Comparison between in-situ surface measurements and global climate model outputs of particle light scattering coefficient as a function of relative humidity

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[7]ExxonMobil Research and Engineering Company
[8]Norwegian Meteorological Institute, Norway

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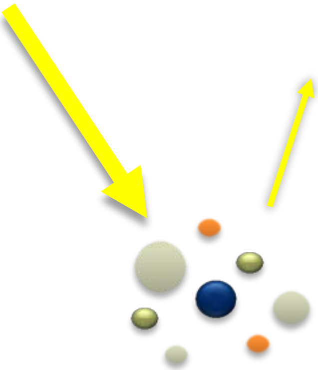
Stockholm University

Aerosols and Climate

- **Direct and indirect effects on the Earth's energy balance**
- Scattering (σ_{sp}) and absorption of solar radiation and the number of cloud condensation nuclei will be affected by aerosol concentration, size and chemical composition



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 **Global Climate Models**

SCATTERING ENHANCEMENT FACTOR

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

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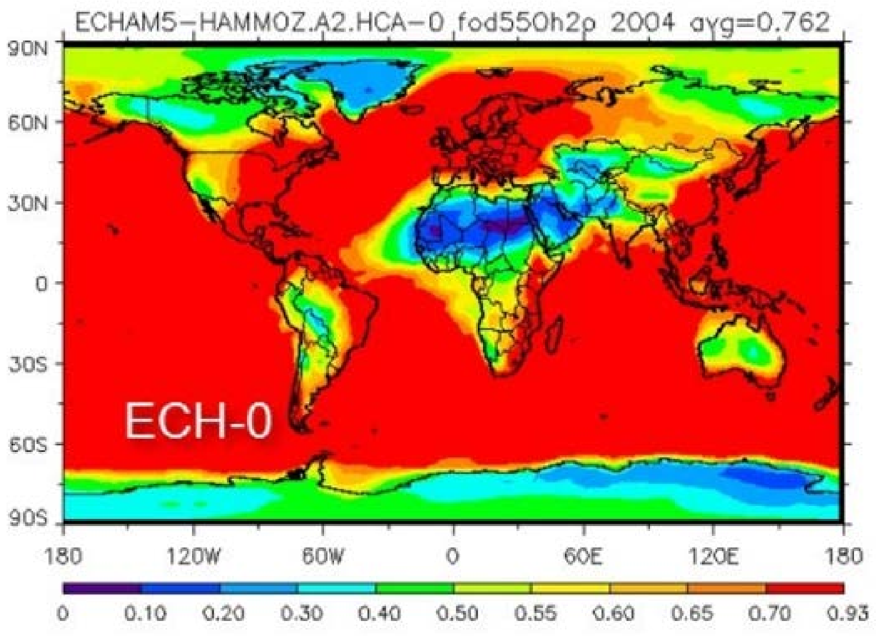
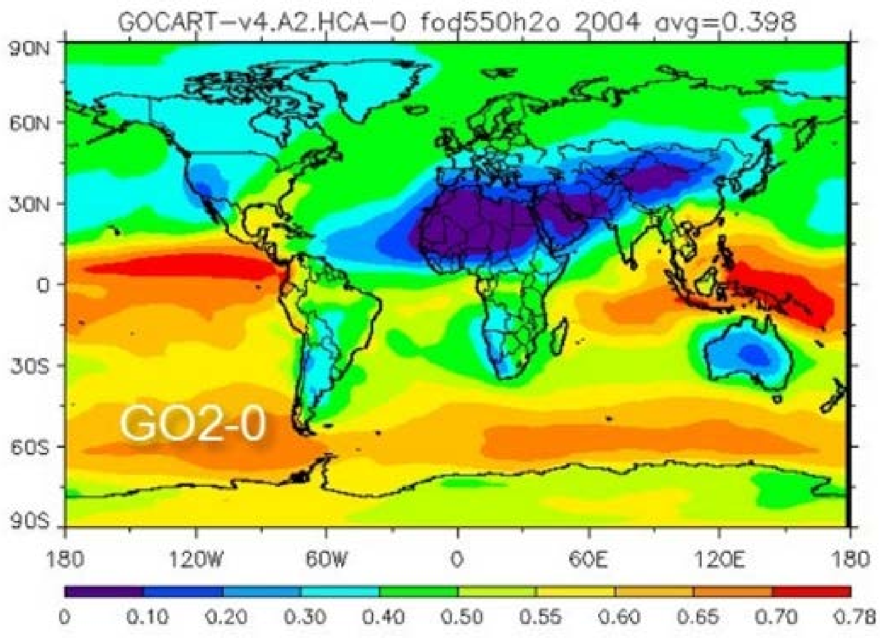
Hygroscopicity in GCM's

Fraction of aerosol optical depth (AOD) due to water in different models:





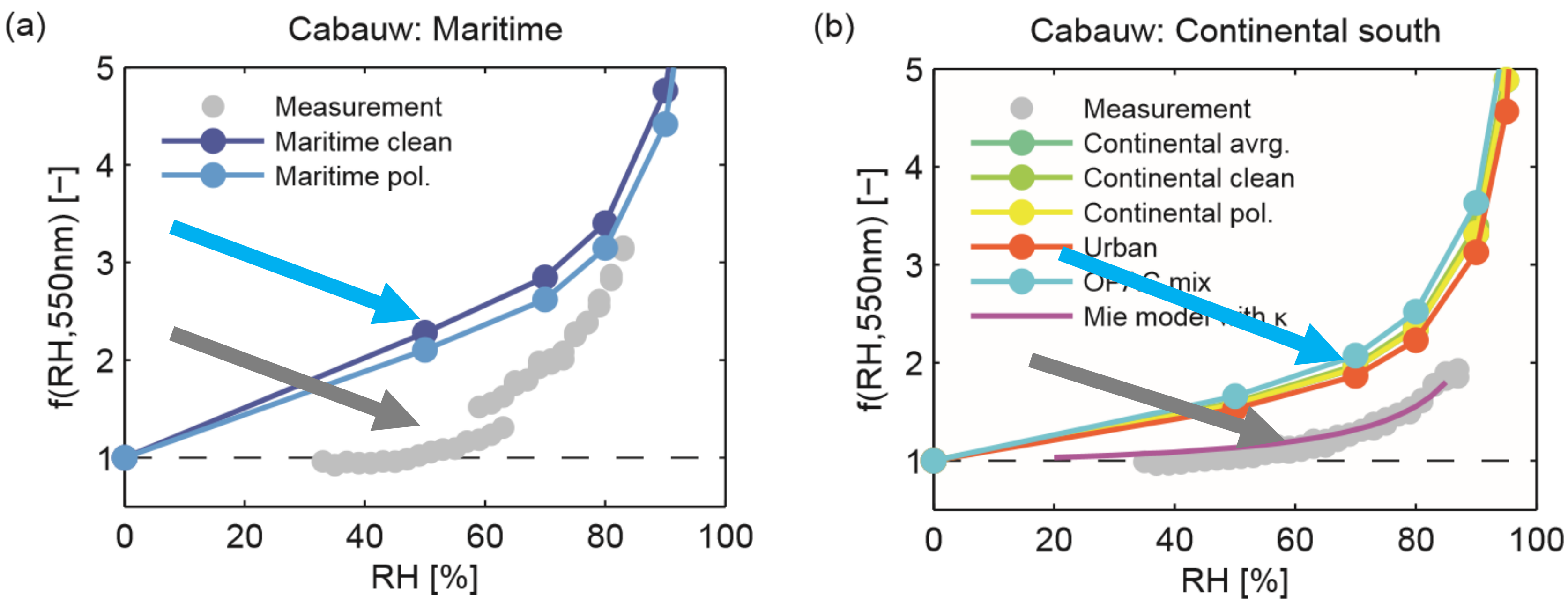
Figures from Mian Chin (NASA Goddard)

ECHAM5: global annual average **76%**

GOCART: global annual average **40%**

Hygroscopicity in GCM's

OPAC: Optical Properties of Aerosol and Clouds (Hess et al., 1998)



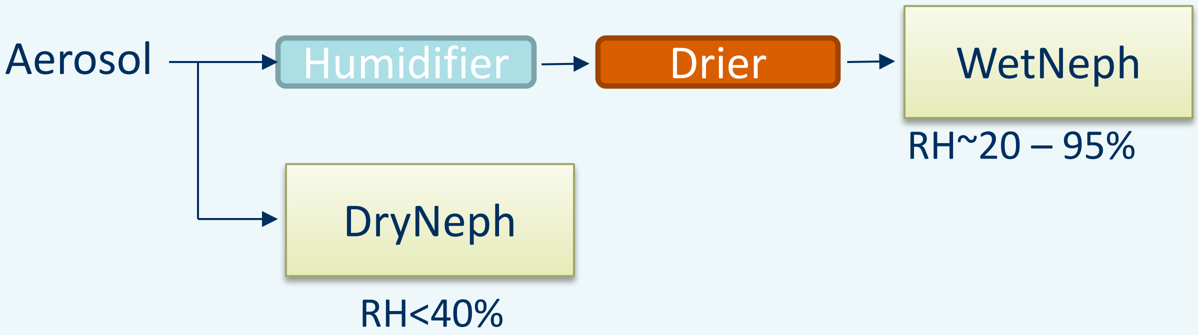
Figures from Zieger et al., 2013

OPAC model generally higher than measurements especially for low-medium RH

Reason: OPAC growth factors for sea salt and sulfate components are too high. Revised growth factors for sea salt published in Zieger *et al.,* 2017.

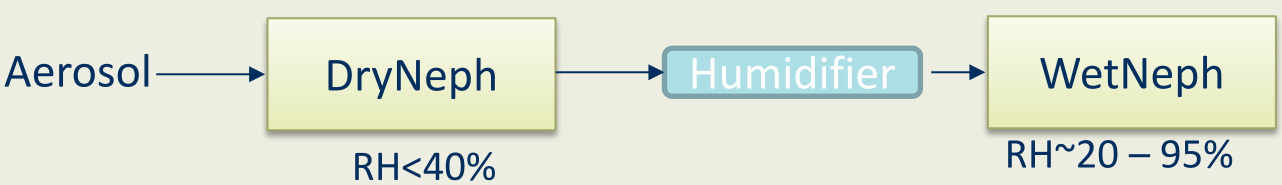
Tandem Humidified Nephelometer

PSI system:



(Fierz-Schmidehauser et al., 2010)

NOAA system:



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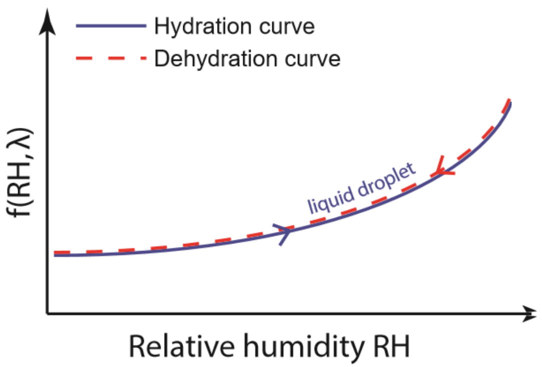
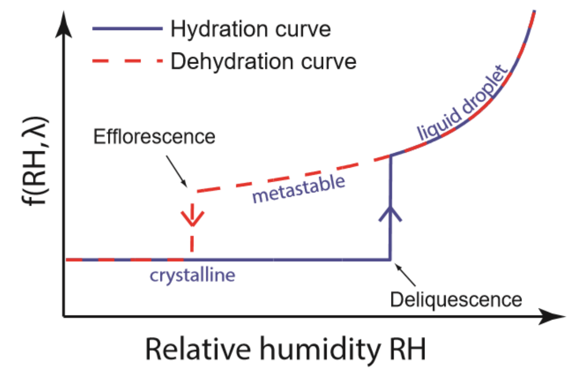


Hygroscopic particles grow or shrink monotonically with ΔRH

Deliquescent aerosols undergo sudden phase transition (hysteresis)

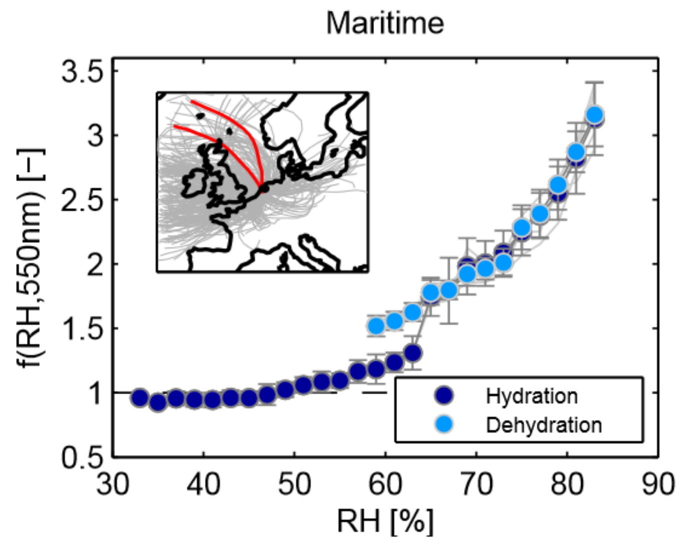


- Humidograms can be parameterized with different equations:

Carrico et al., 2003: $f(RH) = \alpha\,(1 - RH)^{-\gamma}$

→ Problem for sea salt aerosols (deliquescence)

