First global overview on the representation of water uptake by ten Global Climate Models using a new in-situ benchmark hygroscopicity dataset

M. A. Burgos[1,2,*], E. Andrews[3], G. Titos[4], A. Benedetti[5], H. Bian[6,7], V. Buchard[6,8], G. Curci[9,10], A. Kirkevåg[11], H. Kokkola[12], A. Laakso[12], M. Lund[13], H. Matsui[14], G. Myhre[13], C. Randles[6], M. Schultz[11], T. Van Noije[15], K. Zhang[16], L. Alados-Arboledas[4], U. Baltensperger[17], A. Jefferson[3], J. Sherman[18], J. Sun[19], E. Weingartner[17,20] and P. Zieger[1,2]

[1]Department of Environmental Science and Analytical Chemistry, Stockholm University, Stockholm, Sweden
[2]Bolin Centre for Climate Research, Stockholm, Sweden
[3]Cooperative Institute for Research in Environmental Studies, University of Colorado, Boulder, USA
[4]Andalusian Institute for Earth System Research, University of Granada, Granada, Spain
[5]European Centre for Medium-Range Weather Forecasts, Reading, UK
[6]NASA/Goddard Space Flight Center, USA
[7]University of Maryland Baltimore County, Maryland, USA
[8]GESTAR/Universities Space Research Association, Columbia, USA
[9]Dipartimento di Scienze Fisiche e Chimiche, Universita' degli Studi dell'Aquila, L'Aquila, Italy
[10]Centre of Excellence CETEMPS, Università degli Studi dell'Aquila, L'Aquila, Italy
[11]Norwegian Meteorological Institute, Oslo, Norway
[12]Finnish Meteorological Institute, Kuopio, Finland
[13]Center for International Climate Research, Oslo, Norway
[14]Graduate School of Environmental Studies, Nagoya University, Nagoya, Japan
[15]Royal Netherlands Meteorological Institute, De Bilt, Netherlands
[16]Earth Systems Analysis and Modeling, Pacific Northwest National Laboratory, Richland, WA, USA
[17]Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, Villigen, Switzerland
[18]Department of Physics and Astronomy, Appalachian State University, Boone, USA
[19]Key Laboratory of Atmospheric Chemistry of CMA, Chinese Academy of Meteorological Sciences, Beijing 100081, China
[20]Now at: Institute for Sensing and Electronics, University of Applied Sciences, Windisch, Switzerland

Stockholm University

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ASR Atmospheric System Research

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*Maria.Burgos@aces.su.se

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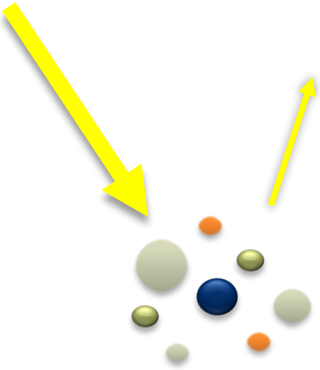
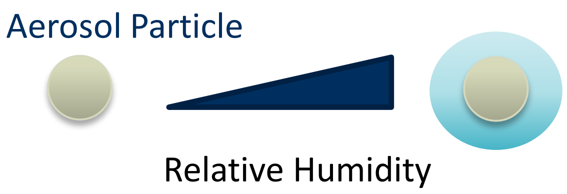
Aerosols and Climate

- **Aerosols have direct and indirect effects on the Earth's energy balance**
  - Scatter (σ_{sp}) and absorb solar radiation
  - Influence the number of cloud condensation nuclei





HYGROSCOPICITY: Since aerosol particles can take up water, they can change in size and chemical composition depending on the ambient relative humidity (RH)

$\boldsymbol{\sigma_{sp}(RH, \lambda)}$**, strongly depends on RH**

The effect of water uptake is **relevant** for **climate forcing calculations** as well as for the comparison or validation of **remote sensing** with in-situ measurements and for the improvement of **Earth System Models**

SCATTERING ENHANCEMENT FACTOR

$$f(RH, \lambda) = \frac{\sigma_{sp}(RH, \lambda)}{\sigma_{sp}(RHdry, \lambda)}$$

How well do Global Climate Models represent aerosol optical hygroscopic growth?

This presentation summarizes our work, which is currently under review in ACP:

Submitted as: research article 20 Jan 2020

A global model-measurement evaluation of particle light scattering coefficients at elevated relative humidity

Review status
This preprint is currently under review for the journal ACP.

María A. Burgos[1,2], Elisabeth J. Andrews[3], Gloria Titos[4], Angela Benedetti[5], Huisheng Bian[6,7], Virginie Buchard[6,8], Gabriele Curci[9,10], Alf Kirkevåg[11], Harri Kokkola[12], Anton Laakso[12], Marianne T. Lund[13], Hitoshi Matsui[14], Gunnar Myhre[13], Cynthia Randles[6], Michael Schulz[11], Twan van Noije[15], Kai Zhang[16], Lucas Alados-Arboledas[4], Urs Baltensperger[17], Anne Jefferson[3], James Sherman[18], Junying Sun[19], Ernest Weingartner[17,a], and Paul Zieger[1,2]

