Response to comments on *The global aerosol-climate model ECHAM6.3-HAM2.3 – Part 2: Cloud evaluation, aerosol radiative forcing and climate sensitivity*, Geosci. Model Dev. Discuss., gmd-2018-307

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We would like to thank the reviewers for the helpful comments and suggestions. They have helped to improve the content of the paper.

The original comments are in black. Responses are in blue. *Modifications to the text are in green and italics*.

# **Anonymous Referee #1**

Neubauer et al. evaluate important aspects of a new version of a three-dimensional global aerosol-atmosphere model. It is well understood that cloud condensation nuclei concentrations vary significantly between strongly and less strongly polluted conditions and that radiative forcing by cloud-aerosol interactions depends non-linearly on cloud condensation nuclei concentration. At a time when box-model studies of cloud-aerosol interactions inform energy budget box-model studies, I find this study by Neubauer et al. a truly laudable effort. My only major concern with this study is that even in the latest model version, the minimum CDNC number is still artificially set to 40 per cubic centimetre. The authors discuss this point in some detail. They show that ERFari+aci and ECS depend on this threshold, but they do not mention this result in the abstract. Although one can certainly argue about this point, I find the reasons for applying this particular threshold unconvincing. Nevertheless, I consider this a very worthwhile study. Unlike some other studies, this study includes a sensitivity run (the E63H23-10CC run) that helps to assess a key uncertainty (partially re-iterating a point made in an earlier study). It also includes another useful sensitivity run (E63H23-LL) that helps to attribute differences between model versions to an individual change in a parameterization. In my opinion, after some revisions, this study clearly deserves to be published in GMD.

Specific comments and suggestions:

1. The fact that the CDNC threshold which leads to a lower ERFari+aci and a lower ECS is still applied should in my opinion definitely be mentioned in the abstract. As is shown in the manuscript, reducing this threshold results in a considerably larger ERFari+aci (first noted in Hoose et al., 2009) and interestingly also a larger ECS. It is not clear to me by how much an improved aerosol would reduce this larger ERFari+aci.

It is now mentioned in the abstract that the value do CDNCmin has an impact on ECS, as well as ERFari+aci:

Experiments with minimum cloud droplet number concentrations (CDNCmin) of 40 cm-3 or 10 cm-3 show that a higher value of CDNCmin reduces ERFari+aci as well as ECS in E63H23.

The experiment GFAS34 gives an indication by how much ERFari+aci would be reduced by an improved aerosol representation in ECHAM-HAM. E63H23-GFAS34 has a 17% lower ERFari+aci than E63H23. Since nitrate aerosol and secondary organic aerosol will occur in further regions, not only where wildfire emission occur, an even stronger decrease than 17% in ERFari+aci by an improved aerosol representation in ECHAM-HAM can be expected.

2. The weaker shortwave ERFari+aci in E63H23 is attributed to the new aerosol activation scheme and sea salt emission parameterization in E63H23 and a more realistic simulation of cloud water. Would it be possible to quantify individual contributions e.g. based on the E63H23-LL sensitivity study from Fig. S5 and perhaps also a run from Tegen et al. (2019)? Or would this require additional sensitivity runs?

We made another sensitivity experiment with E63H23 and the Guelle sea salt parameterization used in E61H22, named E63H23-GUELLE. Using the experiments E63H23, E63H23-LL, E63H23-GUELLE and E61H22 allows to estimate the individual contributions to the change in ERFari+aci from E61H22 to E63H23. But the different code/parameterization changes will not add linearly and there can be interaction between parameterizations, therefore the individual contributions are hard to disentangle. Over land the stronger ERFari+aci is caused about half by the new aerosol activation scheme and about half by other code changes.

Code changes (in particular the ICNC bugfix) cause a decrease by 0.65 Wm<sup>-2</sup> in LW ERFari+aci and the new aerosol activation scheme causes an increase by about 0.15 Wm<sup>-2</sup>.

Over ocean the new aerosol activation scheme makes LW ERFari+aci stronger but has a weak impact on SW ERFari+aci. The new sea salt emission parameterization makes SW ERFari+aci stronger over ocean by about -0.1 Wm<sup>-2</sup> but the majority of the decrease in SW ERFari+aci over ocean by about 0.7 Wm<sup>-2</sup> in SW ERFari+aci is from other code changes (including e.g. the improved stratocumulus cloud cover in the base model ECHAM6.3 or tuning in ECHAM6.3).

We added the results of the E63H23-GUELLE sensitivity experiment to Table S1 and Fig. S5. We updated the text to better quantify individual contributions to changes in ERFari+aci between E61H22 and E63H23:

A sensitivity simulation with the Lin and Leaitch (1997) activation scheme applied in E63H23 shows an ERFari+aci of 0.4 Wm-2 over land, explaining about half of the difference in ERFari+aci over land between E61H22 and E63H23.

The higher natural background aerosol concentrations due to the higher sea salt aerosol number concentrations in E63H23 explain also why ERFari+aci is less negative in E63H23 between 15°N and 45°N over oceans than in E61H22 (Fig. S5). From sensitivity simulations with the Lin and Leaitch (1997) activation scheme or Guelle et al. (2001) sea salt parameterization applied in E63H23 (Table S1) we conclude that the largest part of the change in SW ERFari+aci is actually from changes in the base model ECHAM6.3.

3. It is concluded (p. 1, 1. 27f) that "[t]he decrease in ECS in E63H23 (2.5 K) compared to E61H22 (2.8 K) is due to changes in the entrainment rate for shallow convection (affecting the cloud amount feedback) and a stronger cloud phase feedback". As far as I can see, especially the conclusion regarding a "stronger cloud phase feedback" (see also p. 24, 1. 3) does not seem to be supported by sufficient evidence. Please either explain the existing evidence better, present additional evidence, or else please either preferably remove the statement or at least re-formulate the statement to reflect that this is not a finding but a speculation.

In Fig. 14 it can be seen that the largest difference in cloud feedback between E63H23 and E63H23-10CC is from a different cloud optical depth feedback. The change in non-low clouds optical depth feedback is likely due to a change in the cloud phase feedback. The new Fig. S7 shows the change in in-cloud CDNC from the 1xCO2 to the 2xCO2 climate. Cloud droplets exist at higher altitudes in the warmer climate. The increase in CDNC is stronger in E23H23 than in E63H23-10CC, leading to a larger change (increase) in cloud optical depth in the warmer climate in E63H23 compared to E63H23-10CC.



Figure S7: Zonal mean change in in-cloud CDNC for E63H23 and E63H23-10CC for the change from the  $1xCO_2$  climate to the  $2xCO_2$  climate. Averages for the last 25 years of the  $1xCO_2$  and  $2xCO_2$  experiments were used to create the figure.

### We updated the text to refer to the new Fig. S7:

The optical depth feedback of non-low clouds is less negative in the tropics and in midlatitudes (not shown). This could be an indication of a weaker cloud phase feedback. As there are fewer but larger cloud droplets in E63H23-10CC than in E63H23 (Fig. S7), the cloud droplets have a shorter lifetime and this decreases differences between ice clouds and liquid water clouds.

4. Cloud properties and cloud radiative effects are not only simulated by ECHAM-HAM but also by ECHAM. I wonder whether it would make sense to include ECHAM results in these comparisons. In my opinion, including ECHAM would in general help to better understand which biases in ECHAM-HAM are inherited and which biases are newly introduced by using e.g. different tunings and a different microphysics scheme. Including ECHAM results would also help to understand which differences between versions are due to changes in ECHAM and which differences between versions are due to changes in components that are specific to ECHAM-HAM. The discussion explaining the results frequently refers to changes in ECHAM, and some parts of it might be easier to follow if these changes were shown in tables 2 and 3 and especially also in the plots. On the other hand, the comparison of different ECHAM-HAM versions is useful without an additional focus on attributing the changes to changes in either standard ECHAM or in components that are specific to ECHAM-HAM, and including to many plots would also distract the reader from this comparison. Nevertheless, I think ECHAM plots would potentially be a nice-to-have. In case the authors decide against including ECHAM results, I would recommend to refer even more frequently to the literature documenting these results, especially when common biases are discussed. In some important cases (e.g. p. 20, l. 26), the references are already included.

The focus of the manuscript is on the comparison of different ECHAM-HAM versions and the inclusion of the latest version of the base model ECHAM6.3 in the Figures and Tables would indeed be a distraction for the reader as there are differences between ECHAM6.3 and E63H23 which would need to be discussed. We added more references discussing biases in cloud representation in the base model ECHAM (see answer to the next specific comment). A comparison of the representation of clouds in ECHAM and ECHAM-HAM should be done in a future study.

Other specific comments and suggestions:

p. 1, 1. 18ff "Biases that were identified in E63H23 (and in previous model versions) are a too low cloud amount in stratocumulus regions, deep convective clouds in the Atlantic and Pacific oceans form too close to the continents and there are indications that ICNCs are overestimated": I think that already here it would be good to clearly differentiate between biases that are inherited from ECHAM, biases that are specific to ECHAM-HAM in all HAM versions, and biases that change by the HAM modifications. Also, I think it would be good to clarify which biases were newly identified in this study and which biases are well-known and long-standing biases. Perhaps this can be achieved almost without lengthening the abstract. The resulting sentences could for example read: "Common biases in ECHAM and in ECHAM-HAM are ... . ICNCs are overestimated in ....". If the authors decide against including ECHAM plots (discussed in comment #4 above), as far as ECHAM biases are documented elsewhere, it would be sufficient to point to the corresponding literature somewhere in the text. p. 1, 1. 19f, p. 17, 1. 17f, p. 23, 1. 12f: Based on Figs., 3, 6, and 8, I am not completely sure what is meant by "deep convective clouds in the Atlantic and Pacific oceans form too close to the continents". Is this something that one sees when putting ECHAM results next to ECHAM-HAM results? For example, there seems to be little deep convection over Indonesia in ECHAM and in ECHAM-HAM. Focusing on Indonesia (I think), Mauritsen et al., 2012 (https://doi.org/10.1029/2012MS000154) note that "[a]n interesting and challenging issue in MPI-ESM is the Tropical precipitation distribution over land versus ocean. The model prefers precipitating in the ocean, whereas observations indicate a stronger preference to precipitate on land."

We added references for the base model ECHAM at the suggested places, in particular:

### Section 3.3:

ECHAM underestimates tropical precipitation over land and overestimates tropical precipitation over ocean (Mauritsen et al., 2012; Stevens et al., 2013). This bias can be seen in Fig. 8 also for all ECHAM-HAM versions. Since ECHAM and ECHAM-HAM use the same parameterizations for convective clouds, this bias is very likely inherited from the base model ECHAM.

### Section 3.4:

Deep convective clouds over the Atlantic and Pacific oceans form too close to the continents (see Figs. 3, 6 and 8) in E63H23 and ECHAM (Stevens et al., 2013). Stevens et al. (2013) speculate that the overestimation of tropical precipitation in the Atlantic close to South America may be related to the deficit of precipitation over South America in the tropics.

In Figs. 3, 6 and 8 one can see in the observations that the maximum of cloud cover, IWP and precipitation in the ITCZ in the Pacific and the Atlantic is away from continents. Whereas in the different ECHAM-HAM versions (and also ECHAM). The maximum is of cloud cover, IWP and precipitation in the ITCZ in the Pacific and the Atlantic is close to continents.

Stevens et al. (2013) speculate for the Atlantic that the overestimation of tropical precipitation in the Atlantic close to South America may be related to the deficit of precipitation over South America in the tropics.

## p 1, l. 19f: in the Atlantic and Pacific oceans -> over the Atlantic and Pacific oceans

Changed as suggested.

p. 2, l. 4: resulting in -> substantially contributing

Changed as suggested.

p. 2, l. 5: realistic representation -> increasingly realistic representation (Almost certainly some of the dynamic responses to increased aerosol take place on scales which are too small to resolve by present-day global climate models. This remains a major concern as several studies point out that this potentially leads to an overestimate of ERFaci in coarse-resolution models.)

We agree that this is a concern for global aerosol-climate models (e.g. Mülmenstädt and Feingold, 2018) and changed the phrase as suggested.

# p. 4, l. 5: how does evaporation and sublimation affect aerosol number concentration?

A downward scavenging tracer flux is computed for each model column each model time step. In-cloud and below cloud scavenging are sources for the downward scavenging tracer flux, while evaporation and sublimation of precipitation are sinks for the downward scavenging tracer flux. When the sink term is larger than the source term of the downward scavenging tracer flux in a model level, aerosol mass and number concentrations will be transferred to the respective atmospheric tracers i.e. aerosol is released at this model level below clouds.

CDNC and ICNC used for wet scavenging are after cloud droplet evaporation, ice crystal sublimation and precipitation formation are computed. This is now mentioned in the description and an error in the description of nucleation scavenging has been corrected:

The below cloud collection efficiencies as a function of aerosol and rain drop or snow crystal size are read from a look-up table. The in-cloud scavenging scheme takes nucleation scavenging and impaction scavenging of aerosol particles with cloud droplets and ice-crystals into account. For nucleation scavenging the number of scavenged aerosol particles is computed for liquid cloud droplets from the cloud droplet number concentration (CDNC) (after computation of cloud droplet evaporation and precipitation formation) and the fraction of activated aerosol particles computed by the activation scheme. For ice crystals the aerosol particles are scavenged aerosol particles is equal to the ice crystal number concentration (ICNC) (after the computation of ice crystal sublimation and precipitation formation) of the grid box.

A downward scavenging tracer flux is computed for each model column each model time step. In-cloud and below cloud scavenging are sources for the downward scavenging tracer flux, while evaporation and sublimation of precipitation are sinks for the downward scavenging tracer flux. When the sink term is larger than the source term of the downward scavenging tracer flux in a model level, aerosol mass and number concentrations will be transferred to the respective atmospheric tracers i.e. aerosol is released from evaporating/sublimating precipitation at this model level back to the atmosphere.

# p. 4, l. 28: I don't understand how CDNC from convective clouds is determined. Please try to explain this better.

A description of how CDNC from convective clouds are computed has been moved from section 3.3 to section 2.1.3 and extended:

To obtain CDNC for the detrained condensate, several simplifications are applied. It is assumed that cloud droplets of convective clouds will form at cloud base. The number of activated cloud condensation nuclei (CCN) at the convective cloud base is computed using the vertical velocity from large scale and turbulent fluxes as described in section 2.1.4. It is further assumed that CDNC will be constant throughout the vertical extension of the convective clouds. At the level of detrainment these CDNC from the convective clouds will either evaporate or be added to stratiform clouds if these exist at the level of detrainment. In the latter case a weighted average of the stratiform CDNC and detrained CDNC is computed by weighting stratiform CDNC with the stratiform liquid water content and detrained CDNC with detrained liquid water content. CDNC of the stratiform cloud is not allowed to decrease by this procedure, since cloud droplets will not evaporate in a supersaturated environment.

p. 6, l. 9: please indicate that SALSA2.0 is not used here to avoid confusion.

Done.

# p. 6, l. 18: please refer to my comment regarding p. 4, l. 28.

Section 2.1.5 list changes and improvements in E63H23 but is not meant to provide the details of the changes which are given in other sections (section 2.1.3 for the computation of CDNC from convective clouds). Nevertheless this point was reformulated to avoid confusion: *Changed treatment of detrained cloud water mass and number concentrations from convective clouds: CDNC from detrained cloud water added (weighted average) to CDNC of a stratiform cloud cannot decrease CDNC of the stratiform cloud; split between liquid water and ice of detrained condensate is made consistent between mass and number concentrations* 

### p. 7, l. 8: could you please be slightly more specific?

We list now the most important updates:

Update of default settings, run templates and run organization: the vertical resolution is per default 47 vertical model layers; the reference year and reference period for present day simulations are 2008 and 2003-2012 respectively

p. 8, l. 19: climate system -> atmosphere (if the entire climate system including the ocean were considered, 20 years would be insufficient. Using identical fixed SSTs strongly reduces the influence of internal variability.)

That's a good point. We changed this to:

The simulation time was 20 years to increase the signal of ERFari+aci compared to variations in TOA net radiative flux due to internal variability of the atmosphere. The use of an identical climatology for SST/SIC in all simulations reduces the internal variability compared to simulations with a global climate model (GCM) coupled to a full ocean model.

p. 11, l. 31: over -> cover

Done.

p. 13, l. 7f: The COSP CALIPSO simulator was used here, right? Excluding areas where the cloud products differ by more than five percentage points looks like a good idea to me. The excluded regions are regions in which one might expect problems.

The COSP simulator was not implemented in E55H20 (mentioned in section 3.3). Therefore Fig. 3 shows the direct model output without using the COSP CALIPSO simulator. This is now mentioned in the figure caption of Fig. 3:

... model data is from the PD simulations (direct model output is used without a simulator).

The new Fig. S8 shows the cloud cover from the COSP CALIPSO simulator implemented in E61H22 and E63H23 and a comparison to the direct model output of the cloud cover. Except at high latitudes (where observations are uncertain) and a few areas, there are no large differences between the direct model output and the COSP CALIPSO simulator output:

Since the COSP CALIPSO simulator is not implemented in E55H20, the direct model output is shown for all model versions (see Fig. S8 for COSP CALIPSO simulator output of cloud cover for E61H22 and E63H23).



