Evaluation and improvement of the parameterization of aerosol hygroscopicity in global climate models using in-situ surface measurements

Maria Burgos[1,*], Gloria Titos[2], Elisabeth Andrews[3], Huisheng Bian[4], Virginie Buchard[5], Gabriele Curci[6], Alf Kirkevag[7], Hitoshi Matsui[8], Cynthia Randles[4], Kai Zhang[9], and Paul Zieger[1,*]

[1]Stockholm University and Bolin Centre for Climate Research, Stockholm, Sweden; [2]Andalusian Institute for Earth System Research, University of Granada, Granada, Spain; [3]Cooperative Institute for Research in Environmental Studies (CIRES), University of Colorado, Boulder, USA; [4]NASA/Goddard Space Flight Center, USA; [5]GESTAR/Universities Space Research Association, USA; [6]Universita' degli Studi dell'Aquila, Italy; [7]Norwegian Meteorological Institute, Norway; [8]Nagoya University, Japan; [9]Pacific Northwest National Laboratory, USA; *Contact: maria.burgos@aces.su.se & paul.zieger@aces.su.se

Introduction

Aerosol optical properties are dependent on particle size and chemical composition, which are in turn influenced by the relative humidity (RH) of the surrounding air. Aerosol hygroscopicity, or particle's ability to take up water, will therefore have an effect on the aerosol-radiation interaction and will affect how much do particles absorb or scatter radiation. Our focus in this work is on particle light scattering coefficient, which can be measured at different values of RH and then be compared with the corresponding modelled value. This will allow to assess how well are Global Climate Models (GCMs) representing aerosol optical hygroscopic growth.

Scattering enhancement factor:

$$f(\mathrm{RH}, \lambda) = \frac{\sigma_{\mathrm{sp}}(\mathrm{RH}, \lambda)}{\sigma_{\mathrm{sp}}(\mathrm{RH}_{\mathrm{dry}}, \lambda)} \quad (1)$$

with λ: wavelength, σ_{sp}: scattering coefficient, RH: relative humidity. $f(\mathrm{RH})$ can be directly measured by using a humidified nephelometer system.

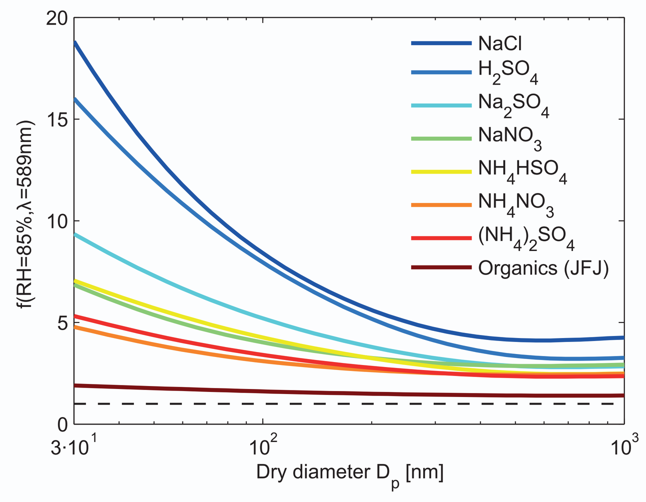

Figure 1 : The scattering enhancement $f(\mathrm{RH})$ at RH=85% and $\lambda = 589$ nm vs. dry particle diameter calculated for different substances (see legend). A monomodal size distribution is assumed. Figure taken from Zieger et al. (2013)