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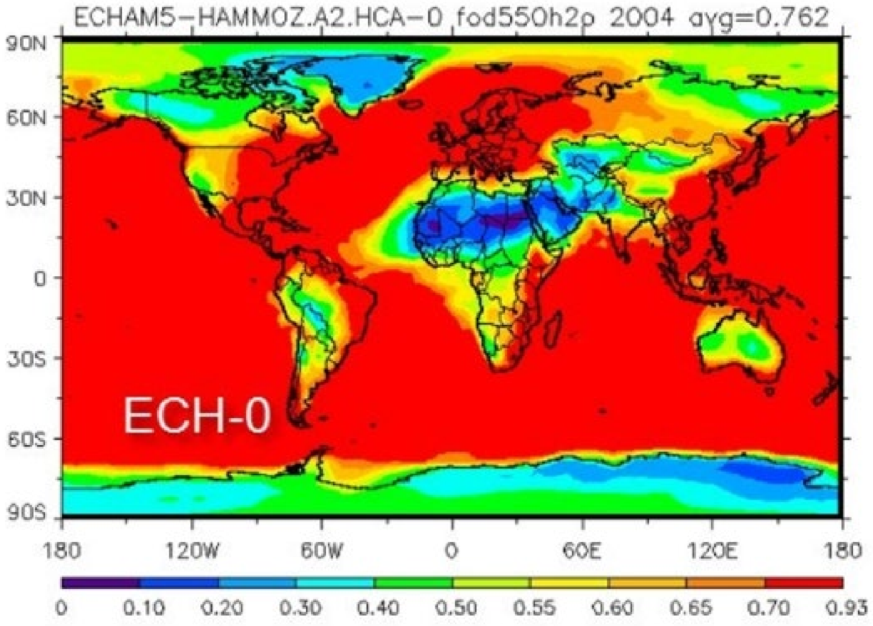
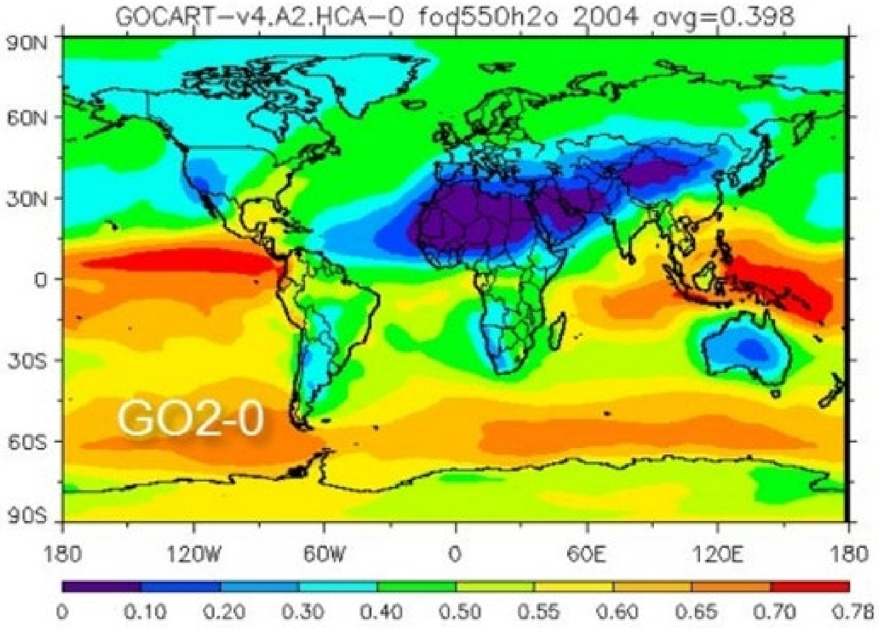
Hygroscopicity in Earth System Models:

Interestingly, most models are doing well in reproducing the total aerosol optical depth (AOD), but a closer look into the individual components reveals discrepancies between them

Fraction of aerosol optical depth due to water:

Figures from Mian Chin (NASA Goddard)





