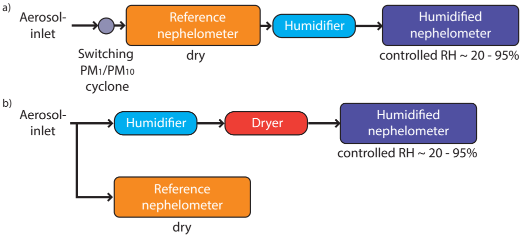
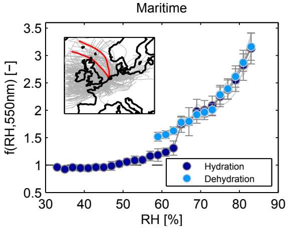
The humidified nephelometer (WetNeph)



The (a) NOAA and (b) PSI system (Fierz-Schmidhauser et al., 2010).



Example humidogram from Cabauw for maritime air (Zieger et al., 2011)

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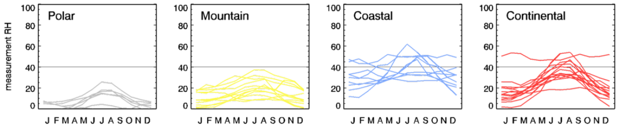
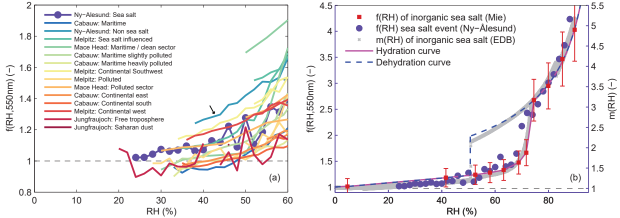
Instrumental differences

- NOAA system only measures lower branch/deliquescence
- PSI system uses active drying after humidifier $\rightarrow$ can measure parts of the upper branch

The dry reference scattering coefficient: What is dry?



RH climatology of various dry nephelometer measurements separated by station type.



(a) Scattering enhancement at various European sites and (b) for inorganic sea salt (modelled and measured). Taken from Andrew et al. (2019, in prep.) and Zieger et al. (2017).

A significant bias could be introduced by insufficient drying of aerosols

- GAW/WMO guideline for aerosol monitoring:

$$\mathrm{RH_{dry}} < 30 - 40\,\%$$

- Not always achieved (e.g. marine sites)
- Important for sea salt (efflorescence RH)
- Ideally $\mathrm{RH_{dry}}$ be much lower

The new benchmark dataset of scattering enhancement

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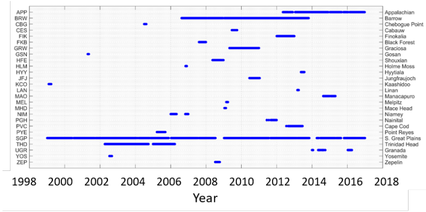

Temporal data coverage of re-analysed sites.

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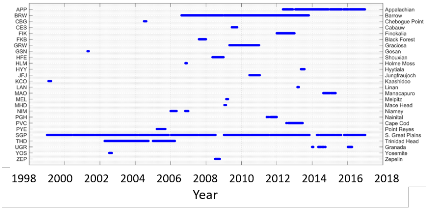

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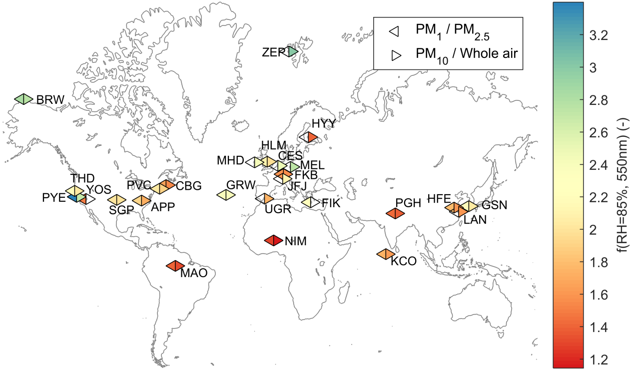
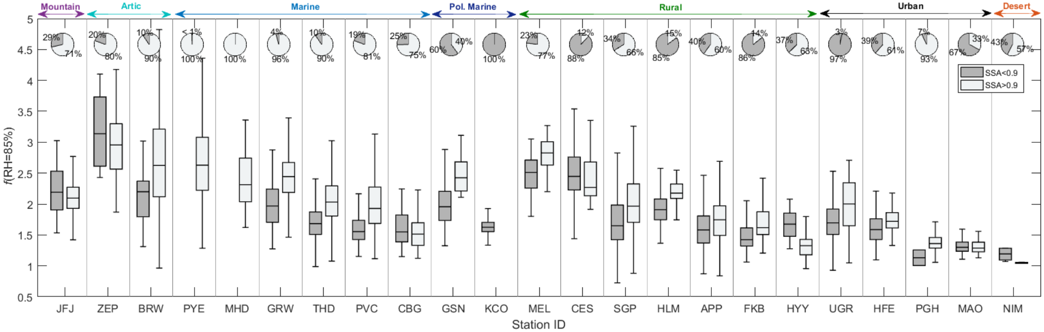