Figure S8: Comparison of annual mean cloud cover of E55H20, E61H22 and E63H23 to CALIPSO GOCCP observations. Areas where the cloud cover of CALIPSO GOCCP, MODIS collection 6.1 and AVHRR-PM differ by more than five percent points are hatched. CALIPSO GOCCP data is for 2006-2010, model data is from the PD simulations. For E61H22 and E63H23 the direct model output, the output of the COSP CALIPSO simulator implemented in those model versions and the difference between direct model output and COSP CALIPSO simulator output is displayed.

p. 14, l. 8f: please refer to my comment regarding p. 4, l. 28.

The description how detrained CDNC are computed and added to an existing stratiform cloud was moved to section 2.1.3 (see our answer to the comment regarding p. 4, 1. 28): *The weighted average of stratiform CDNC and detrained CDNC (see section 2.1.3) may overestimate the CDNC concentration of shallow cumulus clouds.* 

p. 15, l. 32ff: In my opinion this entire discussion would be more interesting if standard ECHAM was included in the comparison.

See our answer to your comment #4.

p. 16, l. 27: where -> which

Done.

p. 16, l. 11f: the authors could mention that such a bias is also found in other models (<u>https://doi.org/10.1029/2012GL053421</u>).

Thanks, this has been added.

p. 17, l. 6: RMS -> RMS error

Done.

p. 18, l. 22: does the statement "due to a smaller gamma\_r" on p. 18, l. 22 indicate that the E63H23-10CC has been retuned? If yes, is this retuning expected to affect ECS? Based on https://doi.org/10.1175/JCLI-D-16-0151.1, I would have perhaps thought brighter clouds in the base state to play a different role for ECS. How about ERFari+aci? I wonder whether the result of Lohmann and Ferrachat (2010) holds also in E63H23.

E63H23-10CC has been retuned. The value of gamma\_r has been added to the text:

# (due to retuning with a smaller $\gamma_r = 2.8$ ; not shown)

The study by Klocke et al. (2011), identified the entrainment rate for shallow convection and the cloud mass flux above the level of neutral buoyancy as tuning factors in ECHAM that affect ECS. They did not test the conversion rate from cloud water to rain in stratiform clouds but the conversion rate from cloud water to rain for convective clouds and found no significant impact on ECS. The results in Figure 6 of Lohmann and Neubauer (2018) indicate no large impact of the tuning parameter gamma\_r on cloud feedback parameters and therefore ECS.

The autoconversion parameterization in E55H20 used by Lohmann and Ferrachat (2010) is the same as in E63H23, so the meaning of gamma\_r and the response to changes in gamma\_r should be similar. Investigating the effect of model tuning on ECS and ERFari+aci would be interesting but is beyond the focus of this study.

# p. 19, l. 26: why "...(Schmidt et al., 2006; Hansen et al., 2005) or Fan et al. (2004)"?

We rephrased this part to make it clear that different models were used in these studies: ... have been found by Fan et al. (2004) (using the Geophysical Fluid Dynamics Laboratory (GFDL) global chemical transport model; Mahlman and Moxim, 1978) and Bauer and Koch (2005) and Bauer et al. (2007) (using the Goddard Institute for Space Studies (GISS) climate model, modelE; Schmidt et al., 2006; Hansen et al., 2005).

# p. 21, lines 4 to 6.: is there a reference for this?

Thanks, unfortunately this was overlooked. These changes are described in Mauritsen et al. (2019), the reference was added.

# p. 22, l. 22: did ECS also decrease in standard ECHAM?

Yes, we added ECS for the three corresponding ECHAM base model versions:

The equilibrium climate sensitivity (ECS) is strongest in E55H20 (3.5 K), weaker in E61H22 (2.8 K) and weakest in E63H23 (2.5 K) (Fig. 13). The corresponding ECS values in the base model versions are: ECHAM5: 3.4 K (Randall et al., 2007; their Table 8.2), ECHAM6.1: 2.8 K (Block and Mauritsen, 2013; Meraner et al., 2013) and ECHAM6.3: 2.8 K (Mauritsen et al., 2019), i.e. changes in ECS between the ECHAM-HAM model versions are driven substantially by changes in the ECHAM base model versions. Note that the ECS value for ECHAM6.3 is from a simulation with abruptly quadrupled CO2 concentrations, in contrast to the CO2 doubling used in the computation of ECS in this study and that ECHAM has a strong state dependency for ECS (see discussion in Mauritsen et al., 2019 and references therein).

# We changed the sentence in p. 22, l: 21-22 to:

The changes and improvements in E63H23 (including changes in the base model version ECHAM6.3) therefore have not only improved the representation of clouds in E63H23 compared to previous model versions, they also have an impact on ERFari+aci and ECS, decreasing both.

# p. 23, l. 12: please refer to my comment regarding p. 1, l. 19f above.

Changed as suggested.

# p. 23, l. 21ff: please refer to my comment #2 above.

This was changed to better reflect the contribution of individual changes:

- *The largest differences between E61H22 and E63H23 in terms of SW ERFari+aci are due to:* 
  - mainly the more realistic simulation of cloud water,
  - but also the new activation scheme in E63H23 and
  - the new sea salt emission parameterization

which lead to a weaker SW ERFari+aci in E63H23. In terms of LW ERFari+aci the difference is mainly due to:

• the removal of an inconsistency in ICNC in cirrus clouds leading to a weaker LW ERFari+aci in E63H23.

# p. 24, l. 4: please refer to my comment #3 above.

See our answer to your comment #3.

# Table 1: what exactly is gamma\_r?

gamma\_r is a unitless scaling factor for the stratiform rain formation rate by autoconversion. The header of Table 2 has been updated to reflect this:

... scaling factor for stratiform rain formation rate by autoconversion ( $\gamma_r$ ), scaling factor for stratiform snow formation rate by aggregation ( $\gamma_s$ ), ...

Table 3: from E61H22 to E63H23 there is a large compensation of changes in SW and LW ERFari+aci. This has also been noted elsewhere (https://doi.org/10.1002/2016GL071975). Are you really sure that here it is mainly due to the removal of a bug?

Test simulations before and after the ICNC bugfix revealed a large change in LW ERFari+aci (not shown). The large change in LW ERFari+aci can be attributed mainly to this bugfix and changes in SW ERFari+aci to other model changes (see our answer to your comment #2).

Fig. 9 is too small. It can't be enlarged because the resolution is too low. Please increase the size and the resolution.

### Done.

Fig. 10: I do not understand the rationale behind excluding areas with little precipitation. While relative errors tend to be large in these areas, absolute errors in these areas tend to be small. Excluding these areas in the bias calculation could in principle hide model deficiencies. Observations of small values may contain important information due to a small absolute error, even where the relative error is large. Using standard deviations already ensures that large relative errors in regions with small values and small absolute errors will not have an overly large influence on the comparison.

The areas hatched in Fig. 8 are indeed areas where the relative error is large but the absolute error is small. Therefore we recomputed the correlation and normalized standard deviation including all areas for precipitation. At the same time we corrected a mistake in the computation of the normalized standard deviation. The discussion of Fig. 10 has been adapted:

The standardized deviations of LWP-LP had to be scaled by a factor of 1/4 so they could fit on the scale. For all variables the root-mean-square error (RMSE) (solid circles in the diagram in Fig. 10) is smaller or equal in E63H23 compared to in E61H22 and E55H20 (note the scaling for LWP-LP).



Figure 10: Taylor diagram for comparison of SW and LW CRE, cloud cover, LWP-low precipitation, Cloud top CDNC, IWP and precipitation of E55H20, E61H22 and E63H23 to observations as REF. The standardized deviations of LWP-low precipitation are scaled by a factor of 1/4 to fit on the diagram. Only areas that are not hatched in Figs. 3 - 6 were used to create the Taylor diagram. Observations are the same as in Figs. 2 - 6 and 8. The correlation coefficient is shown as an angle and quantifies the similarity in pattern between modelled and observed annual mean fields. The standard deviation of the modelled fields (normalized by the standard deviation of the observed fields) is shown as the radial distance from the origin. The RMSE is shown as solid black circles and is the distance from the point marked by REF (the closer a model is to REF the better its skill to reproduce the observations). For E63H23 and the observations for precipitation and LWP-low precipitation an average over the time period 2003 to 2012 was used. For Cloud top CDNC the time period 2003 to 2015, for IWP the time periods in Li et al. (2012), for SW CRE and LW CRE the time period July 2005 to June 2015, for cloud cover the time period June 2006 to December 2010 and for E55H20 and E63H23 the time period 2000 to 2009 were used. Tests with E63H23 showed negligible impact of the different time periods for the data in the Taylor diagram.

Fig. 11: please increase the font size of the variable names (right plot title) and the color bar labels.

Done.

Fig. 14: Amt is amount, right? Please state this somewhere.

Done.

References:

Klocke, D., R. Pincus, and J. Quaas (2011), On constraining estimates of climate sensitivity with present-day observations through model weighting, J. Clim., 24(23), 6092–6099, doi:10.1175/2011JCLI4193.1.

Mülmenstädt, J. and Feingold, G.: The radiative forcing of aerosol-cloud interactions in liquid clouds: Wrestling and embracing uncertainty, Curr. Clim. Change Rep., 4, 23–40, https://doi.org/10.1007/s40641-018-0089-y, 2018.

# Anonymous Referee #2

General comments

The authors present an evaluation of clouds and precipitation simulated in three models ECHAM6.3-HAM2.3, ECHAM6.1-HAM2.2 and ECHAM5.5-HAM2.0 in comparison to global observational datasets. They discuss the performance of each model and the reasons for improvements.

The purpose of the paper is to provide a model documentation in a nutshell and to characterize the quality of clouds and precipitation simulated in the lates model E63H23 as well as in earlier versions. This is valuable information for all users and developers of these models and thus fits well to the scope of GMD. Overall the paper is clearly structured and well written. Figures are of good quality. My main recommendation is to add discussion to the obvious deviation between observed and modeled IWP near the equator and also in the NH storm tracks. Otherwise only minor modifications and corrections are necessary, so that a minor revision seems sufficient before publication.

We added a discussion of the underestimation of cloud ice in the tropics and refer to the discussion of the general low bias of IWP in ECHAM-HAM at the end of section 3.3. See our answers to the specific comments below.

Specific comments

Abstract, L14: low cloud amount  $\rightarrow$  amount of low clouds (?)

Changed to amount of low clouds.

P2 L1: "...Also, the spatial structure of multiple clouds shows a large variability on different scales as it depends not only on large scale motions of the air but also on convective and turbulent motions at different scales. ..." The important point is that the strong diabatic cooling/heating occurring with phase changes of water vapor causes a tight coupling between clouds and circulation, which is much less the case for other constituents.

This is a very good point. We added this to the introduction:

Also, the spatial structure of multiple clouds shows a large variability on different scales as it depends not only on large scale motions of the air but also on convective and turbulent motions at different scales. These convective and turbulent motions in turn are driven to a large part by diabatic heating (and cooling) and radiative cooling (and heating) involving cloud and precipitation hydrometeors, leading to a tight coupling between clouds and circulation.

P4 L12: orographic cirrus cloud  $\rightarrow$  orographic cirrus clouds (2x)

Done.

P5-7 Section "2.1.5 Changes and improvements in E63H23" The preceding section lists already some process models and indicates cases where these are optional but not used in this study. In section 2.1.5 it is not pointed out that SALSA is available but – to my understanding – not used in this study. Is is also the case for other process models, for which improvements are listed here?

It is now explicitly mentioned in section 2.1.5 that SALSA is not used in this study. All other changes and improvements in E63H23 listed in section 2.1.5 are used for the simulations with E63H23 in this study.

P7 L20: use the year  $\rightarrow$  used the year

Done.

P7 L30: ... use a climatology for monthly values of sea surface temperature (SST) and sea ice cover (SIC) ... This setup excludes the influences of El Niño/La Niña on the variability of the atmospheric circulation. Thus the simulated "internal climate variability" is reduced compared to simulations which included El Niño/La Niña. This should be made clear to the reader. This reduction of variability is relevant for the later evaluation, see your comments on P8L18: " ... to increase the signal of ERFari+aci compared to variations in TOA net radiative flux due to internal variability of the climate system. ..."

# That's a good point. We changed this to:

The simulation time was 20 years to increase the signal of ERFari+aci compared to variations in TOA net radiative flux due to internal variability of the atmosphere. The use of an identical climatology for SST/SIC in all simulations reduces the internal variability compared to simulations with a global climate model (GCM) coupled to a full ocean model.

P8 L1: ... the default configuration of these model versions  $\dots \rightarrow \dots$  the default configuration of these ECHAM-HAM model versions ... ECHAM6 on its own uses 47 levels (Stevens et al., 2013).

# Changed as suggested.

P9 L17: ... are described in Mauritsen et al. (2012) ... The recent publication Mauritsen et al. (2019) provides more information for the tuning of ECHAM6.3: Mauritsen, T., Bader, J., Becker, T., Behrens, J., Bittner, M., Brokopf, R., et al. (2019). Developments in the MPI-M Earth System Model version 1.2 (MPI-ESM1.2) and its response to increasing CO2. Journal of Advances in Modeling Earth Systems, 11. https://doi.org/10.1029/2018MS001400

Thanks, this was changed to:

The tuning strategy and parameters for ECHAM6 as well as the impact of these parameters on the model climate are described in Mauritsen et al. (2012, 2019).

P10 L26: ... Table 3 ... compared to observations (OBS) or multi-model mean values (MMM) when observations are not available ... It would be helpful to mark each entry in the OBS/MMM column whether it is OBS or MMM.

Done.

P11 L32: cloud over  $\rightarrow$  cloud cover

Done.

P12 L12: ... The underestimation is particularly large in the tropics. ... Here the authors should add explanations. What is the reason for this major deviation? Microphysics of deep

convection? Cirrus cloud processes? As the authors have expertise in this field they should discuss this obvious modeling problem.

We added a discussion of the underestimation of cloud ice in the tropics:

The underestimation is particularly large in the tropics, which is likely connected to the parameterization of convection in ECHAM(-HAM). ECHAM has a low precipitation bias over land in the tropics (Mauritsen et al., 2012; Stevens et al., 2013). Gasparini et al. (2018) found indications that the level of detrainment from deep convection is too low in altitude in ECHAM-HAM. They lowered the tuning parameter for deep convective entrainment  $\epsilon_d$  to 0.00006, whereas all three ECHAM-HAM versions used here have to use a larger value for this parameter (Table 2) as they use a cirrus scheme in which cirrus clouds can only nucleate homogeneously, which may lead to an overestimation of ICNC and underestimation of IWP by tuning of radiative fluxes (see section 3.3).

P13 L12: ... Over land in the Northern Hemisphere the models overestimate cloud cover ... This seems not correct for the dry continental regions: Sahara, Australia, ..., even when accounting for less observational certainty in the Sahara region. Here the models underestimate the cloud cover.

This was indeed imprecise and was changed to: Over land in the Northern Hemisphere polwards of about 45°N the models overestimate cloud cover ...

P14 L12: ... and in all model version shallow convection is triggered frequently ... What is the role of the markedly increased shallow convection entrainment rate used in E63H23?

The change in shallow convection entrainment rate will shift the level of detrainment to lower altitudes. Since CDNC of detrained condensate are determined from convective cloud base, the CDNC of detrained condensate do not depend on the altitude of the level of detrainment.

P14 L28: ... The regional distribution of IWP of all three model versions agrees in general quite well with the observations. ... I do not agree with this general statement. The general disagreement in the equatorial region and also in the northern storm tracks is clear. Here, or earlier in the presentation of the zonal mean results, there is really the need to address this problem. I do not expect that a full explanations can be given – otherwise the modeling problem could be solved – but the authors should comment on this challenge and provide their insight in the possible reasons. This would make the discussion of the IWP much more interesting.

This statement was reformulated to clarify that the magnitude of IWP does not agree with observations but IWP occurs in the same areas as in observations. For the discussion of the low bias in the tropics we refer now to the discussion in section 3.2. A reason for the low bias of IWP is given at the end of section 3.3.

The regional distribution of occurrence of IWP of all three model versions agrees in general quite well with the observations, although it is biased low in all ECHAM-HAM model versions. This could already be seen in the respective global mean and zonal mean values (see sections 3.1 and 3.2). Similar to what was found in the analysis of zonal mean IWP the underestimation is largest in the tropics (see section 3.2).

We refer to this now also in section 3.4:

Previous studies (Gasparini et al., 2018; Lohmann and Neubauer, 2018) showed that this overestimation of ICNC is (at least partly) due to missing processes in the formation of cirrus

clouds (heterogeneous freezing of ice nucleating particles and/or water vapor deposition on pre-existing ice crystals). These studies also showed that including these processes can reduce the underestimation of IWP in ECHAM-HAM.

P15 L28: ... the areas and magnitude of precipitation differs  $\dots \rightarrow \dots$  the areas and magnitude of precipitation differ ...

Thanks.

P17 L4: ... the root-mean-square (RMS) error  $\dots \rightarrow \dots$  the root-mean-square error (RMSE) ... RMSE seems more useful for the following usage than RMS.