ECHAM5: global annual average **76%**

GOCART: global annual average **40%**

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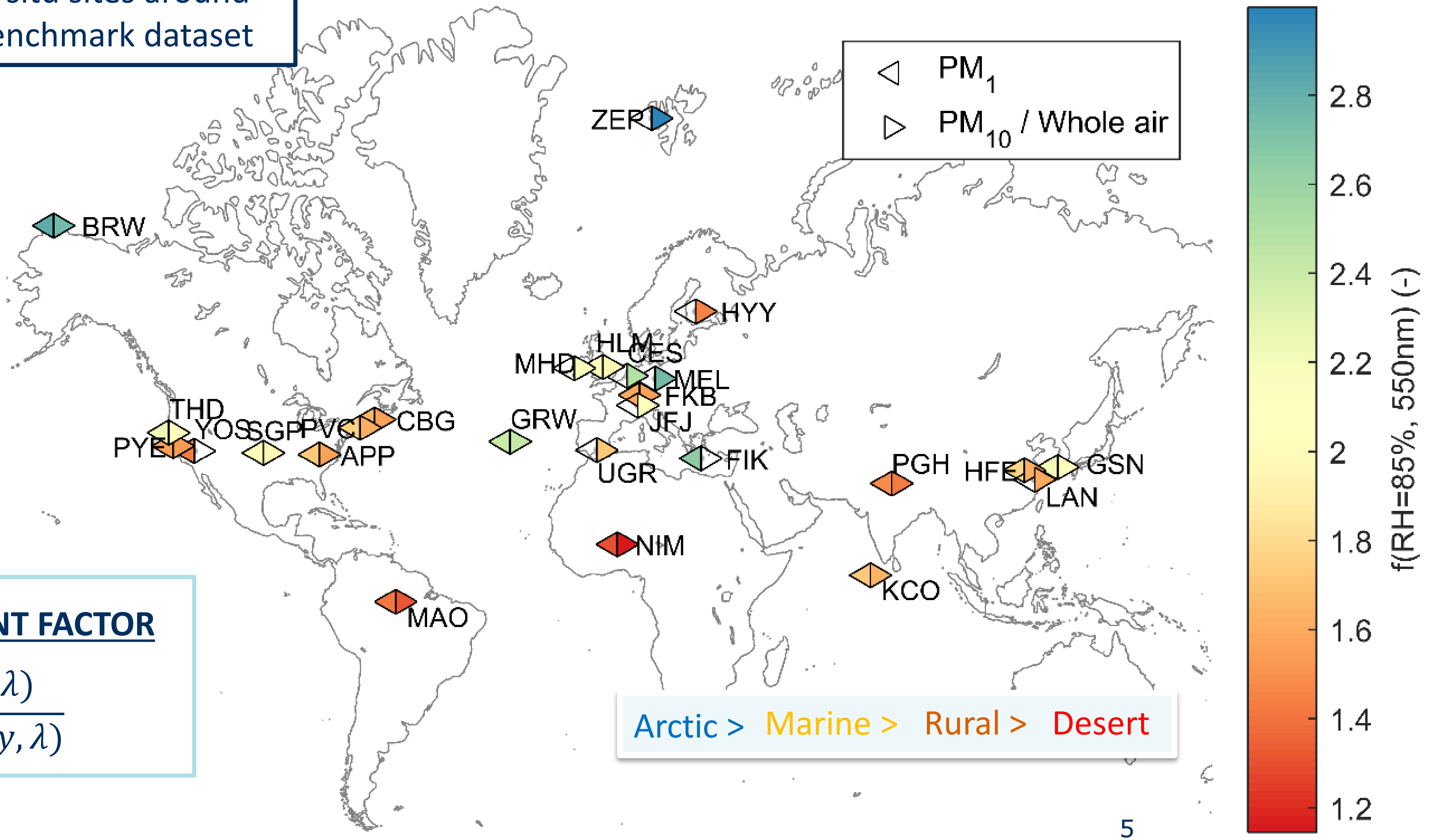
[Burgos et al., 2019:](#)

f(RH) measurements from in-situ sites around the globe used to create a benchmark dataset



SCATTERING ENHANCEMENT FACTOR

$$f(RH,\lambda) = \frac{\sigma_{sp}(RH,\lambda)}{\sigma_{sp}(RHdry,\lambda)}$$

MODEL DATA: INSITU project - AeroCom Phase III

- 10 Models used in this study:
  - Three CAM-family models: CAM-ATRAS, CAM5, CAM-Oslo
  - Three GEOS-family models: GEOS-Chem, GEOS-GOCART, MERRAero
  - Four further models: OsloCTM3, TM5, IFS-AER, SALSA

- We work with the following output:
  - Aerosol optical data, **absorption and extinction** at **RH=0, 40 and 85%**, λ=550 nm
  - Mass mixing ratio for five components: **black carbon, desert dust, organic aerosols, sulfates, and sea salt**

- The **frequency** is hourly or daily values for the **year 2010.**
- An important aspect is that time coverage is not always coincident with measurements

- The extracted model data is for the closest grid point to 22 observational sites

- We have used simulated surface data (regardless of site elevation)

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MODEL	Chemical composition	Mixing State	Hygroscopicity [g(RH=90%)]					
			parameterization	ss	so4	bc	oa	dd
ATRAS	bc,so4,oa,ss,dd + no3/nh4	I	κ-Köhler Theory	2.25	1.87	1.0	1.24	1.0
CAM	bc,so4,oa,ss,dd	I	κ-Köhler Theory	2.25	1.77	1.0	1.24	1.2
CAM-Oslo	bc,so4,oa,ss,dd	I,E	κ-Köhler Theory	2.28	1.77	1.0	1.31	1.2
GEOS-Chem	bc,so4,oa,ss,dd + no3/nh4	E	Modified GADS	2.38	1.64	1.4	1.64	1.0
GEOS-GOCART	bc,so4,oa,ss,dd	E	Modified GADS	1.9-2.1	1.8	1.4	1.6	1.0
MERRAero	bc,so4,oa,ss,dd	E	Modified GADS	1.9-2.1	1.8	1.4	1.64	1.0
OsloCMT3	bc,so4,oa,ss,dd + no3/nh4	I	Own development	2.3-2.4	1.72	1.0	1.46	1.0
TM5	bc,so4,oa,ss,dd + no3/nh4	I, E	Own development	-	-	1.0	1.0	1.0
IFS-AER	bc,so4,oa,ss,dd + no3/nh4	E	Own development	2.36	1.73	1.0	1.64	1.0
SALSA	bc,so4,oa,ss,dd	E	Own development	2.4	1.9	1.0	1.5	1.0

I. Comparison of modelled vs. measured f(RH) (+ organic mass fraction)

II. Importance of temporal collocation: BRW, GRW and SGP sites

III. Graciosa as a test case for modeled sea salt hygroscopicity

IV. Analysis of the implications of the different definitions of RH_{ref}

In this presentation we focus on the sections I and III.