Overview of re-analysed sites with mean values of $f(\mathrm{RH}{=}85\%/\mathrm{RH_{dry}})$ for $\mathrm{PM_1}/\mathrm{PM_{2.5}}$ (left triangles) and $\mathrm{PM_{10}}$/whole-air inlet systems (right triangles).
Source: Burgos et al. (2019, in review)

First global climatology of the scattering enhancement factor



Boxplot of $f(\mathrm{RH}{=}85\,\%)$ at $\lambda = 550\,\mathrm{nm}$ segregated by single scattering albedo (SSA). Source: Titos et al. (2019, in prep.)

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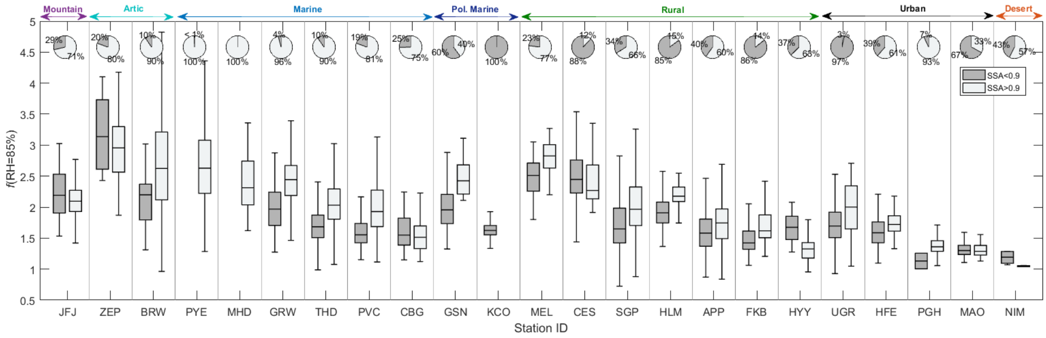

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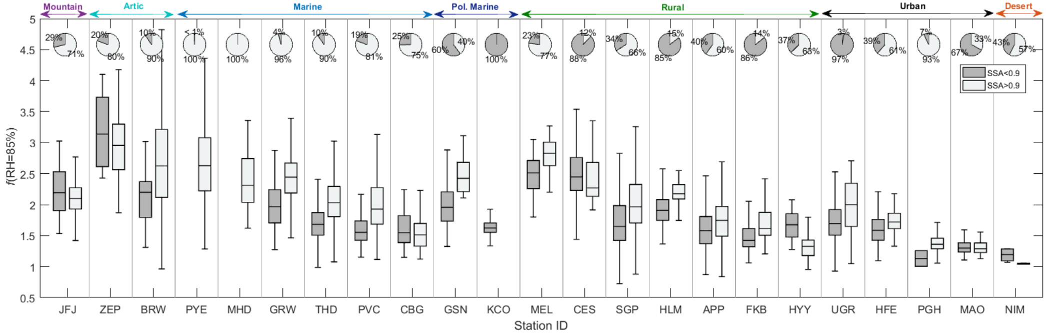

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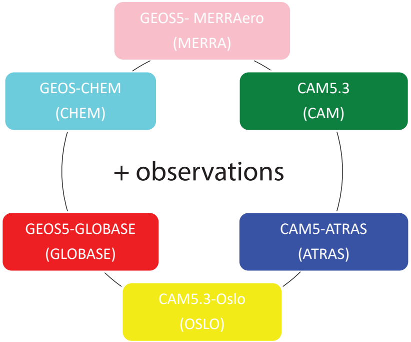
- Most sites show **increased f(RH) for less absorbing aerosol**
- Exceptions for certain sites with possible pronounced **size effect**: smaller & less hygroscopic aerosol may show similar or smaller f(RH) compared to larger but more hygroscopic aerosol (e.g. sea spray, see Zieger et al., 2010)

