In this document we present the Author response to the comments received from the two Anonymous Referees. The Reviewer's comments are in italic and our answers are in normal font. At the end of the Author response we include the marked-up manuscript version showing the changes made in the manuscript in red font.

Author Response to Referee 1

Major comment:
The methods section admits that the MODIS Angstrom Exponent product (used in the C1 ACPD Interactive comment Printer-friendly version Discussion paper Aerosol Index [AI] calculation) is not calculated over land due to its low data quality in these locations. However, Figure 5 still shows MODIS AI values over land. Why is this? In Levy et al., 2013, which describes the collection 6 Dark Target product, it says, "On a global basis, we and others have found little quantitative skill in MODIS-retrieved aerosol size parameters over land (e.g., Levy et al., 2010; Mielonen et al., 2011). We have decided to discontinue further attempts at validating Ångström Exponent (AE) and fine-AOD. A user can still choose to derive AE (from spectral AOD) or fine-AOD (from product of) and evaluate the results themselves."
Levy et al. 2013: https://www.atmos-meas-tech.net/6/2989/2013/amt-6-2989-2013.pdf
Levy et al. 2010: https://www.atmos-chem-phys.net/10/10399/2010/acp-10-10399- 2010.pdf
So did you calculate AE from the spectral AOD over land? Is this any good? Is there any value to compare MODIS AI to the model's AI over land if MODIS AE over land does not have skill? I think that the AI values over land should be removed from Figure 5 and discussion unless these values are tested against e.g. AERONET.

Author response:
We agree with the reviewer that the use of MODIS AI over land has not carefully been explained in the manuscript. The Ångström exponent over land was derived from spectral AOD. Although the resulting AI over land is shown in Figure 5, the values are not used for the calculation of mean values in Table1 not in the calculation of ACI (Figure 8). For these reasons, the author agrees with the reviewer: AI over land is masked out from Figure 5 and from the discussion section. Furthermore, the text now clearly states that AI excludes values over land.

Changes:
Figure 5 and Figure 7 were updated and replaced. The text describing the figures and the results were updated throughout the manuscript.

Specific comments:

P2 L16: I'd remove "primarily" here as climate models serve many purposes.

Author response:
Accepted.

P3 L16-17: This sentence makes it seem like ISCCP is itself a cloud simulator. However, ISCCP is much broader than this, and foremost it has observational data products. Could say "are the simulator developed as part of the of International Satellite. . ."
Author response:
Accepted. Sentence modified as suggested.

P3 L23 and many places throughout: The clause following "which" is a non-restrictive clause (it does not help specify which simulator you're writing about and only provides additional information about it), which means there should be a comma before "which". If it were a restrictive clause, it would continue to not have a comma, e.g., "We use the cloud simulator which was developed as part of CMIP" (the clause after "which" is necessary to know the specific simulator you are referring to). I found many non-restrictive clauses throughout that did not contain commas, so please update. http://www.cws.illinois.edu/workshop/writers/restrictiveclauses/
Author response:
Clause updated at P7, L14; P9, L24; P10, L2, L19 and L24; P14, L9; P15, L18; P18, L34.

P3 L24: Should these acronyms be defined here?
Author response:
Accepted. Acronyms have been included and the sentence has been rephrased as: "COSP is a software tool developed within the CFMIP (Webb20 et al., 2017), which extracts parameters for several spaceborne active sensors, such as the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and the Cloud Profiling Radar (CPR), as well as for passive sensors, such as the Multi-angle Imaging SpectroRadiometer (MISR) and the Moderate Resolution Imaging Spectroradiometer(MODIS).

P3 L33: I'm generally used to AIEs specifically referring to the radiative effects of ACIs rather then being a synonym for ACIs in general, as AIE is presented here.
Author response:
The nomenclature describing the aerosol-cloud-radiation interaction has been changing throughout the years and the IPCC 5AR (IPCC2013) introduced the new terminology which is used in the manuscript.
Changes: the acronym AIE is not used and the abbreviation AIE has been removed from the text.

P4 L20: Although Koren et al., 2007 is cited at the end of the sentence, it seems jargony to list "twilight zone" without definition. May be more clear to replace with "near-cloud impacts on radiative transfer".

Author response:
Accepted. The sentence has been modified as suggested: "The primary artifacts known to affect satellite estimation of aerosol-cloud interactions are related to (1) the inability of untangling aerosol and cloud retrievals from meteorology (e.g. aerosol humidification, entrainment, cloud regimes dependency), (2) inaccuracies in the retrieval algorithms (e.g. near-cloud impacts on radiative transfer, contamination, statistical aggregation) and (3) assumptions in the retrieval algorithms (Koren et al.,2007; Oreopoulos et al., 2017; Christensen et al., 2017; Wen et al., 2007)."

P5: I'm confused as to why the MODIS L2 products are discussed in detail after it is stated that L3 products were using in the paper. I assume it is because the L3 product is built from the L2 product (which is stated), but it would be good to make it clear why the L2 products are discussed in detail.
Author response:
The sentence was expanded to explain why MODIS L2 data are described. The new sentence is: "As the L3 1 x 1 gridded average values of atmospheric properties, along with a suite of statistical quantities, are derived from the corresponding L2 atmosphere data product, a brief description of Level-2 MODIS aerosol and cloud products is now presented."

P5 L22-: Which aerosol product(s) are you using? Just the Dark Target product or also Deep Blue? I assume MAIAC is not used since it says the spatial resolution is 10x10km. It looks like the Dark Target - Deep Blue combined product is used in Figure 5 based on there being AOD information over deserts etc.
Author response:
Indeed the combined product Dark Target - Deep Blue is used in this study.