Zieger et al., 2010: Fit separately for RH>75% or RH<65%
Titos et al., 2016: Several equations, some of them reproduce deliquescence



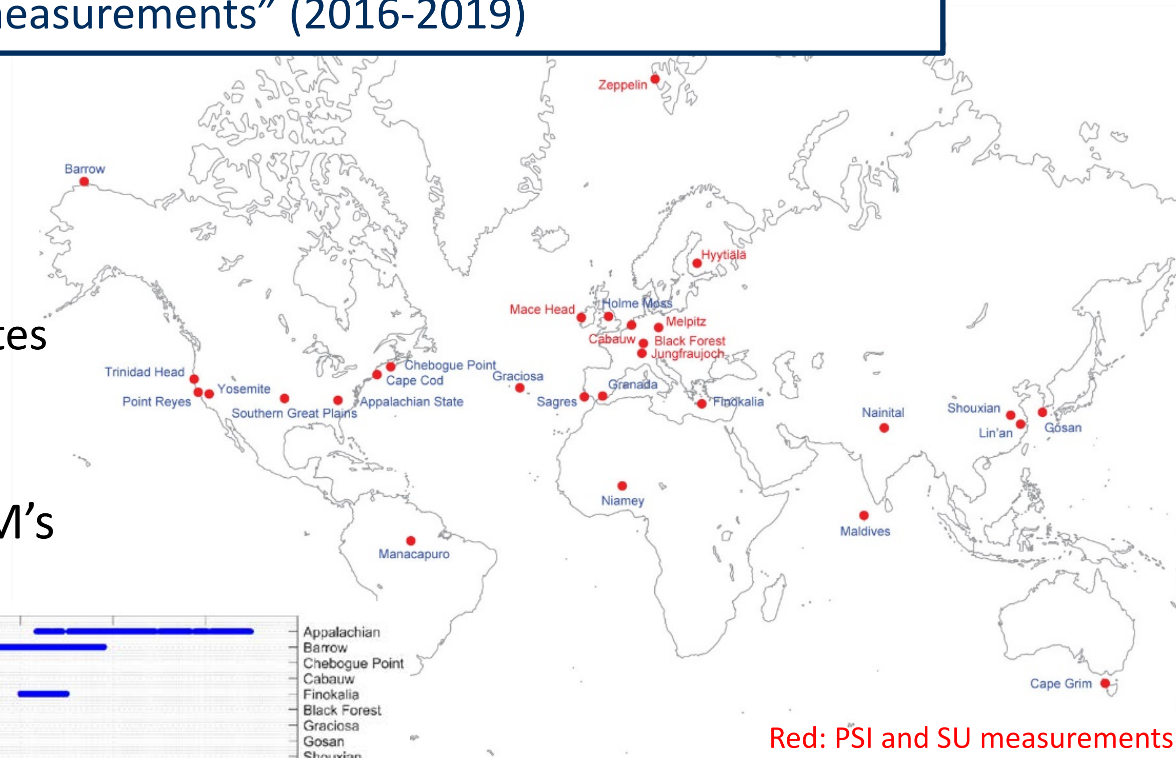
Zieger et al., 2011

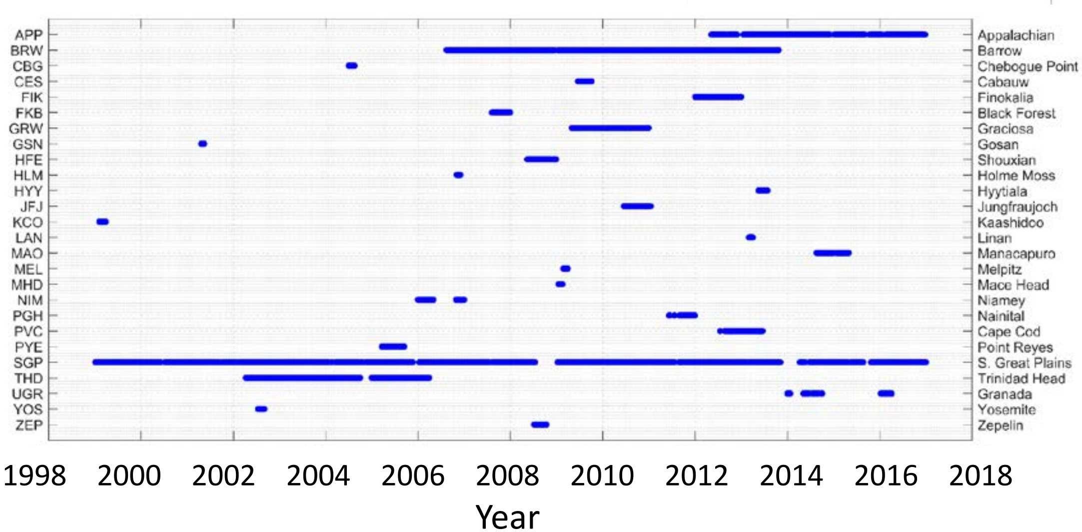
DoE funded project:
"Evaluation and improvement of the parameterization of aerosol hygroscopicity in global climate models using in-situ surface measurements" (2016-2019)

HARMONIZED DATA SET

- DoE/ARM sites, PSI sites and more
- Covering 18 years

compare with GCM's





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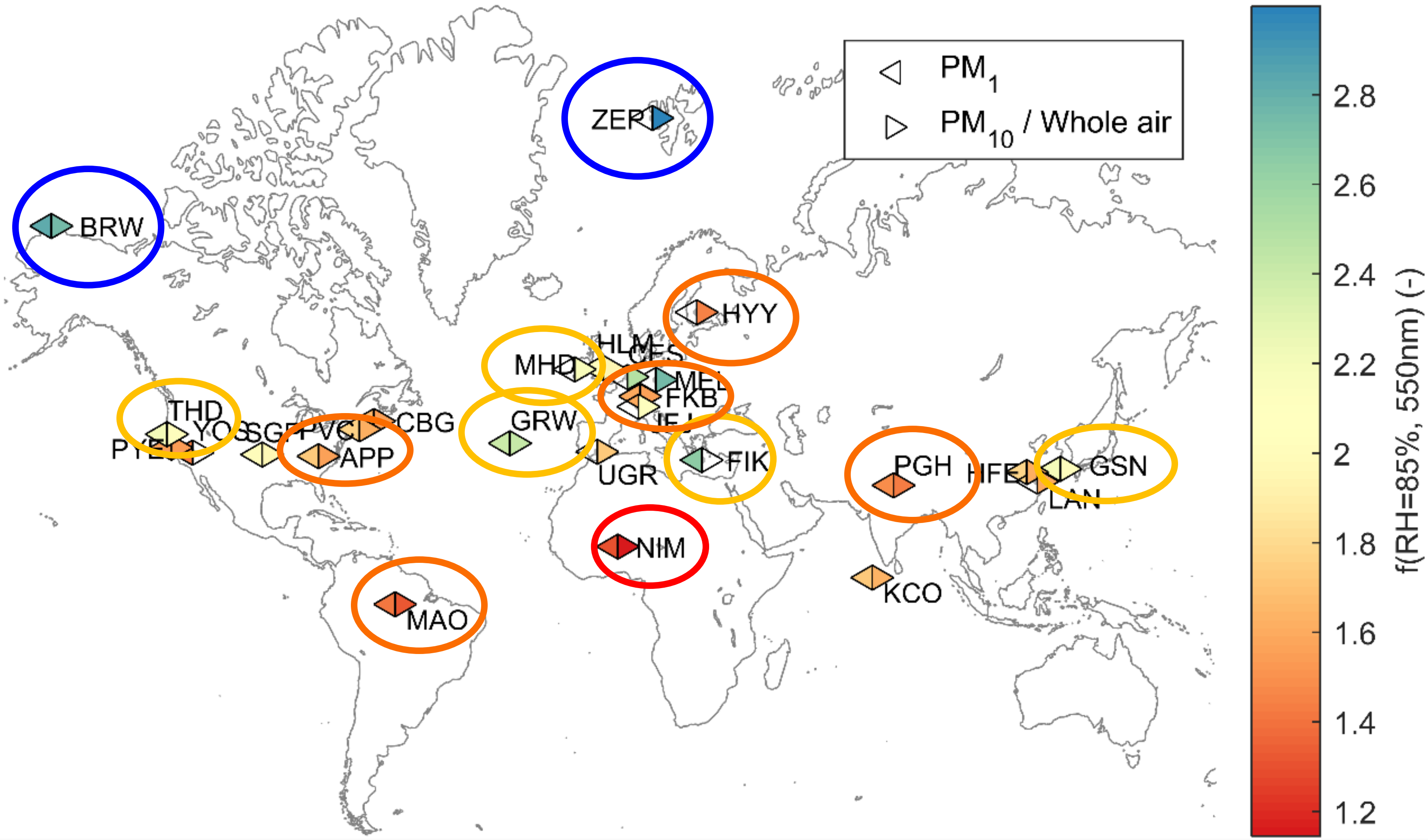


f(RH=85%):

Arctic > Marine > Rural > Desert

MERRA Aerosol Reanalysis (MERRAero):

- **Buchard *et al.* (2015):** “Using the OMI aerosol index and absorption aerosol optical depth to evaluate the NASA MERRA Aerosol Reanalysis”

- **MERRA Aerosol Reanalysis:** reanalysis for the satellite era based on a version of the GEOS-5 model, radiatively coupled to the Goddard Chemistry, Aerosol, Radiation, and Transport (GOCART) aerosol module (bulk (mass) scheme).
  - GEOS-5 -> run in replay mode using 6-hourly atmospheric analysis from MERRA
  - Aerosol species: dust, sea-salt, sulfates, organic and black carbon
  - Assimilation of bias corrected MODIS AOD observations at 550 nm every each 3 hours
  - Provides a aerosol gridded data set covering from 2002 to 2015

CAM5.3-Oslo

- **Kirkevåg et al. (2018):** “A production-tagged aerosol module for earth system models, OsloAero5.3 – extensions and updates for CAM5.3-Oslo ”

- Aerosol module: **OsloAero5.3** implemented in the atmospheric component **CAM5.3-Oslo** of the Norwegian Earth System model (NorESM1.2)

  Improvements: treatment of emissions, aerosol chemistry, particle lifecycle and aerosol-cloud interactions

  New features: improved aerosol sources, aerosol particle nucleation, secondary organic aerosol production, emissions schemes for sea-salt, DMS and marine primary organics…

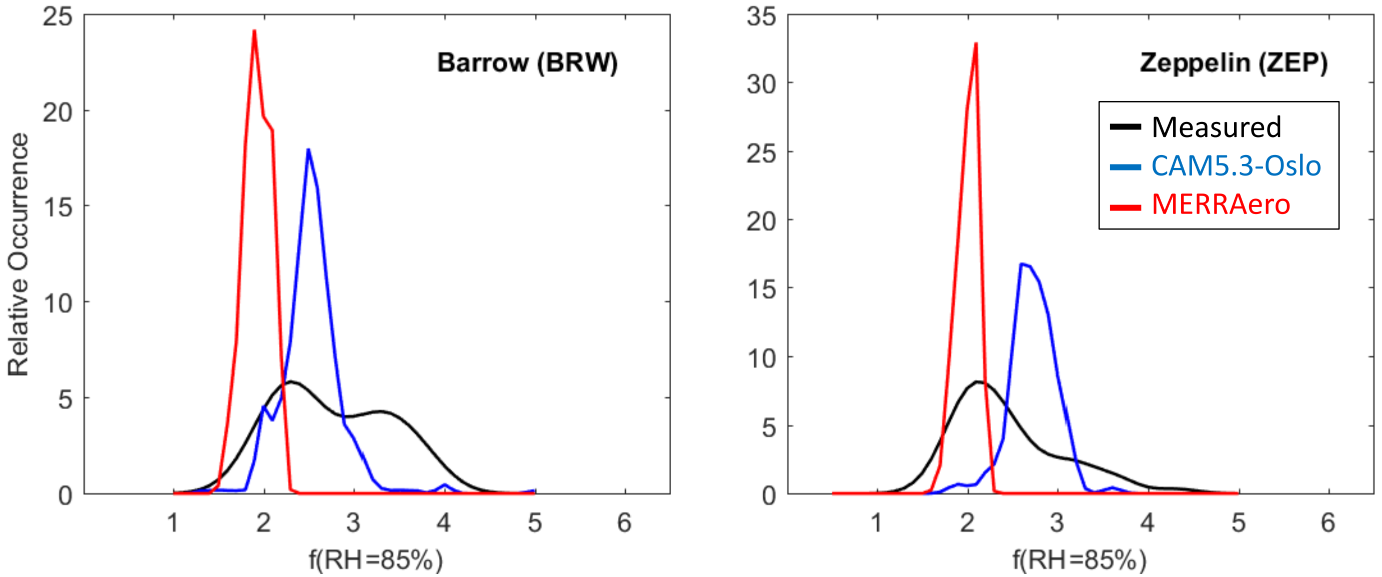
- Model data availability → Daily values
  → Period: January – December, 2010

- **Time coverage** of model data and measurements are **not coincident**.
  For consistency, short-term campaign sites with only a few months of measurements are compared to the same months of the model data.