The humidified nephelometer systems

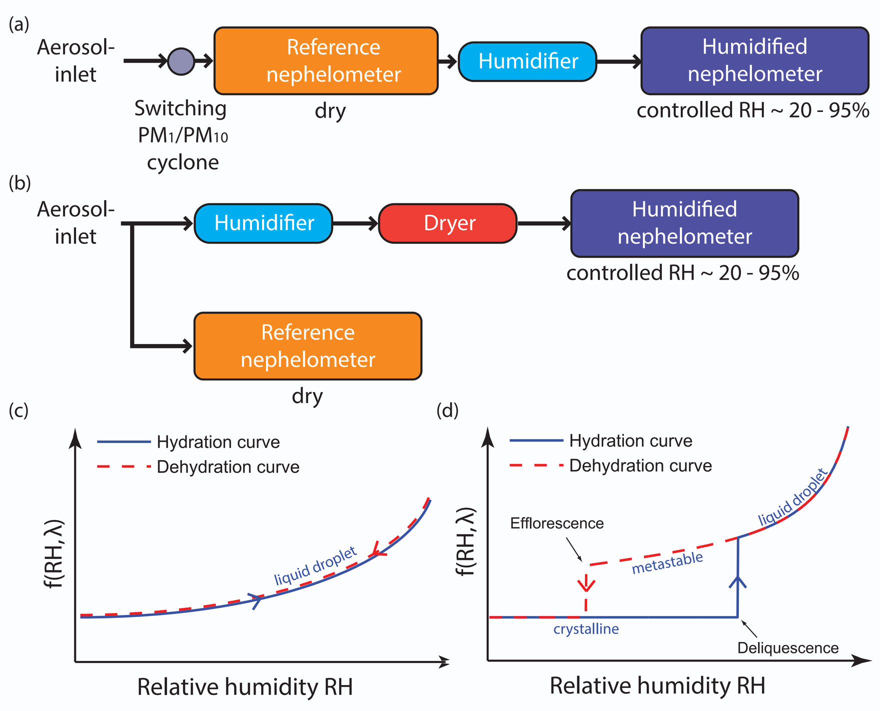

Figure 2 : Overview of the two main instrumental designs. (a) NOAA design and (b) PSI design. This set-up allows to probe particles without (c) and with (d) hysteresis behaviour. Technical details and comparison in Fierz-Schmidhauser et al.(2010a).

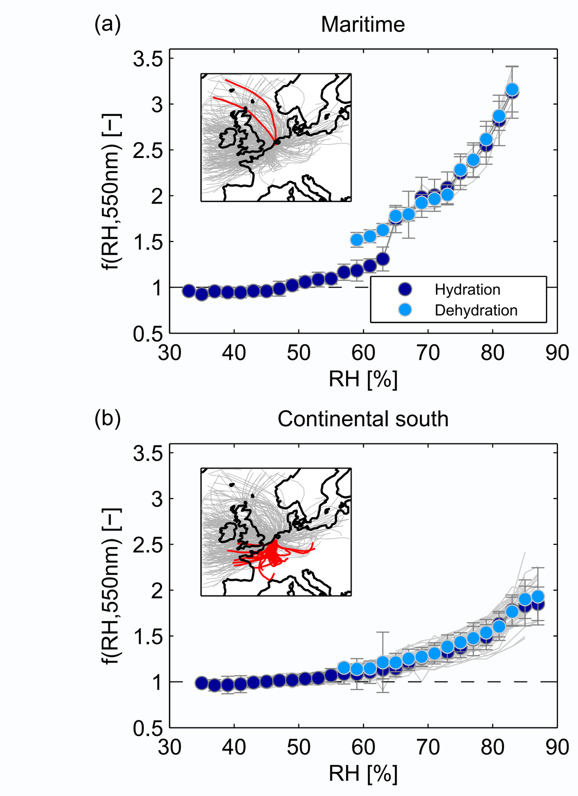

Figure 3 : Examples of $f(\mathrm{RH})$-humidograms measured at Cabauw, The Netherlands, for **(a)** maritime and **(b)** continental air masses (taken from Zieger et al., 2011).