P5 L27: Here it says that AE is only derived over ocean, which is correct, but why does AI have land values in Figure 5? See Major Comment above.
Author response:
See Author response to Major Comment.

P7 L19: How are the model fields downscaled? It seems like there would be a lot of necessary assumptions to break a partially cloudy coarse gridbox into finer subcolumns. These assumptions should affect the results in theory. At a minimum, please add a statement such as, "details of this downscaling process and assumptions are provided in XX", assuming that this process has been documented elsewhere. If these details haven't been documented, please do so here or in the supplement.
Author response:
The following sentence citing the proper documentation has been added at P7, L25-L26: "A comprehensive explanation about the methodology and results of the COSP MODIS simulator is presented in Pincus et al. (2012)."

*P8 L4: "...referred to *here* as. . ."*

Author response:
Accepted.

P8 L23-25: This sentence didn't make sense to me. Please rewrite for clarity.
Author response:
The sentence was to explain that instantaneous output cannot be used with the implementation of the COSP satellite simulator in ECHAM-HAM. The sentence was rewritten to:
"The output of the COSP satellite simulator is also three-hourly. The implementation of the COSP satellite simulator in ECHAM-HAM does not allow instantaneous output. The COSP satellite simulator is called every radiation time step (i.e. every two hours) and the output of the COSP satellite simlator is averaged over the three hourly output period. This means that on average 50% of the values in the output of the COSP satellite simulator are instantenous values (i.e. from only one time step) and 50% of the values are an average over two radiation time steps (i.e. an average over two instantaneous values which are two hours apart)."

P8 L29: M7 should be mentioned/discussed in the previous subsection on ECHAMHAM (without SALSA).
Author response:
A brief description of M7/HAM was added: "Aerosol microphysical processes such as nucleation, coagulation, condensational growth are computed by the modal scheme M7 (Vignati et al, 2004). HAM computes further processes such as emissions, sulfur chemistry (Feichter et al., 1996), dry depositon, wet deposition, sedimentation, aerosol optical properties, aerosol-radiation and aerosol-cloud interactions."

P9 L20: What does "aerosol life cycle scheme which calculations production tagged mass" mean? Is this an aerosol microphysics scheme? Does it track aerosol composition by its emission/process source in addition to chemical composition? Please rewrite for clarity.
Author response:
We have rewritten and expanded the description of the aerosol scheme to improve clarity as follows: "The aerosol microphysics scheme in the NorESM version of CAM, called CAM-Oslo, consists of 12 log-normally shaped background modes which are tagged accord-ing to emission source and chemical composition (Kirkevåg et al., 2018). The shapeof these modes can change due to condensation and coagulation."

P9 L34: Reference is missing a year.
Author response:
Fixed.

P11 L22: "CER" isn't defined until below.
Author response:

The acronym CER is already introduced at P6, L34.

P12 L24-25: Is there a figure that we should be looking at to see these biases?
Author response:
We included the reference to the corresponding figures.

*P13 L26: Do you specifically mean the *model* datasets here, or is the MODIS data being lumped into this comparison too.*
Author response:
Indeed we referred to the model datasets. The sentence was modifed as suggested: "The spatial distribution of the cloud physical and optical properties is remarkably similar among the model datasets with the exception of CER_{ice}, IWP (Fig. 2 d and l)and COT (Fig. 3g,k)."

P15 L22: It seems very subjective to say that a bias of -0.2 is "quite close" given that most of the globe has an AI below 0.2 according to MODIS (so this bias is larger than the AI value in nearly all locations. (Also, most of the locations with AI > 0.2 are over land, where we should not trust the Angstrom Exponent).
Author response:
The sentence was unclear. The author meant that a similar bias is shown by the three models, as each bias is on average about 0.2. The sentence has been rephrased as: "The biases between values of AI from directmodel output and MODIS observations are quite close among the model as their average is about of +0.2."

P17 L9: There is a discussion here about AI over land, but there is no acknowledgement that the MODIS aerosol team does not publish AE over land.
Author response:
The following sentence was added: "As these negative values are derived over land regions, it could be indicative of retrieval biases over bright surfaces (i.e. snow or ice). Furthermore, it is important to inform the readers that MODIS aerosol size parameters over land (i.e. AE or fine-AOD) are no longer official products directly provided by the MODIS aerosol team. The publication of these variable were discontinued due to low quantitative MODIS skill (Mielonen et al.,2011; Levy et al., 2013). Using spectral AOD, we derived AE over land and derived AI on a global scale to allow estimates of ACI on a global scale (Fig.S4). However, the AE values over land were not evaluated."

P18 L25-26: "possibly owing. . ." onward. It is unclear to me what this is saying.
Author response:
The sentence has been updated accordingly to the new version of Figure 7.

P19 L3-4: What is the difference between "model calculation" and "cloud parameterization". These seem like synonyms? Or is the "model calculation" specifically referring to the COSP simulator (rather than the atmospheric model).

Author response: The terms "model calculation" and "model parametrization" describe different aspects of atmospheric modeling. While the term "model calculation" refers in the sentence to the COSP simulator, the term parametrization refers to the climate model. In particular, the latter term is used to describe the approach implemented in any atmospheric model to simplify too complex or too detailed processes to be explicitly resolved withing the model.