Model-measurement comparison



- Part of the AeroCom phase III experiments
- Model output: Scattering coefficient at RH= 0, 40, 85 % and $\lambda = 550\,\text{nm}$ for 2010 for 20 coincident sites with observational data
- Monthly average (note: only 3 sites are co-located in time for 2010)

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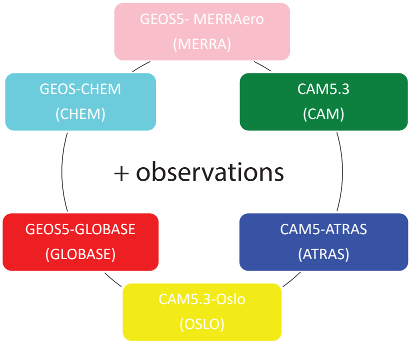

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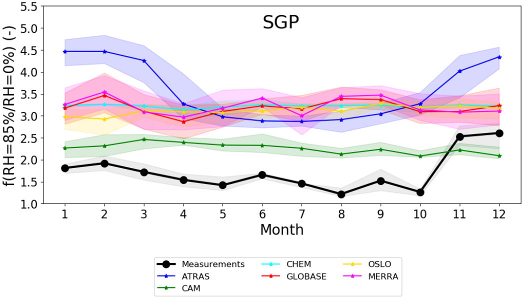
- Note: All models have different parameterizations for hygroscopic growth and particle size (see poster)!

Example for 2010 (co-located in time): Southern Great Plains



Modelled $f(\mathrm{RH})$ for Southern Great Plains with $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ as reference. Measurements are shown at actual measured RH.

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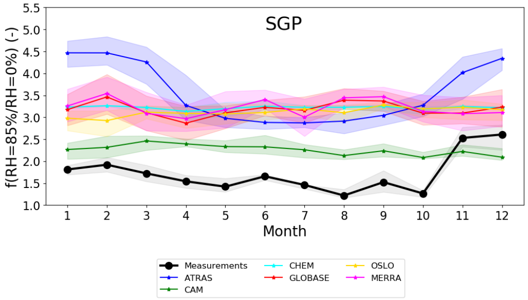

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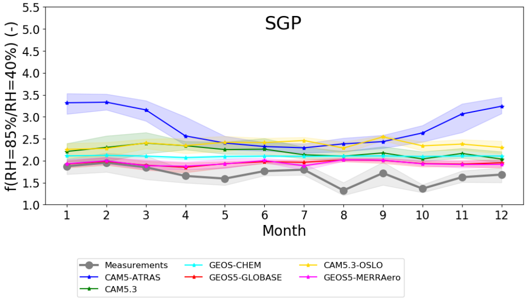
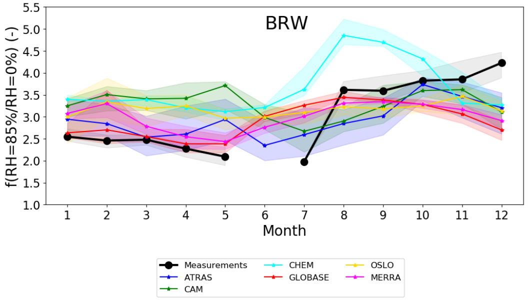


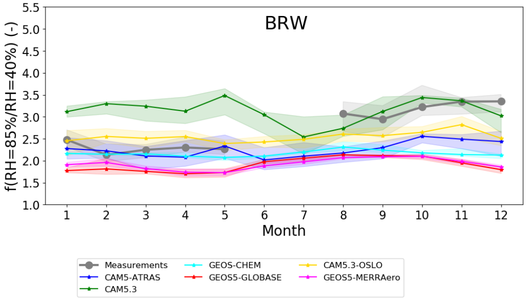
Modelled $f(\mathrm{RH})$ for Southern Great Plains with $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ as reference. Measurements interpolated to $\mathrm{RH}_{\mathrm{dry}}$=40 %.

- Comparison at $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ more suitable to compare at same conditions (not fully dried particles).

Example for 2010 (co-located in time): Barrow / North Slope of Alaska



Modelled and measured $f(\mathrm{RH})$ for Barrow with $\mathrm{RH_{dry}} = 0\,\%$ as reference (measurements not corrected).