- **Uncertainty** in measurements between 20-30%, which has to be taken into account in the measurement-model comparison

Relative Frequency of Occurrence of f(RH=85%)

ARCTIC SITES



- Measurements show higher variability while models present a narrower distribution
- Measurements variability may be affected by the change of particle concentration along the year: Arctic haze in spring/new particle formation in summer/low concentration in winter (Tunved *et al.*, 2013)

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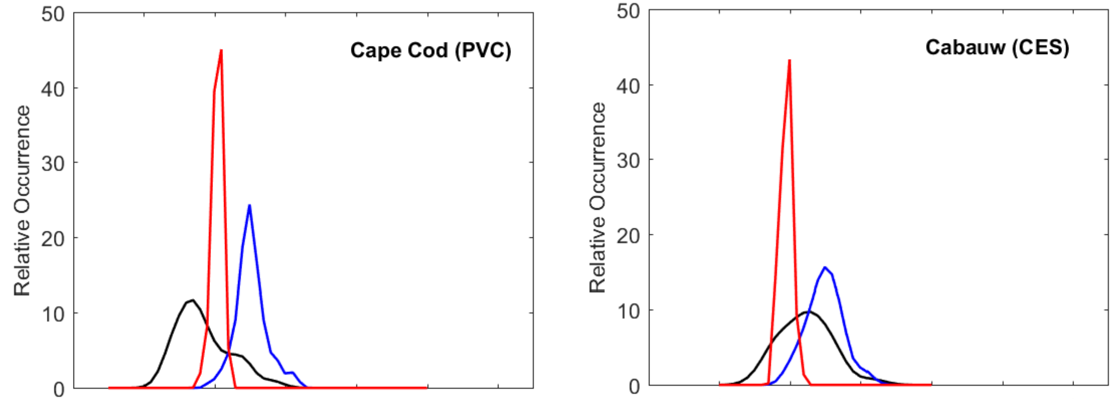
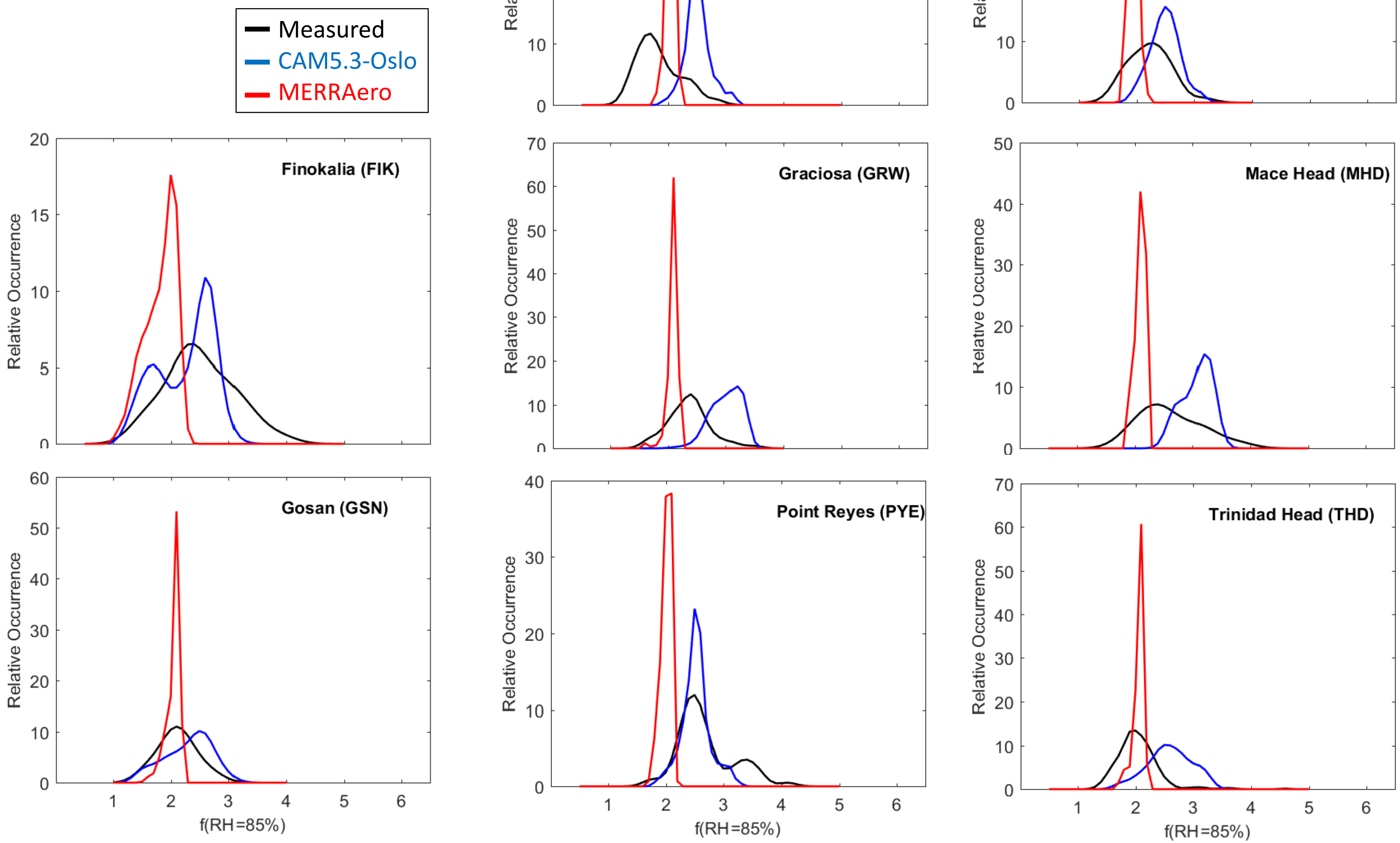
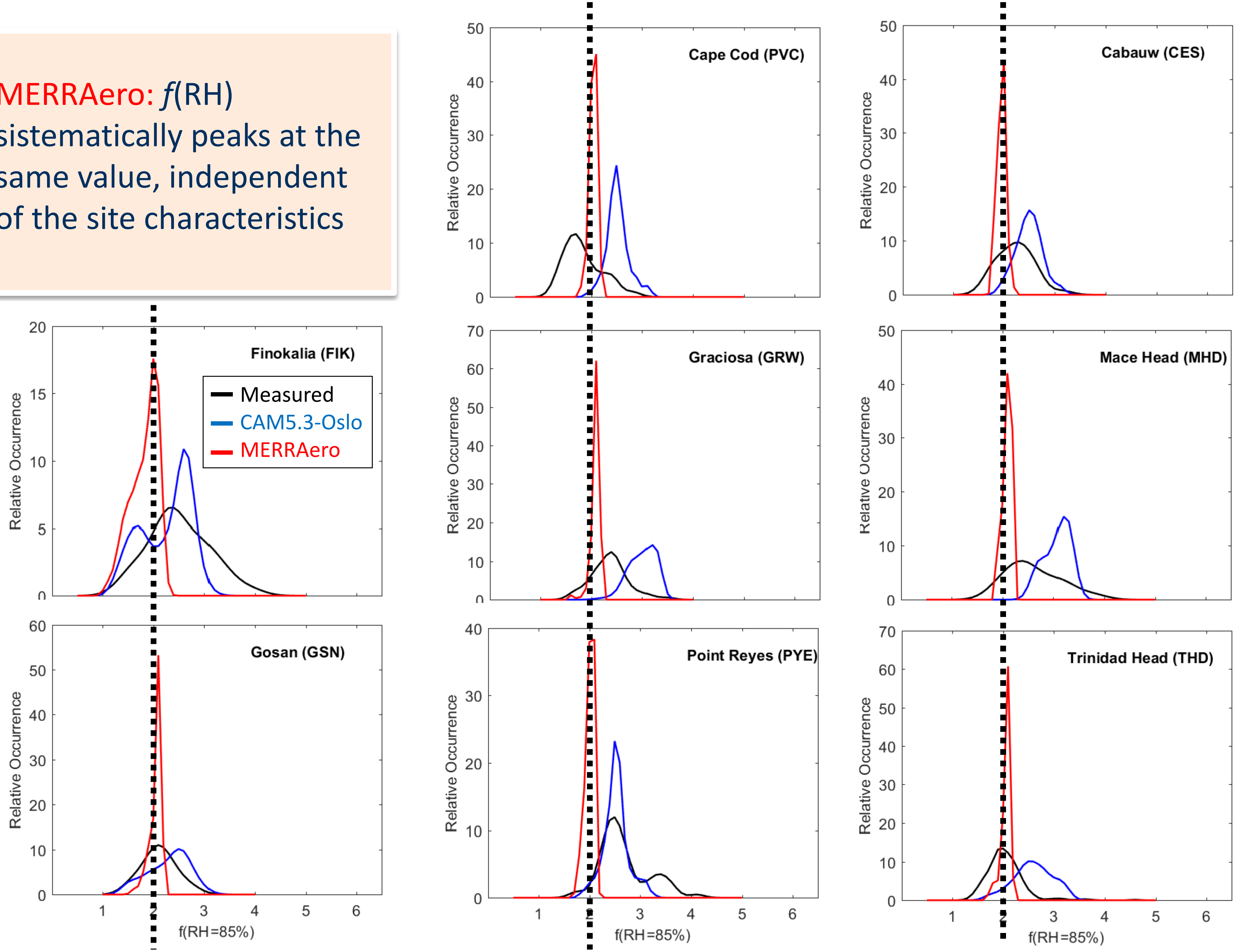
MARINE SITES:

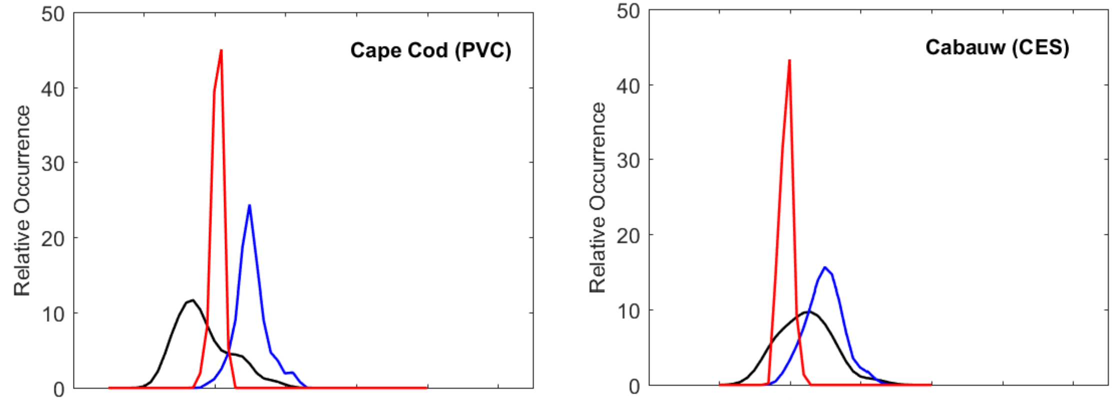


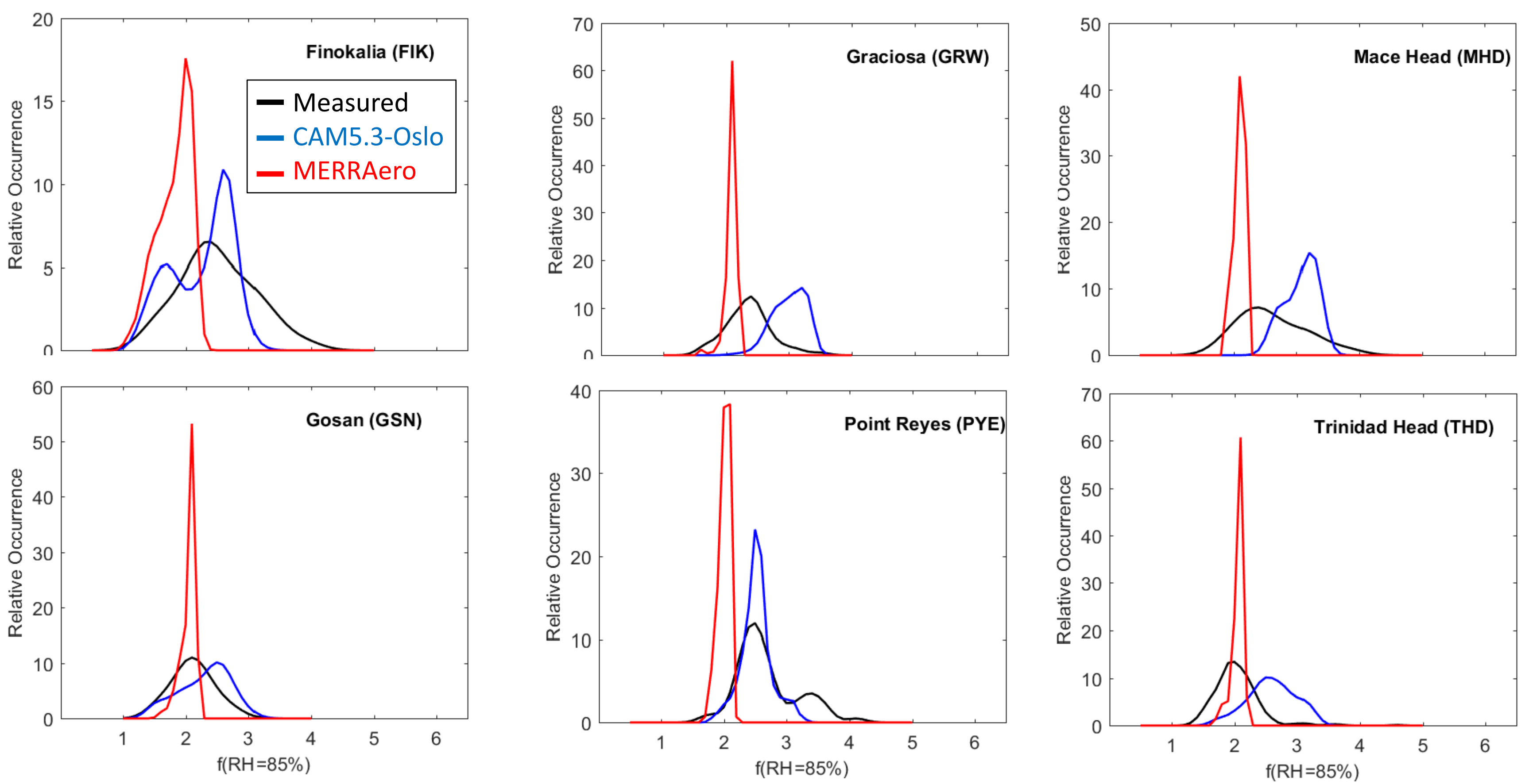


MERRAero: *f*(RH) sistematically peaks at the same value, independent of the site characteristics