The benchmark dataset

- Standardized re-analysis of 26 datasets of RH-dependent scattering and backscattering coefficients, $f(\mathrm{RH})$ covering PM_1, $PM_{2.5}$, PM_{10} and whole-air
- Harmonized dataset openly available on ACTRIS database and EBAS + data descriptor paper (Burgos et al., 2019)

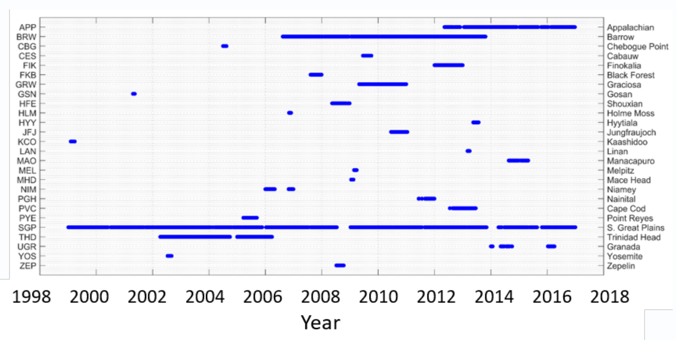

Figure 4 : Temporal data coverage.

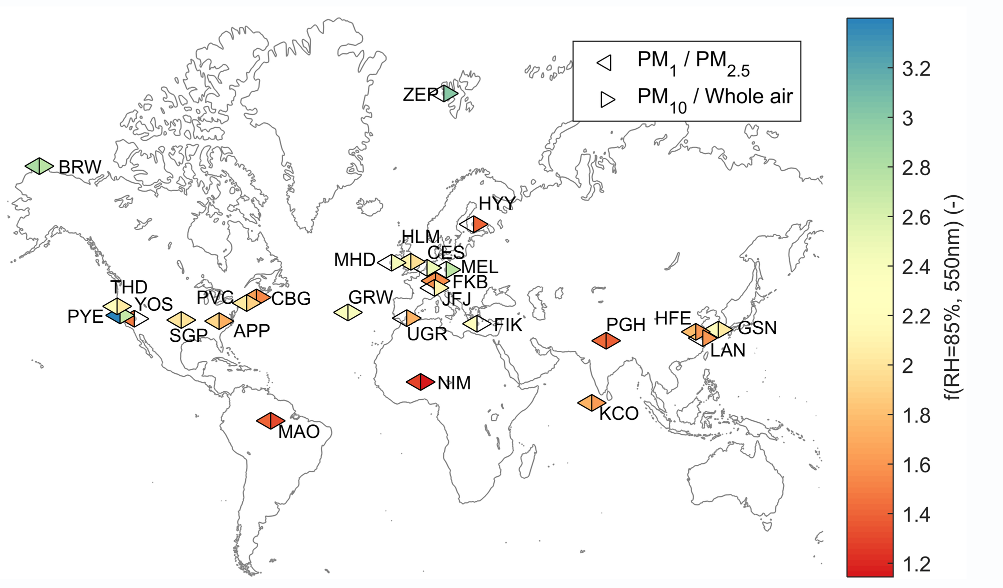

Figure 5 : Overview of re-analysed sites with mean values of $f(\mathrm{RH}{=}85\%/\mathrm{RH}_{\mathrm{dry}})$ for $PM_1/PM_{2.5}$ (left triangles) and PM_{10}/whole-air inlet systems (right triangles). Taken from Burgos et al. (2019).

The dry reference scattering coefficient: What is dry?

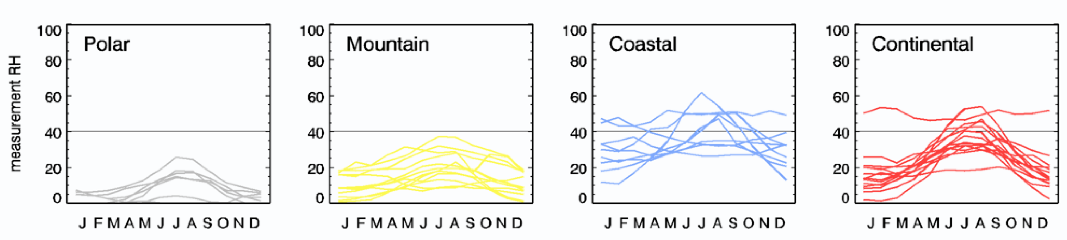

Figure 6 : RH climatology of various dry nephelometer measurements separated by station type. Taken from Andrews et al. (in prep.).

