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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Subgrid-scale variability of clear-sky relative humidity and forcing by aerosol-radiation interactions in an atmosphere model" *by* Paul Petersik et al.

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Response to Reviewer 2

We thank the reviewer for his/her constructive comments and for his/her thorough review. The reviewer comments are in plain font, the responses in *Italics*.

General comments

This study applied a stochastic parameterization of subgrid-scale variability of relative humidity (RH) to a global climate-aerosol model, ECHAM6-HAM2, and examined the impact of the subgrid-scale variability of RH on aerosol optical depth (AOD) and radiative forcing. The authors showed the subgrid-scale variability of RH increased global mean aerosol hygroscopic growth, AOD (by 7.8%), and effective radiative forcing (by 57%) due to the non-linear response of hygroscopic growth to RH. Although this study showed a slight improvement of the estimation of AOD, I don't think this study is suitable for a paper of Atmospheric Chemistry and Physics because the scientific findings, methods, and analysis of this study are not enough as shown below.

To emphasize the scientific importance of applying a stochastic parameterization to clear-sky relative humidity for its application in the aerosol hygroscopic growth scheme we now highlight better in the revised manuscript that our study is the first study that is proposed without strong simplifications about the shape of the used probability density function (PDF) and that is consistent with the cloud cover scheme. This is discussed in more detail further down in this authors response.
Furthermore, we also emphasize in the revision the point that applying a stochastic parameterization is not only a method to estimate uncertainties but leads to a better representation of the mean state of the atmosphere. This was recently summarized in Berner et al. (2017). To highlight this in an example we refer to Tompkins and Berner (2008) that use a method of subgrid-scale variability that is very similar to ours. They investigate its influence when it is applied on the convective scheme of the European Centre for Medium-Range Weather Forecasts (ECMWF) ensemble prediction system. They show that their new stochastic convective scheme generally improves the skill of the operational system for most variables in the short to medium range in mid-latitudes.

Main comments

1) Page 2, lines 7-27
These two paragraphs describe about previous studies. I understand from these paragraphs that the underestimation of radiative forcing by using the grid-box mean RH is

already recognized well in previous studies. In addition, there are some global model studies focused on the subgrid-scale variability of RH previously. Due to these two points, it is hard to understand what was advanced scientifically in this study. This study is new in ECHAM6-HAM2, but I feel that there is no clear advancement both scientifically and technically in the community of aerosol and cloud studies.

We gather that the reviewer refers to our discussion of the studies of Haywood and Shine (1997) and Haywood and Ramaswamy (1998) which apply a subgrid-scale variability of RH in a GCM. We regret that in the previous manuscript version obviously we did not clarify well enough that our study goes substantially beyond this previous work.

1. Haywood and Shine (1997) investigated the effect of subgrid-scale variability in an idealized case. They use globally for each grid cell and height level five fixed RH-values that are taken from a normal distribution around RH = 70%. Hence, they show the gross effect of subgrid-scale variability of RH but do not propose a scheme that is meant to be integrated into an atmosphere model.

2. Haywood and Ramaswamy (1998) use a more sophisticated approach by computing the subgrid-scale variability based on a triangular shaped distribution around grid-box mean RH. However, the shape and width of the distribution is globally constant. In our parameterization the width of the PDF is a function of height (Quaas, 2012). Furthermore, the PDF that Haywood and Ramaswamy implement is artificially generated and inconsistent with the assumptions in the cloud scheme. In contrast, we sample the sub-saturated part of the PDF from the cloud-cover scheme.

Haywood and Shine (1997) and Haywood and Ramaswamy (1998) have in common that they just investigate the effects on RFari by sulphate. We investigate the effect on the entire radiative forcing of aerosols that are included in the ECHAM6-HAM2 model. Summarizing, our study is the first study that investigated the effect of subgrid-scale variability in an approach that does not make idealized assumptions and that is consistent with the cloud cover scheme.

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In response to the reviewer comment, the sentence: "Haywood and Shine (1997) and Haywood and Ramaswamy (1998) include a subgrid-scale variability of RH for the calculation of RFari by sulphate."
in lines 17f of the original manuscript has been amended as follows:
"First attempts to implement a subgrid-scale variability of RH in a GCM for the calculation of RFari by sulphate were made by Haywood and Shine (1997) and Haywood and Ramaswamy (1998). However, these studies make strong simplifications about the shape of the used probability density function (PDF) and are not consistent with the cloud cover scheme."

2) Section 2.2
Please explain why the stochastic treatment was used. Does this mean only single RH value is calculated by Eq. (6) and used in each grid box and each time? I think considering the full range of RH (shown in Figure 1) in each grid box and time is not so difficult, for example by using 5-10 RH bins between RHcls - delta RHcls and RHcls + delta RHcls. This will not increase the computational cost of the model so much. If a random RH is used in the model, does it assure the repeatability of model simulations? For example, when the authors make two simulations which use completely the same inputs and model setups, can the authors obtain the same results from the two simulations?