# Changed as suggested.

P20 L9: ... The equilibrium climate sensitivity (ECS) is strongest in E55H20 (3.5 K), weaker in E61H22 (2.8 K) and weakest in E63H23 (2.5 K) (Fig. 13). ... Here it would be valuable to have explained also the ECS values estimated for the base atmospheric models (ECHAM5, ECHAM6.1 and ECHAM6.3), as discussed in the literature. This would provide a better background for the discussion of the ECS estimates from the ECHAM-HAM models presented here. The recent Mauritsen et al (2019) paper also provides more information to the sensitivity of the ECS to certain model modifications.

We added ECS for the three corresponding ECHAM base model versions:

The equilibrium climate sensitivity (ECS) is strongest in E55H20 (3.5 K), weaker in E61H22 (2.8 K) and weakest in E63H23 (2.5 K) (Fig. 13). The corresponding ECS values in the base model versions are: ECHAM5: 3.4 K (Randall et al., 2007; their Table 8.2), ECHAM6.1: 2.8 K (Block and Mauritsen, 2013; Meraner et al., 2013) and ECHAM6.3: 2.8 K (Mauritsen et al., 2019), i.e. changes in ECS between the ECHAM-HAM model versions are driven substantially by changes in the ECHAM base model versions. Note that the ECS value for ECHAM6.3 is from a simulation with abruptly quadrupled CO2 concentrations, in contrast to the CO2 doubling used in the computation of ECS in this study and that ECHAM has a strong state dependency for ECS (see discussion in Mauritsen et al., 2019 and references therein).

To our knowledge no ECS value for doubling of CO2 using a mixed-layer ocean model of ECHAM6.3 have been published, therefore we refer to the state dependence of ECS in ECHAM, discussed in Mauritsen et al. (2019). Published ECS values of ECHAM5, ECHAM6.1, ECHAM6.3:

5.4 K (Li et al, 2012; ECHAM5/MPIOM 4xCO2; T31L19GR30; simulation thousands of years long)

5.6 K (Li et al, 2012; ECHAM5, 4xCO2 MLO; T31L19)

3.4 K (Randall et al., 2007; their Table 8.2; ECHAM5, 2xCO2 MLO; T63L31)

2.8 K (Block and Mauritsen, 2013; ECHAM6.1, 2xCO2 MLO)

2.8 K (Meraner et al., 2013; ECHAM6.1, 2xCO2 MLO)

3.4 K (Block and Mauritsen, 2013; ECHAM6.1, 4xCO2 MLO)

3.4/3.9 K (Meraner et al., 2013; ECHAM6.1, 4xCO2 MLO; first value from deltaTs change; second value from F/lambda)

3.7 K (Block and Mauritsen, 2013; MPI-ESM-LR (ECHAM6.1) 4xCO2; 150 years after CO2 quadrupling)

3.6 K (Flato et al., 2013; their Table 9.5; MPI-ESM-LR (ECHAM6.1) 4xCO2; 150 years after CO2 quadrupling)

3.7 K (Giorgetta et al. (2013); MPI-ESM-LR (ECHAM6.1) 4xCO2; 150 years after CO2 quadrupling)

2.8 K (Mauritsen et al, 2019; MPI-ESM1.2-LR 4xCO2; 150 years after CO2 quadrupling) 3.6 K (Mauritsen et al, 2019; MPI-ESM1.2-LR 4xCO2; 100-1000 years after CO2 quadrupling) 2.8 K (Mauritsen et al, 2010; MPI-ESM1.2-LR 2xCO2; 100-1000 years after CO2

2.8 K (Mauritsen et al, 2019; MPI-ESM1.2-LR 2xCO2; 100-1000 years after CO2 quadrupling)

We added the reference to Mauritsen et al. (2019) for the change in the entrainment rate for shallow convection and its impact on ECS:

This seems to be related to the stronger entrainment rate for shallow convection in E63H23 (Mauritsen et al., 2012, 2019). Mauritsen et al. (2019) describe the increase of entrainment rate for shallow convection in ECHAM6.3 as a measure to reduce ECS in ECHAM6.3.

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# The global aerosol-climate model ECHAM6.3-HAM2.3 – Part 2: Cloud evaluation, aerosol radiative forcing and climate sensitivity

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Abstract. The global aerosol-climate model ECHAM6.3-HAM2.3 (E63H23) and the previous model versions ECHAM5.5-

- 15 HAM2.0 (E55H20) and ECHAM6.1-HAM2.2 (E61H22) are evaluated using global observational datasets for clouds and precipitation. In E63H23 low eloud amount of low clouds, liquid and ice water path and cloud radiative effects are more realistic than in previous model versions. E63H23 has a more physically based aerosol activation scheme, improvements in the cloud cover scheme, changes in detrainment of convective clouds, changes in the sticking efficiency for accretion of ice crystals by snow, consistent ice crystal shapes throughout the model, changes in mixed phase freezing and an inconsistency
- 20 in ice crystal number concentration (ICNC) in cirrus clouds was removed. Biases that were identified<u>Common biases</u> in <u>ECHAM and in E63H23</u> (and in previous model<u>ECHAM-HAM</u> versions) are a too low cloud amount in stratocumulus regions, and that deep convective clouds inover the Atlantic and Pacific oceans form too close to the continents and there(while tropical land precipitation is underestimated). There are indications that ICNCs are overestimated in E63H23. Since clouds are important for effective radiative forcing due to aerosol-radiation and aerosol-cloud interactions (ERF<sub>aritaci</sub>).
- 25 and equilibrium climate sensitivity (ECS), also differences in ERF<sub>ari+aci</sub> and ECS between the model versions were analyzed. ERF<sub>ari+aci</sub> is weaker in E63H23 (-1.0 W m<sup>-2</sup>) than in E61H22 (-1.2 W m<sup>-2</sup>) (or E55H20; -1.1 W m<sup>-2</sup>). This is caused by the weaker shortwave ERF<sub>ari+aci</sub> (new aerosol activation scheme and sea salt emission parameterization in E63H23, more realistic simulation of cloud water) overcompensating the weaker longwave ERF<sub>ari+aci</sub> (removal of an inconsistency in ICNC in cirrus clouds in E61H22).
- 30 The decrease in ECS in E63H23 (2.5 K) compared to E61H22 (2.8 K) is due to changes in the entrainment rate for shallow convection (affecting the cloud amount feedback) and a stronger cloud phase feedback.

Experiments with minimum cloud droplet number concentrations (CDNCmin) of 40 cm<sup>-3</sup> or 10 cm<sup>-3</sup> show that a higher value of CDNCmin reduces ERF<sub>ari+aci</sub> as well as ECS in E63H23.

#### **1** Introduction

Clouds are the largest modulators of radiation in Earth's atmosphere. Cloud hydrometeors are generally shorter-lived than other modulators of radiation in the atmosphere like aerosol particles, greenhouse gases or changes in surface albedo through changes in land use. Also, the spatial structure of multiple clouds shows a large variability on different scales as it depends

- 5 not only on large scale motions of the air but also on convective and turbulent motions at different scales. <u>These convective</u> and turbulent motions in turn are driven to a large part by diabatic heating (and cooling) and radiative cooling (and heating) involving cloud and precipitation hydrometeors, leading to a tight coupling between clouds and circulation (e.g. Wood, 2012; Voigt et al., 2014; Vial et al., 2016). The range of microphysical properties of cloud droplets and ice crystals adds to the complexity of clouds in Earth's atmosphere. This complexity makes clouds difficult to observe and to simulate using
- 10 models, resulting insubstantially contributing to the current large uncertainties in future climate projections. Therefore, it is necessary to have an increasingly realistic representation of clouds in global climate models to be able to study past and present climate forcings and to strengthen the reliability of climate projections. It is crucial to evaluate clouds in these models with reliable observations and account for the complexity in clouds in the process.
- In this study we use current satellite products to evaluate the aerosol-climate model ECHAM6.3-HAM2.3 and the two precursor model versions ECHAM6.1-HAM2.2 and ECHAM5.5-HAM2.0. One problem in using satellite products is that they are produced with retrieval algorithms that have to make assumptions about the nature of the clouds (e.g. assumptions about the vertical cloud profile; Miller et al., 2016) (and other modulators of radiation) which will not always fit optimally for every cloud in the observed satellite pixels. Accordingly, current satellite products include measures of uncertainty in the retrieved cloud properties. We use these uncertainty measures to limit the evaluation only to regions where the observations
- 20 are reliable. Furthermore, we apply the CFMIP (Cloud Feedback Model Intercomparison Project) Observation Simulator Package (COSP) where appropriate, to account for limitations in the satellite observations (e.g. clouds cannot be observed below the level of full lidar signal attenuation by spaceborne lidar; Chepfer et al., 2010), the different scales of the model grid compared to the satellite data and ensure to compare exactly the same variables in the model output as in the satellite products.
- 25 To further limit the impact of observational uncertainties we use several products from independent instruments and aim at identifying model biases in several of them. We also perform some of the analysis for different regions to study biases for different cloud types.

For studying past and present climate forcings it is indispensable to constrain the effective anthropogenic aerosol forcing due to aerosol-radiation and aerosol-cloud interactions (ERF<sub>ari+aci</sub>). Because of the large impact of clouds on radiation, the

30 representation of clouds in a global model can have an impact on ERF<sub>ari+aci</sub>. Therefore, we also investigate the difference in ERF<sub>ari+aci</sub> in the three ECHAM-HAM model versions and how they compare to differences in the simulations of clouds. As cloud feedbacks will have a large impact on temperature in a warmer climate we compare equilibrium climate sensitivity (ECS) and cloud feedbacks of the different model versions.

Section 2 gives a short description of the representation of clouds in ECHAM6.3-HAM2.3 and of the observational products applied in the model evaluation. In section 3 the results of the cloud evaluation, the comparison of  $ERF_{ari+aci}$ ,  $RF_{ari}$  and ECS in the ECHAM-HAM model versions are presented and discussed. The results are summarized in section 4 and conclusions are drawn.

#### 5 2 Methodology

#### 2.1 Model description

The global aerosol-climate model ECHAM-HAM is the combination of the global climate model ECHAM with the aerosol microphysics module HAM (Stier et al., 2005). The ECHAM5 and ECHAM6 model versions used in this study are described in Roeckner et al. (2003) and Stevens et al. (2013) respectively. The ECHAM-HAM model versions and configurations used in this study are described in separate studies: ECHAM5.5-HAM2.0 in Zhang et al. (2012), ECHAM6.1-HAM2.2 in Neubauer et al. (2014) and ECHAM6.3-HAM2.3 in Tegen et al. (20182019). For the sake of brevity, in the following ECHAM5.5-HAM2.0 will be referred to as E55H20, ECHAM6.1-HAM2.2 as E61H22 and ECHAM6.3-HAM2.3 as E63H23. In contrast to the 1-moment cloud microphysics scheme in the ECHAM base model (Lohmann and Roeckner, 1996), ECHAM-HAM uses a 2-moment cloud microphysics scheme. The 2-moment cloud microphysics scheme is described

- 15 in Lohmann et al. (2007), Lohmann and Hoose (2009) and recent changes and improvements applied in E63H23 in Lohmann and Neubauer (2018). A 2-moment cloud microphysics scheme is required to study aerosol-cloud interactions. In ECHAM-HAM cloud droplet activation and ice crystal nucleation from cloud condensation nuclei and ice nucleating particles are computed as well as in-cloud and below cloud scavenging. Therefore, ECHAM-HAM simulates aerosol-cloud interactions in liquid, mixed-phase and ice clouds. However, a 2-moment cloud microphysics scheme is not only a prerequisite for
- 20 simulating aerosol-cloud interactions but the additional information from the prognostic cloud droplet and ice crystal number concentrations can also improve the simulation of clouds compared to a 1-moment cloud microphysics scheme. The general representation of clouds in ECHAM-HAM is described in the literature given in this section but is briefly repeated in the subsections below for the convenience of the reader.

#### 2.1.1 Liquid stratiform clouds and wet scavenging

- 25 The scheme for stratiform clouds comprises prognostic variables for water vapor, cloud liquid and cloud ice, a cloud microphysics scheme and a diagnostic cloud cover scheme (based on Sundqvist et al., 1989). Cloud microphysics is represented by a 2-moment scheme described in Lohmann et al. (2007), Lohmann and Hoose (2009) and Lohmann and Neubauer (2018). Optionally available but not used in this study is the 1-moment scheme by Lohmann and Roeckner (1996). In ECHAM6.3 changes were made in the diagnostic cloud cover scheme to enhance the cloud cover for marine
- 30 stratocumulus clouds (Mauritsen et al., 2019). Condensation of cloud liquid water is based on moisture convergence (from transport by advection, turbulence and convection) and subsequent saturation adjustment. Evaporation of cloud liquid water

(or sublimation of cloud ice) occurs when the cloud cover decreases, or by transport of cloud liquid (or ice) mass into the cloud free part of a gridbox. For aerosol activation in liquid clouds the Köhler theory-based Abdul-Razzak and Ghan (2000) scheme is used. Its implementation is described in Stier (2016). Optionally available is the Lin and Leaitch (1997) aerosol activation scheme. Precipitation is computed diagnostically. Autoconversion of cloud droplets to rain follows Khairoutdinov

- 5 and Kogan (2000). Accretion of cloud droplets by rain (Khairoutdinov and Kogan, 2000) and evaporation of rain below clouds (based on Rotstayn, 1997) are also computed. Size dependent wet scavenging of aerosol particles in-cloud and below cloud follows Croft et al. (2009, 2010). The below cloud collection efficiencies as a function of aerosol and rain drop or snow crystal size are read from a look-up table. The in-cloud scavenging scheme takes nucleation scavenging and impaction scavenging of aerosol particles with cloud droplets and ice-crystals into account. For nucleation scavenging the number of
- 10 scavenged aerosol particles is computed for elouds warmer than <u>35°Cliquid cloud droplets</u> from the cloud droplet number concentration (CDNC), the ice crystal number concentration (ICNC) (after computation of cloud droplet evaporation and the numberprecipitation formation) and the fraction of activated aerosol particles computed by the activation scheme. For elouds colder than <u>35°Cice crystals</u> the aerosol particles are scavenged progressively from the largest to the smallest modes until the number concentration of scavenged aerosol particles is equal to the ice crystal number concentration (ICNC) (after
- 15 the computation of ice crystal sublimation and precipitation formation) of the grid box.

A downward scavenging tracer flux is computed for each model column each model time step. In-cloud and below cloud scavenging are sources for the downward scavenging tracer flux, while evaporation and sublimation of precipitation are sinks for the downward scavenging tracer flux. When the sink term is larger than the source term of the downward scavenging tracer flux in a model level, aerosol mass and number concentrations will be transferred to the respective atmospheric tracers i.e. aerosol is released from evaporating/sublimating precipitation at this model level back to the

atmosphere.

#### 2.1.2 Mixed-phase and cirrus stratiform clouds

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The cirrus scheme follows Kärcher and Lohmann (2002) and details are given in Lohmann et al. (2008). The ice crystals in cirrus clouds form by homogenous nucleation of supercooled liquid droplets. The scheme by Joos et al. (2010) for
 orographic cirrus clouds can optionally be applied to account for the higher updraft velocities in orographic cirrus clouds, therefore the depositional growth equation is solved for cirrus ice crystals (Kärcher and Lohmann, 2002). For mixed-phase clouds heterogeneous nucleation of supercooled cloud droplets is computed via immersion and contact freezing following Lohmann and Diehl (2006). Depositional growth of cloud ice in mixed-phase clouds is computed analogous to liquid clouds based on moisture convergence and subsequent saturation adjustment. In addition, the growth of ice crystals at the expense of cloud

droplets via the Wegener-Bergeron-Findeisen process (Wegener 1911; Bergeron 1935; Findeisen 1938) is implemented following Korolev (2007). Snow forms by aggregation of ice crystals, riming of cloud droplets by snow and accretion of ice crystals by snow. Sedimentation of ice crystals follows Rotstayn (1997). Sublimation and melting of ice crystals and snow

below clouds is also computed. Ice multiplication via rime splintering (Hallet-Mossop process) following Levkov et al. (1992) is optional (not used in this study).

#### 2.1.3 Convective clouds

The convective parameterization from Tiedtke (1989) with modifications for deep convection from Nordeng (1994) and for the triggering of convection from Stevens et al. (2013) is used. The convective parameterization uses only a 1-moment cloud microphysics scheme. Detrained condensate of convective clouds is added to stratiform clouds if they exist at the level of detrainment. Whether the condensate is detrained as liquid or ice is based on the same criteria as in the 2-moment stratiform cloud microphysics scheme in ECHAM-HAM. To obtain CDNC from-for the detrained condensate, several simplifications are applied. It is assumed that cloud droplets of convective clouds is weighted by detrained cloud water-will form at cloud

- 10 base. The number of activated cloud condensation nuclei (CCN) at the convective cloud base is computed using the vertical velocity from large scale and turbulent fluxes as described in section 2.1.4. It is further assumed that CDNC will be constant throughout the vertical extension of the convective clouds. At the level of detrainment these CDNC from the convective clouds will either evaporate or be added to stratiform clouds is weighted by if these exist at the level of detrainment. In the latter case a weighted average of the stratiform cloud water when addingCDNC and detrained CDNC to the CDNC of the
- 15 stratiform cloud in E63H23 (is computed by weighting stratiform CDNC with the stratiform liquid water content and detrained CDNC with detrained liquid water content. CDNC of the stratiform cloud is not allowed to decrease by this procedure). since cloud droplets will not evaporate in a supersaturated environment. The detrained ICNC is computed from the temperature dependent empirical relationship of Boudala et al. (2002). An alternative convection scheme based on the Convective Cloud Field Model (CCFM) (Wagner and Graf, 2010) with representation of aerosol-convection interactions is
- 20 available (Kipling et al., 2017; Labbouz et al. 2018) but not used in this study.