P19 L22: How does one select dry aerosols when using satellite-derived properties? Or is this a statement of when using modelled properties only?
Author response:
The sentence refers to the study that Neubauer performed using the model ECHAM-HAM.
The sentence has been rephrased in the manuscript as follow: "The results highlight that a minimum distance between cloud and aerosol gridded data should be taken into account in the anaylsis of satellite data, and that dry aerosols should be selected to reduce the influence of aerosol growth due to humidity for model simulations when comparing satellite-based and model estimates for ACI."

P19 L34-35: What property is being underestimated?
Author response:
Cloud fraction is the parameter omitted in the sentence. The sentence has been rephrased as: "We highlighted many discrepancies in cloud spatial and vertical representations and the results showed that the three models overall similarly underestimate cloud fraction for the stratocumulus cloud regime being when compared to MODIS."

Author Response to Referee 2

During the review of the manuscript, two errors were found in the text:

1. We selected the maximum column value of CDNC of direct output of the models and not the values at cloud top. The text has been revised accordingly at page 11, line 29.
2. We refer in the text to AI, and consequently, ACI estimates, for land areas only. Clearly, this is incorrect as AI values are shown over ocean only. The text is revised:
   - at page, 4 line 35;
   - at page 5, line 1;
   - in the caption of Figure 5 and Figure 7;
   - in the caption of Table 1.

General comment:

The authors simply mention that the MODIS-derived CDNC and the direct model output are different, which is true, but offer no explanation of why those difference appear. Intuitively, the authors assume that the CDNC at cloud top should correspond to the MODIS retrieval, but omit the fact that MODIS does not produce CDNC directly. Instead using the method from Bennartz et al. (2007) one may argue that CDNC at cloud base or even the maximum CDNC in the column should be used. Also since the assumption of adiabaticity and vertically constant CDNC is embedded in the MODIS-derived CDNC the authors should limit the analysis only to regions where those assumption are valid (probably mostly over ocean).

Author response:

We present some possible explanation regarding the differences between MODIS-derived CDNC and the direct model output for CDNC (page 15, line 1-10). However, we do not enter in details as it would go beyond the scope of the paper. The result, being the direct modeled CDNC being lower than the COSP-derived CDNC, is supported by a similar conducted by Ban-Weiss et al. (2014). Therefore, this is an interest topic that should be investigated in future work.

The paragraph has been modified as:

"Consequently, the CDNC from direct model output is lower than MODIS-COSP diagnostics. Possible explanations could be either related to the COSP method for deriving $\mathrm{CER_{liq}}$ and $\mathrm{COT_{liq}}$ or the approach used for deriving CDNC from $\mathrm{CER_{liq}}$ and $\mathrm{COT_{liq}}$ or related to the fact that the computation of the direct CDNC requires a minimum number of CDNC set to 40 $\mathrm{cm^{-3}}$. This outcome is very similar to what has been found by Ban-Weiss et al. (2014), where the CDNC satellite-simulated values are higher than the standard CAM5 model output near clout top as well as the column maximum value. Therefore, this result represent an interesting topic that extends beyond the scope of this paper but it should further developed in future work. "

Further discussion on the different results between the model direct CDNC output and the COSP-derived CDNC is included in section 4.1, page 15, lines 4-20. Considerations regarding the limitations of MODIS-derived CDNC are presented in the manuscript. Alternative methodologies exists for deriving CDNC at cloud base such as the approach developed by Rosenfeld et al. (2016). However, this method is specifically design for Suomi NPP satellite retrievals and based on adiabacity assumptions. Ban-Weiss et al. (2014) showed that the modelled CDNC is lower than values derived from satellite observation regardless the selection of CDNC at cloud top or the maximum CDNC in the column.

MODIS provides three separate products of the cloud properties (i.e. cloud effective radius, optical depth and water path) using three different water absorbing channels (1.6 μm, 2.1 μm and 3.6 μm). The signal of the these three wavelengths implies different penetration depths in the cloud system, which has clearly obvious consequences on the capabilities of the wavelengths to be responsive to cloud top or near-cloud-top microphysics as shown in Rosenfeld et al. (2003), for example. The 3.6 μm has the least in cloud penetration depth, therefore the most representative of cloud top microphysics, and showed the best agreement with in situ value (King et al. 2013). Furthermore, Zhang at al.

(2012) showed that the 3.6 μm has shown is less sensitive to the plane-parallel cloud assumption.

It is explicitly mentioned (i.e. Page 1, Line 9-11; Page 3, Line 31-33) that the CDNC is not a product of MODIS and that the CDNC is identically derived from MODIS and COSP values of CER and COT following the methods of Bennartz et al. (2007), which is described in the dedicated Section 3.2. MODIS-derived CDNC is compared with the CDNC derived from COT and CER simulated from COSP as well as with the CDNC modeled direct output.

Regarding the spatial coverage of the CDNC bias distribution, the authors would prefer including both land and ocean. In order to clarify the possible implications of the assumptions embedded in the methodology used in the CDNC calculation, a new sentence has been added in the manuscript (see Specific Comment below). The discrepant results from the comparison of COSP-derived CDNC and the modeled direct output of CDNC, where the MODIS data represents solely the reference dataset rather than "truth" data, stem from the aerosol model set-up and this discrepancy has potentially important implications for the modelling community, using satellite observations to evaluate standard (not satellite-simulated) model output.

References:

Ban-Weiss, G. A., et al. (2014), Evaluating clouds, aerosols, and their interactions in three global climate models using satellite simulators and observations,J. Geophys. Res. Atmos.,119,10,876–10,901, doi:10.1002/2014JD021722.