At each time step and each grid box we apply Eq. (6) using a newly generated random number. This is computationally cheaper than subsampling the entire PDF in each time step while on average one expects a very similar result. It should be noted that our scheme is intended for use in a 3-D climate model and not just constructed to show that taking into account subgrid scale variability decreases ERFari. Applying a binning approach for the RH values means, in other words, increasing the resolution of the model for the hygroscopic growth scheme. This results in additional computational

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cost compared with the approach suggested here.
Up until now, we used a random number generator that starts always with a new seed. In general it would be possible to start always with the same seed. The repeatability of our study is ensured by integrating the model over a rather long time (10 years). Hence, we expect to find results that do not differ from our results larger than within the given error bars (95%). In the current setting, the integration is not fully deterministic anymore. We clarify this now in the revised text.

3) Treatment of aerosol absorption
How does the model calculate aerosol absorption? Please describe the method and the treatment of absorption enhancement by water. The treatment of absorption enhancement of black carbon by water will be a key in the calculations of single scattering albedo and radiative forcings. The authors show negative values of radiative forcings, but I suspect the authors do not consider the positive forcings by the absorption enhancement. The absolute values of radiative forcings will be smaller when the absorption enhancement is treated properly, and the total effect of the subgrid-scale variability of RH will be less important.

Aerosol radiative properties are calculated using Mie theory. The model uses volume-averaging for each of the seven aerosol modes to calculate the refractive indices where aerosol water is included using the ambient relative humidity. The effective complex radiative indices and the Mie size parameter is then used for the aerosol radiative properties, namely extinction cross section, single scattering albedo, and asymmetry parameter in the radiation scheme (see Zhang et al. (2012) section 2.6). The explanation is now extended in the revised manuscript.
For the version 2 of the aerosol module HAM the refractive indices for black carbon were updated to reduce the negative bias aerosol absorption enhancement (Stier et al., 2007). Based on findings of Stier et al. (2006) it is argued in Stier et al. (2007)

that the absorption enhancement of BC due to mixing with hydrophilic aerosols is compensated by the lower life time of and abundance of BC.
In contrast to Jacobson (2012) and Bond et al. (2013), HAM2 does not include a very strong absorption enhancement for absorbing particles inside clouds. This is because the hypothesis of Jacobson (2012) is very controversial and not supported by most other studies (e.g. Twohy et al., 1989; Chýlek et al., 1996; Liu et al., 2002).
We include values for the AAOD and AAOD of BC. In PD simulations AAOD increases by +0.12 ($\pm$0.4) $\cdot$ 10^{-3} ($\sim$ 4.7%) were the AAOD by BC increased by +0.11 ($\pm$0.3) $\cdot$ 10^{-3} ($\sim$5.1%). This shows that our parameterization leads to a stronger absorption of solar light by BC aerosols. However, the overall increase of AOD in PD simulations due to our new parameterization is +9.0 ($\pm$2.2) $\cdot$ 10^{-3} ($\sim$7.8%). That highlights that in our simulations the contribution of absorption to AOD is rather low although absorption is enhanced.
For point 3, the reviewers concerns are to our understanding mostly based on our insufficient description of the aerosol-module HAM2. Therefore, we include a new subsection, "2.1 The aerosol module HAM2" into our methods that briefly summarizes the properties of HAM2.

Other comments

Page 1, lines 23-24:
I think Ginoux [2017] is for mineral dust only. References for primary and secondary organic aerosols and aerosols from biomass burning should be added.

We included Bond et al.(2013) and Myhre et al. (2013) for biomass burning, Shindell et al. (2013) for SOA as well as a reference to the AR5 from the IPCC.

Page 2, line 7:
"However" is better to move to the next sentence (before "General circulation").

Changed.

Page 2, lines 34-35:
I cannot understand what the authors mean in this sentence ("RHcls is chosen. . .") and next sentence ("That means, . . ."). I don't think "aerosol-radiation interactions are negligible in the cloud part". Some studies (e.g. Jacobson) have shown the importance of this issue.