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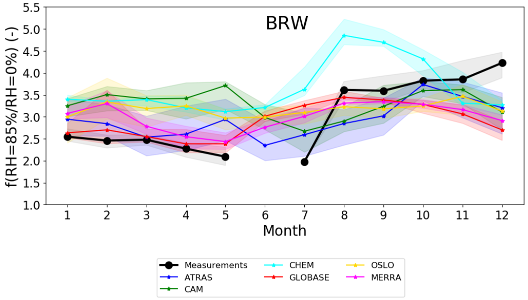

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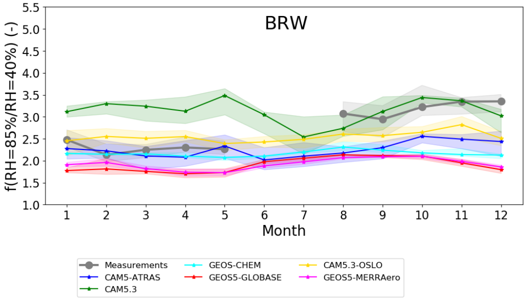

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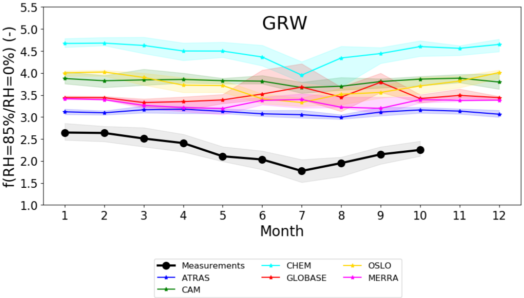
- Measurements in Barrow should be less affected by remaining water (lower $\mathrm{RH}_{\mathrm{dry}}$)
- Some models show large change in $f(\mathrm{RH})$ if $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ or $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ is taken as reference

Example for 2010 (co-located in time): Graciosa



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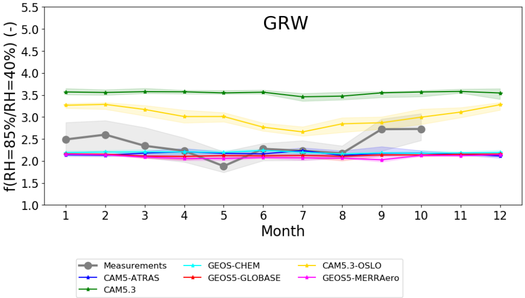

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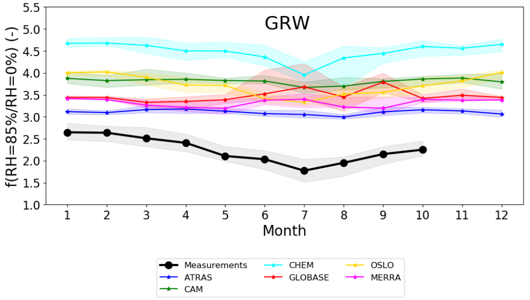

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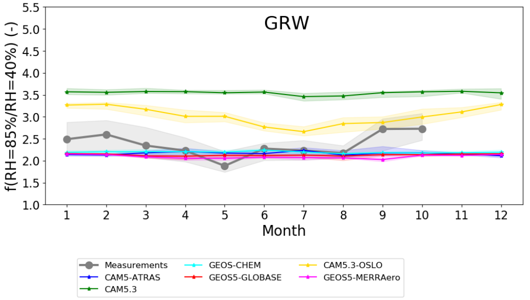

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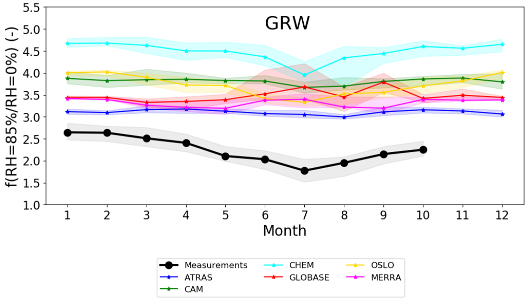

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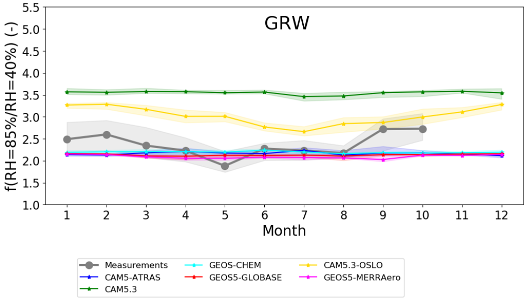