#### 2.1.4 Other processes

The sulfur cycle model of Feichter et al. (1996) forms the base of the sulfur chemistry module. Three sulfur species are treated prognostically: sulfur dioxide, dimethyl sulfide (DMS) and sulfate (the latter not only in the gas phase but also as an

- 25 aerosol). Three-dimensional climatological fields for oxidants are used i.e. ozone (O<sub>3</sub>), OH, H<sub>2</sub>O<sub>2</sub>, NO<sub>2</sub> and NO<sub>3</sub>. The nucleation scheme was implemented by Kazil et al. (2010) and is based on Kazil and Lovejoy (2007). Organic nucleation following Kulmala et al. (2006) or Kuang et al. (2008) can optionally be used. Sea salt, dust and DMS emissions are computed online based on near surface wind speeds (Stier et al, 2005; Tegen et al., 2002). The Long et al. (2011) sea salt parameterization (temperature dependent; Sofiev et al., 2011) is used in E63H23 and the Guelle et al. (2001) sea salt
- 30 parameterization is used in E55H20 and E61H22. Aerosol water uptake is computed via kappa-Köhler theory (Petters and Kreidenweis, 2007) as described in Zhang et al. (2012).

Radiative transfer is computed with the two-stream model PSrad (Pincus and Stevens, 2013). Turbulent fluxes in the atmosphere are computed with the turbulent kinetic energy (TKE) scheme of Brinkop and Roeckner (1995). The subgrid-scale vertical velocity that is needed for many cloud microphysical processes (e.g. cloud droplet activation, ice crystal nucleation, Wegener-Bergeron-Findeisen process) is computed from the TKE (Lohmann et al., 2007). Next to a single

5 characteristic updraft velocity for a gridbox which is based on TKE, there is also the option to represent the subgrid-scale variability of updraft velocities by a Gaussian probability density function (pdf) of updraft velocities (West et al., 2014). The subgrid-scale variability is again assumed to be due to turbulence and the width of the Gaussian pdf is therefore a function of TKE. The impact of the width of the Gaussian pdf on ERF<sub>ari+aci</sub> is discussed in West et al. (2014). The pdf-approach by West et al. (2014) is optionally available but not used in this study. The physics part of ECHAM6.3 as well as the 2-moment microphysics scheme for stratiform clouds in E63H23 are now energy conserving.

#### 2.1.5 Changes and improvements in E63H23

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Changes and improvements in E63H23 are also described in Lohmann and Neubauer (2018) and Tegen et al. (20182019) and are repeated here shortly for the convenience of the reader. From ECHAM6.1 to ECHAM6.3 the following improvements were made: (Mauritsen et al., 2019):

- New PSrad radiation scheme (Pincus and Stevens, 2013), which uses the Monte Carlo independent column
  approximation for fractional cloudiness and has the option for spectrally sparse but temporally dense calculations
  - Update of fractional cloud cover scheme, which improves the low-bias of marine stratocumulus clouds; this is
    motivated by the difficulty of representing the strong inversions of stratocumulus-topped marine boundary layers in
    global climate models (Mauritsen et al., 2019)
- Update of land model JSBACH (Reick et al., 2013), which uses a new five layer soil hydrology scheme
  - Removal of inconsistencies in the convection scheme, convective detrainment and the vertical diffusion scheme to conserve the atmospheric energy budget

The aerosol microphysics scheme HAM2.3 received the following improvements compared to HAM2.0:

- Update of mineral dust emission parameterization which makes use of a satellite-based source mask for Saharan dust sources (Heinold et al., 2016)
- New sea salt emission parameterization based on Long et al. (2011) which uses a temperature dependence following Sofiev et al. (2011)
- The latest version of the sectional aerosol module SALSA2.0 is implemented (described in Kokkola et al., 2018) (not used in this study)
- New emission datasets have been made available in an input file distribution for E63H23 for anthropogenic aerosol emissions (Global Fire Assimilation System (GFAS): Kaiser et al., 2012; Community Emissions Data System (CEDS): Hoesly et al., 2018; the latter is not used in this study)

Aerosol-cloud interactions were improved from HAM2.0 to HAM2.3 by the following changes:

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- The Köhler-theory based Abdul-Razzak and Ghan (2000) cloud droplet activation scheme (described in Stier, 2016) replaces the empirical Lin and Leaitch (1997) activation scheme
- The in-cloud scavenging scheme by Croft et al. (2010) combines a diagnostic nucleation scavenging scheme with a size-dependent impaction scavenging parameterization and replaces prescribed (size-dependent) aerosol scavenging fractions
- Changed treatment of detrained cloud water mass and number concentrations from convective clouds: CDNC is weighted byfrom detrained cloud water when adding itadded (weighted average) to theCDNC of a stratiform cloud cannot decrease CDNC of the stratiform cloud; split between liquid water and ice of detrained condensate is made consistent between mass and number concentrations
- In mixed-phase clouds the heterogeneous freezing by immersion freezing of black carbon particles is limited to
  particles in the accumulation mode and coarse mode

The 2-moment stratiform cloud microphysics scheme in ECHAM-HAM received the following improvements from Lohmann and Hoose (2009) to E63H23:

- Ice crystals are assumed to have a shape of hexagonal plates which covers the whole size range of ice crystals and the shape is consistent in all modules
  - Sticking efficiency used in the accretion of ice crystals by snow has been changed to the one used in Seifert and Beheng (2006)
  - Two settings for minimum CDNC are available: 40 cm<sup>-3</sup> or 10 cm<sup>-3</sup>
- 20 Further technical improvements, bugfixes and minor corrections in E63H23 include:
  - Removal of an inconsistency in the fractional cloud cover and cloud microphysics schemes in ECHAM6.3, which had led cloud cover to be either 0 or 1 in ECHAM6.1
  - Removal of inconsistencies in the kappa-Köhler water uptake in HAM2.3
  - Modularization of the 2-moment stratiform cloud microphysics scheme
- Removal of an inconsistency for convective detrainment in the 2-moment stratiform cloud microphysics scheme to conserve the atmospheric energy budget
  - Removal of an inconsistency in the 2-moment stratiform cloud microphysics scheme which led to homogeneous freezing of dry aerosol particles, independent of availability of water vapor below -35°C
  - CDNC/ICNC can no longer grow and in the same timestep evaporate or sublimate
- No more CDNC at temperatures colder than 238.15K, no more ICNC at temperatures warmer than 273.15K
  - Update of default settings, run templates and run organization: the vertical resolution is per default 47 vertical model layers; the reference year and reference period for present day simulations are 2008 and 2003-2012 respectively

#### 2.2 Experiment description

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For each of the three model configurations, E55H20, E61H22 and E63H23 three types of experiments were conducted to evaluate the clouds in present day climate,  $ERF_{ari+aci}$  and the equilibrium climate sensitivity (ECS) (Table 1). The simulation setup was chosen to be as similar as possible for the three model versions to minimize the impact of boundary conditions on the model version comparison. The setup represents the standard setup of E63H23 which is a compromise between which processes are represented in the model and the computational performance. Climatological monthly mean mixing ratios of

oxidants from an eight-years (2003-2010) mean Monitoring Atmospheric Composition and Climate (MACC) reanalysis (Inness et al., 2013) are used in E61H22 and E63H23. For E55H20 the climatological monthly mean mixing ratios of oxidants are from simulations with the MOZART model for present day conditions (Horowitz et al., 2013).

#### 10 2.2.1 Present day climate simulation (PD)

10-year simulations for PD conditions were done for all model versions. Previous studies using E55H20 or E61H22 often useused the year 2000 as the reference year or 2000-2009 as the reference period for present day simulations (Zhang et al., 2012; Neubauer et al., 2014), therefore we also use the period 2000-2009 for the PD simulations of E55H20 and E61H22. For E63H23 the default model setup has changed and the reference year and reference period for present day simulations are

- 15 now 2008 and 2003-2012 respectively. This has become necessary because of the relatively large changes in aerosol emissions in recent years in several regions (Hoesly et al., 2018) and was aided by the availability of new boundary condition datasets for the Coupled Model Intercomparison Project (CMIP) Phase 6 (CMIP6). Time varying (RCP8.5) ACCMIP MACCity (AEROCOM II ACCMIP) aerosol emissions were used for E63H23 and E61H22. The biomass burning emissions are based on observations until 2008 in ACCMIP MACCity, afterwards the biomass burning emissions are from
- 20 the RCP8.5 emission scenario. For E55H20 the AEROCOM I emission for the year 2000 are applied for all years. The greenhouse gas concentrations are set to year 2008 (RCP8.5) concentrations in all model versions. All model versions also use a climatology for monthly values of sea surface temperature (SST) and sea ice cover (SIC) derived from AMIP data (Taylor et al., 2000) of the years 2000-2015. The spectral horizontal resolution is T63 (~1.9°x1.9°) in all model versions. For E55H20 and E61H22 31 vertical model layers (L31) are used (as in the default configuration of these <u>ECHAM-HAM</u> model versions), the model top is 10 hPa. For E63H23 47 vertical model layers (L47) are used (new default configuration of the set of the section of the section of the set of the section of the section of the set of the section of the section of the section of the section of the set of the section o
- 25 versions), the model top is 10 hPa. For E63H23 47 vertical model layers (L47) are used (new default configuration of E63H23), with a model top at 0.01 hPa. The lowermost levels of L31 and L47 (up to about 100 hPa) are identical. A comparison of E63H23 simulations with L31 and L47 showed only minor differences (see Table S1 and Fig. S1). Therefore, we expect also no large differences by using different vertical grids for the different model versions.

#### 2.2.2 ERF<sub>ari+aci</sub> and RF<sub>ari</sub> simulations (PD<sub>aer</sub>/PI<sub>aer</sub>)

30 To compute  $ERF_{ari+aci}$  two 20-year simulations were conducted, one with present day aerosol emissions (PD<sub>aer</sub>) and one with pre-industrial aerosol emissions (PI<sub>aer</sub>). The simulations were otherwise identical. For the PD<sub>aer</sub> simulation the PD simulation

was extended to cover the years 2000-2002 and 2013-2019 (or the years 2010-2019 for E55H20 and E61H22). The same greenhouse gas concentrations and SST and SIC climatology as in the PD simulations have been used. For the time period 2013-2019 the ACCMIP MACCity (AEROCOM-II-ACCMIP) aerosol emissions for the year 2008 were used for all years for E63H23 and E61H22. For E55H20 the AEROCOM I emission for the year 2000 are applied for all years (2000-2019) for

5 the PD simulation. For the PI<sub>aer</sub> simulation the aerosol emission for the year 1850 from ACCMIP MACCity (AEROCOM-II-ACCMIP) were used for E63H23 and E61H22 and the ones for the year 1750 from AEROCOM I for E55H20. ERF<sub>ari+aci</sub> is computed as the difference in top of atmosphere (TOA) net radiative flux (net<sup>*TOA*</sup>) between the PD<sub>aer</sub> and the PI<sub>aer</sub> simulation:

$$ERF_{ari+aci} = \operatorname{net}_{PDasr}^{TOA} - \operatorname{net}_{Plasr}^{TOA}.$$
(1)

- 10 The simulation time was 20 years to increase the signal of  $ERF_{ari+aci}$  compared to variations in TOA net radiative flux due to internal variability of the elimate systematmosphere. The use of an identical climatology for SST/SIC in all simulations reduces the internal variability compared to simulations with a global climate model (GCM) coupled to a full ocean model. The radiative forcing due to aerosol-radiation interactions (RF<sub>ari</sub>) is computed from the same pair of simulations as  $ERF_{ari+aci}$ (PD<sub>aer</sub>/PI<sub>aer</sub>). The direct aerosol radiative effect is computed by double calls to the radiation, once with the prognostic aerosol
- 15 and once without aerosol. RF<sub>ari</sub> is computed as the difference of the direct aerosol radiative effect between the PD<sub>aer</sub> and the PI<sub>aer</sub> simulations at TOA, once for all-sky and once for clear-sky (CS) conditions:

$$RF_{ari} = \left(\operatorname{net}^{TOA} - \operatorname{net}^{TOA}_{no\_aer}\right)_{PD_{aer}} - \left(\operatorname{net}^{TOA} - \operatorname{net}^{TOA}_{no\_aer}\right)_{PI_{aer}},$$
(2a)

$$RF_{ari,CS} = \left(\operatorname{net}^{TOA,cs} - \operatorname{net}^{TOA,cs}_{no\_aer}\right)_{PD_{aer}} - \left(\operatorname{net}^{TOA,cs} - \operatorname{net}^{TOA,cs}_{no\_aer}\right)_{PI_{aer}}.$$
(2b)

20 Note that we did not follow the protocol in Myhre et al. (2013) for all-sky conditions, therefore our all-sky RF<sub>ari</sub> is somewhat affected by changes in clouds from pre-industrial to present day simulations. This has no large impact on the regional analysis for RF<sub>ari</sub> our study. The reason why we did not follow Myhre et al. (2013) is that we include indirect aerosol effects in our simulations.

#### 2.2.3 ECS simulations (1xCO<sub>2</sub>/2xCO<sub>2</sub>)

25 To compute ECS, ECHAM-HAM was coupled to a mixed-layer ocean to compute two 50-year simulations, one with preindustrial CO<sub>2</sub> concentrations (1xCO<sub>2</sub>) and one with doubled pre-industrial CO<sub>2</sub> concentrations (2xCO<sub>2</sub>). The first 25 years of the simulations were used as spin-up time for the (50 m deep) mixed-layer ocean. ECS was then computed from the difference in global mean surface temperature ( $T_s$ ) between 2xCO<sub>2</sub> and 1xCO<sub>2</sub> from the last 25 years of the simulations:

$$ECS = T_s^{2 \times CO_2} - T_s^{1 \times CO_2}.$$
(3)

Pre-industrial concentrations for well-mixed greenhouse gases other than  $CO_2$  were used in all simulations as well as preindustrial aerosol emissions (similar to  $PI_{aer}$ ). The ocean heat flux corrections required by the mixed-layer ocean to maintain present-day sea surface temperatures were computed for each model version by extending the respective  $PD_{aer}$  simulations another 5 years to a total of 25 years.

#### 5 2.3 Tuning strategy

Following Hourdin et al. (2017) who argue that estimating uncertain parameters in model development is an important process that should be made transparent we document here our tuning strategies and targets. Tuning is needed mainly to ensure that the TOA radiative fluxes are balanced, and model tuning is limited to adjusting global mean properties. We start from the ECHAM6.3 parameter settings and adapt mainly parameters related to the cloud and convection scheme for tuning

- 10 E63H23. The tuning strategy and parameters for ECHAM6 as well as the impact of these parameters on the model climate are described in Mauritsen et al. (2012, 2019). The tuning parameters for ECHAM6-HAM2 and their impact on climate are described in Lohmann and Ferrachat (2010). The parameters that were used in the tuning of the ECHAM-HAM versions and that have different values in E55H20, E61H22 and E63H23 are shown in Table 2.
- The primary tuning target for E63H23 is to match the global mean observed shortwave (SW) and longwave (LW) TOA fluxes within the range of uncertainty of the observations and that the net radiative imbalance TOA is close to the observed present day value. The secondary tuning target is that the SW, LW and net cloud radiative effect (CRE) TOA are within the range of uncertainty of the observations. Also cloud cover (CC), liquid water path (LWP), ice water path (IWP), total precipitation (P) and aerosol optical depth (AOD) should be close to the range of observed values (see Table 3).
- The tuning is done with short one-year simulations with a climatology for SST and sea ice. When a set of parameters has 20 been found one or more ten-year simulations are done to minimize the uncertainty in net radiative imbalance TOA. For E61H22 the default parameter values are used (Neubauer et al., 2014). For E55H20 it was necessary to retune the model with the tuning strategy described above as the tuning in Zhang et al. (2012) was undertaken for nudged simulations and we performed free simulations to compare the three ECHAM-HAM model versions. The largest differences in tuning between the three model versions are in the tuning parameters for autoconversion of cloud droplets to rain and entrainment for
- 25 shallow convection. The latter was adopted from the base model ECHAM6.3 (see Mauritsen et al., 2019 for a discussion of the impact of the change in this tuning parameter on climate sensitivity). In E63H23 the stratiform rain formation by autoconversion will be faster than in the other two model versions. This is due to the larger value of the respective tuning parameter leading to reduced LWP, CC, SW CRE and more positive net radiative imbalance TOA in E63H23 (Lohmann and Ferrachat, 2010). The larger value of the tuning parameter for entrainment for shallow convection in E61H22 and the even
- 30 larger value in E63H23 have the opposite effect, increased LWP, CC, SW CRE and more negative net radiative imbalance TOA (Mauritsen et al., 2012). For E63H23 there is a compensation by changing both tuning parameters, the most pronounced net effect is a reduced LWP compared to the two other model versions (we hypothesize that LWP is reduced since entrainment for shallow convection affects mainly low, thin clouds, whereas the autoconversion rate affects all liquid

clouds; since the reflectivity of clouds depends non-linearly on their thickness, an increase in thin low clouds can compensate the SW CRE change by the decrease in thicker clouds but lead to a lower global mean LWP).