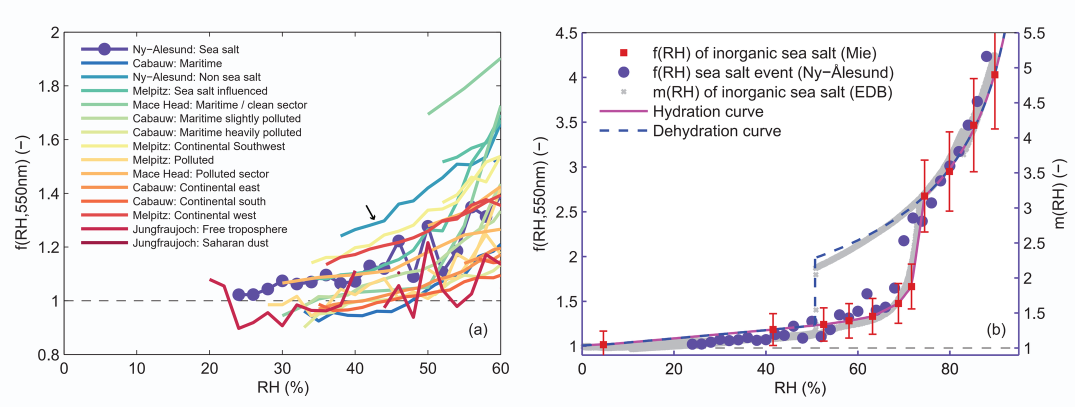

Figure 7 : (a) Scattering enhancement at various European sites and (b) for inorganic sea salt (modelled and measured). Taken from Andrews et al. (in prep.).

A significant bias could be introduced by insufficient drying of aerosols (especially with deliquescent aerosol such as sea spray, see Zieger et al., 2017)*.

- GAW/WMO guideline for aerosol monitoring: $\mathrm{RH}_{\mathrm{dry}} < 30 - 40\,\%$
- Not always achieved (e.g. marine sites)
- Important for sea salt (efflorescence RH)
- Ideally be much lower

*Side note: This paper also shows how small changes in hygroscopic growth factor within model parameterizations directly translate into changes in aerosol optical depth which motivates our endeavour.

References

Bey, I., et al.: *Journal of Geophysical Research: Atmospheres*, 106(D19):23073–23095 (2001)
Buchard, V., et al.: *Atmospheric Chemistry and Physics*, 15(10):5743–5760 (2015)
Burgos, M., et al.: *Scientific Data*, (in review) (2019)
Chin, M., et al.: *Journal of the Atmospheric Sciences*, 59(3):461–483 (2002)
D'Almeida, G. A., et al.: *Atmospheric aerosols: global climatology and radiative characteristics*. A Deepak Pub (1991)
Fierz-Schmidhauser, R., et al.: *Atmos. Meas. Tech.*, 3(1):39–50 (2010)
Kirkevåg, A., et al.: *Geoscientific Model Development*, 11(10):3945–3982 (2018)
Liu, C., et al.: *Geophys. Res. Lett.*, 39(13) (2012)
Martin, R. V., et al.: *Journal of Geophysical Research: Atmospheres*, 108(D3) (2003)
Matsui, H., et al.: *Journal of Geophysical Research: Atmospheres*, 116(D19) (2011)
Matsui, H., et al.: *Atmospheric Chemistry and Physics*, 14(18):10315–10331 (2014)
Tang, I. N.: *Journal of Geophysical Research: Atmospheres*, 102(D2):1883–1893 (1997)
Zieger, P., et al.: *Atmos. Chem. Phys.*, 11(6):2603–2624 (2011)
Zieger, P., et al.: *Atmos. Chem. Phys.*, 13(21):10609–10631 (2013)
Zieger, P., et al.: *Nature Communications*, 8(15883) (2017)

Acknowledgements: We thank all data providers and co-authors from previous studies. This work is funded by the U.S. Department of Energy (contract no. DE-FO-0001430).