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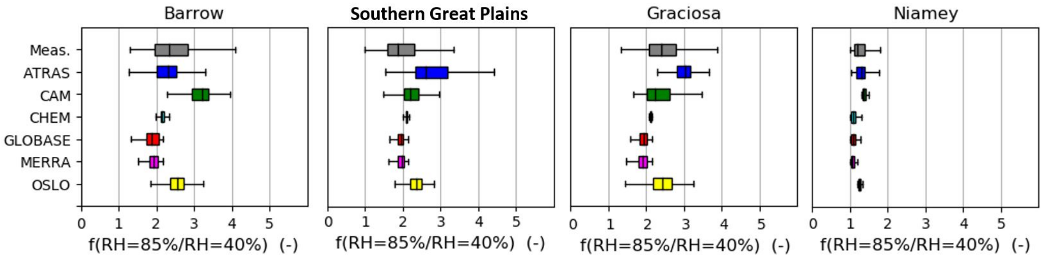
- Models show a **large site-specific diversity**

Comparison of entire data set with 2010 model data



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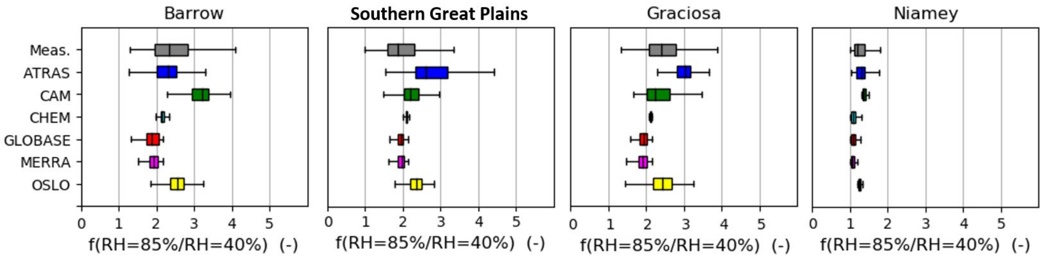

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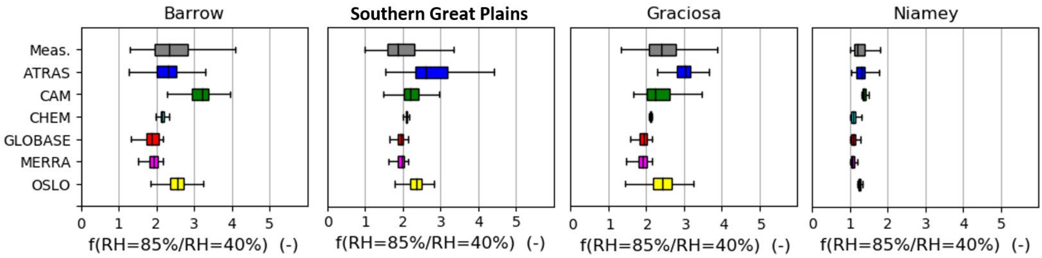

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- Dust dominated sites are captured well by models (low hygroscopic growth)
- Consistent model biases even among various different site types (rural vs. marine)

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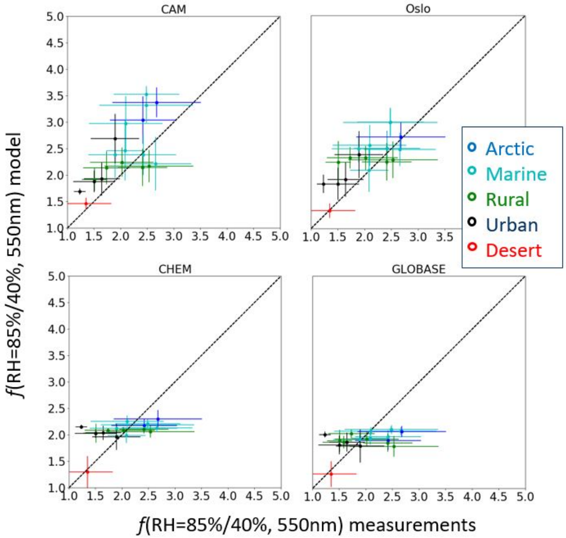