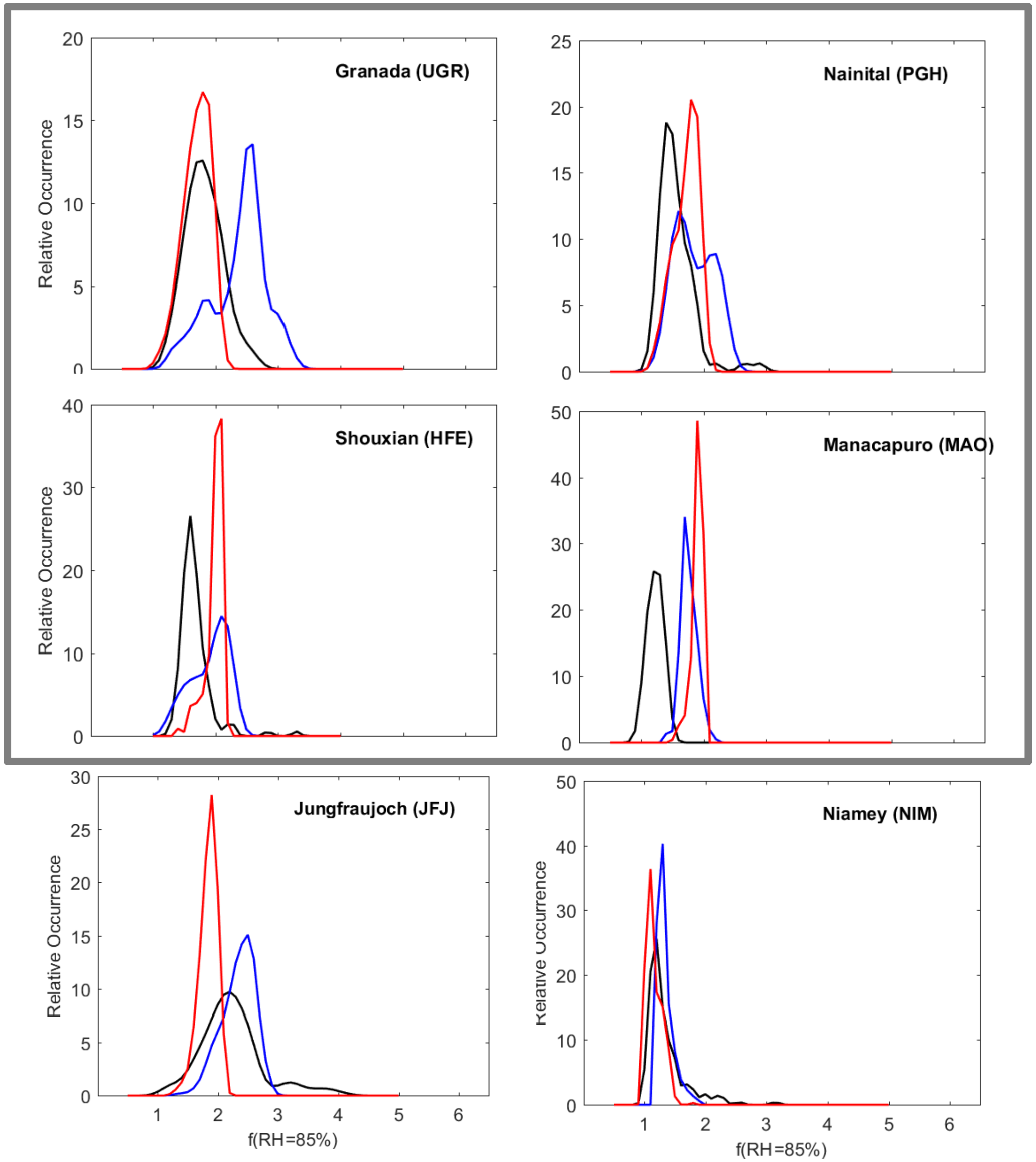
CAM5.3-Oslo does better in reproducing the observed shape, though it tends to overestimate the measured values





Urban-Mountain-Desert Sites

- **Urban** Sites:
  Models reproduce observed *f*(RH) for Granada and Nainital, but overestimate in Shouxian and Manacapuro



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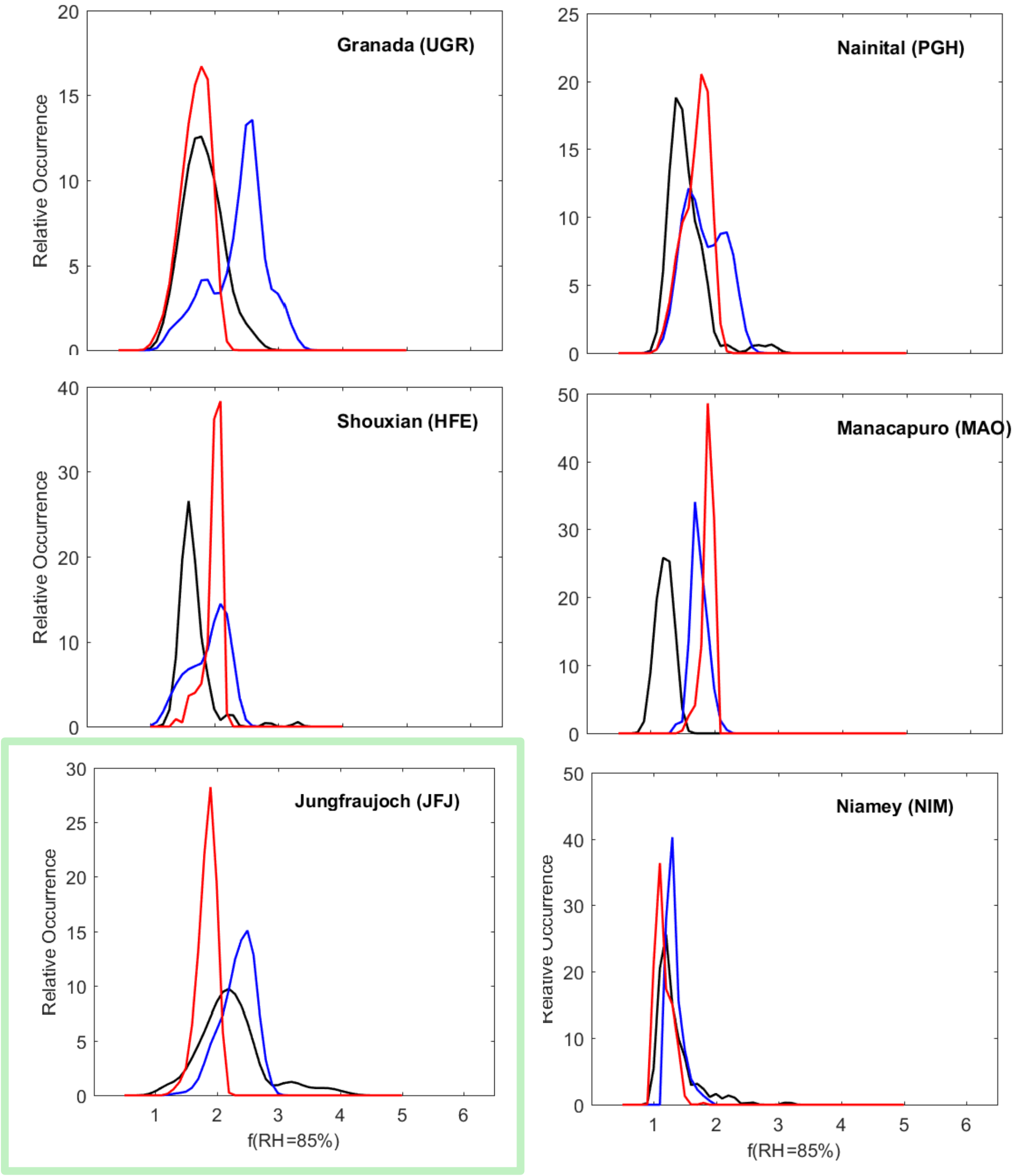
- **Mountain** site (Jungfraujoch):
  Model surface is not the same as measurement surface, so wouldn't expect models to do well necessarily



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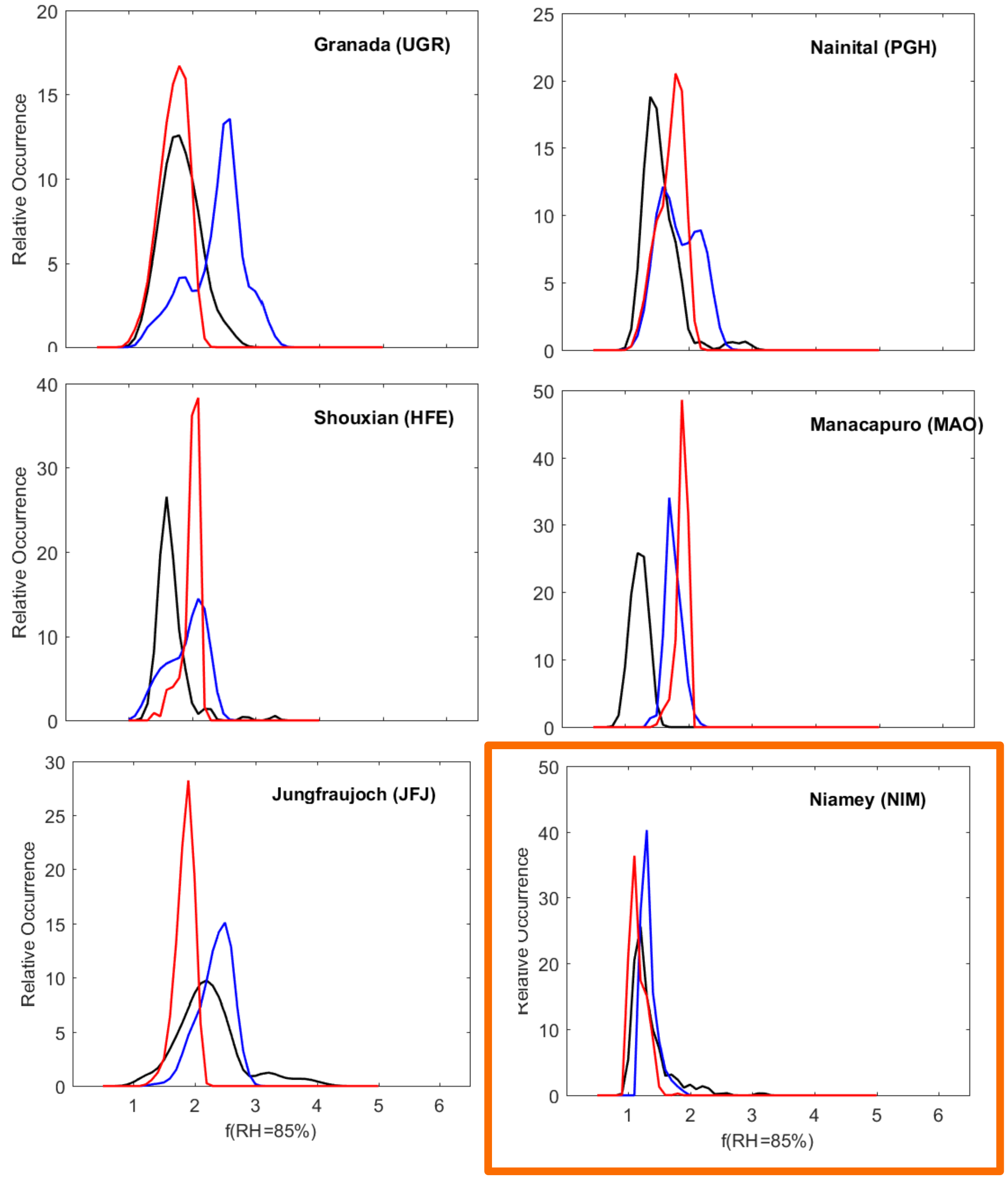
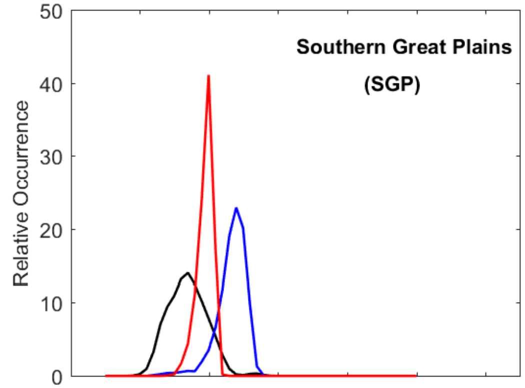
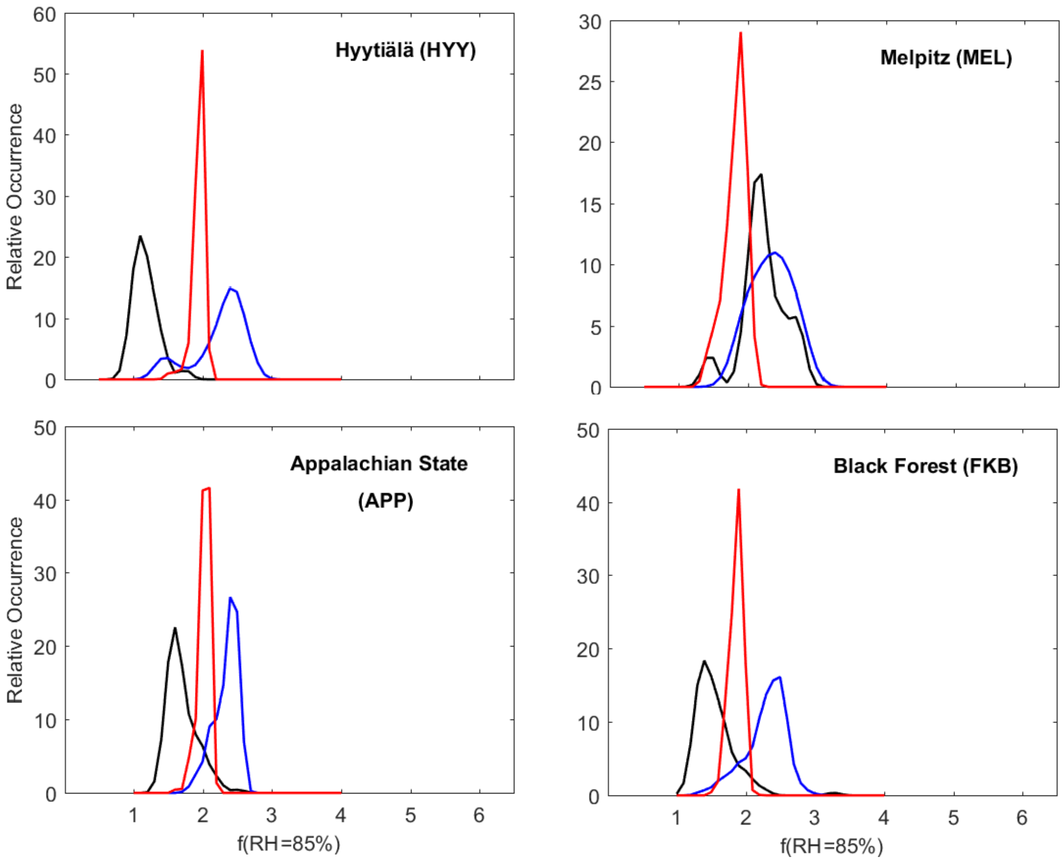
- **Desert** site (Niamey): models reproduce the measurements of *f*(RH) quite well