Project website:
https://www.aces.su.se/research/projects/evaluation-and-improvement-of-the-parameterization-of-aerosol-hygroscopicity-in-global-climate-models-using-in-situ-surface-measurements/

Model-measurement comparison: Overview

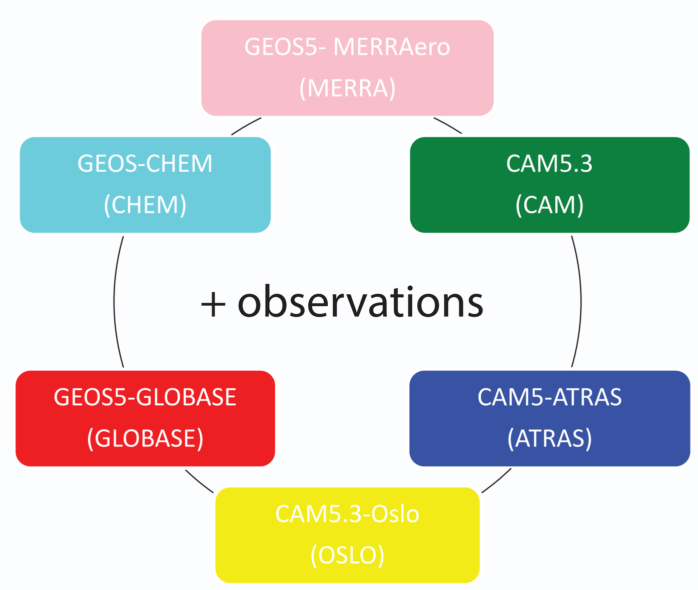

Figure 8 : Overview of the compared models.

- Part of the AeroCom phase III experiments
- Model output: Hourly scattering coefficient at $\lambda = 550$ nm and RH$= 0, 40, 85\,\%$ for 2010 for 20 coincident sites with observational data
- Monthly averages are compared (note: only 3 sites are sufficiently co-located for 2010)

Future possible dedicated AeroCom experiment:

- Corresponding particle size distributions plus chemical composition for Mie closure study
- Further wavelengths and additional years

Table 1 : Overview of contributing models with main reference paper and aerosol parameterizations.

Model	Main reference	Hygroscopicity	Hygroscopicity for marine aerosols	Mixing state	Size distribution
CAM5.5	Liu et al. (2012)	κ-Köhler	$\kappa = 1.16$ (sea salt)	Internal and external mixing	Aitken, accumulation and coarse
CAM5-ATRAS	Matsui et al. (2014); Matsui et al. (2011)	κ-Köhler	$\kappa = 1.16$ (Na and Cl)	Multiple mixing states for each size bin	128 aerosol bins
CAM5.3-Oslo	Kirkevåg et al. (2018)	κ-Köhler	2 for RH=80 %, sea salt	Internal and external mixing	44 size-bins with radii (r) ranging from 0.001 to 20 m
GEOS5-Globase	Chin et al. (2002)	Global Aerosol Data Set (GADS) and D'Almeida et al. (1991)	2 for RH=80 %, sea salt	External mixing	Sulfate, BC and OC (2 bins each), dust and sea salt (5 bins each)
GEOS-Chem	Bey et al. (2001)	Martin et al. (2003)	2.4 for RH=90 %, sea salt	External mixing	Sulfate-nitrate-ammonium, OC, BC (bulk-mass approach) Dust (4 bins), sea salts (2 bins)
GEOS5-MERRAero	Buchard et al. (2015)	OPAC and Tang (1997)	Figure 1 in Tang (1997)	External mixing	OC and BC (2 bins), sulfate, dust (5 bins), sea salt (5 bins)