#### 2.4 Observational products

We list here the products and the respective references for the observational products used in the model evaluation. From Moderate-resolution Imaging Spectroradiometer (MODIS-AQUA) collection 6.1 (Platnick et al., 2015, 2017) and from the ESA Cloud Climate Change Initiative (CCI)-Advanced Very-High-Resolution Radiometer (AVHRR-PM) v3.0 (prototype; Stengel et al., 2017a, 2017b), histograms of cloud top pressure vs. cloud optical depth and CC are taken and from MODIS also LWP. Histograms of cloud top pressure vs. cloud optical depth were also taken from the International Satellite Cloud Climatology Project (ISCCP; Rossow and Schiffer, 1999) D1 data. Cloud-Aerosol Lidar and Infrared Pathfinder Satellite

- 10 Observations (CALIPSO) data for CC is from the GCM-Oriented CALIPSO Cloud Product (GOCCP) dataset (Chepfer et al., 2010). Cloud radiative effect data is from the Clouds and the Earth's Radiant Energy System (CERES) Energy Balanced and Filled (EBAF) TOA edition-4.0 data product (Loeb et al., 2018). Precipitation data is from the Global Precipitation Climatology Product (GPCP) 2.3 (Adler et al., 2018). Cloud top CDNC are from the climatology of Bennartz and Rausch (2017). LWP data are from the Multi-Sensor Advanced Climatology of LWP (MAC-LWP; Elsaesser et al., 2017), an
- 15 updated version of the University of Wisconsin LWP climatology. IWP is from satellite observations compiled by Li et al. (2012).

#### **3 Results and Discussion**

#### 3.1 Global mean comparison to observations

- Table 3 includes global mean values of radiation, cloud and aerosol related variables of the PD simulations of E55H20, E61H22 and E63H23 compared to observations (OBS) or multi-model mean values (MMM) when observations are not available. The global mean values of the radiative fluxes shown in Table 3 are tuning targets (see section 2.6) and therefore cannot be used directly for model evaluation. For E63H23 the SW and LW TOA fluxes as well as the SW and LW CRE TOA are within the range of the observations. The net TOA flux of E63H23 is also close to the observations (additional tuning could bring it closer to the observed value but was not attempted given the large uncertainty in e.g. SW and LW TOA
- 25 fluxes). The SW, LW and net TOA fluxes of E61H22 and E55H20 are outside the range of observations. This reflects the change in the tuning targets/strategy in E63H23 and the availability of better observations. The net CRE of E63H23 (and also E55H20 and E61H22) is outside the observed range. It was not possible to find parameter settings which bring the net CRE within the range of observations without bringing one or more of the other radiative fluxes outside of the range of observations. This is a first indication of a structural problem in ECHAM-HAM, which could be related to how ice crystals
- 30 nucleate in (warming) cirrus clouds or an underestimation of (cooling) stratocumulus. This will be further discussed in the evaluation. CC, P and cloud top cloud droplet number concentration (CDNC) of all three model versions agree fairly well

with observations (for cloud-top CDNC of the model simulations we selected CDNC over ocean only of the topmost layer of clouds with a cloud top temperature > 273.15 K). For LWP a climatology based on microwave sensors (Elsaesser et al., 2017) provides reliable observations as long as the ratio of LWP to LWP+rain water path is large (>0.8 is used here), i.e. in regions with relatively low precipitation. The values of LWP only in this low precipitation regions (LWP-LP) are also shown

- 5 in Table 3. Whereas the mean values over the global oceans for LWP and LWP-LP of E61H20 and E55H20 are higher than observed, E63H23 shows values within the observational range (71 and 76 g m<sup>-2</sup> respectively). This is due to the more physically based activation scheme in E63H23 and improvements in ECHAM6.3 like energy conservation in the physics part and improvements in the cloud cover scheme for marine stratocumulus clouds, which allow to increase the tuning parameter for autoconversion (see Table 2). Similarly, the global mean value of IWP in E63H23 with 15 g m<sup>-2</sup> is only slightly below
- 10 the observational range (18-32 g m<sup>-2</sup>), whereas in E61H22 and E55H20 IWP is considerably smaller (10 and 8 g m<sup>-2</sup> respectively). This is because in the accretion of ice crystals by snow, the sticking efficiency follows Seifert and Beheng (2006) in E63H23, whereas in E55H20 and E61H22 it followed Levkov et al. (2012). The Seifert and Beheng (2006) sticking efficiency leads to a less efficient removal of ice crystals by snow. Furthermore, the changed sticking efficiency allows to reduce the stratiform snow formation rate by aggregation compared to earlier model versions (see Table 2), which
- 15 also increases IWP. The aerosol mass burdens of the five prognostic aerosol species in ECHAM-HAM are within the range of AeroCom models (Textor et al., 2006), except for particulate organic matter (POM). This may be related to the simplistic treatment of secondary organic aerosol (SOA) in all three model versions in the experiments for this study. Details of the evaluation of E63H23 with respect to the atmospheric aerosol are given in Tegen et al. (20182019).

#### 3.2 Zonal mean comparison to observations

- 20 Although the global mean values are tuning targets (see section 2.6), biases in net CRE and IWP in the ECHAM-HAM versions, which could not be brought in agreement with observations via tuning, were identified in the previous section. Zonal mean values of observable variables can nevertheless be used for model evaluation because tuning targets the global mean quantities. Fig.1 shows zonal mean distributions of several quantities for the three model versions and observations. The zonal distribution of SW CRE and LWP-LP of E63H23 agree relatively well with observations, whereas in E61H22 and
- 25 E55H20 the magnitude of both quantities is overestimated in mid-latitudes. The cloud overcover distribution of E63H23 also agrees well with observations, whereas E61H22 and E55H20 show an underestimation by up to 10 percentage points in the subtropics. Biases in cloud top CDNC are more complex and retrievals of cloud top CDNC are only possible for specific clouds (e.g. horizontally homogeneous, unobscured, optically thick clouds) and rely on assumptions like that the liquid water content increases with altitude like in an adiabatically rising cloud parcel (or at least like a constant fraction of this liquid
- 30 water content) and that CDNC are constant throughout the cloud and further assumptions which together make cloud top CDNC retrievals uncertain (Grosvenor et al., 2018). We therefore expect larger differences between observations and models for cloud top CDNC than for other variables. E55H20 agrees well with MODIS observations in the tropics but overestimates cloud top CDNC in the subtropics on both hemispheres and mid latitudes in the Southern Hemisphere. E61H22

overestimates cloud top CDNC in the tropics and subtropics but underestimates it at mid latitudes in the Northern Hemisphere. E63H23 also overestimates cloud top CDNC in the subtropics, however less than E61H22, and also in the tropics. The liquid phase of clouds is therefore better represented in E63H23 than in the previous model versions. IWP is underestimated in all three model versions. E63H23 has the smallest bias, followed by E61H22 and E55H20 shows the

- 5 largest deviation from observed zonal mean IWP. The underestimation is particularly large in the tropies. The underestimation is particularly large in the tropics, which is likely connected to the parameterization of convection in ECHAM(-HAM). ECHAM has a low precipitation bias over land in the tropics (Mauritsen et al., 2012; Stevens et al., 2013). Gasparini et al. (2018) found indications that the level of detrainment from deep convection is too low in altitude in ECHAM-HAM. They lowered the tuning parameter for deep convective entrainment  $\epsilon_d$  to 0.00006, whereas all three
- 10 ECHAM-HAM versions used here have to use a larger value for this parameter (Table 2) as they use a cirrus scheme in which cirrus clouds can only nucleate homogeneously, which may lead to an overestimation of ICNC and underestimation of IWP by tuning of radiative fluxes (see section 3.3). For LW CRE (and precipitation and AOD, not shown) all three model versions differences are within the range of different observational products. In the tropics E55H20 has a rather strong LW CRE, whereas E63H23 and E61H22 have a rather weak LW CRE but all are within the range of observations.

#### 15 3.3 Regional comparison to observations

The comparison of CRE of the different model versions with CERES CRE reveals several biases in the representation of clouds. We start therefore by identifying biases in CRE and use then observations for other quantities to identify causes of the model biases. In Fig. 2 the differences in SW, LW and net TOA CRE of all model versions to CERES observations are shown. In all three model versions the (negative) SW CRE is too weak in the marine stratocumulus regions west of the continents (the average bias in the wider stratocumulus regions is 1.1, 8.1 and 7.0 W m<sup>-2</sup> in E63H23, E61H22 and E55H20 respectively). In addition the SW CRE is too weak in some land areas in E63H23 and E61H22, in the Southern Ocean in E63H23 and in the tropical oceans in E55H20 (3.3 W m<sup>-2</sup> average bias over ocean between 15°N and 15°S excluding wider stratocumulus regions). These biases are compensated by a stronger SW CRE over large parts of the oceans and mid and high latitude land areas in the Northern Hemisphere. The bias in stratocumulus regions is smaller in E63H23 than in the 25 older model versions and so are the compensating negative biases. This is due to improvements in ECHAM6.3 like

- improvements in the cloud cover scheme for marine stratocumulus clouds. The (positive) LW CRE is too weak in the tropics in E63H23 and E61H22 (-7.7 and -4.8 W m<sup>-2</sup> average bias respectively between 20°N and 20°S excluding wider stratocumulus regions) and too strong in the tropics (in particular over land) in E55H20 (1.2 W m<sup>-2</sup>). Together with the biases in the tropical SW CRE this points to problems with the parameterization of deep convective clouds, detrained
- 30 condensate or the representation of anvils from detrained condensate in all three model versions. In all three model versions the LW CRE in mid-latitudes is too weak (except over land in the Northern Hemisphere). At high latitudes it is stronger than in the CERES data in all model versions (but also the uncertainty of CERES CRE is larger at high latitudes, Loeb et al., 2018). Only few of the biases in SW and LW CRE compensate, therefore the biases in net CRE are as large as or larger than

in the SW (LW) CRE. In the net CRE the positive biases in stratocumulus regions as well as in the Southern Ocean in E63H23 and E61H22, over land in E61H22 and in the tropical oceans in E55H20 are compensated in the global mean by negative biases in all other regions. The negative biases are caused by adjusting cloud parameters to bring the global mean values in agreement with observations. Therefore, if the biases in stratocumulus regions and the Southern Ocean (and the tropics) could be reduced, the negative biases in SW and net CRE would also be smaller.

In Fig. 3 the cloud cover of the CALIPSO GOCCP product and all three model versions are shown. The hatched areas in Fig. 3 are the regions where the cloud cover of CALIPSO GOCCP, MODIS collection 6.1 and ESA Cloud CCI (AVHRR-PM) differ by more than five percentage points. We use therefore only the areas not marked by hatching in Fig. 3 for the model evaluation. Since the COSP CALIPSO simulator is not implemented in E55H20, the direct model output is shown for all

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- 10 model versions (see Fig. S8 for COSP CALIPSO simulator output of cloud cover for E61H22 and E63H23). The cloud cover of all three model versions agrees fairly well with the observations. The largest biases are in stratocumulus regions west of the continents (-10, -18 and -22 percentage points in E63H23, E61H22 and E55H20 respectively in the wider stratocumulus regions), where the models underestimate the cloud cover. Over land in the Northern Hemisphere poleward of about 45°N the models overestimate cloud cover and in the Indonesian warm pool region the cloud cover is biased high in the three
- 15 model versions. The underestimation of cloud cover in stratocumulus regions is less severe in E63H23 than in the other two model versions (the cloud cover scheme in ECHAM6.3 was improved to better represent cloud cover in these regions). Fig. 4 shows LWP from the MAC-LWP climatology (Elsaesser et al., 2017) and the three model versions. The retrieval of LWP has biases both from visible/near infrared sensors as well as microwave sensors (Seethala and Horváth, 2010; Lebsock and Su, 2014). Visible/near infrared sensors such as MODIS have problems when the solar zenith angle is large or at
- 20 detecting pixels of clouds at low altitudes (Lebsock and Su, 2014). Retrievals of microwave sensors such as AMSR-E may retrieve LWP in cloud free scenes and the split between LWP and rain water path is difficult (Lebsock and Su, 2014). Elsaesser et al. (2017) corrected the retrieval bias of LWP of microwave sensor based products in cloud free scenes. And they recommend to use regions with low precipitation (LWP/(LWP+rain water path)>0.8) for model evaluation. The regions where precipitation could influence the LWP retrieval are therefore hatched in Fig. 4. This leaves the stratocumulus regions
- 25 west of the continents and the storm tracks over ocean in the Northern and Southern Hemisphere as the most reliable areas for the evaluation of LWP. All three model regions show a fairly good agreement of LWP in the stratocumulus regions except west off South America and southwest Africa, where all model versions tend to underestimate LWP. In the storm tracks over ocean in the Northern and Southern Hemisphere on the other hand E61H22 and even more E55H20 overestimate LWP. E63H23 shows instead a rather good agreement of LWP in the storm tracks compared to observations. This is likely
- 30 the result of different model tuning in E63H23 (see section 2.3), which was possible due to a more realistic geographic pattern of cloud cover and SW CRE in E63H23.

To further characterize the simulation of liquid clouds in the ECHAM-HAM model versions we also compare cloud top CDNC of warm (cloud top warmer than 0°C) liquid clouds to the cloud top CDNC from Bennartz and Rausch (2017), which is based on MODIS Aqua data (Fig. 5). The hatched area marks regions where the relative uncertainty in the observations is

larger than 75%. The general geographical distribution and magnitude of cloud top CDNC of all model versions agree with the observations, although there are certain areas where model biases are apparent. E61H22 has lower cloud top CDNC concentrations in mid latitude ocean regions than E63H23, in E55H20 they are higher than in the other two model versions. In E55H20 the higher cloud top CDNC concentrations can be explained by reduced entrainment of deep convection (see

- 5 Table 2) compared to the other model versions which leads to higher relative humidity in the upper tropical troposphere which subsequently leads to increased aerosol nucleation, more Aitken mode particles and increased eloud condensation nuclei (CCN) concentrations. E63H23 uses the Abdul-Razzak and Ghan (2000) activation scheme and the Long et al. (2011) sea salt emission parameterization (temperature dependent; Sofiev et al., 2011) which lead to higher cloud top CDNC concentrations than in E61H22 (which uses the Lin and Leaitch, 1997 activation scheme and the Guelle et al, 2001 sea salt
- 10 emission parameterization) despite the lower LWP in E63H23. Furthermore, in subtropical regions where the cloud cover and LWP are low (see Figs. 3 and 4), cloud top CDNC are higher in all three model versions than in the observations. In these regions, shallow trade-wind cumulus clouds occur frequently (Medeiros and Stevens, 2011) and in all model version shallow convection is triggered frequently (see Fig. S2). For convective clouds a 1 moment cloud microphysics scheme is used in ECHAM HAM and these clouds are considered to be short lived. But the detrained condensate of these clouds can
- 15 add to existing large scale clouds. To obtain CDNC for the detrained condensate, the number of activated CCN at the convective cloud base (computed using the vertical velocity from large scale and turbulent fluxes as described in section 2.1.4) is propagated as CDNC to the level of detrainment. The detrained CDNC are then added to an existing stratiform cloud (otherwise they dissipate) by taking an average of the stratiform CDNC and detrained CDNC (weighted by their respective LWP). This weighted average may overestimate the CDNC concentration of shallow cumulus clouds. The
- 20 weighted average of stratiform CDNC and detrained CDNC (see section 2.1.3) may overestimate the CDNC concentration of shallow cumulus clouds. The use of a 2-moment cloud microphysics scheme for convective clouds (e.g. Lohmann, 2008) so that CDNC in convective clouds can be reduced by collision-coalescence or a different way to account for detrained CDNC could help to alleviate this model bias. All three model versions also underestimate cloud top CDNC at high latitudes. As retrievals of visible/near infrared sensor often have biases at large zenith angles (see above) this may be a problem with the
- 25 observations.

In Fig. 6 the IWP of all three model versions and IWP satellite observations compiled by Li et al. (2012) are shown. Li et al. (2012) used three different CALIPSO plus CloudSat ice water products and two different methods to remove the contribution of convective clouds and precipitation from the products. Fig. 6 displays the compiled mean IWP of the datasets of Li et al. (2012) and areas where the relative standard deviation of the different datasets is larger than 75% are hatched.