Rural Sites:

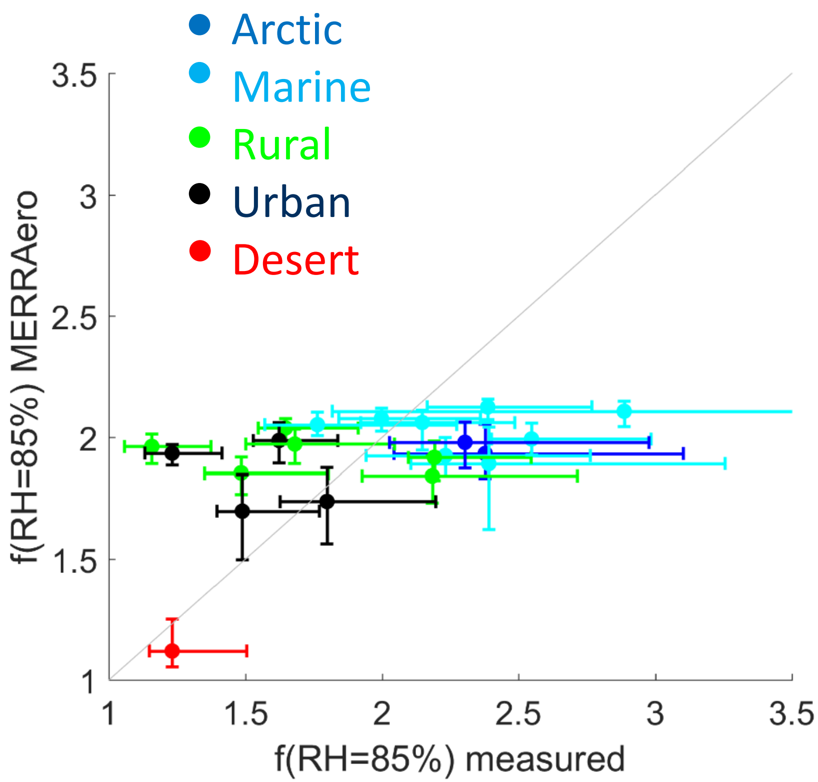


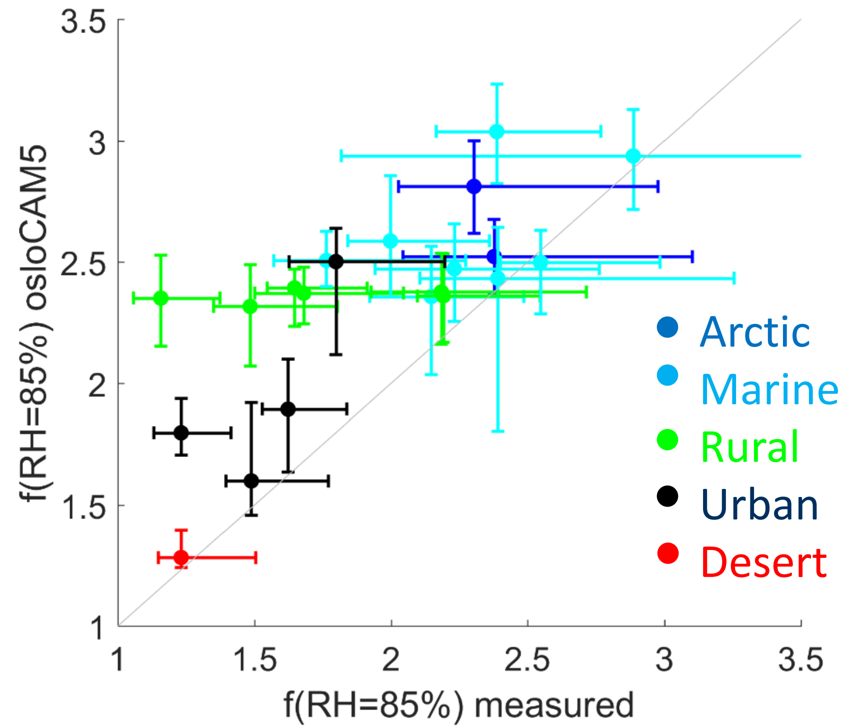


- Each model exhibits consistent **peak** values of *f*(RH=85%):
    - ~2 for MERRAero
    - ~2.5 for CAM5.3-Oslo
- Models systematically **overestimate** *f*(RH=85%) except for Melpitz (MEL), where the measurements peak is shifted towards larger values relative to the other sites

Median Values and 25 and 75 Percentiles

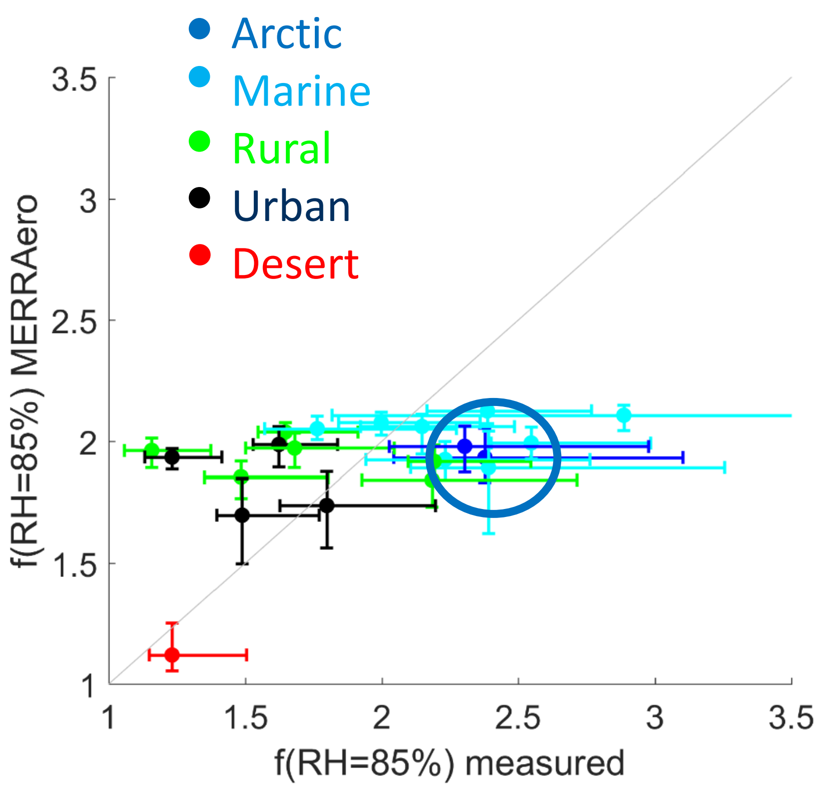
MERRAero CAM5.3-Oslo





Median Values and 25 and 75 Percentiles

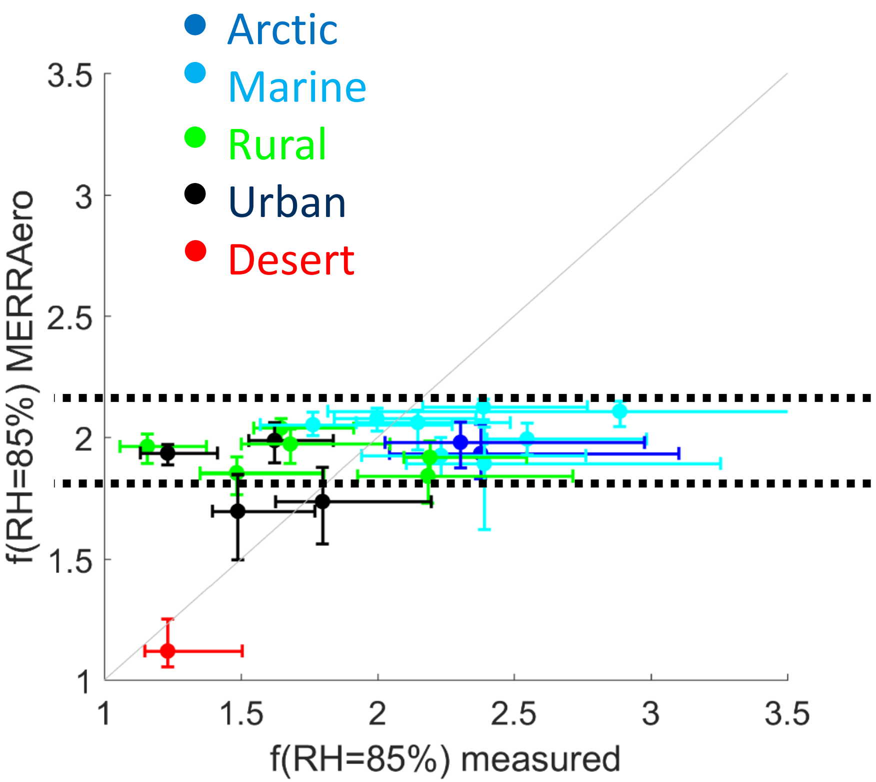
MERRAero



- **Underestimates** *f*(RH=85%) observations for Arctic sites

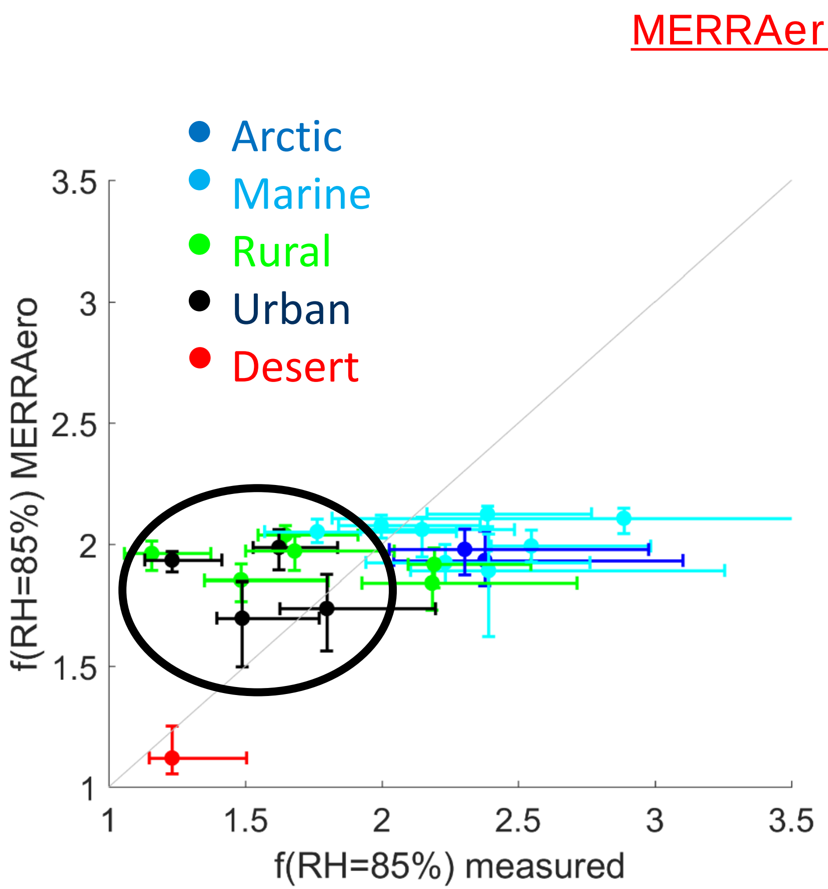
Median Values and 25 and 75 Percentiles

MERRAero



- **Underestimates** *f*(RH=85%) observations for Arctic sites
- Exhibits similar values for most Marine and Rural sites