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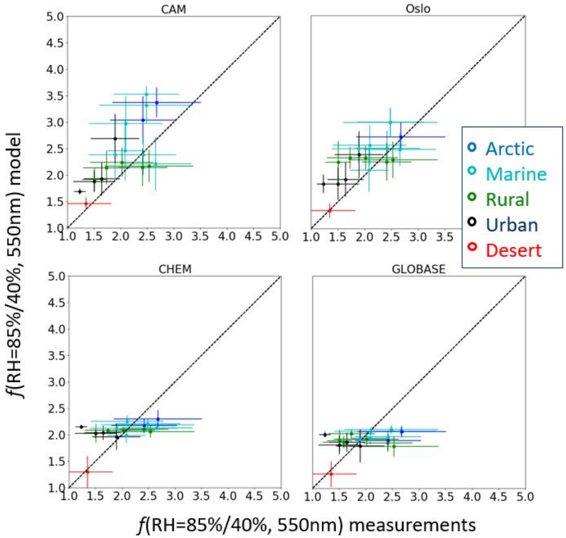

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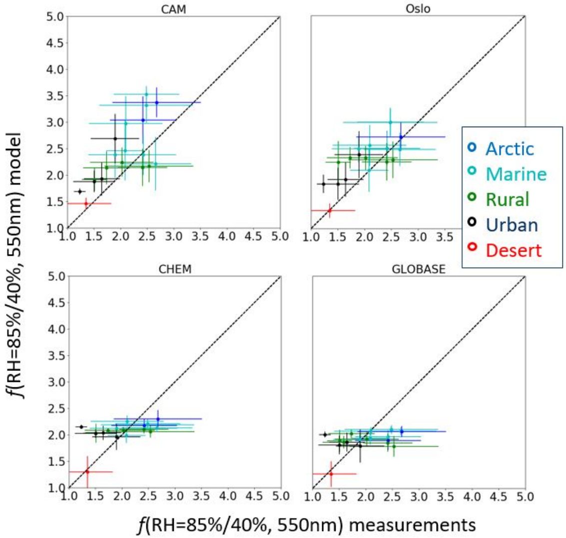

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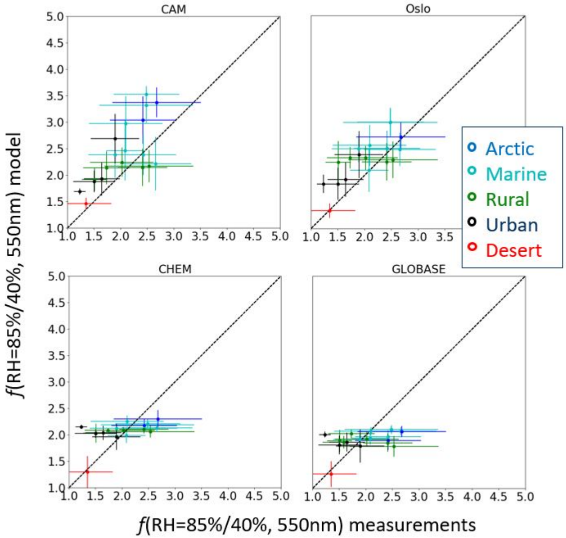

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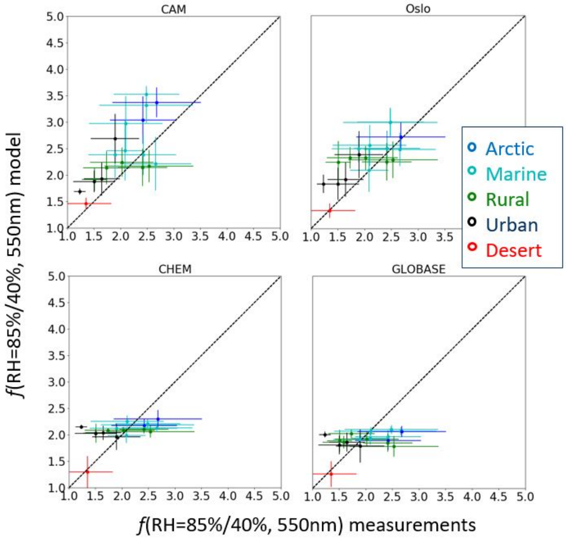

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- More details at the poster!