30 The regional distribution of <u>occurrence of IWP</u> of all three model versions agrees in general quite well with the observations. <u>IWP</u> although it is biased low in all <u>ECHAM-HAM</u> model versions, as was found for. This could already be seen in the respective global mean and zonal mean values (see sections 3.1 and 3.2). Similar to what was found in the analysis of zonal mean IWP the underestimation is largest in the tropics. (see section 3.2). Cloud ice mass vertical profiles can be obtained from CALIPSO plus CloudSat observations. The global mean vertical profile of IWC is shown in Fig. 7 for all three model versions and the compiled mean IWC from Li et al. (2012). IWC is underestimated above 700 hPa in all model versions. In E63H23 the maximum of IWC is at the same pressure level as in the observations, at about 350 hPa, whereas in E61H22 and E55H20 the maximum of IWC is at higher altitudes at about 300 to

- 5 250 hPa. This can be explained by changes in ICNC and subsequent changes in precipitation formation and ice crystal sedimentation. ICNC changed between the model versions because of the way detrained ice crystals are added to existing stratiform clouds was changed since E61H22. The shape of the ice crystals was made consistent in all modules since E61H22 and a bug in E61H22 was removed in E63H23 which led to homogeneous freezing of dry aerosol particles, independent of availability of water vapor below -35°C (the latter improvement was most important as it doubled the ICNC
- 10 burden in E61H22 compared to the two other model version; see Table 3). Below 700 hPa the three model versions are close to the observed IWC, with E63H23 showing an overestimation of IWC and E55H20 an underestimation. The regions where IWP is underestimated in the three model versions correspond to the regions where the three model versions underestimated LW CRE in Fig. 2 (in particular in the tropics). There are also regions where LW CRE is overestimated (see Fig. 2) in the three model versions although IWP is underestimated (see Fig. 6). This is an indication that
- 15 ICNC is too large in the three model versions (the vertical profile of IWC agrees fairly well with observations although the IWC magnitude is underestimated in all three model versions). As IWP is larger in E63H23 than in E61H22 and E55H20 but the overestimation in LW CRE is smaller in E63H23 than in E61H22 and E55H20, this is an indication that ICNC and the size of the ice crystals are closer to reality in E63H23 than in E61H22 and E55H20. The overestimation of LW CRE in E61H22 around 60°N and 60°S can be explained by the high ICNC in E61H22 (see Table 3) caused by the ICNC bug mentioned above.
- 20 mentioned above.

Next to E61H22 there is also a bias of net CRE south of 60°S in E63H23 (see Fig. 2). This is not due to too high ICNC as LW CRE of E63H23 agrees well with CERES observations in this region. In E63H23 there is an underestimation of SW CRE south of 60°S. Cloud cover and IWP of E63H23 agree well with observations in this region. LWP is slightly underestimated, but cloud top CDNC is strongly underestimated. Either there is a problem with the satellite retrievals at

- 25 these high southern latitudes or E63H23 is missing liquid clouds in this region. In E61H22 and E55H20 this possible bias south of 60°S is hidden by the overestimation in LWP which leads to a stronger SW CRE. In Fig. 8 the total precipitation of all model versions and GPCP2.3 (Adler et al., 2018) is shown. Areas where the relative uncertainty of the GPCP2.3 data is larger than 75% are hatched. Despite the biases in the representation of clouds in the
- three model versions identified above, the geographical distribution and magnitude of the annual mean precipitation of all
  model versions agrees well with the observations. Only in the intertropical convergence zone (ITCZ) and South Pacific
  convergence zone (SPCZ) the areas and magnitude of precipitation differsdiffer from the observations, corresponding to
  differences in cloud cover and IWP (Fig. 3 and Fig. 6, respectively). Cloud cover, IWP and precipitation are low in the
  central Pacific and central Atlantic ITCZ but relatively large in the ITCZ west off Central America, east off South America,
  over the Philippines and west off Southeast Asia. In the SPCZ cloud cover, IWP and precipitation are relatively large

compared to the respective observations. <u>ECHAM underestimates tropical precipitation over land and overestimates tropical precipitation over ocean (Mauritsen et al., 2012; Stevens et al., 2013). This bias can be seen in Fig. 8 also for all ECHAM-HAM versions. Since ECHAM and ECHAM-HAM use the same parameterizations for convective clouds, this bias is very likely inherited from the base model ECHAM.</u>

- 5 In Fig. 9 histograms of cloud top pressure vs. cloud optical depth of ECHAM-HAM are compared to ISCCP, AVHRR-PM and MODIS observations. The COSP simulator (Bodas-Salcedo et al., 2011) was not implemented in E55H20 so we only compare E61H22 and E63H23 to the observations. We applied the COSP-ISCCP-simulator for E61H22 and E63H23 for comparison to ISCCP and AVHRR-PM. The COSP-MODIS-simulator is only implemented in E63H23 so we compare only E63H23 to MODIS. The histograms were produced for four regions (shown in Fig. 2): wider stratocumulus regions, mid-
- 10 latitudes, tropics and 60°N to 60°S. The five marine stratocumulus regions are west off North and South America, west off northern and southern Africa and west off Australia. The marine stratocumulus regions are extended to the west to cover approximately the regions were the three model versions underestimate SW CRE (see Fig. 2). The mid-latitudes regions are 60°N to 20°N and 20°S to 60°S, excluding the areas covered by the wider stratocumulus regions. The tropics are between 20°N to 20°S, excluding the areas covered by the wider stratocumulus regions. The region 60°N to 60°S is the sum of the
- 15 wider stratocumulus, mid-latitudes and tropics. 60°N to 60°S was chosen because retrievals of visible/near infrared sensors often have biases at large zenith angles (see above). Several of the biases described above are also seen in the histograms in Fig. 9. In the region 60°N to 60°S E63H23 and E61H22 simulate too many optically thick clouds and too few optically thin clouds at low and mid-levels compared to the three satellite datasets. Stevens et al. (2013) found a similar biasNam et al. (2012) found a similar bias in several Fifth phase of the Coupled Model Intercomparison Project (CMIP5) models and
- 20 Stevens et al. (2013) in ECHAM6.1 ("too few, too bright"). This bias can also be seen in the mid-latitudes and in the tropics. In the wider stratocumulus region on the other hand the occurrence of low-level optically thick clouds of E61H22 and E63H23 agrees rather well with those of the three observational datasets. In the wider stratocumulus regions the optically thin low level (and mid-level) clouds are missing. This agrees with the analysis of SW CRE that the underestimation in the stratocumulus regions is compensated by a stronger SW CRE (clouds being optically thicker) in other regions (by model
- 25 tuning). Therefore, if the bias in stratocumulus regions could be reduced, also the biases in SW CRE and cloud optical depth in other regions could be reduced. In the mid-latitudes the optical depth of low, mid- and high level clouds is larger in E61H22 than in E63H23 and ISCCP and AVHRR-PM. This is related to the stronger compensation by tuning for the lack of clouds in stratocumulus regions (removal of LWP by autoconversion is slower in E61H22 than in E63H23; see Table 2), and to the ICNC bug in E61H22 mentioned above. In the mid-latitudes and the tronics there is also a lack of high level clouds
- 30 with optical depth between 1.3 and 23 in E63H23 and E61H22. This lack of cirrus clouds corresponds to the underestimation of IWP and LW CRE. Gasparini et al. (2018) evaluated the cirrus clouds in a version of E61H22 that included a cirrus cloud scheme which accounts for a competition in cirrus cloud formation by homogeneous nucleation of solution droplets, heterogeneous freezing of ice nucleating particles and water vapor deposition on pre-existing ice crystals (Kuebbeler et al, 2014). With this cirrus scheme E61H22 could be tuned such that the global mean IWP agrees with the observations compiled

by Li et al. (2012). Similarly Lohmann and Neubauer (2018) made an experiment wherein which cirrus clouds could only form by heterogeneous freezing of ice nucleating particles in E63H23. In their experiment this caused the global mean IWP to agree with the observations compiled by Li et al. (2012). These studies and our analysis indicate that the IWP bias in the three model versions occurs because cirrus clouds can only nucleate homogeneously and therefore ICNC in cirrus clouds and hence their optical properties are misrepresented.

#### 3.4 Summary of model evaluation

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Fig.\_10 shows a Taylor diagram (Taylor, 2001) comparing SW and LW CRE, cloud cover, LWP-LP, cloud top CDNC, IWP and precipitation of the three model versions to the respective observations. The standardized deviations of eloud top CDNCLWP-LP had to be scaled by a factor of 1/34 so they could fit on the scale. For all variables except IWP and

- 10 precipitation, the root-mean-square (RMS) error (RMSE) (solid circles in the diagram in Fig. 10) is smaller or equal in E63H23 compared to in E61H22 and E55H20. For IWP, the RMS of E55H20 is smaller than for E63H23 but the bias in global mean IWP is much larger in E55H20 than in E63H23. (note the scaling for LWP-LP). The changes in the geographical pattern between the three model versions are rather small. E63H23 has somewhat higher correlations except for LW CRE, IWP and precipitation where E55H20 has higher correlations than E63H23 and E61H22. E55H20 has higher
- 15 correlations of LW CRE, IWP and precipitation because the ratio of the peaks in these variables in the tropics compared to mid-latitudes is better represented in E55H20 (see Fig.1). Overall E63H23 is an improvement over earlier model versions. Several biases in the representation of clouds in the three ECHAM-HAM model versions could be identified. The common problem of global elimate models (GCMs) in their representation of convective and boundary layer clouds is also present in the three ECHAM-HAM model versions. Stratocumulus clouds are underestimated in all three model versions. Shallow
- 20 convective clouds are underestimated in E61H22 and E55H20. In E63H23 the cloud cover and LWP in regions where shallow convective clouds are common agree well with observations but the cloud top CDNC are overestimated, leading to an overestimation of SW CRE in these regions. Deep convective clouds inover the Atlantic and Pacific oceans form too close to the continents (see Figs., 3, 6 and 8), in E63H23 and ECHAM (Stevens et al., 2013). For the tropical Atlantic this is a common bias in GCMs with coarse horizontal resolution (Siongco et al. 2014). Siongco et al. (2017) discuss different
- 25 ways how this bias in the tropical Atlantic precipitation could be reduced in ECHAM6. IWP is underestimated in all three model versions, in particular in the tropics, whereas LW CRE and the vertical profile of cloud ice agree rather well with observations. This indicates that ICNC may be overestimated in all three model versions (since LW CRE depends on the cloud temperature (~ cloud altitude) and cloud optical depth (∝ ICNC, IWP)). As E63H23 has the smallest bias in IWP also the bias in ICNC should be smaller than in the previous versions of ECHAM-HAM. Previous studies (Gasparini et al., 2018;
- Lohmann and Neubauer, 2018) showed that this overestimation of ICNC is (at least partly) due to missing processes in the formation of cirrus clouds (heterogeneous freezing of ice nucleating particles and/or water vapor deposition on pre-existing ice crystals). These studies also showed that including these processes can reduce the underestimation of IWP in ECHAM-HAM. South of 60°S LWP and cloud top CDNC of E63H23 could be underestimated, although there could also be problems

with the satellite retrievals at these high latitudes. In the previous model versions this possible bias was hidden by the overestimation of LWP.

#### 3.5 Simulation of ERF<sub>ari+aci</sub>, RF<sub>ari</sub> and ECS

- In Fig. 11 global maps of SW and LW ERF<sub>ari+aci</sub> for E63H23, E61H22 and E55H20 are shown. An important difference exists in the setup of E55H20 and the two other model versions. E55H20 uses AeroCom I aerosol emission data and the years 1750 and 2000 for pre-industrial and present day aerosol emissions. E63H23 and E61H22 on the other hand use AeroCom II aerosol emissions and the years 1850 and 2008 for pre-industrial and present day aerosol emissions. The stronger SW ERF<sub>ari+aci</sub> in the east of North America and Europe and the weaker SW ERF<sub>ari+aci</sub> in South and East Asia in E55H20 compared to the two other model versions are therefore predominantly the result of the different representative
- 10 emission years and inventories (see supplementary Fig. S3). We keep these emissions years as they were used as the default in previous studies (e.g. Zhang et al., 2012; Neubauer et al., 2014). The treatment of surface albedo over land, ocean and sea ice has changed substantially from ECHAM5 to ECHAM6 (see
- Stevens et al., 2013), which has an impact on SW ERF<sub>ari+aci</sub> (Stier et al., 2013). Because of the differences in setup and surface albedo treatment of E55H20 we focus here the comparison of ERF<sub>ari+aci</sub> on differences between E61H22 and E63H23. ERF<sub>ari+aci</sub> is stronger over land and weaker over oceans in E63H23 compared to E61H22 (Fig. S4). In the global mean 50% (-0.5 Wm<sup>-2</sup>) of ERF<sub>ari+aci</sub> originate over land in E63H23 and 50% (-0.5 Wm<sup>-2</sup>) over ocean. This is in contrast to E61H22 where 27% (-0.3 Wm<sup>-2</sup>) of ERF<sub>ari+aci</sub> originate over land and 73% (-0.9 Wm<sup>-2</sup>) over ocean. E63H23 uses the Köhler-theory based Abdul-Razzak and Ghan (2000) activation scheme, while E61H22 applies the empirical Lin and Leaitch (1997) activation scheme which depends only on the number of aerosol particles and updraft velocities. The increase in ERF<sub>ari+aci</sub> in the set of the number of aerosol particles and updraft velocities. The increase in ERF<sub>ari+aci</sub> in the set of the number of aerosol particles and updraft velocities. The increase in ERF<sub>ari+aci</sub> in the number of aerosol particles and updraft velocities. The increase in ERF<sub>ari+aci</sub> in the number of aerosol particles and updraft velocities. The increase in ERF<sub>ari+aci</sub> in the number of aerosol particles and updraft velocities.
- 20  $\frac{\text{E63H23 over land mayA sensitivity simulation with the Lin and Leaitch (1997) activation scheme applied in E63H23 shows$ an ERF<sub>ari+aci</sub> of 0.4 Wm<sup>-2</sup> over land, explaining about half of the difference in ERF<sub>ari+aci</sub> over land between E61H22 andE63H23. The increase in ERF<sub>ari+aci</sub> in E63H23 over land may also be related to the higher rate of autoconversion in E63H23(Table 2). While Lohmann and Ferrachat (2010) found no strong dependence of ERF<sub>ari+aci</sub> (or a small decrease) to theautoconversion tuning parameters in the global mean, the ratio of autoconversion to the total rain formation rate has a strong
- 25 regional dependence (see e.g. Sant et al., 2015) which could lead to regional differences in ERF<sub>ari+aci</sub>. It is interesting to note that although biases in the simulation of clouds in stratocumulus regions are reduced in E63H23, there seems to be no increase in ERF<sub>ari+aci</sub> in these regions compared to E61H22. Over the remote oceans, the largest differences in ERF<sub>ari+aci</sub> between E63H23 and E61H22 occur between 15°N and 45°N, where E61H22 simulates a strong ERF<sub>ari+aci</sub> in tradewind cumulus clouds (Zhang et al., 2016). ERF<sub>ari+aci</sub> in these shallow convective clouds regions is weaker in
- 30 E63H23, although more clouds are simulated in E63H23 in these regions. LWP and cloud cover in E63H23 are closer to observations in these regions (Figs. 3 and 4), and cloud top CDNC are rather too high in E63H23 (Fig. 5). A smaller LWP could lead to a weaker ERF<sub>ari+aci</sub> (Lohmann and Ferrachat, 2010). To better understand the differences over oceans between E63H23 and E61H22, we compare E63H23 also with a simulation with E63H23 where CDNCmin was lowered from 40 cm<sup>-</sup>
<sup>3</sup> to 10 cm<sup>-3</sup> (E63H23-10CC) as this simulation has a higher LWP (due to <u>retuning with</u> a smaller  $\gamma_r$ ;=2.8; not shown). Although the smaller CDNCmin of 10 cm<sup>-3</sup> leads to a stronger ERF<sub>ari+aci</sub> everywhere (Hoose et al., 2009; -1.7 Wm<sup>-2</sup> in the global mean; Table S1), this simulation still provides useful information. In the Northern Hemisphere Pacific there is an increase in ERF<sub>ari+aci</sub> in the E63H23-10CC compared to E63H23 (Fig. S5). This may be due to the larger LWP or the change

- in CDNCmin itself. In the Northern Hemisphere Atlantic however, ERF<sub>ari+aci</sub> does not increase. The weaker ERF<sub>ari+aci</sub> in the Northern Hemisphere Pacific in E63H23 could therefore be due to the smaller LWP in this simulation, while the smaller ERF<sub>ari+aci</sub> in the Northern Hemisphere Atlantic is due to a different reason. <u>TheE63H23 uses the Köhler theory based Abdul-Razzak and Ghan (2000) activation scheme</u>, while E61H22 applies the empirical Lin and Leaiteh (1997) activation scheme which depends only on the number of acrossl particles and undraft velocities. A sensitivity simulation with the Lin and
- 10 Leaitch (1997) activation scheme applied in E63H23 shows a negative ERF<sub>ari+aci</sub> between 15°N and 45°N in the Atlantic (Fig. S5). Therefore, the stronger ERF<sub>ari+aci</sub> in E61H22 <u>over oceans</u> can be partly explained by the different activation scheme. Another reason for the stronger ERF<sub>ari+aci</sub> in E61H22 over oceans between 15°N and 45°N is that different sea salt parameterizations are used in E61H22 and E63H23. Tegen et al. (20182019) show that the Long et al. (2011) sea salt parameterization (temperature dependent; Sofiev et al., 2011) used in E63H23 leads to higher aerosol number concentrations
- 15 over ocean compared to the Guelle et al. (2001) sea salt parameterization used in E61H22, improving the agreement with measured sea salt surface concentrations and particle size distributions at different marine sites (see also the comparison of sea salt parameterizations in Zieger et al., 2017). The higher natural background aerosol concentrations due to the higher sea salt aerosol number concentrations in E63H23 explain also why ERF<sub>ari+aci</sub> is less negative in E63H23 between 15°N and 45°N over oceans than in E61H22 (Fig. S5). From sensitivity simulations with the Lin and Leaitch (1997) activation scheme
- 20 or Guelle et al. (2001) sea salt parameterization applied in E63H23 (Table S1) we conclude that the largest part of the change in SW ERF<sub>ari+aci</sub> is actually from changes in the base model ECHAM6.3.