Median Values and 25 and 75 Percentiles



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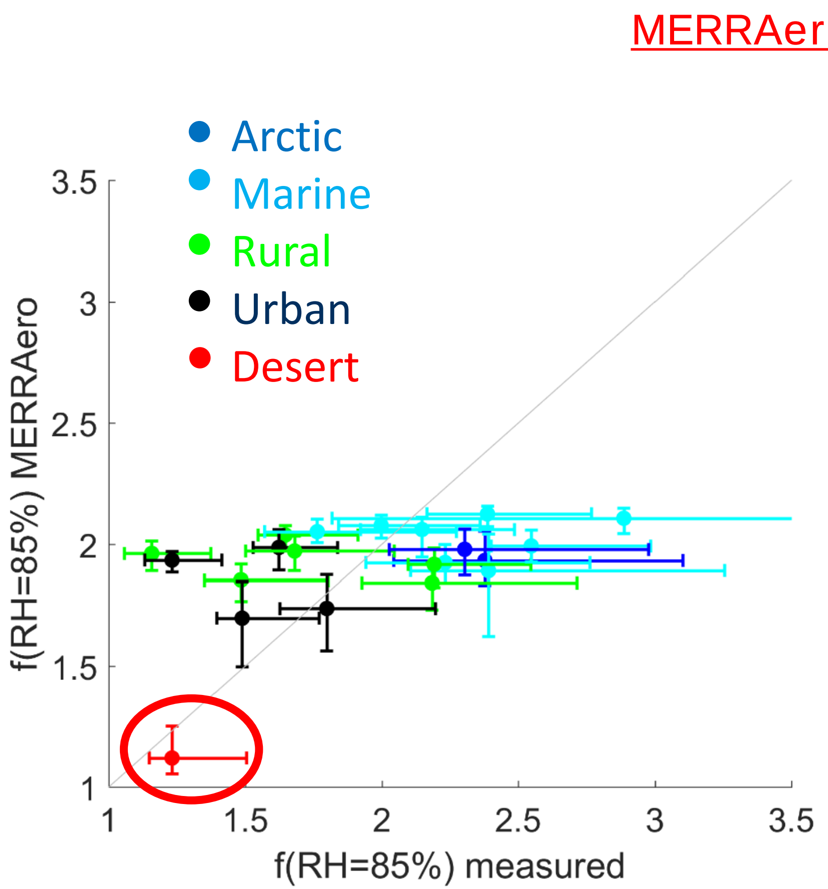
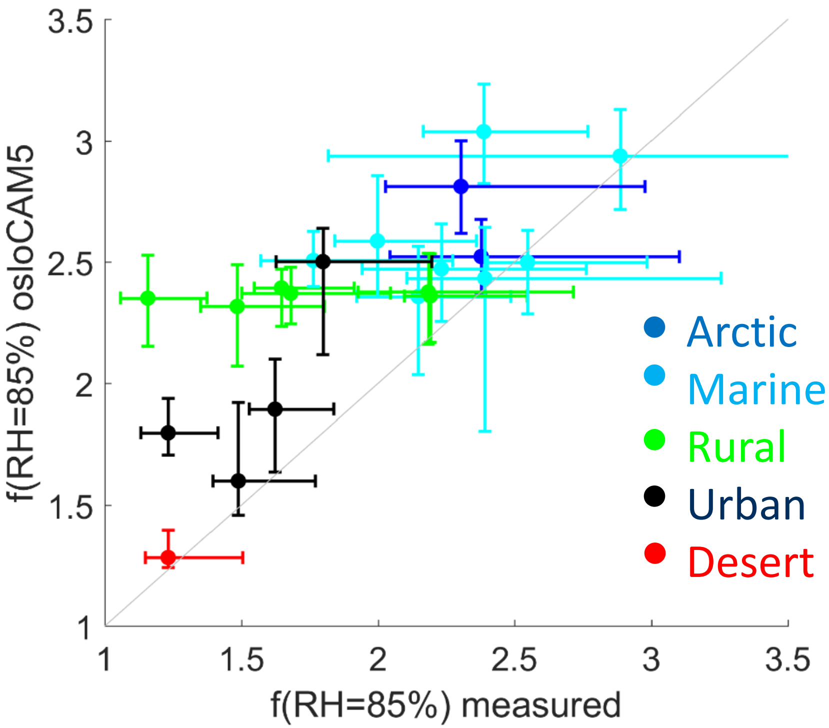


- **Underestimates** *f*(RH=85%) observations for Arctic sites
- Exhibits similar values for most Marine and Rural sites
- Inconsistent for **Urban** sites
- Does well for Desert site

Median Values and 25 and 75 Percentiles

- Overestimates *f*(RH=85%) relative to observations, but better reproduces the diversity of observations

CAM5.3-Oslo



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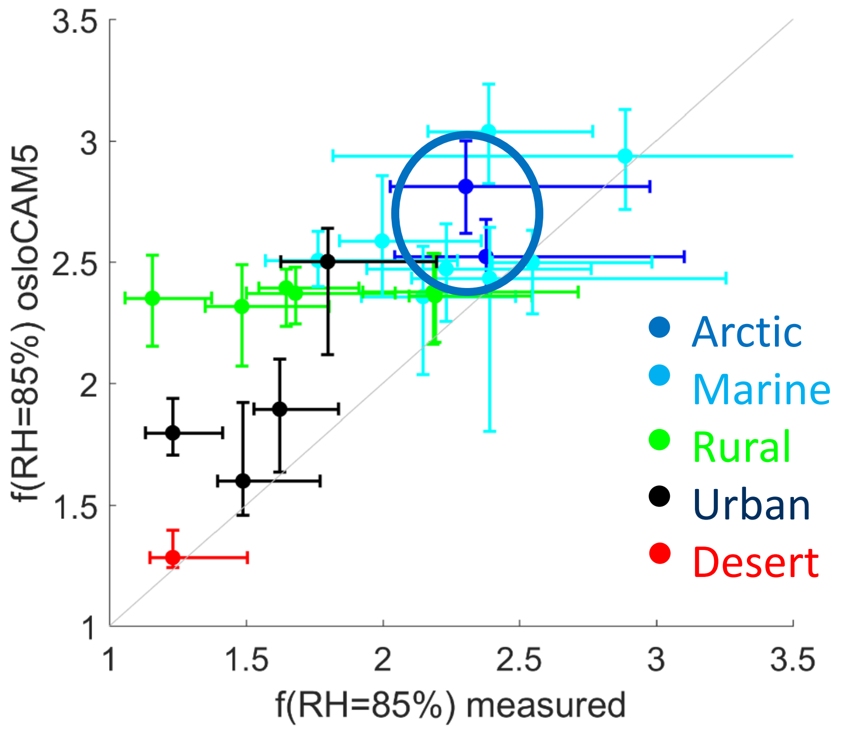
CAM5.3-Oslo



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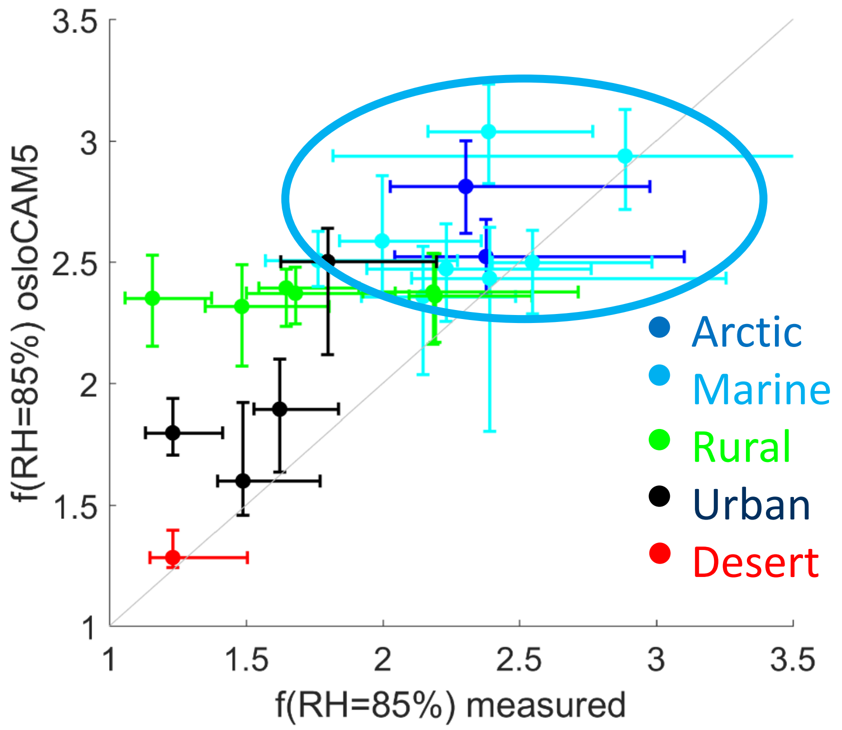
- Reproduces the **diversity** in Marine sites with a general overestimation

CAM5.3-Oslo



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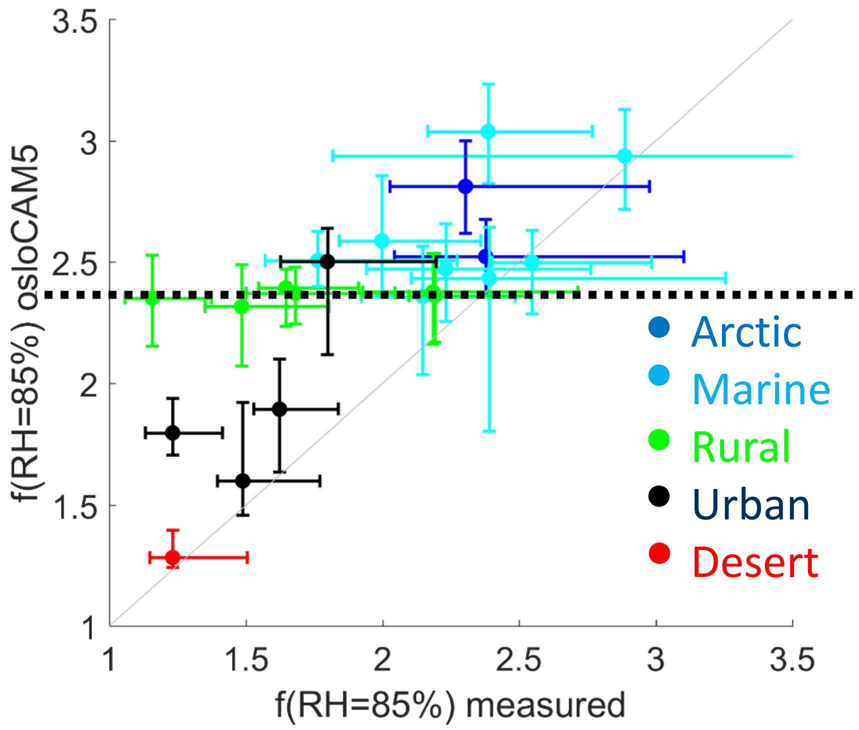
- Exhibits approximately constatn *f*(RH=85%) at Rural sites – does NOT capture observed diversity