To see the rest of the results, please take a look at our paper (ACPD)

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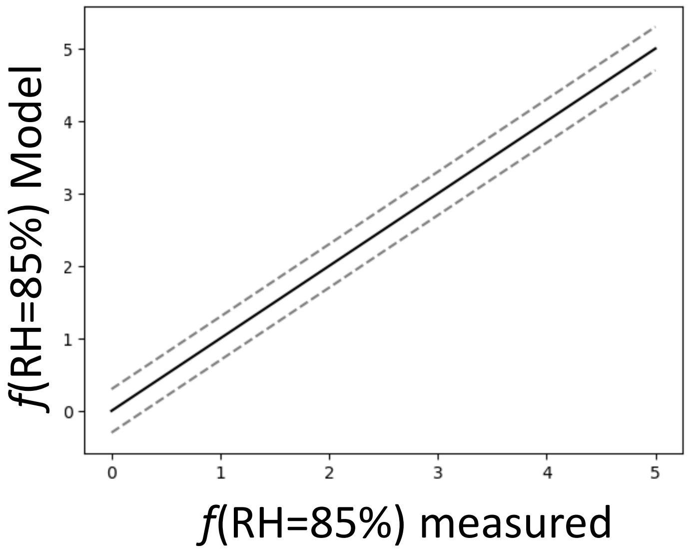
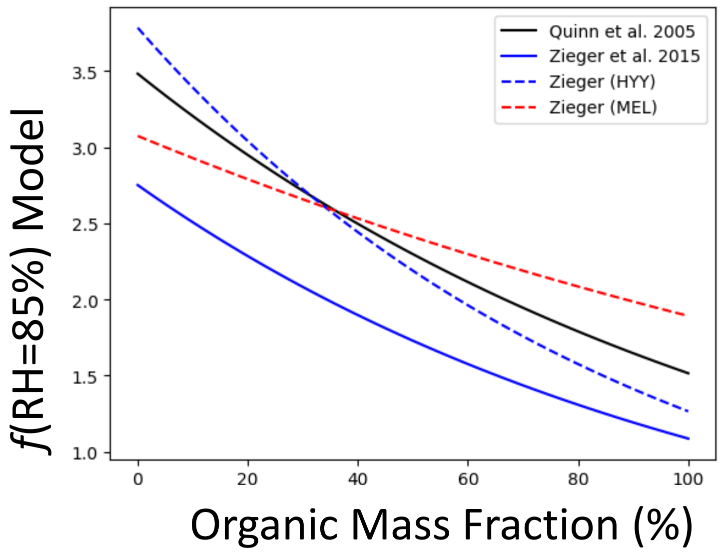


$$f(RH, \lambda = 550nm) = \frac{\sigma_{sp}(RH = 85\%)}{\sigma_{sp}(RH = 40\%)} *$$

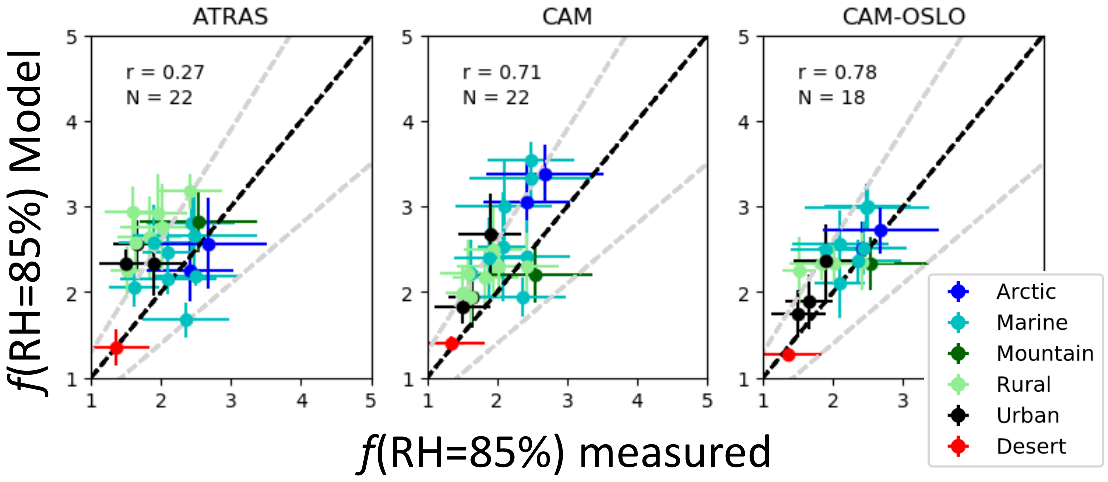
*Chose RH=85% to minimize potential issues with hysteresis