Model-measurement comparison: Co-located annual cycles for 2010

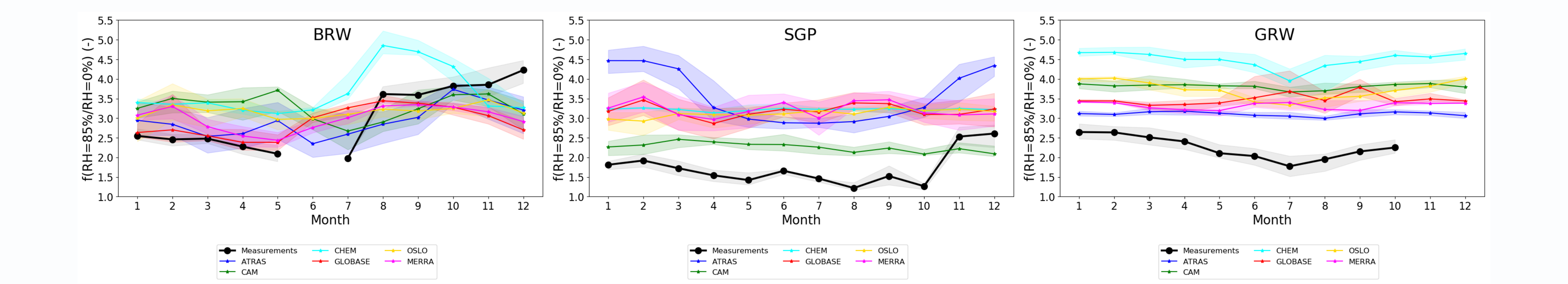

Figure 9 : Annual cycle of modelled and measured $f(\mathrm{RH})$ for Barrow, Southern Great Plains and Graciosa with $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ as reference.

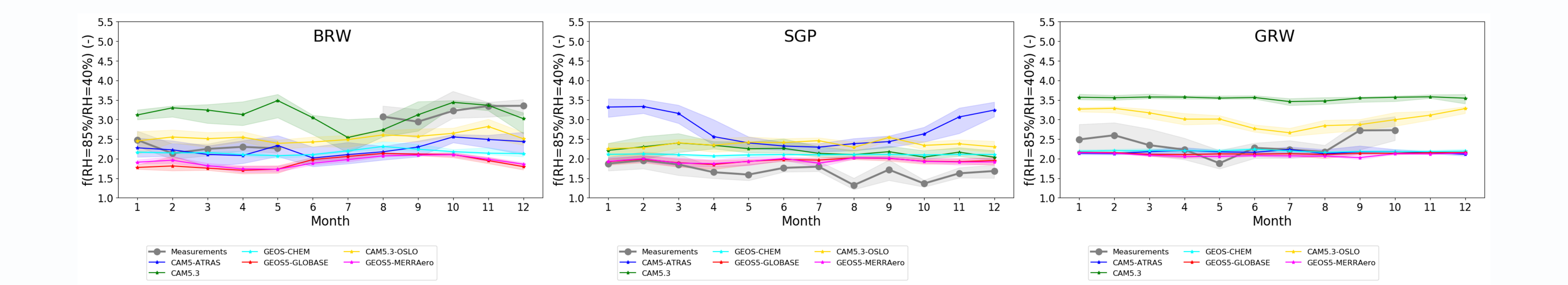

Figure 10 : Annual cycle of modelled and measured $f(\mathrm{RH})$ for Barrow, Southern Great Plains and Graciosa with $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ as reference.