King, N. J., et al. (2013): Evaluating MODIS cloud retrievals with in situ observations from VOCALS-REx, Atmos. Chem. Phys., 13, 191-209, https://doi.org/10.5194/acp-13-191-2013.

Rosenfeld, D. and C.W. Ulbrich, (2003): Cloud Microphysical Properties, Processes, and Rainfall Estimation Opportunities. Meteorological Monographs, 52, 237–258, https://doi.org/10.1175/0065-9401(2003)030¡0237:CMPPAR¿2.0.CO;2

Rosenfeld, D., et al. (2016), Satellite retrieval of cloud condensation nuclei concentrations by using clouds as CCN chambers, Proceedings of the National Academy of Sciences of the United States of America, 113(21), 5828-5834, doi:10.1073/pnas.1514044113.

Zhang, Z., et al (2012): Effects of cloud horizontal inhomogeneity and drizzle on remote sensing of cloud droplet effective radius: Case studies based on large-eddy simulations. J. Geophys. Res., 117, no. D19, D19208, doi:10.1029/2012JD017655.

The global CNDC-aerosol index sensitivity calculations (what the authors mistakenly call ACI throughout the manuscript) are also mostly descriptive and even though some speculation is given on the possible causes for discrepancy I imagine there is enough model output to go deeper (see specific comments). Also, in principle calculating the "ACI" from 2D vertically integrated fields makes little sense. It is not clear what role the assumptions of overlap are, or even whether the cloud and the aerosol occupy the same space.

Author response:

We thank the Referee for addressing some well-known critical points in the satellite-based assessment of the ACI parameter. Some aspects are presented in the conclusion section (Page 19, Lines 10-30). However, in section 3.3 we added a paragraph that acknowledges the possible implications of the approach used to infer the CDNC-aerosol index sensitivity, or ACI.

The following paragraph is now added in Sect. 5:

"Some of the challenges and limitations in assessing ACI are now highlighted. MODIS AOD retrievals are limited to cloud-free conditions, which creates challenges to studying the ACI where the intention is to study collocated aerosol and cloud observations. Unless height-resolving instruments (i.e. lidars) are considered, the vertical location of the aerosol is unknown. Aerosol and cloud measurements may contain retrieval errors, which are further propagated to ACI estimates, as well as they reciprocally may bias the respective retrievals (Jia et al., 2019). The interpretation of the observed aerosol-cloud relationships is further complicated by the effect of meteorology (Quaas et al., 2010; Gryspeerdt et al., 2014; Gryspeerdt et al., 2016; Brenguier2003). As cloud formation happens in high humidity conditions, aerosol humidification can severely affect the assessment of ACI by causing positive correlation between AOD and cloud properties (Myhre et al., 2007; Quaas et al.,2010; Gryspeerdt et al., 2014; Gryspeerdt et al., 2016). Additionally to aerosol particles, water vapour also affects precipitation (Boucher et al., 2013), obviously linked to the presence of clouds, and consequently causes spurious correlations between aerosols and clouds (Koren et al, 2012)."

The following additional references have been included in the manuscript:

Brenguier, J.-L., Pawlowska, H., and Schüller, L.: Cloud micro-physical and radiative properties for parameterization and satellite monitoring of the indirect effect of aerosol on climate, Journalof Geophysical Research: Atmospheres, 108, https://doi.org/10.1029/2002JD002682, URLhttps://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/200
2003.

Gryspeerdt, E., Stier, P., and Grandey, B. S.: Cloud fraction mediates the aerosol optical depth-cloud top height relationship, Geo-physical Research Letters, 41, 3622–3627, https://doi.org/10.1002/2014GL059524, URLhttps://agupubs.onlinelibrary.wiley.com/
2014.

Gryspeerdt, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, Journal of Geophysical Research: Atmospheres, 121, 3566–3583, https://doi.org/10.1002/2015JD023744, URLhttps://agupubs.onlinelibrary.wiley.com/doi/abs/
2016.

Jia, H., Ma, X., Quaas, J., Yin, Y., and Qiu, T.: Is positive correlation be-tween cloud droplet effective radius and aerosol optical depth over land due to retrieval artifacts or real physical processes?, Atmospheric Chemistry andPhysics, 19, 8879–8896, https://doi.org/10.5194/acp-19-8879-2019, URLhttps://www.atmos-chem-phys.net/19/8879/2019/, 2019.

Koren, I., Oreopoulos, L., Feingold, G., Remer, L. A., and Altaratz, O.:Aerosol-induced intensification of rain from the tropics to the mid-latitudes,Nature Geoscience, 4, https://doi.org/10.1038/ngeo1364, URLhttps://doi.org/10.1038/ngeo1364, 2012.

Myhre, G., Stordal, F., Johnsrud, M., Kaufman, Y. J., Rosenfeld, D.,Storelvmo, T., Kristjansson, J. E., Berntsen, T. K., Myhre, A., and Isak-sen, I. S. A.: Aerosol-cloud interaction inferred from MODIS satellite data and global aerosol models, Atmospheric Chemistry and Physics, 7,3081–3101, https://doi.org/10.5194/acp-7-3081-2007, URLhttps://www.atmos-chem-phys.net/7/3081/2007/, 2007.

Finally there is the issue of "the ACI". Aerosol-cloud interactions encompass many processes occurring in clouds as a result of the presence of aerosols. As a noun, aerosol cloud interactions is an area of study, not a metric. So it is troubling, and in many places grammatically incorrect, that ACI are reduced to a single number and equated to the CDNC-AI sensitivity. Please correct this and be precise in the terminology used.