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- Further evaluation needs the **addition of the size & chemical composition** to the analysis

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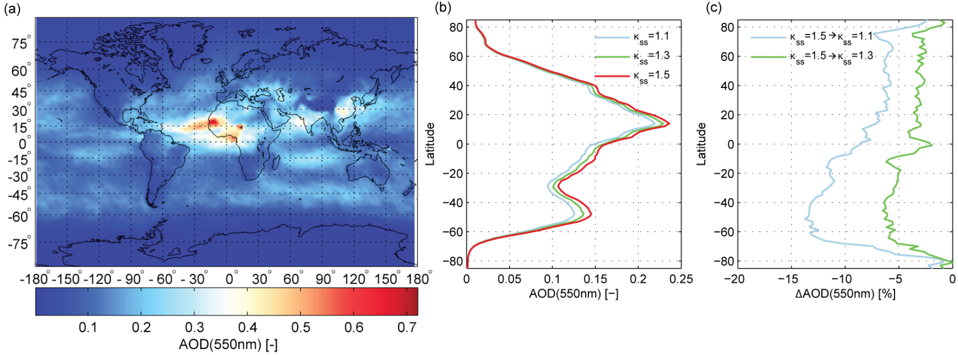
- **Models generally overestimate** $f(\mathrm{RH})$ but comparison improves if $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ is taken as reference RH
- Models show a **large diversity** in $f(\mathrm{RH})$ with respect to magnitude and temporal evolution (e.g. seasons).**Reasons are manifold**: differences in model paramerizations of e.g. hygroscopicity, size, sources + strength, mixing state, removal processes, etc.
- Further evaluation needs the **addition of the size & chemical composition** to the analysis
- Importance of **sufficient drying** for continuous field observations

Is it worth the effort? Yes, small changes matter!



Impact of reduced inorganic sea salt hygroscopicity within a general circulation model. Model results for different κ-values. (a) Global map of AOD. (b) Latitudinal mean of the AOD(550nm) (c) Percental change in AOD. Taken from Zieger et al., 2017.

Inorganic sea spray: Reduction of hygroscopic growth factor by $\approx 10\,\%$ $\rightarrow$ reduction in aerosol optical depth (AOD) by $\approx 10 - 15\,\%$.

Outlook

- Further **AeroCom modelling experiment** with additional information on **size** and **chemistry** and **closure/sensitivity study** using Mie theory
- Finalization of **papers**:
  - Data descriptor paper
  - Model-measurement comparison
  - What is dry?
  - f(RH) climatology
- Global comparison to CALIOP extinction coefficients to evaluate lidar ratio scheme (similar to Tesche et al. (2014), depending on funding)

Thank you for your attention! Questions?

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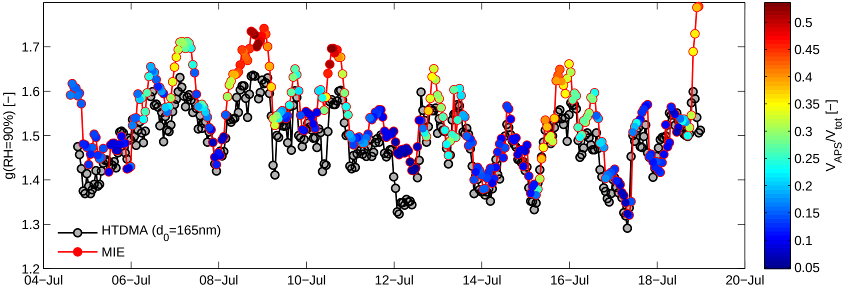
The 'core' team meeting at Stockholm University (2017).

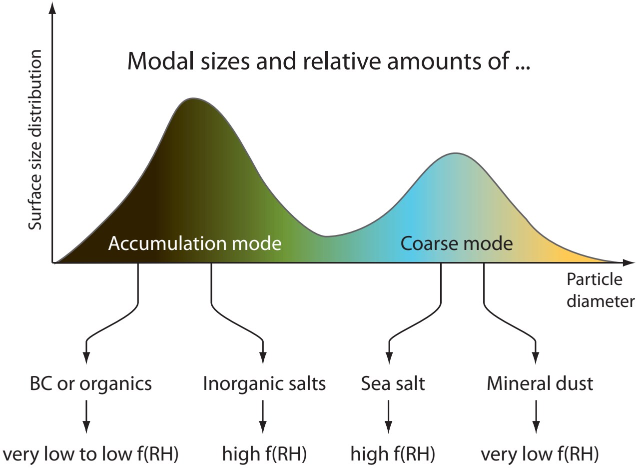
Advantages of humidified nephelometer measurements

- HTDMA captures size dependent hygroscopic growth & mixing state; limited to submicrometer size range
- WetNeph captures entire optical important size range; usually no size cut (or PM_1 and PM_{10} cyclone)