$$OMF = \frac{organics}{(organics + sulfate)}$$

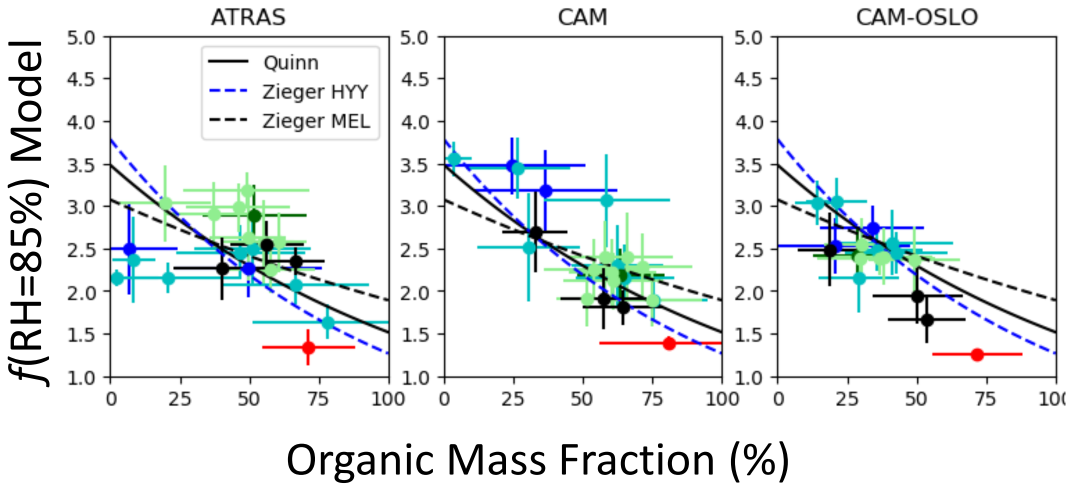
- Quinn et al. 2005: parameterization based on measurements at CBG, GSN, KCO
- Zieger et al. 2015: same approach for MEL and HYY sites.
- Zieger et al. 2015: Solid line including nitrate, black carbon, ammonia, and Cl

CAM-family models



<u>*f*(RH=85%) model vs measured:</u>

- Models reproduce the range in measured *f*(RH)
- Good correlation coefficients for CAM and CAM-Oslo



<u>*f*(RH=85%) model vs OMF:</u>

- CAM and CAM-Oslo exhibit similar relationship between *f*(RH) and Organic Mass Fraction as suggested by Quinn and Zieger parameterizations

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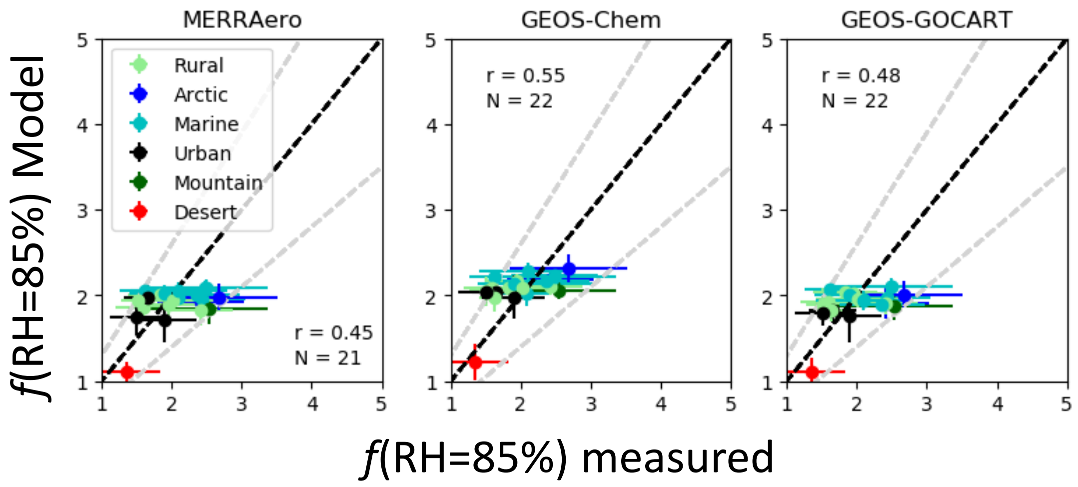
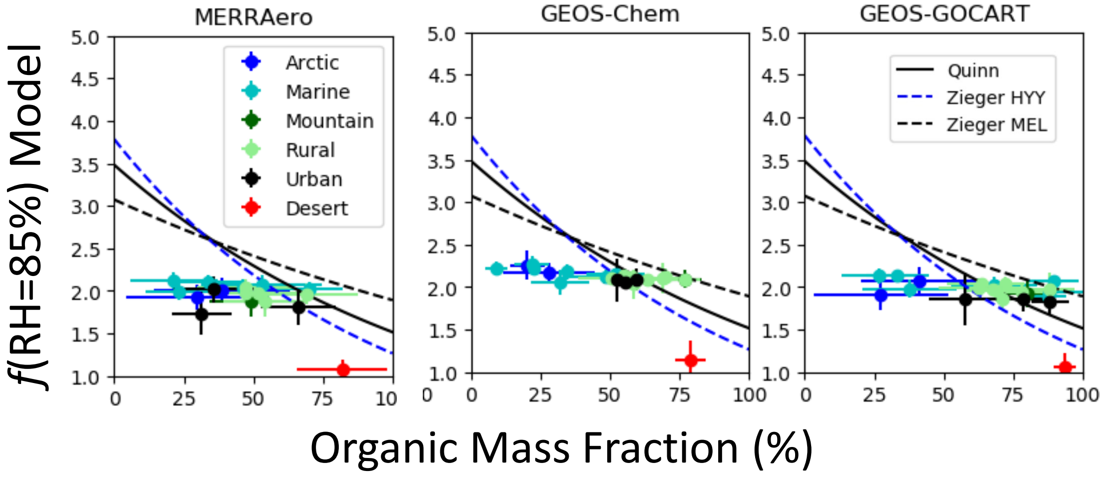