Author response:

We agree with the Reviewer that by definition the aerosol-cloud interactions encompasses different processes. The original text at Page 4, Lines 1-14, has been rephrase and expanded as:

"Aerosol-cloud interactions are based on the role of aerosol particles in changing cloud properties, which involves several processes (Bellouin et al., 2019). In this study we focus one of these processes, known as the first aerosol indirect effects or Twomey effect, which can be quantified by an indicator parameter, called ACI, defined as the change in an observable cloud property (e.g., cloud optical depth, cloud effective radius, cloud droplet number concentration) to a change in a cloud condensation nuclei proxy (e.g. aerosol optical depth, aerosol index, or aerosol particle number concentration)." The analysis of aerosol-cloud interactions has been reported in literature by a variety of methods: studies presenting results from global scales (Feingold et al., 2001; Quaas et al., 2010) to regional scales(i.e Saponaro et al., 2017; BanWeiss et al., 2014; Liu et al., 2017; Liu et al., 2018) and in-situ observations (i.e. Sporre et al., 2014), using different approaches, i.e. observations from satellites, airborne and ground based instrumentation, or modelling. Nonetheless, the quantification of the aerosol-cloud interactions is still a major uncertainty in understanding climate change (eg. Lohmann et l., 2007; Quaas et al., 2009; Storelvmo et al., 2012; Boucher et al., 2013; Lee et al., 2015; Seinfeld et al., 2016; Bellouin et al., 2019).

The authors argue that the acronym ACI has already been used extensively in literature to purely define the mathematical relationship representing the sensitivity of clouds to changes in aerosol loading, eg. $\mathrm{ACI} = \mathrm{dln}(\mathrm{N_d})/\mathrm{dln}(\mathrm{AI})$. The authors agree that using the acronym ACI for both the metric and the topic can be confusing for the readers, thus it is important to adopt one terminology for the mathematical expression and one for the more general discussions. Therefore, the manuscript is updated by using the term $\mathrm{ACI_{CDNC}}$ only for the mathematical parameter representing the first aerosol indirect effect, as it common in literature, and the term "aerosol-cloud interactions" when the topic is discussed in general.

The following additional references have been included in the manuscript:

Bellouin, N. et al (2019). Bounding global aerosol radiative forcing of climate

change. Reviews of Geophysics, 57. https://doi.org/10.1029/2019RG000660
Seinfeld J. H. et al. (2016). Improving our fundamental understanding of the role of aerosol-cloud interactions in the climate system. Proceedings of the National Academy of Sciences May 2016, 113 (21) 5781-5790; DOI: 10.1073/pnas.1514043113

Specific comments:

Page 3, Line 34. Why 2008? The horizontal resolution is low enough that it should be easy to run a couple of years at least. MODIS data spans at least 15 years as well.
Author response:
When the simulations were made, 2008 was the default year for AEROCOM model experiments. We also adopted this year to support the intercomparison of the results.

Page 3, Line 35. No, the aerosol- cloud interaction is not a metric, and certainly not confused with the aerosol indirect effect. Even saying "the aerosol cloud interaction" is probably incorrect. Moreover, ACI is referred here as a metric and later on as a topic. Please be precise in the terms used.
Author response:
The sentence has been rephrased as:
See General Comment above.

Page 4 Line 32. This is the CDNC-AI sensitivity. Also I am not convinced that the meteorological component is completely removed, please explain.
Author response: We do not claim that meteorologic influence are completely removed (Page 4, Line 21-23) and this is clearly stated in the added paragraph in Section 3.3 as well as in the conclusive section (Page 20, Lines 15-30).

Page 4 Line 32. Why should the liquid water path remain constant? Is there a clear connection between the CDNC-AI sensitivity and the first aerosol indirect effect?
Author response:
The aerosol-cloud interaction is a concept introduced to compare how changes in aerosol loading affect cloud properties. The ACI can be defined as:

$$\mathrm{ACI} = \frac{\mathrm{dlnCOT}}{\mathrm{dln}(\sigma)} = -\frac{\mathrm{dlnCER}}{\mathrm{dln}(\sigma)} = \frac{1}{3}\frac{\mathrm{dlnCDNC}}{\mathrm{dln}(\sigma)} \qquad (1)$$

where σ is a proxy for the CCN concentration. From the theory (Twomey, 1977; Feingold et al., 2001; MCcomiskey and Feigold, 2008) for the first two parts of the equations, the LWP needs to be constant in the calculation of the partial derivatives while the third part of the equation (that is the one we use in our study) does not require any restriction of the LWP. Answering the Referee's second point: the CDNC-AI sensitivity is indeed an analytic approach to quantify the first aerosol indirect effect alternatively to the models described in the first two term of the Eq. (1).

Page 9, Line 17. Why is the vertical resolution different than when running ECHAM-HAM?

Author response:

The version of ECHAM-HAMMOZ with SALSA was the default configuration at the time when the simulations were run. However, it has to be noted that the difference between L31 of HAM and L47 of SALSA does not increase the vertical resolution of lowest grid-points but rather adds levels in the mid-atmosphere. The lowermost levels of L31 and L47 (up to about 100 hPa) are identical and Neubauer et al. (2019) have shown that the difference in cloud properties between the two vertical resolutions are minor.