CAM5.3-Oslo



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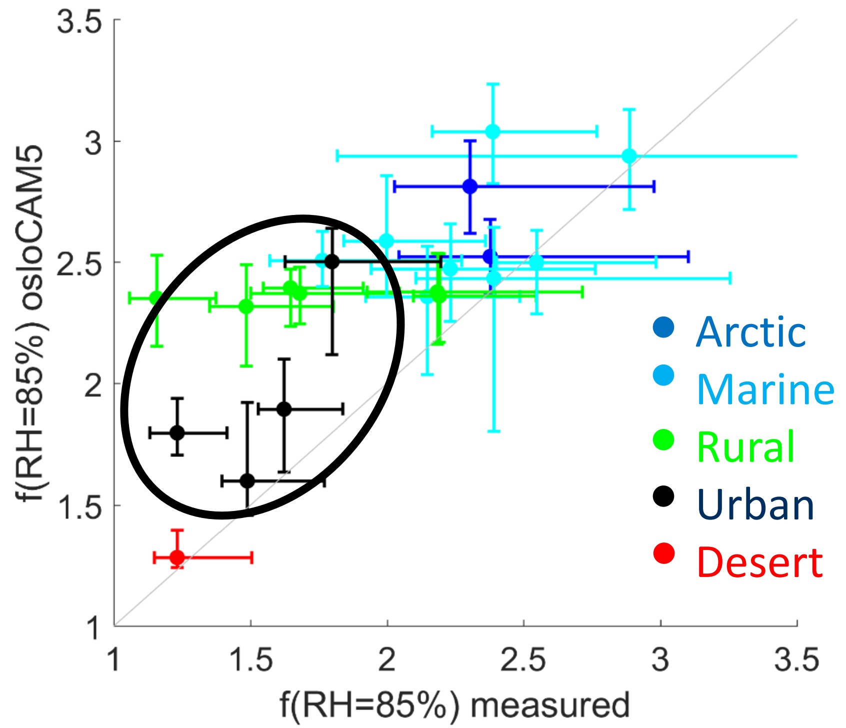
- Inconsistent results for **Urban** sites, with a tendency to overestimate

CAM5.3-Oslo



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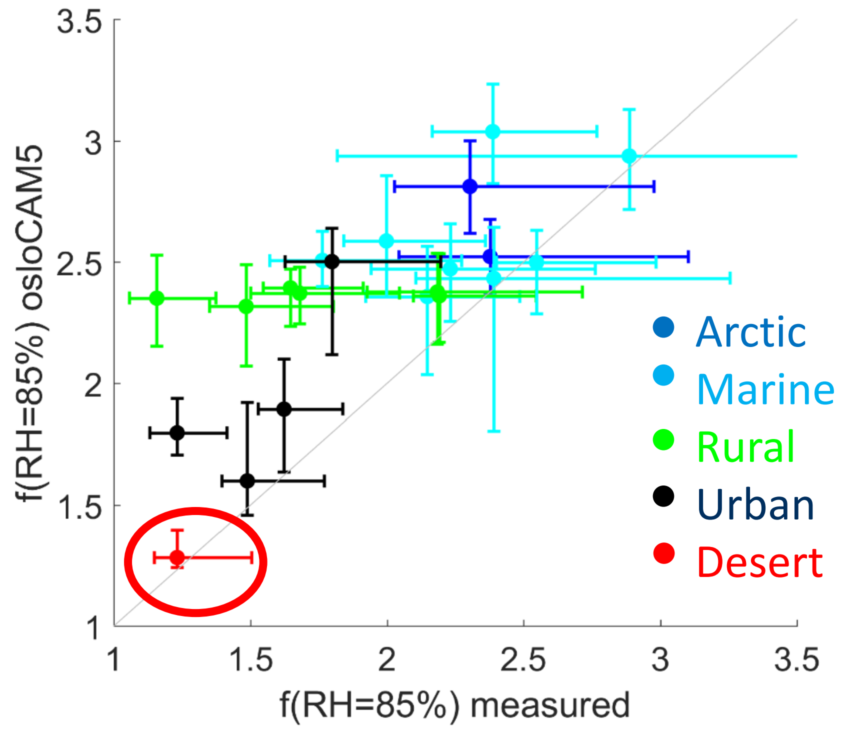
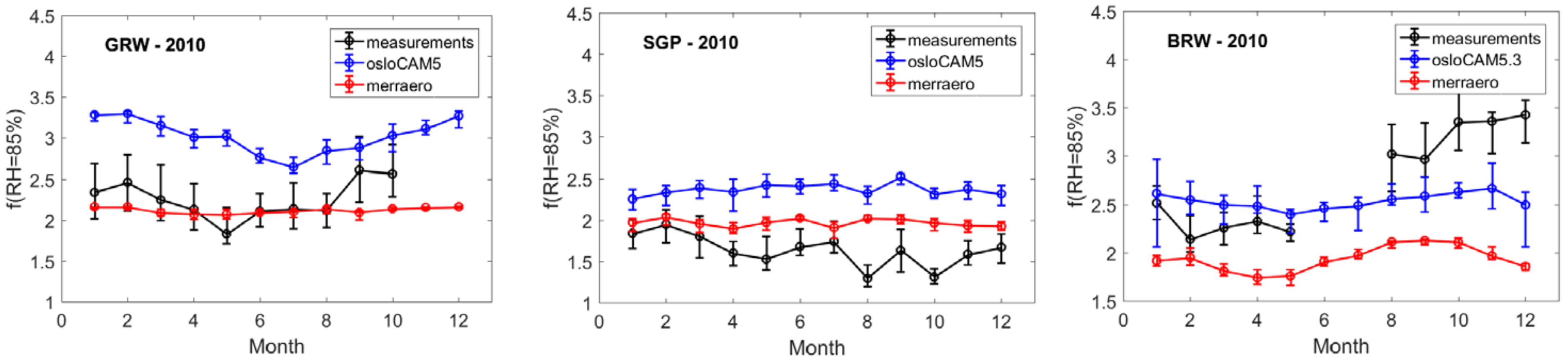
CAM5.3-Oslo



Annual Cycles

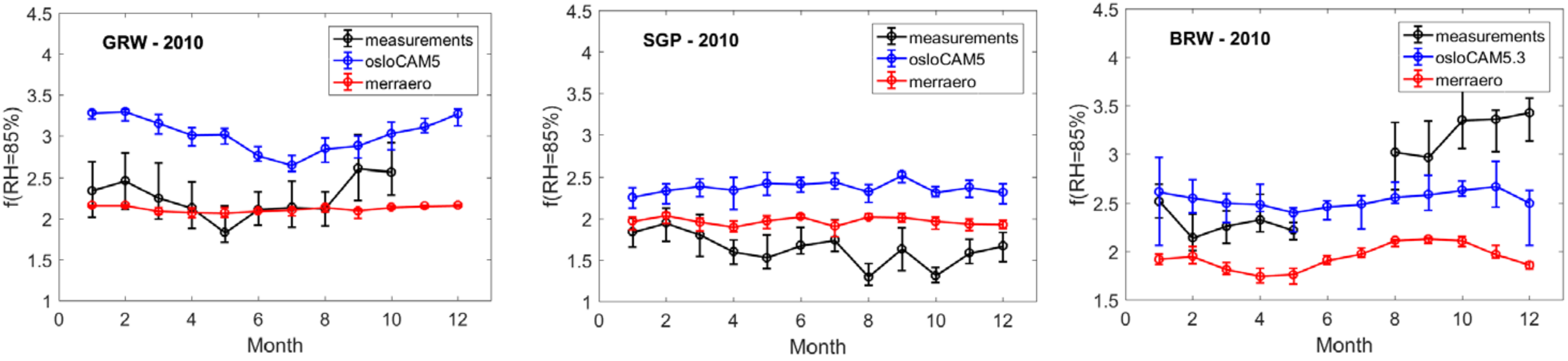


- GRW (Marine):
  - the value of f(RH=85%)=2 simulated by MERRAero is constant throughout the year
  - CAM5.3-Oslo simulates a similar cycle to the observations with a bias towards larger values

- SGP (Rural):
  - both models overestimate f(RH =85%) throughout the year.
  - CAM5.3-Oslo tracks the observed annual cycle better than MERRAero

- BRW (Arctic):
  - Both models track observed annual cycle (higher in autumn, lower in spring)

Annual Cycles



- Differences suggests some seasonal chemistry that models are not reproducing
  - → Possibility to compare model and measurement chemistry at some sites to further assess
  - → Study how number, surface and volume size distributions affect scattering

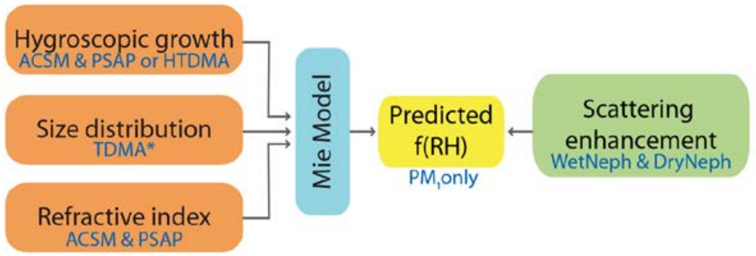
- Re-analysis of data from 26 sites measuring different aerosol types to built a **benchmark, harmonized and reliable database**

- Comparison of *f*(RH=85%) between **measurements** and **model** outputs (MERRAero and CAM5.3-Oslo) highlights that:
  - Constraint values of the model output for several aerosol types
  - Overall, CAM5.3-Oslo reproduces better the variability of measurements while MERRAero present less variability
  - The *f*(RH=85%) values are coincident with measurements for some sites
  - Differences in seasonal chemistry may not be well represented in models

Next Steps...

- Optical closure studies can help to reduce uncertainties (not possible at all sites due to measurement restrictions)

- Study the covariance of aerosol hygroscopic growth with other intensive properties such as SAE or SSA



- Study what is considered a valid definition of "dry RH" and the changes in optical properties at low RH conditions and its implications (Poster Andrews, P02)

We encourage you to provide model data!!

Questionaire to AeroCom modelling community to collect metadata and a description of growth parameterization

Variables requested:

- Aerosol extinction, 550 nm, 40%, 55%, 65%, 75%, 85% RH + ambient
- Aerosol absorption, 550 nm, 40%, 55%, 65%, 75%, 85% RH + ambient
- AOD speciated

Years of simulation/emission:

- 2010
- Optimal: 2000-2014

Please participate!
Description of data request can be found at:

https://wiki.met.no/_media/aerocom/INSITU_AeroComPIII_description.pdf

REFERENCES:
Buchard et al., 2015: Using the OMI aerosol index and absorption aerosol optical depth to evaluate the NASA MERRA Aerosol Reanalysis, Atmos Chem Phys
Chin, M. *et al.,* 2002: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sun photometer measurements, J.A.S.
Fierz-Schmidehauser *et al.,* 2010: Measurements of relative humidity dependent light scattering of aerosols, Atmos meas tech
Hess, M., *et al.,* 1988: Optical Properties of Aerosols and Clouds: The Software Package OPAC, A.M.S.
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Randles, C. A. *et al.,* 2013: Direct and semi-direct aerosol effects in the NASA GEOS-5 AGCM: aerosol-climate interactions due to prognostic versus prescribed aerosols, J.G.R.
Tang, I. N. *et al.,* 1997: Thermodynamic and optical properties ofvmixed-salt aerosols of atmospheric importance, J.G.R.
Titos et al., 2016: Effect of hygroscopic growth on the aerosol light-scattering coefficient: A review of measurements, techniques and error sources, Atmos environ
Zieger et al., 2010: Effect of relative humidity on aerosol light scattering in the Arctic, Atmos Chem Phys
Zieger *et al.,* 2017: Revising the hygroscopicity of inorganic sea salt particles, Nature Communications.

THAT YOU for your ATTENTION!

BackUp slides

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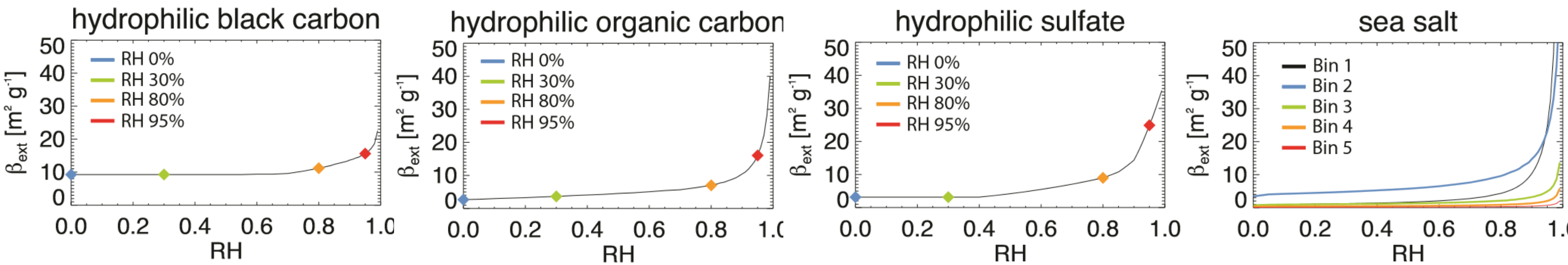
MERRA Aerosol Reanalysis (MERRAero):

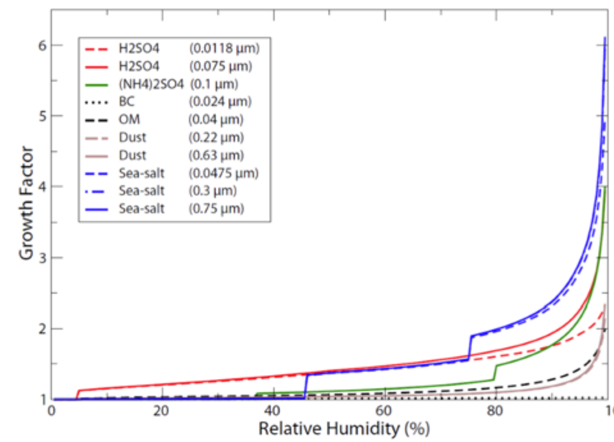
Implementation of hygroscopic growth (Randles, C. A. *et al.,* 2013):