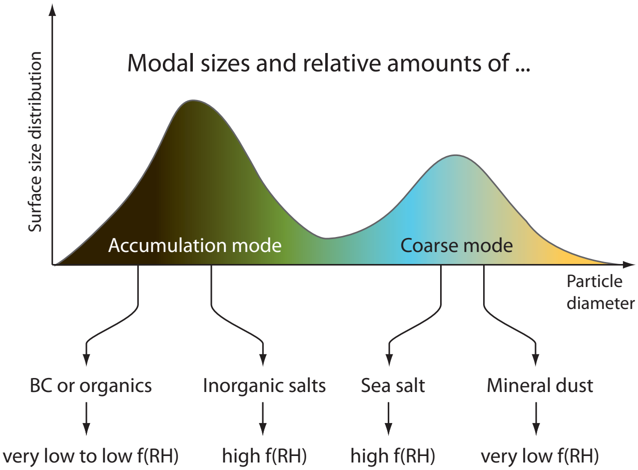


Time series of the hygroscopic growth factor measured by the H-TDMA (black line) and retrieved from WetNeph, DryNeph, SMPS, APS measurements and Mie theory (red line). The color code denotes the coarse mode volume fraction measured by the APS and SMPS (Zieger et al., 2011).

What determines the scattering enhancement?

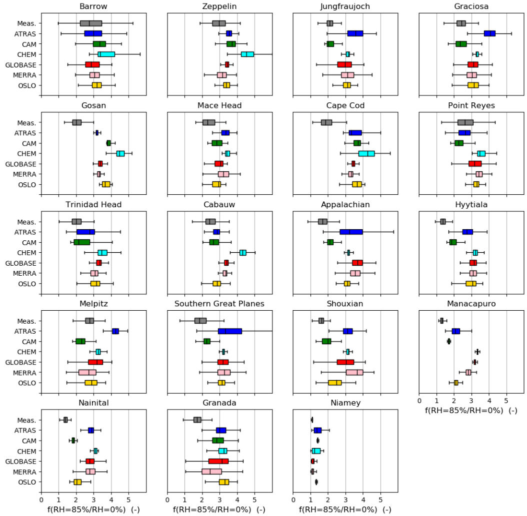


What determines the scattering enhancement?

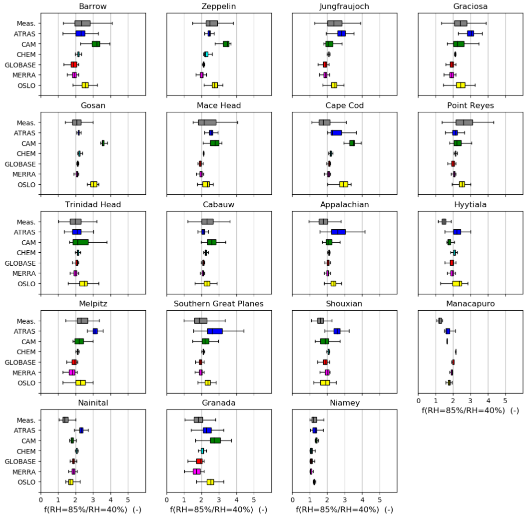


- Fine mode: e.g. Aerosol Mass Spectrometer (AMS)
- Coarse mode: e.g. filter techniques

Comparison of entire data set with 2010 model data

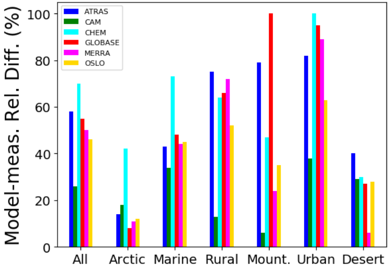


Comparison of entire data set with $RH_{dry} = 0\,\%$ as reference.

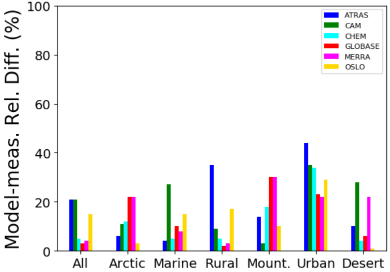


Comparison of entire data set with $RH_{dry} = 40\,\%$ as reference.

Comparison of entire data set with 2010 model data



Relative difference between modelled and measured $f(\mathrm{RH})$ with $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ as reference.



Relative difference between modelled and measured $f(\mathrm{RH})$ with $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ as reference.

- Improvement in comparison if $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ is taken as reference
- Models mainly over-estimate $f(\mathrm{RH})$
- Large diversity among models
- Caution: Site-specific and temporal characteristics are masked out but can still be significant

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