Reference:

Neubauer, D., Ferrachat, S., Siegenthaler-Le Drian, C., Stier, P., Partridge, D. G., Tegen, I., Bey, I., Stanelle, T., Kokkola, H., and Lohmann, U.: The global aerosol–climate model ECHAM6.3–HAM2.3 – Part 2: Cloud evaluation, aerosol radiative forcing, and climate sensitivity, Geosci. Model Dev., 12, 3609–3639, https://doi.org/10.5194/gmd-12-3609-2019, 2019.

Page 11, Line 6-8. This seems incorrect and could be a major flaw. COSP should account for the fact that MODIS only sees in-cloud values. It makes little sense to divide two column-integrated, 2D values. If COSP cannot account for it, then the 3D model calculation should be converted to in-cloud values before sending it to COSP to calculate MODIS-like values. Please clarify.

Author response:

The calculation are correct. COSP does account for that MODIS only see in-cloud values. However, since the model output of cloud parameters from ESMs most often are grid-box averages, the in-cloud values from COSP are multiplied with cloud fraction before they are written as output. To get the in-cloud values one therefore needs to divide the output by cloud fraction in the post processing.

Page 11, Line 18. Is there a MODIS algorithm to retrieve CDNC? It may be more correct to say in MODIS cloud effective radius is biased towards cloud top values, which may propagate to the CDNC calculation. The method used to estimate CDNC (equation 1) should approximate better cloud-base values.

Author response:

See General Comment above.

Page 12, Section 3.2. Please label this equation, and explain the assumptions behind it. Given than clouds are 3D, to what vertical level should this calculation correspond? Also, if this applies well in the stratiform marine boundary layer why are the authors applying it globally? What would be the error incurred in applying it to a shallow convective trade cumulus for example?

Author response:

The authors thank the Referee for pointing out the absence of an explanation of the assumption behind the equation, now labelled in the manuscript as Eq.1.

The following additional paragraph has been added in Sect.3.2.: "The assumption of not accounting for temperature effect and setting γ as a bulk costant applies rather well to the warm stratiform clouds in the marine boundary layer but less for convective clouds (Bennartz, 2007; Rausch et al., 2010; Grosvenor et al, 2018). The equation represents the "Idealized Stratiform Boundary Layer Cloud" (ISBLC) model (Bennartz and Rausch, 2017) which is based on the following assumptions:

- the cloud is horizontally homogeneous;
- the LWC increases linearly from the cloud base to the cloud top;
- the CDNC is constant throughout the vertical extent of the cloud.

While the ISBLC model describes important aspects of stratiform boundary layer clouds, its assumption will never be fully valid for any real cloud. Issues related to the ISBLC model assumptions are extensively elaborated in (Bennartz, 2007; Bennartz and Rausch, 2017) and references therein."
The following reference was added to the references:
Grosvenor et al, (2018): Remote Sensing of Droplet Number Concentration in-Warm Clouds: A Review of the Current State of Knowledge and Per-spectives, Reviews of Geophysics, 56, 409–453, https://doi.org/10.1029/2017RG000593, URLhttps://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2017RG000593.

Page 12, Line 15. This is the CDNC-AI sensitivity, not "the ACI".
Author response:
Please, see Author response to the corresponding General Comment.

Page 12, Lines 16-28. Please expand this. Is this done using a linear regression of the CDNC and AI daily time series? Given that this is a global calculation why would the number of data points be different? Also, it is not clear what the "ACI" from 2D vertically integrated fields represents (see general comments).
Author response:
When calculating the ACI for the 1 x 1 degree grid points the ACI metric is applied to data for a given season and grid box. This methodology can be thought of as calculating the linear regression slope of a scatter plot of ln(CDNC) vs. ln(AI), where each point represents a day for which both aerosol and cloud data exist for this grid box. As we follow directly the methodology presented by Grandey and Stier (2010), we do not repeat the details. The reader can find additional information in the mentioned reference. The number of data are different only when the dataset is divided accordinly to the season. A clarification regarding the ACI calculation is presented in the General Comment.
The following sentence was included in the manuscript:
"When calculating the ACI_{CDNC} for the 1 degree x1 degree gridboxes Eq.1 is applied to data for the selected season and grid box. This methodology (Grandey and Stier, 2010) can be thought of as computing the linear regression slope of a scatter plot of ln(CDNC) versus ln(AI), where each point represents a day for

which both aerosol and cloud data exist for the considered grid box."

Page 13, Line 18. Does COSP produce COP CF or CF all? Maybe the discussion should be limited to that one.
Author response:
COSP simulates CF_{all}. However, the author consider important to show also the results of COP-CF because it is valuable for modelling and satellite communities.

Page 15, Lines 1-10. This is very vague and should be explored in more detail (see comments above).
Author response:
Please, see Author response to the corresponding General Comment.

Page 15, Lines 17-20. Is the difference due to the activation scheme or to the aerosol models?
Author response:
The difference is due to the difference in aerosol models since they both use the same ICNC scheme.

Page 15, Lines 25-30. These two sentences contradict each other.
Author response:
The sentence has been rephrased as follows:
"Despite the higher CDNC, CER_{liq} is larger in ECHAM-HAM-SALSA than in ECHAM-HAM. Although it is expected that CER_{liq} decreases with increasing CDNC, higher LWP in ECHAM-HAM-SALSA in turn results in larger CER_{liq} and outweighs the CDNC effect on CER_{liq}. The causes for differences in LWP between the two model versions are very difficult to diagnose but candidates for causing them are differences in wet scavenging, convective detrainment or freezing."

Page 15, Lines 30-35. This is purely speculative.
Author response:
The sentence has been deleted. See previous comment.