Most of the differences between the model versions discussed above are differences in SW ERF<sub>ari+aci</sub>. There is one important difference in LW ERF<sub>ari+aci</sub> between the model versions. LW ERF<sub>ari+aci</sub> is more than twice as large as in E61H22 than in E55H20 and E63H23 (Table 3). The stronger LW ERF<sub>ari+aci</sub> in E61H22 occurs in Northern Hemisphere mid-latitudes and in

25 the tropics (Fig. 12). In Northern Hemisphere mid-latitudes also LW CRE is larger in E61H22 due to the ICNC bug (see Fig. 2; also ICNC itself is higher in E61H22, see Table 3). The strong LW ERF<sub>ari+aci</sub> in E61H22 is therefore likely an artefact which was removed in the latest model version.

Tegen et al. (20182019) found an improved aerosol representation in biomass burning regions when GFAS biomass burning emissions, multiplied by a scaling factor of 3.4, as recommended by Kaiser et al. (2012), replace the default ACCMIP

30 biomass burning emissions. Therefore, we performed a E63H23 simulation with GFAS biomass burning emissions multiplied by 3.4 (E63H23-GFAS34). E63H23-GFAS34 has a weaker  $\text{ERF}_{ari+aci}$  (-0.9 W m<sup>-2</sup>) than E63H23 (-1.0 Wm<sup>-2</sup>), because the pre-industrial aerosol burden is higher in E63H23-GFAS34 and  $\text{ERF}_{ari+aci}$  is sensitive to pre-industrial aerosol

concentrations (Carslaw et al., 2013). Also, the present day aerosol burdens in E63H23-GFAS34 agree better with the mean aerosol burden of the AeroCom models (Textor et al., 2006) than in E63H23 (see Table 3 and S1).

We would like to point out that our simulations include interactions between sulfate and mineral dust. On the one hand (anthropogenic and natural) gaseous sulfate may coat mineral dust particles which leads to a transfer of dust from insoluble

- 5 modes to soluble modes in the models, which increases the wet deposition of dust (and leads to decreased present day mineral dust burdens, see Table S2), while on the other hand mineral dust particles provide surfaces where (anthropogenic and natural) gaseous sulfate may condensate, leading to a dampening of the nucleation of new particles. Similar interactions between sulfate and mineral dust have been found by Bauer-Fan et al. (2004) (using the Geophysical Fluid Dynamics Laboratory (GFDL) global chemical transport model; Mahlman and Moxim, 1978) and Bauer and Koch (2005) and Bauer et al. (200
- al. (2007) (using the Goddard Institute for Space Studies (GISS) climate model, modelE–(; Schmidt et al., 2006; Hansen et al., 2005) or Fan et al. (2004). The forcing from these interactions between sulfate and mineral dust is included in our estimates for  $\text{ERF}_{ari+aci}$  and  $\text{RF}_{ari}$  (these interactions will make  $\text{ERF}_{ari+aci}$  and  $\text{RF}_{ari}$  less negative but they are difficult to quantify).
- RF<sub>ari</sub> is shown in Fig. 12 for all-sky and clear-sky conditions for E63H23, E61H22 and E55H20 (since RF<sub>ari</sub> is computed by double calls to the radiation scheme, many values in Fig. 12 are statistically significant). RF<sub>ari</sub> is strong in the east of North America, Europe, South Asia, East Asia and the tropical Atlantic and Indian Oceans. The differences in the strength of RF<sub>ari</sub> between E55H20 and E63H23 and E61H22 in these regions are predominantly due to different emission years (and a different emission dataset) used in E55H20, as described above for ERF<sub>ari+aci</sub>. Differences in aerosol water uptake can explain the stronger RF<sub>ari</sub> over land in E63H23 than in E61H22. Absorbing aerosol above clouds leads to a positive RF<sub>ari</sub>.
- 20 This can be seen in all three model versions in the all-sky RF<sub>ari</sub> fluxes west off Africa (in particular in the Southern Hemisphere) and to a lesser extent also west of South America. The significant positive RF<sub>ari</sub> in the Saharan region and the Arabian Peninsula in E55H20 is due to a coding error in E55H20 (the refractive index of POM was used for sulfate aerosol) which was fixed in later model versions. The small positive RF<sub>ari</sub> in the Saharan region, the Arabian Peninsula and Pakistan in E61H22 and E63H23 is due to a decrease in dust load, which is caused by interaction with sulfate aerosol as described
- 25 above (also present in E55H20 but shadowed by the coding error). RF<sub>ari</sub> is weaker over ocean in E63H23 than in E61H22 and E55H20. One reason is that the dust burden is larger in E63H23 than in the other model versions and also the decrease in dust burden is larger in E63H23, leading to a positive RF<sub>ari</sub> which compensates the negative RF<sub>ari</sub> from the increase in anthropogenic aerosol. But there are also differences in aerosol water uptake (aerosol water increases less over oceans in E63H23 than in the other two model versions).
- 30 The equilibrium climate sensitivity (ECS) is strongest in E55H20 (3.5 K), weaker in E61H22 (2.8 K) and weakest in E63H23 (2.5 K) (Fig. 13). The corresponding ECS values in the base model versions are: ECHAM5: 3.4 K (Randall et al., 2007; their Table 8.2), ECHAM6.1: 2.8 K (Block and Mauritsen, 2013; Meraner et al., 2013) and ECHAM6.3: 2.8 K (Mauritsen et al., 2019), i.e. changes in ECS between the ECHAM-HAM model versions are driven substantially by changes in the ECHAM base model versions. Note that the ECS value for ECHAM6.3 is from a simulation with abruptly quadrupled

<u>CO<sub>2</sub> concentrations, in contrast to the CO<sub>2</sub> doubling used in the computation of ECS in this study and that ECHAM has a strong state dependency for ECS (see discussion in Mauritsen et al., 2019 and references therein).</u> For E61H22 and E63H23 we computed the cloud feedback parameter using the cloud radiative kernel method of Zelinka et al. (2016) (Fig. 14; in E55H20 the COSP-ISCCP simulator is not implemented, therefore the cloud feedback parameter could not be computed). In

- 5 addition, we computed ECS and cloud feedback for the E63H23-GFAS34 and E63H23-10CC simulations. E63H23-GFAS34 and E63H23 have very similar ECS and cloud feedback. In E63H23-10CC, on the other hand, ECS is stronger and the cloud feedback is more positive than in E63H23 (leading to more warming in agreement with the stronger ECS). The optical depth feedback of low clouds is more positive between 40°N and 40°S in E63H23-10CC. The optical depth feedback of non-low clouds is less negative in the tropics and in mid-latitudes- (not shown). This could be an indication of a weaker cloud phase
- 10 feedback. As there are fewer but larger cloud droplets in E63H23-10CC than in E63H23, (Fig. S7), the cloud droplets have a shorter lifetime and this decreases differences between ice clouds and liquid water clouds. A similar less negative cloud optical depth feedback of non-low clouds (weaker cloud phase feedback) occurs in E61H22 (CDNC are higher and the representation of supercooled liquid in mixed-phase clouds is improved in E63H23 compared to E61H22, see Fig. S6). Furthermore, in E61H22 also the cloud amount feedback of low clouds is more positive than in E63H23. This is because in
- 15 E63H23 in regions of low cloud cover where shallow convective clouds are simulated, the cloud amount feedback is negative whereas in E61H22 it is positive. This seems to be related to the stronger entrainment rate for shallow convection in E63H23 (Mauritsen et al., 2012), 2019). Mauritsen et al. (2019) describe the increase of entrainment rate for shallow convection in ECHAM6.3 as a measure to reduce ECS in ECHAM6.3. When more of the water vapor remains in the boundary layer as in E63H23, the increased water vapor in the warmer climate can lead to increased cloud cover. The overall more positive cloud feedback in E61H22 than in E63H23 agrees with the stronger ECS in E61H22.

The largest differences between E61H22 and E63H23 in terms of  $\text{ERF}_{ari+aci}$  are therefore due to a more realistic simulation of cloud water, the removal of a bug in ICNC, the new activation scheme and the new sea salt emission parameterization in E63H23, whereas for ECS they are due to a more realistic simulation of cloud water, and to model tuning.

## 3.6 Impact of changes and improvements in E63H23

25 The liquid phase of clouds is better represented in E63H23 than in the previous model versions because the low-bias in cloud cover in the subtropics is reduced and the zonal distribution of LWP agrees with observations. This leads also to a better agreement of the SW CRE with observations in E63H23. Important reasons for these improvements are the change in the fractional cloud cover scheme for marine stratocumulus clouds and the removal of an inconsistency which had led to either 0 or 1 cloud cover in ECHAM6.3 and subsequent changes in model tuning. (Mauritsen et al., 2019). Furthermore E63H23 uses 30 the Abdul-Razzak and Ghan (2000) activation scheme and the Long et al. (2011) sea salt emission parameterization (temperature dependent; Sofiev et al., 2011) which leads to higher CDNC concentrations where LWP is large. The Abdul-Razzak and Ghan (2000) activation scheme is more physically realistic as the empirical Lin and Leaitch (1997) activation scheme used in the previous model versions as it is Köhler-theory based and therefore takes into account the size of the

aerosol particles and their chemical composition. Although the Abdul-Razzak and Ghan (2000) activation scheme has limitations under certain conditions (the assumption that the aerosol particles are in equilibrium with its environment is not valid in all conditions; Phinney et al., 2003) and does not account for pre-existing cloud droplets during cloud droplet activation (Barahona et al., 2010), it certainly helps to improve the representation of cloud droplets in E63H23. The

- performance of the new Long et al. (2011) (temperature dependent; Sofiev et al., 2011) and the old Guelle et al. (2001) sea
   salt parameterizations in E63H23 was analyzed by Tegen et al. (20182019). The new temperature dependence leads to increased sea salt emissions where the sea surface temperature is warmer than 20°C and a decrease at colder temperatures. The new sea salt parameterization performs better, particular with respect to number concentrations, than the previous sea salt parameterization compared to measurements on research cruises and research stations. This is another indication that
- 10 CDNC concentrations are more realistic in E63H23 than in the previous model versions. The higher CDNC concentrations in E63H23 allowed us to increase the tuning parameter for autoconversion of cloud droplets to rain. Together these changes led to a better representation of the liquid phase of clouds in E63H23 and to a reduction of the SW component of ERF<sub>ari+aci</sub> in E63H23 compared to E61H22 (because CCN concentrations from natural background aerosol are higher in E63H23).
- Also the ice phase of clouds has improved in E63H23 compared to previous model versions. The low-bias in IWP is reduced in E63H23 and the global mean vertical IWC is within the observational range (Fig. 7). This is because the Seifert and Beheng (2006) sticking efficiency used in E63H23 leads to a less efficient removal of ice crystals by snow. A subsequent reduction in the tuning parameter for stratiform snow formation by aggregation further increases IWP in E63H23. Only few laboratory studies for sticking efficiency have been conducted and even fewer theories for sticking efficiency were developed (Phillips et al, 2015). We find that the simple formulation of Seifert and Beheng (2006) for sticking efficiency for
- 20 accretion of ice crystals by snow improves the simulation of cloud ice in E63H23. Furthermore, the altitude of the global mean maximum IWC agrees well with observations in E63H23 whereas in E61H22 and E55H20 it is at higher altitudes than observed. This can be explained by changes in ICNC described in section 2.1.5 such as the use of a consistent ice crystal shape (hexagonal plates), removal of an ICNC bug or the changed treatment of detrained ice crystals. The subsequent changes in precipitation formation and ice crystal sedimentation can then lead to a different vertical distribution of cloud ice.
- 25 In E61H22 the global ICNC burden is considerably higher than in the two other model versions because of an inconsistency between cloud droplet activation, condensation, vertical transport of CDNC and homogeneous freezing of cloud droplets in cirrus clouds, which led to homogeneous freezing of aerosol particles even when the water vapor pressure was too low for homogeneous nucleation. The higher ICNC are also responsible for the LW component of ERF<sub>ari+aci</sub> in E61H22 being more than twice as large as in the other two model versions. The good agreement of the global mean vertical distribution of IWC
- 30 and LW CRE with observations in E63H23 is an indication that ICNC and the size of ice crystals are closer to reality in E63H23 than in the previous model versions. In a future version of the model we want to include also the competition for water vapor between homogeneous freezing of solution droplets, heterogeneous freezing of ice nucleating particles and pre-existing ice crystals in cirrus cloud formation as has been done by Kuebbeler et al. (2014) or Gasparini et al. (2018), which should further improve the simulation of ICNC.

While the global mean values of  $RF_{ari}$  are quite similar between E63H23 and the previous model versions, there are regional differences. These are caused by the removal of inconsistencies in the model code and for E63H23 also by the new emission parameterization for mineral dust which uses new satellite-based data for dust sources, which increases the confidence in the simulation of  $RF_{ari}$  in E63H23.

- 5 The weaker ECS in E63H23 compared to E61H22 can be linked to changes in cloud feedbacks. There are indications for a stronger cloud phase feedback in non-low clouds due to increased CDNC and changes in cloud water in E63H23. A stronger (cooling) cloud phase feedback will lead to less warming in the future. Similarly the less positive cloud amount feedback of low clouds (related to model tuning in ECHAM6.3) in E63H23 contributes to the weaker ECS in E63H23 compared to E61H22.
- 10 The changes and improvements in E63H23 <u>(including changes in the base model version ECHAM6.3)</u> therefore have not only improved the representation of clouds in E63H23 compared to previous model versions, they also have an impact on ERF<sub>ari+aci</sub> and ECS, decreasing both.

## 4 Summary and conclusions

Clouds in the current (E623H23) and previous (E55H20 and E61H22) model versions of the ECHAM-HAM global aerosolclimate model were evaluated using a suite of global observational datasets for clouds and precipitation. Improvements in E63H23 compared to previous model versions for cloud water include:

- a more physically based activation scheme, (Abdul-Razzak and Ghan, 2000).
- changes in the treatment of CDNC detrained from convective clouds, (as described in section 2.1.5).
- an increase in low clouds, (Mauritsen et al., 2019),
- which together lead to a more realistic LWP globally.

For cloud ice the improvements include:

- a different sticking efficiency for accretion of ice crystals by snow, (Seifert and Beheng, 2006),
- consistent ice crystal shapes throughout the model, (Lohmann and Neubauer, 2018),
- changes in mixed phase freezing; (as described in section 2.1.5),
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- the removal of an inconsistency in ICNC in cirrus clouds,
- which together lead to a more realistic IWP globally.

The sum of the changes leads to improved cloud radiative effects. Although the representation of shallow convective clouds has improved in E63H23, stratocumulus clouds are still underrepresented. The comparison of the different model versions showed that the misrepresentation of certain cloud types can lead to compensating biases in other clouds via model tuning.

30 Therefore, if the bias in stratocumulus clouds in E63H23 can be reduced, this could also improve the representation of other cloud types. Reasons for the bias in stratocumulus clouds identified by Neubauer et al. (2014) in E61H22 were e.g. too

strong turbulent mixing at cloud top, the shallow convection scheme triggering too often or a lack of vertical resolution. Future work will focus on addressing these difficult issues.

Deep convective clouds inover the Atlantic and Pacific oceans form too close to the continents, which leads to biases in the geographical distribution of precipitation. in E63H23 and ECHAM (while tropical land precipitation is underestimated).

- 5 While the geographical (except for deep convective clouds) and vertical distribution of cloud ice agree well with observations in E63H23, IWP remains biased low. The combination of observations of IWP, LW CRE and the vertical distribution of cloud ice indicate that ICNC may be overestimated in ECHAM-HAM. Previous studies with ECHAM-HAM showed that the bias in ICNC and IWP can be reduced when heterogeneous freezing of ice nucleating particles and/or water vapor deposition on pre-existing ice crystals are accounted for in cirrus clouds.
- 10 Estimates of ERF<sub>ari+aci</sub> and ECS of E55H20, E61H22 and E63H23 were compared since the representation of clouds is important for both ERF<sub>ari+aci</sub> and ECS. The largest differences between E61H22 and E63H23 in terms of SW ERF<sub>ari+aci</sub> are due to:
  - mainly the more realistic simulation of cloud water,
  - <u>but also</u> the new activation scheme in E63H23, and
  - the new sea salt emission parameterization,
  - the more realistic simulation of cloud water

which lead to a weaker SW ERF<sub>ari+aci</sub> in E63H23. In terms of LW ERF<sub>ari+aci</sub> the difference is mainly due to:

• the removal of an inconsistency in ICNC in cirrus clouds leading to a weaker LW ERF<sub>ari+aci</sub> in E63H23.