In the standard configuration ECHAM6-HAM2 uses the mean clear-sky relative humidity ($\overline{RH}_{\text{cls}}$) instead of the grid-box mean relative humidity ($\overline{RH}$) to compute aerosol hygroscopic growth. Note, that $\overline{RH}_{\text{cls}}$ is by definition smaller than $\overline{RH}$:

$$\overline{RH} = f \cdot \overline{RH}_{\text{cloud}} + (1-f) \cdot \overline{RH}_{\text{cls}} = f + (1-f) \cdot \overline{RH}_{\text{cls}}$$

Here, f is the fractional cloud cover. It is assumed that the radiative effect of the hygroscopic growth of aerosol is more important in the cloud-free part than for interstitial aerosol in clouds where cloud radiative effects are dominant. If not RH_{cls} but $\overline{RH}$ would be used to calculate the aerosol hygroscopic growth for the entire grid-box, aerosols would grow to strong in the cloud free part where aerosol-radiation interactions are of higher importance than in the cloudy sky. Thus, it is reasoned in the ECHAM literature to use RH_{cls} instead $\overline{RH}$ (see section 2.6 in Stier et al., 2005).
We added to the method section:
"RH_{cls} is chosen, instead of grid-box mean relative humidity RH, because cloud processing and cloud radiative effects are dominant in the cloudy part of a grid box as reasoned in Stier et al. (2005) for ECHAM5-HAM1."

Page 3, line 28:
I don't think "age due to internal mixing" is good explanation. Did the authors mean that internally-mixed particles are made by aging processes such as condensation and coagulation?

Yes, that is what we meant. We changed the sentence to:
"DU and BC are considered as non-hygroscopic on emission. However, they can merge with hygroscopic particles due to internal mixing by ageing processes such as condensation and coagulation."

Page 4, line 15:
Because the authors used "usually" here, it looks there are some previous studies considering the subgrid-scale variability.

We just want to highlight that the idea of subgrid-scale variability is not new to model formulations (e.g. vertical velocity) but was not applied (besides in idealized studies as mentioned further up) in global circulation models to RH or RH_{cls}*. We deleted the "usually" changed the sentence to:*
"Several global atmosphere models including ECHAM6-HAM2 already make assumptions to account for the subgrid-scale variability of atmospheric variables, e.g. for vertical velocity when computing droplet activation rates (Ghan et al., 1997; Lohmann et al., 2007; Golaz et al., 2011). However, subgrid-scale variability of RH or RH_{cls} *is not taken into account when computing hygroscopic growth of interstitial aerosols besides in some studies that made gross simplification regarding the shape and variation of the used PDF (Haywood and Shine, 1997; Haywood and Ramaswamy 1998)."*

Page 4, Line 25:

The values of cs, ct, and nx should be given after the equation (2) (at line 8).

Changed.

Page 5, line 2:
What is “Rhcls,old”?

We changed the expression to $\overline{RH}_{\text{cls}}$*. This variable is used earlier to indicate the grid-box mean clear-sky relative humidity that was actually meant by* $RH_{\text{cls,old}}$*.*

Page 6, line 6:
“c” values (Equation (9)) are not useful in the current manuscript. Discuss more or remove from the manuscript.

We agree with the referee and removed them from the manuscript.

Page 6, line 13:
How important the different growth factors between CS/AS and KS/NS modes is?

In Figure 2a one can see that particles swell stronger for bigger aerosol modes due to the implementation of the new parameterization. For more clarity, we changed the sentence:
“Thus, the effect is stronger for particles from CS and AS mode than for particles from KS and NS mode. “
to:
“Thus, the effect is strongest for particles from the CS mode (red line in Fig. 2a) and

weakest for particles from the NS mode (black line in Fig. 2a)"

Page 6, line 18:
Why did AOD increase especially in the tropics?

Our parameterization leads to an on average stronger growth of hygroscopic aerosols, especially for very hygroscopic aerosols that are sulphate and sea salt.
Sea salt is most abundant in lower latitudes in the model. This is indicated in Figure 1 that we attached to this response. Depicted is the AOD by non-hydrated sea salt aerosols. Furthermore, anthropogenic emissions of sulphate are very high in China, India and over the Arab Peninsula and contribute in addition to the peak of increased AOD in the northern tropics.

Page 6, line 25:
Why don't you show the results of "c" by using a figure?

We excluded the c-value completely from the paper in response to the previous reviewer comment on "Page 6, line 6".

Page 6, line 29:
"w" means single scattering albedo?

Yes. But we use term "ratio of scattering to total extinction" when we refer to results from the radiation transfer equation to compute the ratio between scattering/extinction. We do this in order to make the mentioned variable clearly distinguishable from the single scattering albedo as a property of a certain particle type (that is constant).

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Page 6, line 32:
The alpha change shown here is for wet particles? Please clarify. Please show the percentage of the change.

Yes it is the parameter for wet particles. We added this and the percentage of this change.

Page 7, line 14:
Did the authors show the definition of ERFaer?

We added a description of how ERFaer was computed.

What is the difference between ERFari-cls and ERFaer?

In response to the comments of Steven Ghan, we now discuss our results in terms of radiative forcing due to aerosol-radiation interactions (RFari) to avoid confusion due to the earlier used $ERFari_{cls}$*, that was a idealized value that depicted the radiative inbalance at the top of atmosphere in a hypothetical atmosphere without clouds.*

The effect of total and anthropogenic aerosols is shown, respectively?