Page 17, Line 15-25. Could this be corroborated? It seems odd that the sensitivity would be negative.
Author response:
Several studies, some of which we mention in the manuscript, have found negative values of ACI.

Page 17, Line 25. Global maps of CDNC-AI sensitivity should show this better.
Author response:
A bar plot provides a more concise elaboration of the ACI estimates including the 95% confidence intervals. Additionally, we argue that the plot bar enables a quick comparison with similar studies. For these reasons, the authors wish to

maintain Fig. 7 in its original plot.

Evaluation of aerosol and cloud properties in three climate models using MODIS observations and its corresponding COSP simulator, and their application in aerosol-cloud interactions

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Abstract.

The evaluation of modeling diagnostics with appropriate observations is an important task that establishes the capabilities and reliability of models. In this study we compare aerosol and cloud properties obtained from three different climate models ECHAM-HAM, ECHAM-HAM-SALSA, and NorESM with satellite observations using MOderate Resolution Imaging Spectrometer (MODIS) data. The simulator MODIS-COSP version 1.4 was implemented into the climate models to obtain MODIS-like cloud diagnostics, thus enabling model to model and model to satellite comparisons. Cloud droplet number concentrations (CDNC) are derived identically from MODIS-COSP simulated and MODIS-retrieved values of cloud optical depth and effective radius. For CDNC, the models capture the observed spatial distribution of higher values typically found near the coasts, downwind of the major continents, and lower values over the remote ocean and land areas. However, the COSP-simulated CDNC values are higher than those observed, whilst the direct model CDNC output is significantly lower than the MODIS-COSP diagnostics. NorESM produces large spatial biases for ice cloud properties and thick clouds over land. Despite having identical cloud modules, ECHAM-HAM and ECHAM-HAM-SALSA diverge in their representation of spatial and vertical distribution of clouds. From the spatial distributions of aerosol optical depth (AOD) and aerosol index (AI), we find that NorESM shows large biases

for AOD over bright land surfaces, while discrepancies between ECHAM-HAM and ECHAM-HAM-SALSA can be observed mainly over oceans. Overall, the AIs from the different models are in good agreement globally, with higher negative biases on the Northern Hemisphere. We evaluate the aerosol-cloud interactions by computing the sensitivity parameter ACI_{CDNC} = dln(CDNC)/dln(AI) on a global scale. However, one year of data may be considered not enough to assess the similarity or dissimilarities of the models due to large temporal variability in cloud properties. This study shows how simulators facilitate the evaluation of cloud properties and expose model deficiencies, which are necessary steps to further improve the parametrization in climate models.

1 Introduction

A climate model is a powerful tool for investigating the response of the climate system to various forcings, enabling climate forecasts on seasonal to decadal time scales, and therefore can be used for estimating projections of the future climate over the coming centuries based on future greenhouse gas and aerosol forcing scenarios (Flato, 2011). Based on physical principles, climate models reproduce many key aspects of the observed climate and aid to understand the dynamics of the physical components of the climate systems.

The evaluation of modeling diagnostics is an important task that establishes the capabilities and reliability of models. When key properties of the atmosphere (e.g., clouds, aerosols) are considered, the model assessment is relevant to assure that the climate model correctly captures key features of the climate system. The interest in the reliability of climate models reaches outside the scientific community, as these simulations will form the basis for future climate assessments and negotiations. Therefore, understanding the level of reliability is a necessary step to strengthen the robustness of climate projections and, if necessary, improve the model parametrizations for the relevant processes.

For the evaluation of parametrizations of aerosol indirect effects in global models, satellite data have been proven to be useful (Quaas et al., 2009; Boucher et al., 2013) as they provide large spatial coverage at suitable temporal resolution. Satellite instruments measure the intensity of radiation coming from a particular direction in a selected wavelength range. From the observed radiances, the geophysical quantities are then inferred by inverse modeling using a retrieval algorithm.

The compensation of modeling errors, the intrinsic uncertainties of observational data, and the possible discrepant definitions of variables between models and observational data are major issues affecting the crucial task of model evaluation. For that, satellite simulators have been developed to mimic the retrieval of observational data and to avoid ambiguities in the definition of variables mentioned above. Simulators recreate what the satellite would retrieve when observing the modeled atmosphere. By reprocessing model fields using radiative transfer calculations, they generate physical quantities fully consistent with the satellite retrievals. By including microphysical assumptions, which usually differ between models, inconsistencies in the simulators are avoided. Hence, simulators represent a robust and consistent approach not only for the application of satellite data to evaluate models, but also for model-to-model comparisons. Simulators have been widely used, and their implementation in several models enables intercomparison studies on atmospheric variables, such as clouds, aerosols (Quaas et al., 2009; Williams and Bodas-Salcedo, 2017; Zhang et al., 2010; Luo et al., 2017), and upper atmospheric humidity (Bodas-Salcedo et al., 2011).

Two prominent examples of simulators are the simulator developed as part of the International Satellite Cloud Climatology Project, ISCCP, (Klein and Webb, 2009; Yu et al., 1996) and the CFMIP (Cloud Feedback Model Intercomparison Project) Observation Simulator Package, COSP (Bodas-Salcedo et al., 2011). CFMIP is part of The Coupled Model Intercomparison Project (CMIP) (Eyring et al., 2016b; Webb et al., 2017), which is a framework providing the modeling community with guidelines for the development, tuning and evaluation of models (Eyring et al., 2016a, c). COSP is a software tool developed within the CFMIP (Webb et al., 2017), which extracts parameters for several spaceborne active sensors, such as the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) and the Cloud Profiling Radar (CPR), as well as for passive sensors, such as the Multi-angle Imaging SpectroRadiometer (MISR) and the Moderate Resolution Imaging Spectroradiometer(MODIS).