- Models are usually higher than measurements and show large site-specific variations

Model-measurement comparison: All data

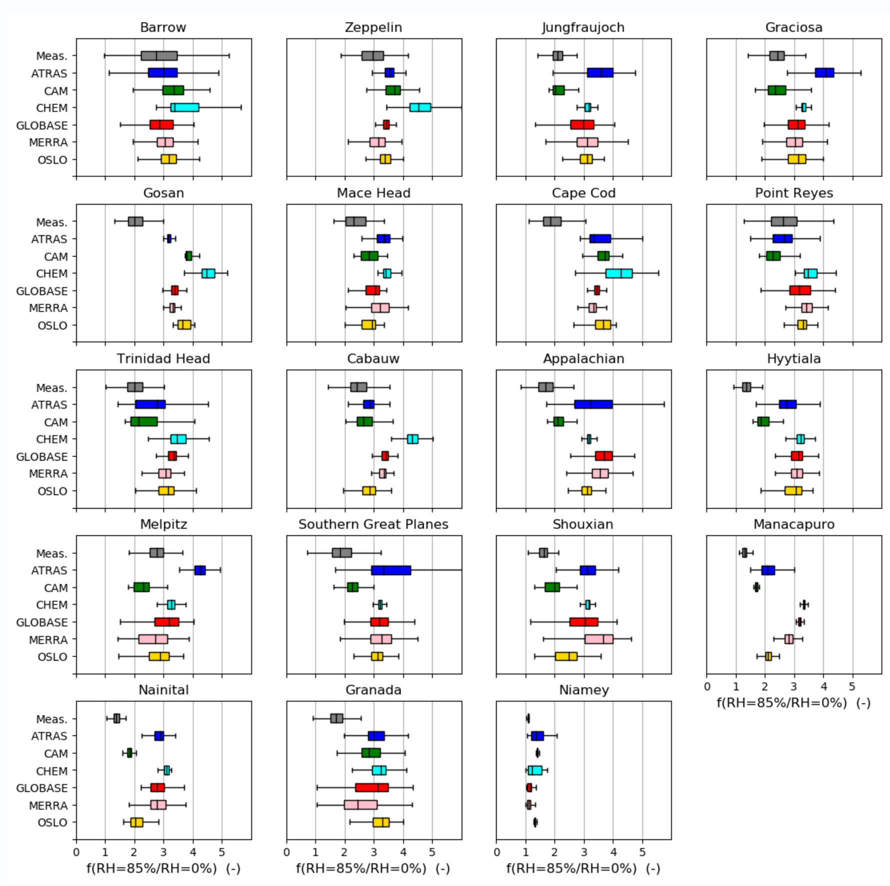

Figure 11 : Box and whisker plots for the entire data set with $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ as reference.