- Carbonaceous species and sulfate: parameterized based on **OPAC** (Hess *et al.,* 1998) as in Chin *et al.* (2002)
- Sea salt: parameterized based on observations of mixed-salt aerosol growth from ***Tang et al.,* (1997)**



CAM5.3-Oslo

- **Hygroscopic growth factors** for aerosol components at some typical dry radii and for relative humidities up to RHmax = 99.5%



Checking the time series of BRW for the measurements

Wet/dry

N.data(2009)=[0,0,0,0,0,0,0,0,5,13,33,11]

N.data(2010)=[51,78,39,17,0,1,36,28,32,19,52]

N.data(2011)=[24,23,34,24,3,0,0,0,19,0,3,73]

N.data(2012)=[34,63,63,0,11,11,5,4,15,26,19,60]

N.data(2013)=[92,33,75,54,9,6,8,5,14,2,0,0]

85%/40%

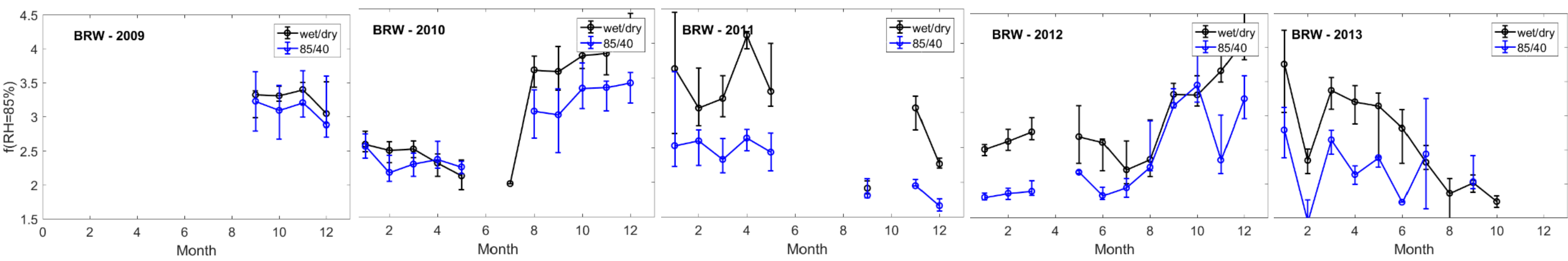
N.data(2009)=[0,0,0,0,0,0,0,0,2,5,16,9]

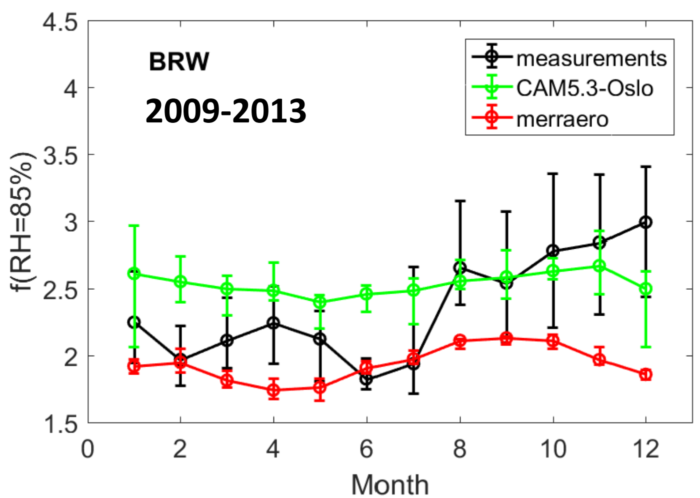
N.data(2010)=[19,43,58,15,10,0,0,20,19,19,16,49]

N.data(2011)=[21,20,25,24,2,0,0,0,5,0,3,25]

N.data(2012)=[19,34,41,0,2,4,2,3,5,4,12,40]

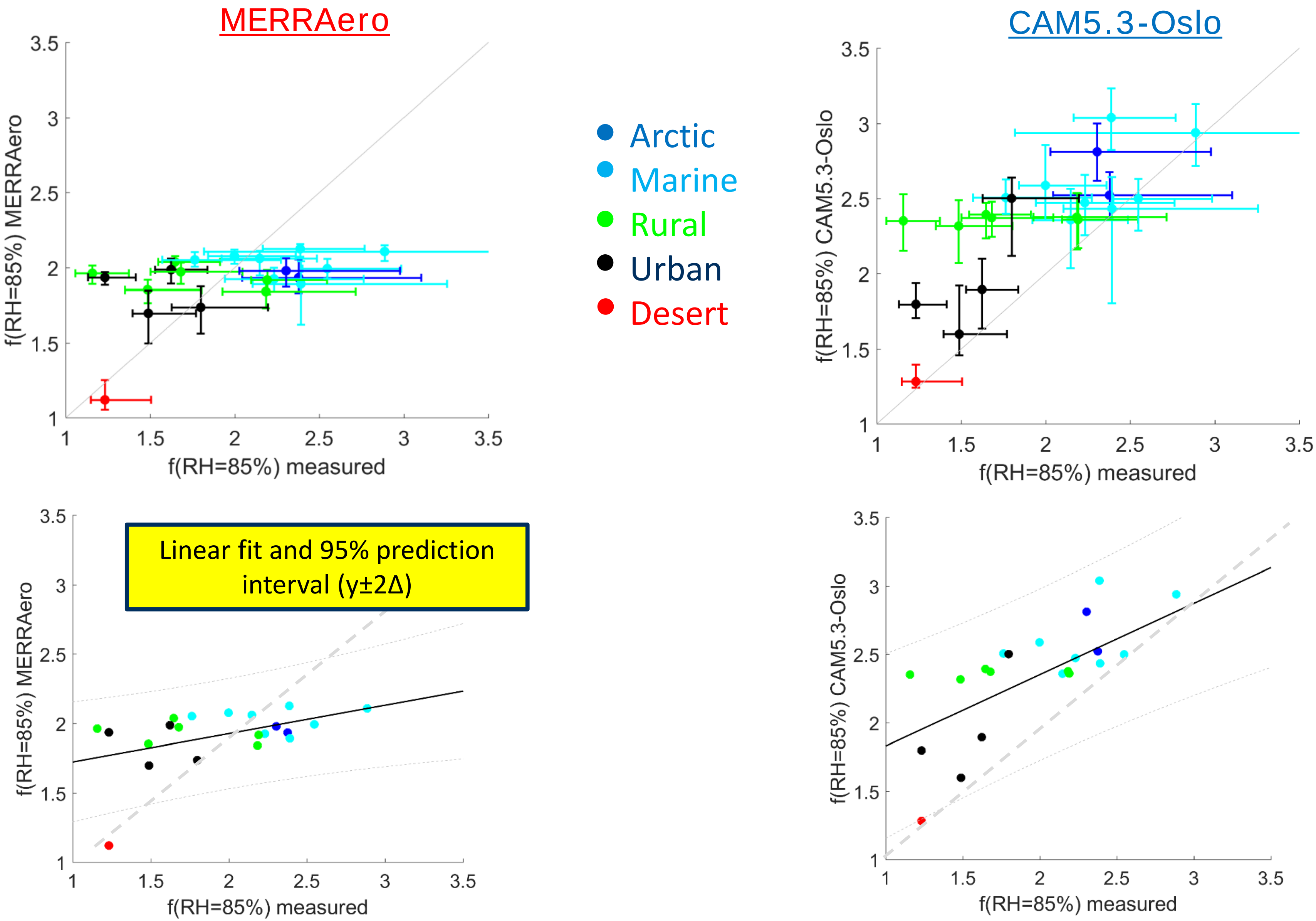
N.data(2013)=[36,12,50,40,6,1,2,0,3,0,0,0]



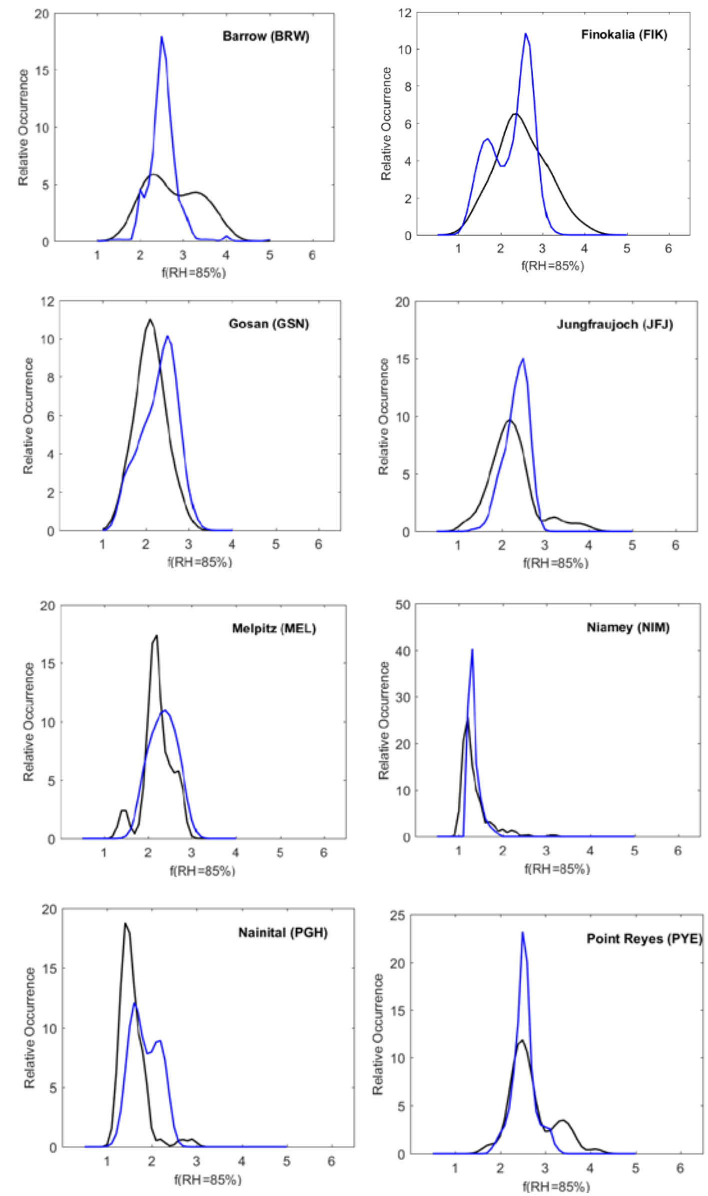


Stockholm University

Median Values and Percentiles 25 and 75



SITE	Measurements	CAM5.3-Oslo
Appalachian	1.7±0.4	2.3±0.2
Barrow	**2.4±0.6**	**2.5±0.3**
Cabauw	2.2±0.6	2.5±0.3
Finokalia	**2.5±0.6**	**2.3±0.5**
Black Forest	1.5±0.4	2.3±0.3
Graciosa	2.3±0.6	3.0±0.3
Gosan	**2.1±0.4**	**2.3±0.4**
Shouxian	1.6±0.3	1.9±0.3
Hyytiälä	1.2±0.3	2.3±0.3
Jungfraujoch	**2.3±0.8**	**2.3±0.3**
Manacapuro	1.2±0.1	1.8±0.2
Mace Head	2.5±1.0	2.9±0.3
Melpitz	**2.3±0.5**	**2.3±0.3**
Niamey	**1.3±0.5**	**1.3±0.1**
Nainital	**1.5±0.4**	**1.7±0.3**
Cape Cod	1.9±0.5	2.5±0.2
Point Reyes	**2.6±0.7**	**2.5±0.3**
Southern Great Plains	1.7±0.6	2.3±0.2
Trinidad Head	2.0±0.7	2.6±0.4
Granada	1.8±0.4	2.4±0.4
Zeppelin	2.5±1.3	2.8±0.3



SITE	Measurements	MERRAero
Appalachian	1.7±0.4	2.0±0.1
Barrow	2.4±0.6	2.0±0.1
Cabauw	**2.2±0.6**	**1.9±0.1**
Finokalia	2.5±0.6	1.9±0.2
Black Forest	1.5±0.4	1.9±0.1
Graciosa	**2.3±0.6**	**2.1±0.1**
Gosan	**2.1±0.4**	**2.1±0.1**
Shouxian	1.6±0.3	2.0±0.1
Hyytiälä	1.2±0.3	2.0±0.1
Jungfraujoch	2.3±0.8	1.8±0.1
Manacapuro	1.2±0.1	1.9±0.1
Mace Head	2.5±1.0	2.1±0.1
Melpitz	2.3±0.5	1.9±0.1
Niamey	**1.3±0.5**	**1.2±0.1**
Nainital	**1.5±0.4**	**1.7±0.2**
Cape Cod	**1.9±0.5**	**2.1±0.1**
Point Reyes	2.6±0.7	2.0±0.1
Southern Great Plains	1.7±0.6	2.0±0.1
Trinidad Head	**2.0±0.7**	**2.1±0.1**
Granada	**1.8±0.4**	**1.7±0.2**
Zeppelin	2.5±1.3	2.0±0.1