GEOS-family models

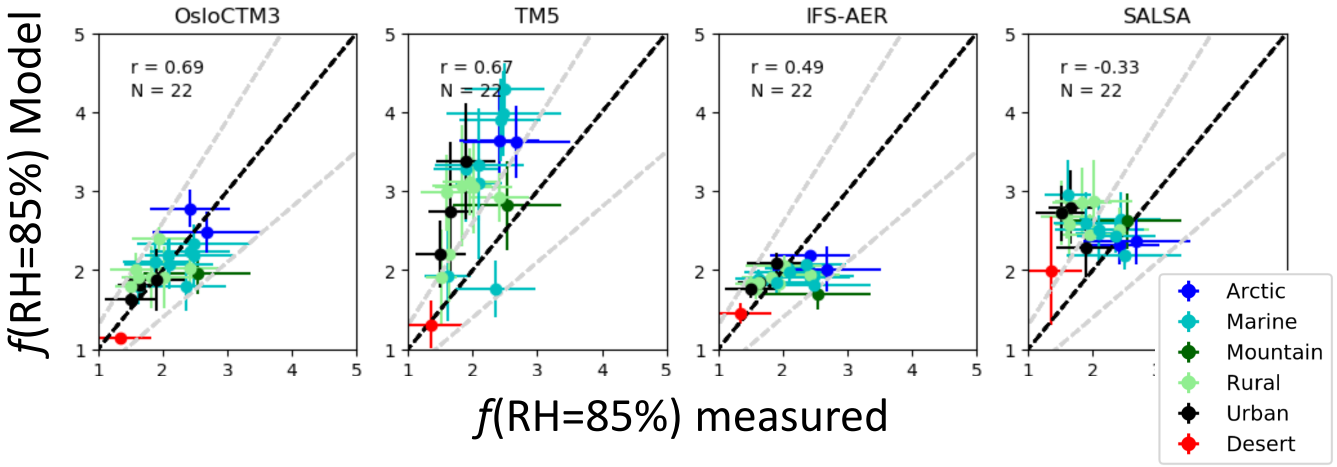
f(RH=85%) model vs measured:

- Models do not reproduce the range in measured *f*(RH) but values fall within 30% uncertainty
- Lower correlation coefficients than for CAM-models

f(RH=85%) model vs OMF:

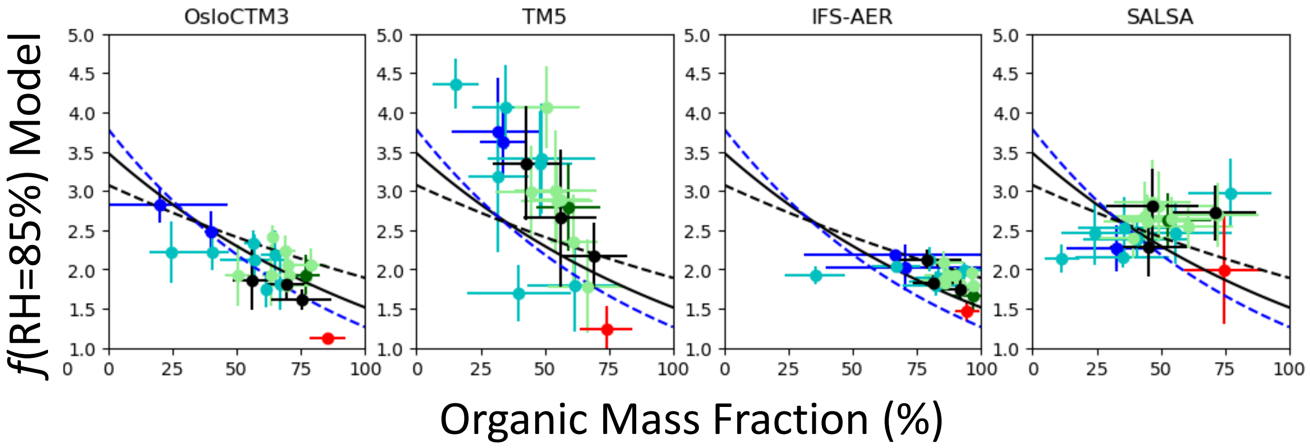
- Models do not exhibit same Organic Mass Fraction - *f*(RH) relationship as observations

OsloCTM3, TM5, IFS-AER, SALSA



Diversity of behaviors:

- Good correlation for OsloCTM3 and TM5
- Inverse correlation for SALSA



- OsloCTM3 and IFS-AER agree well with parameterizations
- IFS-AER simulates aerosol dominated by organics
- TM5 exhibits same tendency as paramerterizations but overestimates *f*(RH) relative to Organic Mass Fraction
- SALSA is different