We just show the ERF due to anthropogenic aerosols (ERFaer) since we compute the difference between PD and PI emissions. We now mention this in the methods section in the manuscript.

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Page 7, line 15:
Why did cloud cover increase in PD simulations but decrease in PI simulations?

We ascribe this to internal variability. That can be seen by overlapping confidence intervals:
TCC_{PD}: -0.08 $\pm$ 0.14%
TCC_{PI}: 0.17 $\pm$ 0.14%

We added a sentence about internal variability to the discussion.

Page 9, line 13:
The authors focus on sulfate and sea salt here, but how about nitrate, ammonium, and secondary organic aerosol? How does the global model treat these aerosol species?

We focus on sulphate and sea salt because they are very hygroscopic (for HAM2 κ_{SS} = 1.12 and κ_{SO4} = 0.6, see Zhang et al., 2012) in comparison to SOA (κ_{SOA} = 0.037). Aerosols that are more hygroscopic are more sensitive to changes in relative humidity than aerosols that are less hygroscopic. This characteristic is what we use for the explanation of the observed profile of the change in the growth factor. Note, that the κ-value for sulphate in HAM2 is in the range of the observed value for ammonium sulphate (0.33 - 0.72) (Petters and Kreidenweise, 2007). Furthermore, the model that we use currently does not simulate nitrate aerosols (Stier et al., 2005). We highlight in our paper now that the addition of nitrate aerosols will introduce very hygroscopic aerosols into the model that would alter our results.

Page 10, lines 1-5:
Please show the simulation results obtained by the authors rather than citing previous

studies.

We now integrate a profile plot that shows that the mixing ratio of hygroscopic aerosols. The plot shows that the mixing ratio of sea salt decreases stronger with height than for other aerosols. Hence, the overall aerosols composition becomes less hygroscopic.

Page 10, line 8:
Please explain why the function of height is used. The authors explain the treatment in the cloud scheme but do not explain whether the treatment is realistic.

RH_{crit} is a function of height in the general formulation of ECHAM6-HAM2. Implicitly we already stated that in the introduction when we referred to Quaas (2012). However, now clearly formulate that Quaas (2012) found RH_{crit} to be a function of height.

Page 10, line 11:
"Eq. (Equation 1)" should be "Eq. (1)".

Changed.

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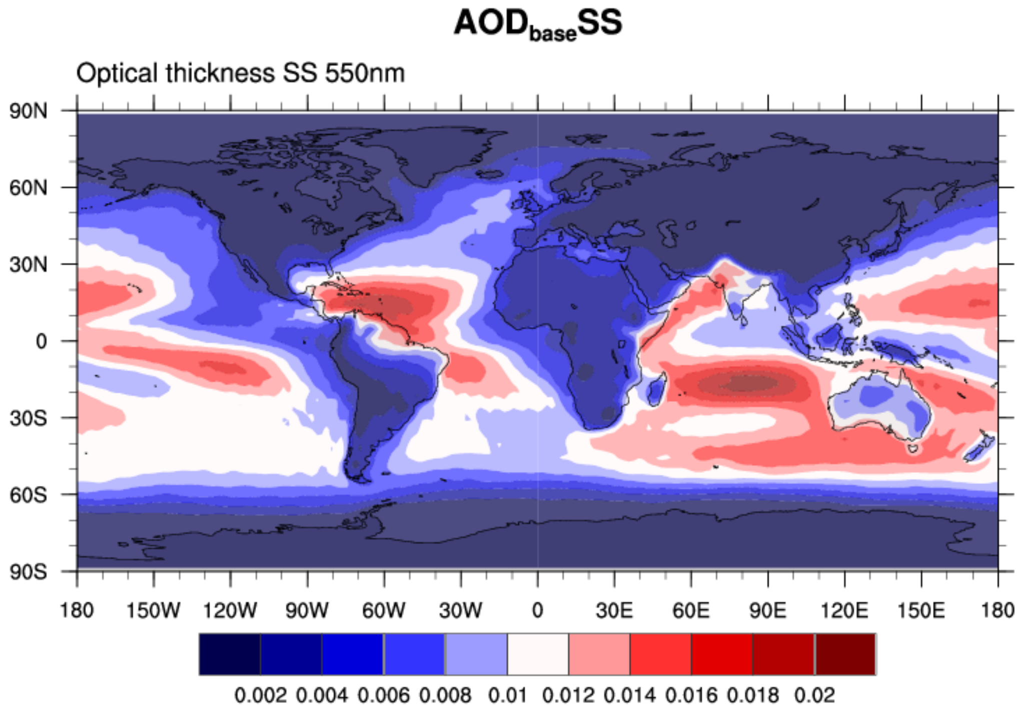

Fig. 1. AOD by sea salt (SS) in the control run of the model.

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