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

- Ambient aerosol particles experience hygroscopic growth at enhanced relative humidity (RH)
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- Aerosol **particle light scattering** is strongly dependent on RH
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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

\[ f(RH, \lambda) = \frac{\sigma_{sp}(RH, \lambda)}{\sigma_{sp}(RH_{dry}, \lambda)} \]

with \( \lambda \): wavelength, \( \sigma_{sp} \): 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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\( f(RH) \) can be measured using humidified nephelometer systems

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Source: Zieger et al. (2013)
Introduction

Experimental: Scattering enhancement

Model-measurement comparison

Conclusions

Outlook

The humidified nephelometer (WetNeph)

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

Scattering enhancement factor

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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

Example humidogram from Cabauw for maritime air (Zieger et al., 2011)
The dry reference scattering coefficient: What is dry?

A significant bias could be introduced by insufficient drying of aerosols

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

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).
The new benchmark dataset of scattering enhancement

- Standardized re-analysis of 26 datasets (mostly DoE and ACTRIS) of RH-dependent scattering and backscattering coefficients, $f(RH)$, $f_b(RH)$
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- Harmonized dataset openly available + data descriptor paper

Temporal data coverage of re-analysed sites.
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Overview of re-analysed sites with mean values of $f(RH=85%/RH_{dry})$ for PM$_1$/PM$_{2.5}$ (left triangles) and 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 $f(\text{RH}=85\%)$ at $\lambda = 550\text{ nm}$ segregated by single scattering albedo (SSA). Source: Titos et al. (2019, in prep.)
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- Most sites show increased $f(\text{RH})$ for less absorbing aerosol
Boxplot of $f(RH=85\%)$ at $\lambda = 550\,\text{nm}$ segregated by single scattering albedo (SSA). Source: Titos et al. (2019, in prep.)

- 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 $\lambda = 550$ 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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- Monthly average (note: only 3 sites are co-located in time for 2010)
- 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 $f(RH)$ for Southern Great Plains with $RH_{\text{dry}} = 0\%$ as reference. Measurements are shown at actual measured RH.
Example for 2010 (co-located in time): Southern Great Plains

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

- Comparison at $RH_{dry} = 40\%$ 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 $f(RH)$ for Barrow with $RH_{dry} = 0\%$ as reference (measurements not corrected).

- Measurements in Barrow should be less affected by remaining water (lower $RH_{dry}$).

Modelled and measured $f(RH)$ for Barrow with $RH_{dry} = 40\%$ as reference (measurements interpolated).
Example for 2010 (co-located in time): Barrow / North Slope of Alaska

![Graphs showing modelled and measured $f(RH)$ for Barrow with RH$_{dry}$ = 0% and RH$_{dry}$ = 40% as reference.]

- Measurements in Barrow should be less affected by remaining water (lower RH$_{dry}$).
- Some models show large change in $f(RH)$ if RH$_{dry}$ = 0% or RH$_{dry}$ = 40% is taken as reference.
**Example for 2010 (co-located in time): Graciosa**

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

Modelled and measured $f(RH)$ for Graciosa with $RH_{dry} = 40\%$ as reference (measurements interpolated).
Example for 2010 (co-located in time): Graciosa

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- Models for GRW, SGP and BRW and 2010 are usually higher than measurements
Example for 2010 (co-located in time): Graciosa

- Models for GRW, SGP and BRW and 2010 are usually **higher** than measurements
- Models show a **large site-specific diversity**
Comparison of entire data set with 2010 model data

Comparison of the entire dataset for Barrow (North Slope of Alaska), Southern Great Plains, Graciosa and Niamey.
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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)
Comparison of entire data set with 2010 model data

- Some models correlate with measurements better than others

![Graphs showing comparison between model predictions and measurements](image-url)
Comparison of entire data set with 2010 model data

- Some models correlate with measurements better than others
- Models mainly over-estimate $f(\text{RH})$
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- Caution: Airmass-specific and temporal characteristics are masked out but can still be significant
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- More details at the poster!
Conclusions

- The **new benchmark dataset** of RH-dependent particle light scattering coefficients and scattering enhancement factors $f(RH)$ has been **finalized** and **successfully tested against** six GCM’s.
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- Models show a **large diversity** in $f(\text{RH})$ with respect to magnitude and temporal evolution (e.g. seasons).
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• Models show a **large diversity** in $f(\text{RH})$ with respect to magnitude and temporal evolution (e.g. seasons). **Reasons are manifold**: differences in model parameterizations of e.g. hygroscopicity, size, sources + strength, mixing state, removal processes, etc.
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- 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 $\kappa$-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?  
  - $f(\text{RH})$ 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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Acknowledgements: To all co-authors and data providers of the data descriptor paper. This work is funded by the U.S. Department of Energy (contract no. DE-FO-0001430).

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?

Modal sizes and relative amounts of ...
What determines the scattering enhancement?

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

Reference:

Modal sizes and relative amounts of...

- Accumulation mode
- Coarse mode

- BC or organics
  - very low to low f(RH)
- Inorganic salts
  - high f(RH)
- Sea salt
  - high f(RH)
- Mineral dust
  - very low f(RH)
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 $f$(RH) with RH$_{dry}$ = 0 % as reference.

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

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