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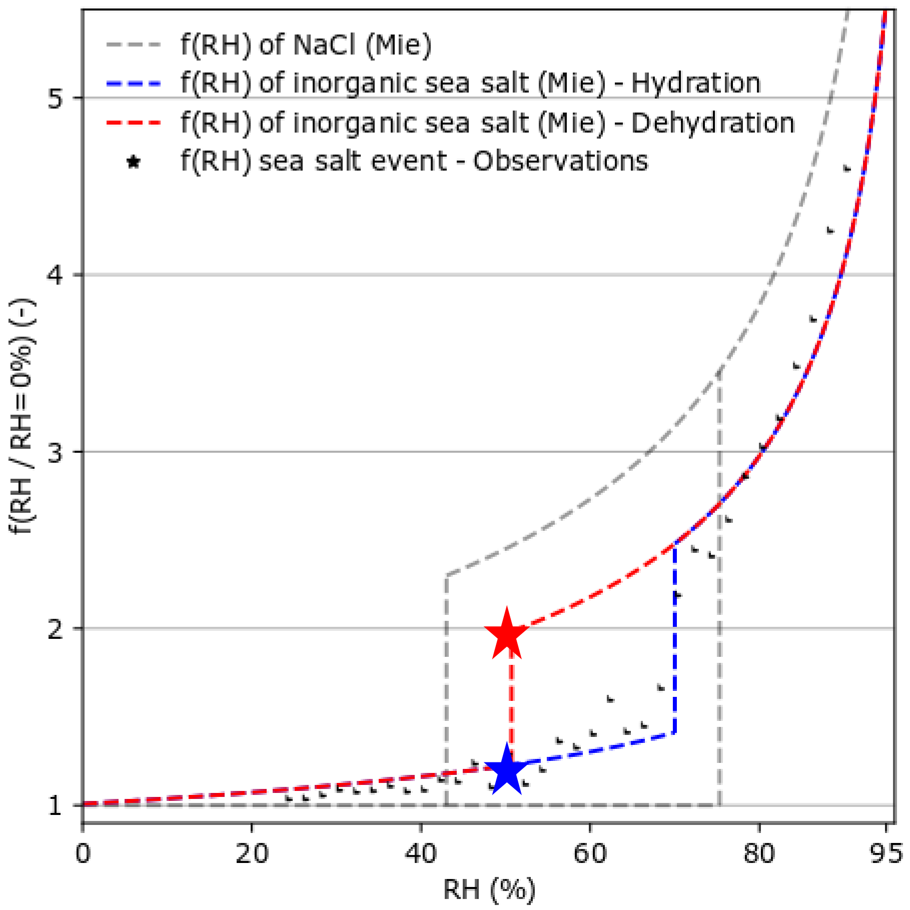

Observational data and theoretical curves for inorganic sea salt and NaCl

(calculated using Mie theory as described in Zieger et al., 2013, and the revised hygroscopic growth factors of inorganic sea salt and NaCl determined by Zieger et al., 2017)

Zieger et al., 2017 has shown that inorganic sea salt:
★ Hydration curve: f(RH=40%) ≈ 1.2
★ Dehydration curve: f(RH=40%) = 2.0

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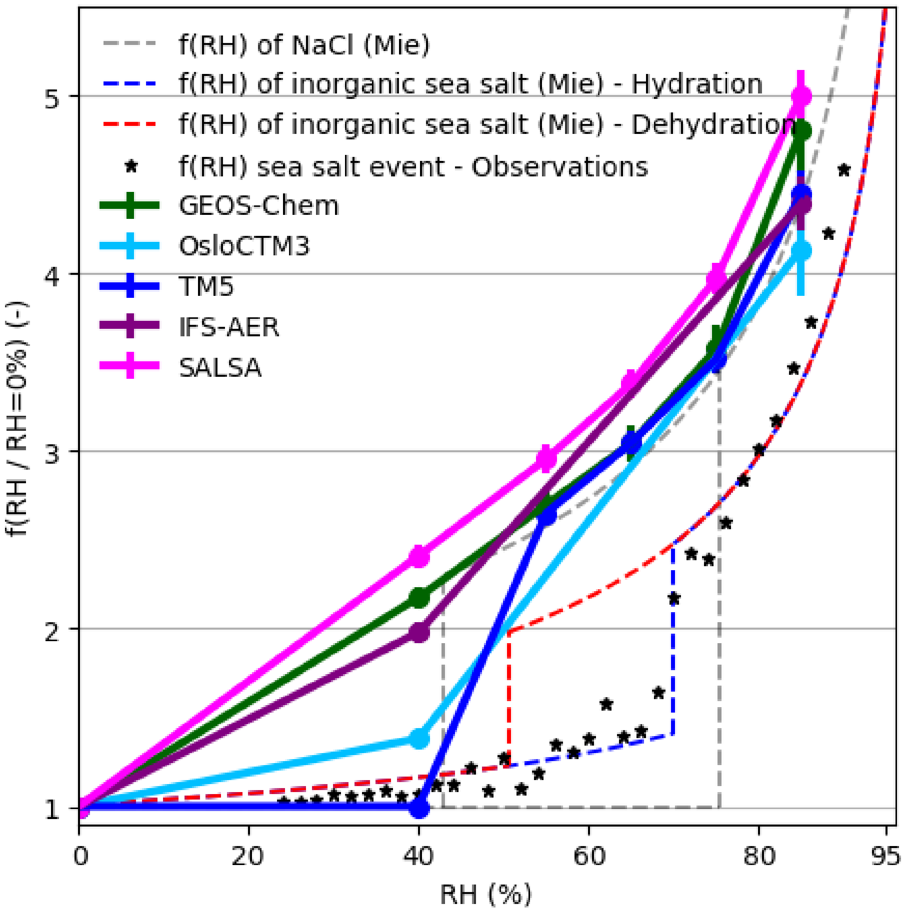
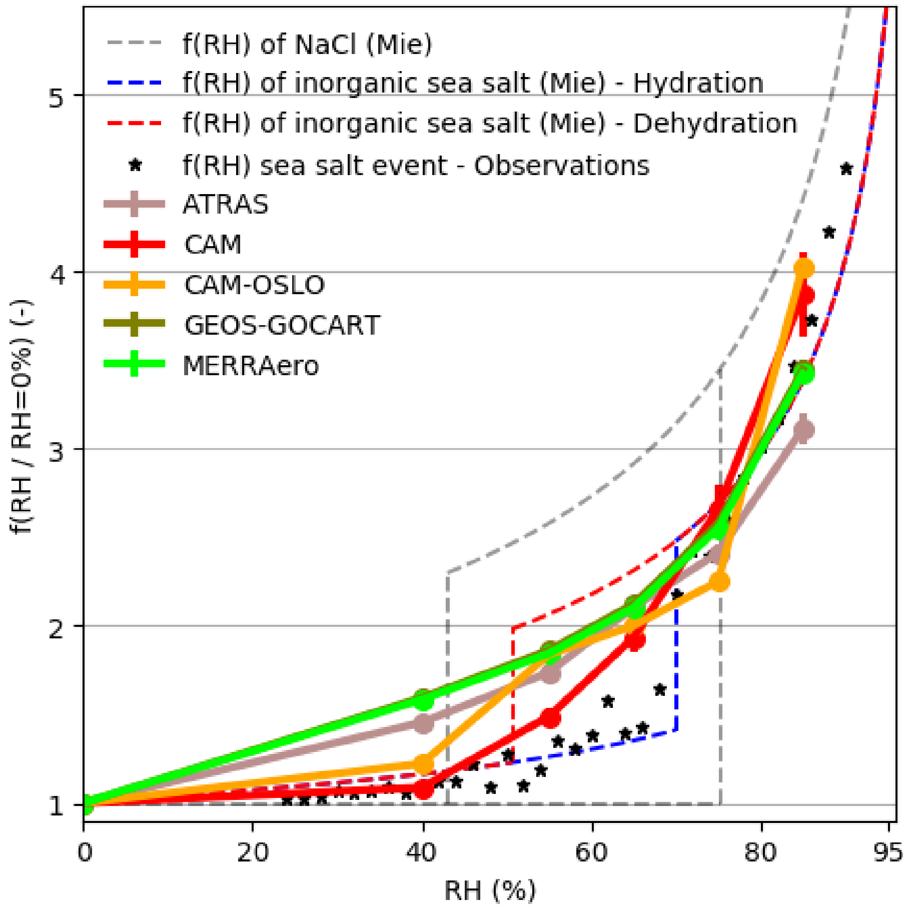