Since there are reductions in both SW and LW ERF<sub>ari+aci</sub> the net ERF<sub>ari+aci</sub> is only slightly weaker in E63H23 (-1.0 W m<sup>-2</sup>). A sensitivity simulation where CDNCmin was lowered to 10 cm<sup>-3</sup> leads to a stronger ERF<sub>ari+aci</sub> everywhere (-1.7 W m<sup>-2</sup>) showing that the necessary usage of CDNCmin (Lohmann and Neubauer, 2018) has a strong impact on ERF<sub>ari+aci</sub>. Another sensitivity simulation with increased biomass burning emissions (E63H23-GFAS34) indicates that ERF<sub>ari+aci</sub> in E63H23 would be weaker (-0.9 W m<sup>-2</sup>) when the representation of biomass burning aerosol could be improved.

ECS is weaker in E63H23 (2.5 K) than in E61H22 (2.8 K) (and E55H20; 3.5 K). The decrease compared to E61H22 is due

25 to:

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- changes in the entrainment rate for shallow convection <u>adopted from the base model ECHAM6.3</u> (which leads to a less positive feedback of cloud amount of low clouds in some regions),
- a stronger cloud phase feedback.

Although the differences in both ERF<sub>ari+aci</sub> and ECS between E63H23 and E61H22 can be explained by changes in the 30 representation of clouds, not the same changes in the clouds that affect ERF<sub>ari+aci</sub> also affect ECS and vice versa. Therefore, many aspects of clouds in GCMs will need to be improved to increase the confidence in computations of ERF<sub>ari+aci</sub> and ECS.

Code availability. The ECHAM-HAMMOZ model is made freely available to the scientific community under the HAMMOZ Software Licence Agreement, which defines the conditions under which the model can be used. More information can be found at the HAMMOZ Website (https://redmine.hammoz.ethz.ch/projects/hammoz). Scripts can be found at http://doi.org/10.5281/zenodo.2553892 (Neubauer et al., 2019a).

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Data availability. Data can be found at http://doi.org/10.5281/zenodo.2541937 (Neubauer et al., 2019b). ESA Cloud CCI data can be downloaded from: http://www.esa-cloud-cci.org/?q=data download. MODIS products are available for download from Level 1 and Atmosphere Archive and Distribution System (LAADS) https://ladsweb.modaps.eosdis.nasa.gov/search/. ISCCP histogram data and CALIPSO-GOCCP product can be obtained 10 from http://climserv.ipsl.polytechnique.fr/cfmip-obs/. Cloud top CDNC can be downloaded from https://doi.org/10.15695/vudata.ees.1. MAC-LWP data is available at the Goddard Earth Sciences Data and Information Services Center (GES DISC, current hosting; http://disc.sci.gsfc.nasa.gov). CERES satellite data can be obtained from the NASA Langley Research Center Atmospheric Science Data Center https://ceres.larc.nasa.gov/order data.php. The IWP satellite data from Li et al. (2012) was obtained from the authors. GPCP data can be downloaded from https://www.esrl.noaa.gov/psd/data/gridded/data.gpcp.html.

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Simulation	Configuration	E55H20	E61H22	E63H23		
All	Resolution	T63L31 T63L31		T63L47		
	Sea salt emissions	Guelle et al. (2001) Guelle et al. (2001)		Long et al. (2011) / Sofiev et al. (2011)		
	Onidante	MOZART	MACC	MACC		
	Oxidants	(present day, ~2000) (2003-2010 mean)		(2003-2010 mean)		
$PD/PD_{aer}/PI_{aer}$	Greenhouse gases		RCP8.5 (year 2008)			
PD	Simulation period	2000-2009	2000-2009	2003-2012		
	Aerosol emissions	AEROCOM I	ACCMIP MACCity	ACCMIP MACCity		
		(year 2000)	(historic and RCP8.5)	(historic and RCP8.5)		
	SST and SIC		AMIP 2000-2015 mean			
PD <sub>aer</sub>		Identical to PD except for simulation period 2000-2019 Identical to PD <sub>aer</sub> except for aerosol emissions				
	Simulation period					
PIaer						
	A group amiggiong	AEROCOM I	ACCMIP MACCity	ACCMIP MACCity		
	Aerosol emissions	(year 1750)	(year 1850)	(year 1850)		
1xCO <sub>2</sub> /2xCO <sub>2</sub>	Simulation duration	50 years (25 years spin-up, 25 years for analysis)				
	Aerosol emissions	Same as in PI <sub>aer</sub> Mixed-layer ocean (50 m deep)				
	SST and SIC					
	Heat flux corrections	Computed from extended PD <sub>aer</sub> (2000-2024)				
1		Year 1850				
TXCO <sub>2</sub>	Greennouse gases	CO <sub>2</sub> : 284.7 ppm				
2 60	C 1	Year 1850				
$2xCO_2$	Greennouse gases	CO <sub>2</sub> : 569.5 ppm				

Table 1. Setup of the simulations for E55H20, E61H22 and E63H23.

	Table 2. Parameter settings for E55H20, E61H22 and E63H23. The parameters used to tune the ECHAM-HAM versions are
	scaling factor for stratiform rain formation rate by autoconversion ( $\gamma_r$ ), scaling factor for stratiform snow formation rate by
	aggregation ( $\gamma_s$ ), critical relative humidity at the surface which is used in the cloud cover scheme ( $\gamma_c$ ), entrainment rate for
	shallow convection ( $\epsilon_s$ ), entrainment rate for deep convection ( $\epsilon_d$ ), convective conversion rate from cloud water to rain
5	$(\gamma_{cr})$ , inhomogeneity factor for ice clouds ( $\gamma_i$ ) and the minimum cloud droplet number concentration (CDNCmin).

Parameter	E55H20	E61H22	E63H23	
γ <sub>r</sub>	3	4	10.6	
γ <sub>s</sub>	1200	1200	900	
γ <sub>c</sub>	0.9	0.9	0.975	
$\epsilon_s (\text{m}^{-1})$	0.0003	0.0008	0.003	
$\epsilon_d (\text{m}^{-1})$	0.0001	0.00035	0.0002	
$\gamma_{cr}$ (s <sup>-1</sup> )	0.0001	0.0009	0.0009	
γ <sub>i</sub>	0.85	0.7	0.7	
CDNCmin (cm <sup>-3</sup> )	20	40	40	

Variable	OBS/MM	Á	E55H20	E61H22	E63H23	Exemption Table
			(2000-2009)	(2000-2009)	(2003-2012)	
$SW(Wm^2)$	241 (238 to 244) <sup>a</sup>	<u>OBS</u>	232	236	238	Inserted Cells
$LW (W m^{-2})$	-240 (-237 to -241) <sup>a</sup>	<u>OBS</u>	-232	-236	-238	
Net(W m <sup>-2</sup> )	$0.7 \pm 0.1^{6}$	<u>OBS</u>	-0.1	0.4	0.4	
SW CRE (W m <sup>-2</sup> )	$-46 (-44 \text{ to } 53.3)^{\circ}$	<u>OBS</u>	-53	-52	-50	
LW CRE (W m <sup>-2</sup> )	28 (22 to 30.5) <sup>c</sup>	<u>OBS</u>	28	27	24	
Net CRE (W m <sup>-2</sup> )	-18 (-17.1 to 22.8) <sup>c</sup>	<u>OBS</u>	-25	-25	-26	
CC (%)	$68\pm5^{d}$	<u>OBS</u>	64	64	69	
LWP (ocean) (g m <sup>-2</sup> )	42.9 to 89.4 <sup>e</sup>	<u>OBS</u>	85	94	71	
LWP-LP (ocean) (g m <sup>-2</sup> )	$73.5 \pm 5.5^{f}$	<u>OBS</u>	104	96	76	
IWP $(g m^{-2})$	25±7 <sup>g</sup>	<u>OBS</u>	8	10	15	
Cloud-top CDNC (ocean; 60°N-60°S) (cm <sup>-3</sup> )	72±37 <sup>h</sup>	<u>OBS</u>	80	76	78	
$\text{CDNC}_{\text{burden}} (10^{10} \text{ m}^{-2})$	-		3.1	3.2	3.1	
$ICNC_{burden} (10^{12} \text{ m}^{-2})$	-		8.9	17.9	8.0	
$P (mm d^{-1})$	$2.7 \pm 0.2^{i}$	<u>OBS</u>	3.0	3.0	3.0	
Sulfate burden (Tg)	2.0(±25%) <sup>j</sup>	<u>MMM</u>	2.6	1.9	2.2	
Black carbon burden (Tg)	0.2(±42%) <sup>j</sup>	<u>MMM</u>	0.13	0.15	0.14	
Particulate organic matter burden (Tg)	1.7(±27%) <sup>j</sup>	<u>MMM</u>	1.1	1.1	1.0	
Sea salt burden (Tg)	7.5(±54%) <sup>j</sup>	MMM	12.6	10.8	4.1	
Mineral dust burden (Tg)	19.2(±40%) <sup>j</sup>	<u>MMM</u>	8.0	10.9	18.2	
Aerosol water burden (Tg)	27.7(±46%) <sup>j</sup>	<u>MMM</u>	48.4	48.9	23.0	
RF <sub>ari</sub> (all-sky) (W m <sup>-2</sup> )	-0.27±0.15 <sup>k</sup>	<u>MMM</u>	-0.04	-0.06	0.00	
RFari (clear-sky) (W m <sup>-2</sup> )	$-0.67 \pm 0.18^{k}$	<u>MMM</u>	-0.41	-0.30	-0.27	
ERF <sub>ari+aci</sub> (W m <sup>-2</sup> )	-0.9 (-1.9 to -0.1) <sup>1</sup>	<u>OBS/</u> MMM	-1.1	-1.2	-1.0	
SW ERF <sub>ari+aci</sub> (W m <sup>-2</sup> )	-		-1.3	-2.0	-1.3	
LW ERF <sub>ari+aci</sub> (W m <sup>-2</sup> )	-	<del>0.2</del>	0.2	0.8	0.3	Inserted Cells
ECS (K)	1.5 to 4.5 <sup>m</sup>	MMM	3.5	2.8	2.5	Inserted Cells

Table 3. Global mean values of the PD simulations. Radiative fluxes are at the top of atmosphere. Values from observations (OBS) and multi-model means (MMM) for aerosol burdens are shown next to those of the three model versions.  $ERF_{ari+aci}$  and ECS are from the  $PD_{acr}/PI_{acr}$  and 1xCO2/2xCO2 simulations respectively.

5 <sup>a</sup> Central values from Loeb et al. (2018), range from Stevens and Schwartz (2012). <sup>b</sup> Loeb et al. (2018) and Johnson et al.

(2016). <sup>c</sup> Central values from Loeb et al. (2018), the range takes into account values from Loeb et al. (2009) and Matus and

L'Ecuyer (2017). <sup>d</sup> Stubenrauch et al. (2013). <sup>e</sup> Platnick et al. (2015, 2017), ATSR2-AATSR v2.0 (Stengel et al., 2017a; Poulsen et al., 2017), Elsaesser et al. (2017). <sup>f</sup> Elsaesser et al. (2017). <sup>g</sup> Li et al. (2012). <sup>h</sup> Bennartz and Rausch (2017). <sup>i</sup> Central value from Adler et al. (2018), uncertainty from Adler et al. (2012). <sup>j</sup> Taken from Table 10 of Textor et al. (2006). <sup>k</sup> Taken from Table 3 of Myhre et al. (2013). <sup>1</sup> Boucher et al. (2013). <sup>m</sup> Collins et al. (2013), Knutti et al. (2017).



Figure 1: Comparison of zonal annual mean values of E55H20, E61H22 and E63H23 to observations, (a) SW CRE, (b) LWP-low precipitation over oceans, (c) LW CRE, (d) IWP, (e) total cloud cover, (f) Cloud top CDNC of clouds between 268 and 300 K over 5 occans. Observations of IWP are from Li et al. (2012), LWP-low precipitation over oceans from Elsaesser et al. (2017), Cloud top CDNC over oceans from Bennartz and Rausch (2017). The solid SW and LW CRE lines are from CERES (Loeb et al., 2018), the dashed ones from ERBE (Barkstrom, 1984) and the dotted one for LW CRE is from TOVS satellite data (Susskind et al., 1997). Total cloud cover from CALIPSO GOCCP (solid line; Chepfer et al., 2010), AVHRR-PM (dashed line; Stengel et al., 2017b) and

<sup>10</sup> MODIS collection 6.1 (dotted line; Platnick et al., 2015, 2017).



Figure 2: Comparison of annual mean SW, LW and net CRE of E55H20, E61H22 and E63H23 to CERES 4.0 (Loeb et al., 2018) observations. CERES data is for 2005-2015, model data is from the PD simulations. In the top left panel the regions used for cloud top pressure vs. cloud optical depth histograms are shown by green lines.



Figure 3: Comparison of annual mean cloud cover of E55H20, E61H22 and E63H23 to CALIPSO GOCCP observations. Areas where the cloud cover of CALIPSO GOCCP, MODIS collection 6.1 and AVHRR-PM differ by more than five percent points are hatched. CALIPSO GOCCP data is for 2006-2010, model data is from the PD simulations- (direct model output is used without a simulator).



Figure 4: Comparison of annual mean LWP of E55H20, E61H22 and E63H23 to MAC-LWP observations. Areas where precipitation could influence the LWP retrieval (LWP/(LWP+rain water path)≤0.8) are hatched. MAC data is for 2003-2012, model data is from the PD simulations.



Figure 5: Comparison of annual mean Cloud top CDNC of E55H20, E61H22 and E63H23 to MODIS observations from Bennartz and Rausch (2017). Areas where the relative uncertainty in the observations is larger than 75% are hatched. The MODIS data is for 2003-2015, model data is from the PD simulations.



Figure 6: Comparison of annual mean IWP of E55H20, E61H22 and E63H23 to CALIPSO/CloudSat observations from Li et al. (2012). Areas where the relative standard deviation of the different datasets compiled in Li et al. (2012) is larger than 75% are hatched. The CALIPSO/CloudSat data covers the years 2006-2010, model data is from the PD simulations.



Figure 7: Comparison of global annual mean IWC as a function of pressure of E55H20, E61H22 and E63H23 to CALIPSO/CloudSat observations from Li et al. (2012). Gray shading indicates the uncertainty in the CALIPSO/CloudSat observations. The CALIPSO/CloudSat data covers the years 2006-2010, model data is from the PD simulations.



Figure 8: Comparison of annual mean precipitation (stratiform + convective) of E55H20, E61H22 and E63H23 to GPCP2.3 observations. Areas where the relative uncertainty of the GPCP2.3 data is larger than 75% are hatched. The GPCP2.3 data is for 1979-2017, model data is from the PD simulations.







Figure 9: Histograms of cloud top pressure vs. cloud optical depth of E61H22 and E63H23 as compared to ISCCP, MODIS and AVHRR-PM observations for different regions. The definition of the four regions shown is described in the text and the regions are shown in Fig. 2. The ISCCP data is for 2000-2008, MODIS data is for 2003-2012, AVHRR-PM data is for 2003-2012 and the model data is from the PD simulations.





Figure 10: Taylor diagram for comparison of SW and LW CRE, cloud cover, LWP-low precipitation, Cloud top CDNC, IWP and precipitation of E55H20, E61H22 and E63H23 to observations as REF. The standardized deviations of LWP-low precipitation are scaled by a factor of 1/34 to fit on the diagram. Only areas that are not hatched in Figs.  $3_x = 6$  and 8 were used to create the Taylor diagram. Observations are the same as in Figs.  $3_x = 6$  and 8. The correlation coefficient is shown as an angle and quantifies the similarity in pattern between modelled and observed fields) is shown as the radial distance from the origin. The RMS errorRMSE is shown as solid black circles and is the distance from the point marked by REF (the closer a model is to REF the better its skill to reproduce the observations). For E63H23 and the observations for precipitation and LWP-low precipitation an average over the time period 2003 to 2012 was used. For Cloud top CDNC the time period 2003 to 2015, for IWP the time periods in Li et al. (2012), for SW CRE and LW CRE the time period 2000 to 2009 were used. Tests with E63H23 showed negligible impact of the different time periods for the data in the Taylor diagram.





5 Figure 11: Global maps of SW, LW and net ERF<sub>arri+aci</sub> of E55H20, E61H22 and E63H23 from 20 year free simulations with present day minus pre-industrial aerosol emissions (PD<sub>aer</sub>-PI<sub>aer</sub>). Hatching indicates statistically significant differences at the 95% significance level. The false discovery rate is controlled following Wilks (2016).


5 Figure 12: Global maps of all-sky and clear-sky net RF<sub>ari</sub> of E55H20, E61H22 and E63H23 from 20 year free simulations with present day minus pre-industrial aerosol emissions (PD<sub>aer</sub>-PI<sub>aer</sub>). Hatching indicates statistically significant differences at the 99% significance level. The false discovery rate is controlled following Wilks (2016).



Figure 13: Global mean ERF<sub>ari+aci</sub> and ECS of E55H20, E61H22, E63H23, E63H23-GFAS34 and E63H23-10CC. The coefficient of determination between ERF<sub>ari+aci</sub> and ECS is also displayed.



Figure 14: Components of the net global mean cloud feedback parameter of E61H22 and E63H23 for low (cloud top pressure (CTP)>680hPa) and non-low (CTP<680 hPa) clouds.