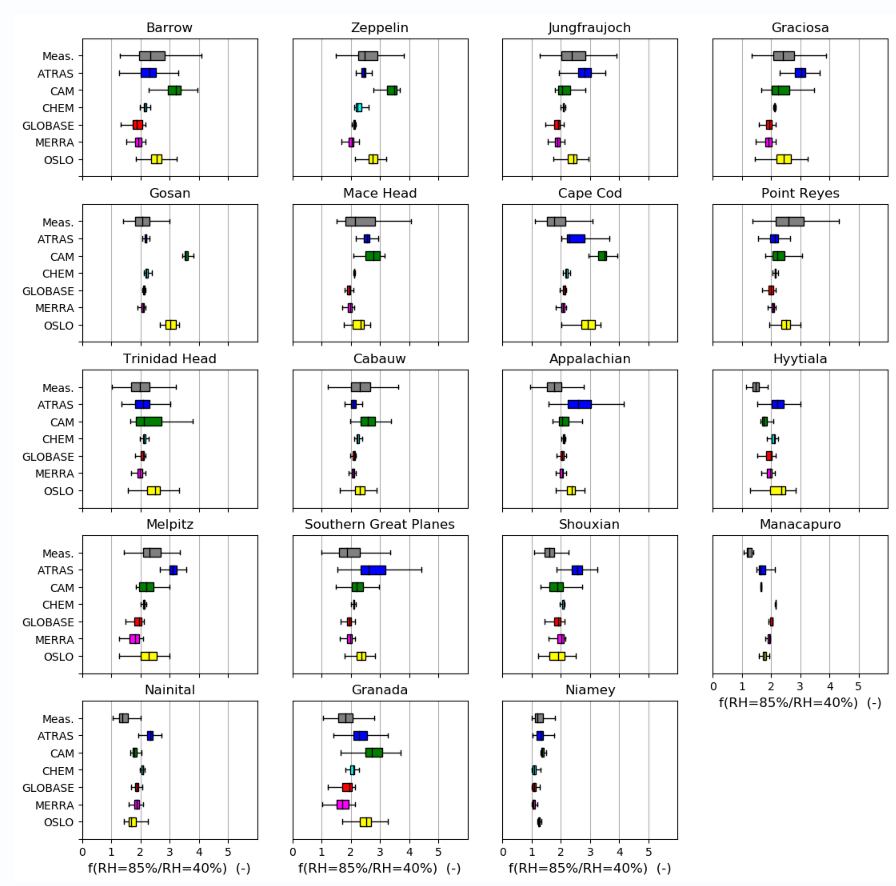

Figure 12 : Box and whisker plots for the entire data set with $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ as reference.

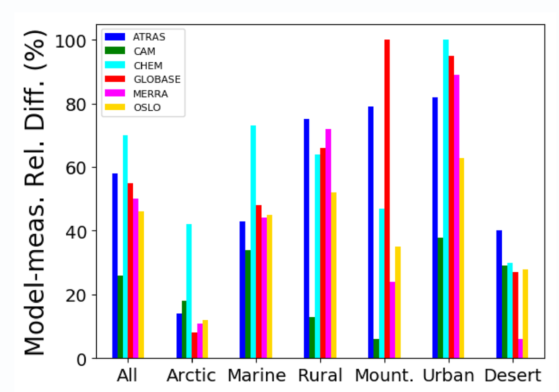

Figure 13 : Relative difference between modelled and measured $f(\mathrm{RH})$ with $\mathrm{RH}_{\mathrm{dry}} = 0\,\%$ as reference.

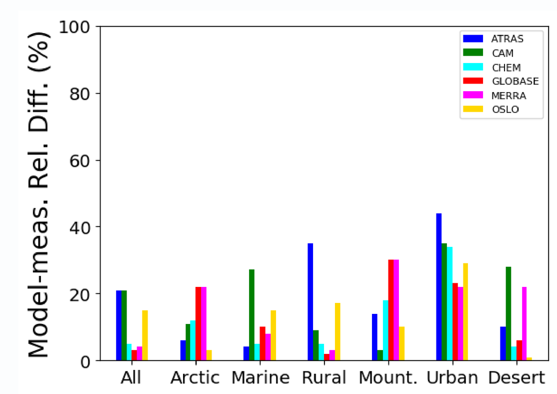

Figure 14 : Relative difference between modelled and measured $f(\mathrm{RH})$ with $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ as reference.

- Co-located monthly averages for all available years
- Large differences between models
- Significant improvement when $\mathrm{RH}_{\mathrm{dry}} = 40\,\%$ is taken as reference

Conclusions

- The **new benchmark dataset** of RH-dependent particle light scattering coefficients and scattering enhancement factors $f(\mathrm{RH})$ has been **finalized** and **successfully tested against six GCM's**
- **Models generally overestimate** $f(\mathrm{RH})$ (similar to the results by Zieger et al. (2013) for OPAC)
- Models still show a **large variability** in $f(\mathrm{RH})$
- Further evaluation needs the **addition of the size & chemical composition** to the analysis

Outlook

- Finalization of papers: model-measurement comparison (lead Maria), What is dry? (lead Betsy & Paul) and $f(\mathrm{RH})$ climatology (lead Gloria: no results shown here)
- Second AeroCom experiment with additional information on size and chemistry and closure/sensitivity using Mie theory
- Comparison to CALIOP extinction coefficients (depending on funding)