GEOS-Chem, OsloCTM3, TM5, IFS-AER, and SALSA:
Are modelling sea salt as NaCl

- TM5: no hygroscopic growth up to RH=45%
- GEOS-Chem, IFS-AER, SALSA: don’t assume the aerosol to be solid at RH=40%
- SALSA: estimates slightly larger values -> smaller particle sizes



ATRAS, CAM, CAM-Oslo, GEOS-GOCART, and MERRAero:
Are modelling inorganic sea salt

At RH=40%:
- CAM, CAM-Oslo: values closer to the hydration curve
- ATRAS, GEOS-GOCART, MERRAero: values closer to the dehydration curve

Hysteresis range: always in between hydration/dehydration curves

At higher RH:
- ATRAS lowest value
- GEOS-GOCART, and MERRAero: best match
- CAM and CAM-Oslo: include hysteresis

Summary of main results:

- Model assumptions about water uptake at low RH are a significant factor
- Different assumptions about the hygroscopicity of sea salt explain some model variation at a marine location
  -> some models assume sea salt can be represented by NaCl, while others do not
- **GEOS-family** models assign too much hygroscopicity to **all species** (except dust)
  -> (almost) regardless of simulated composition the resulting *f*(RH) will be high (exception dust dominated site)
  -> narrow *f*(RH) range
- **GEOS models** all use **Global Aerosol Data SET (GADS)*** to parameterize growth so this high f(RH) is **consistent** with findings by Zieger et al., 2013 showing overestimation at low RH

* GADS is a popular database on aerosol and cloud optical properties that is widely used by the scientific community since it provides a comprehensive set of microphysical and optical data of aerosol and clouds

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In conclusion:

1. Measurements of **particle light scattering enhancement factors** have been compared to a set of 10 Earth System Models

2. We see a **high diversity** in the comparison between models and measurements due to the variability in the different assumptions related to hygroscopic growth and chemical composition

3. **Organic Mass Fraction** can be used as a constraint or "sanity check" for the modelled f(RH)

4. Aerosol mixing size and mixing state, as prescribed in the models, may have an important influence too. Accounting for the exact contribution of each of these factors is a **challenge** and more research needs to be carried out

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Further results… check out our paper currently in ACPD

1. **Temporal collocation** between models and measurements was done for three sites.
   - Did not appear to improve the comparison of model simulations and observations relative to climatology
   - Model diversity was larger than the variability in the observed long-term climatology

2. Model and measurement assumptions about 'What is dry' are different and need to be considered in these types of comparisons

Check out our project website

Maria Burgos,
Stockholm University
For Questions, feel free to contact me at:
Maria.Burgos@aces.su.se

Thanks for your attention!

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