In this study the COSP version 1.4 was implemented in three climate models, namely ECHAM-HAM, ECHAM-HAM-SALSA and NorESM, and the diagnostic outputs of the MODIS simulator were compared to MODIS observational data collected during the year 2008. The main goal of this study is to evaluate the models' capability to realistically represent clouds by employing MODIS satellite observations and its corresponding COSP simulator. A secondary goal of the study is to estimate the aerosol-cloud interactions through the application of the cloud droplet number concentration (CDNC) derived from observed and COSP simulated values of cloud optical thickness and effective radius. Aerosol-cloud interactions are based on the role of aerosol particles in changing cloud properties, which involves several processes (Bellouin et al.). In this study we focus one of these processes, known as the first aerosol indirect effects or Twomey effect, which can be quantified by an indicator parameter, called ACI, defined as the change in an observable cloud property (e.g., cloud optical depth, cloud effective radius, cloud droplet number concentration) to a change in a cloud condensation nuclei proxy (e.g. aerosol optical depth, aerosol index, or aerosol particle number concentration). The analysis of aerosol-cloud interactions has been reported in literature by a variety of methods: studies presenting results from global scales (Feingold et al., 2001; Quaas et al., 2010) to regional scales (e.g. Saponaro et al., 2017; Ban-Weiss et al., 2014; Liu et al., 2017, 2018) and in-situ observations (e.g. Sporre et al., 2014), using different approaches, i.e. observations from satellites, airborne and ground based instrumentation, or modelling. Nonetheless, the quantification of the aerosol-cloud interactions is still a major uncertainty in understanding climate change (e.g. Lohmann et al., 2007; Quaas et al., 2009; Storelvmo, 2012; Flato et al., 2013; Lee et al., 2016; Seinfeld et al., 2016; Bellouin et al.).

The choice of observations and spatial scale of a study presents intrinsic uncertainties when quantifying aerosol-cloud interactions, and some of them relate to spatial or temporal limitations or artifacts (McComiskey and Feingold, 2012). When considering satellite observations, cloud and aerosols properties are provided at a quite comprehensive spatial and temporal coverage; however several aspects bring challenges in the analysis of these observations. The primary artifacts known to affect satellite estimation of aerosol-cloud interactions are related to (1) the inability of untangling aerosol and cloud retrievals from meteorology (e.g. aerosol humidification, entrainment, cloud regimes dependency), (2) inaccuracies in the retrieval algorithms (e.g. near-cloud impacts on radiative transfer, contamination, statistical aggregation) and (3) assumptions in the retrieval algorithms (Koren et al., 2007; Oreopoulos et al., 2017; Christensen et al., 2017; Wen et al., 2007).

In this work, the Cloud Feedback Model Intercomparison Project (CFMIP) Observation Simulator Package (COSP) (Bodas-Salcedo et al., 2011) is implemented in three climate models to obtain satellite-like diagnostics that enable a direct comparison with satellite retrieval fields. In particular, we focus on liquid cloud properties, which are used to derived CDNC. Cloud droplet number concentration is computed for both satellite observations and satellite-simulated values in a consistent way using an algorithm presented in Bennartz (2007). Aerosol-cloud interactions are quantified over ocean by parameter ACI_{CDNC} = dln(CDNC)/dln(AI). By considering the changes in CDNC, it is possible to isolate the microphysical component of the ACI without the need for constraining the liquid water path.

In Section 2 we provide details of the MODIS data, the models, and the COSP simulator. Section 3 presents the methods used in the analysis of the data. The evaluation of the simulator cloud diagnostics with MODIS satellite data on a global scale is presented in subsections 4.1 and 4.2, while the ACI results are shown in subsection 4.3. Conclusions are summarised in Section 5.

2 Data

2.1 MODIS

The Moderate Resolution Imaging Spectrometer (MODIS) is a 36-channel radiometer flying aboard the Terra and Aqua platforms since 2000 and 2002, respectively, which views the entire Earth's surface every 1 to 2 days, thus representing an extensive data set of global Earth observations. MODIS delivers a wide range of atmospheric products including aerosol properties, water vapour, cloud properties, and atmospheric stability variables.

We consider data for the year 2008 from MODIS-Aqua since its equatorial crossing time (13:30 local time) ensures a more complete development of the cloud during its daily cycle. MODIS Level-1 (L1) products are geo-located brightness and temperature values, which are elaborated into geophysical data products at Level-2 (L2), and aggregated onto a uniform space-time grid at Level-3 (L3). We used the latest Collection 6.1 daily MODIS/Aqua MYD08L3, which is a regular gridded Level-3 daily global product (Hubanks et al., 2016). The dataset is limited to observations made during daytime, because these contain a richer set of retrievals and better accuracy in cloud detection. As the L3 1° x 1° gridded average values of atmospheric properties, along with a suite of statistical quantities, are derived from the corresponding L2 atmosphere data product, a brief description of Level-2 MODIS aerosol and cloud products is now presented.

The Level-2 MODIS aerosol products provide information regarding the aerosol loading and aerosol properties over cloud-, snow-, and ice-free land and ocean surfaces at a spatial resolution of 10 km x 10 km. The primary aerosol product is the aerosol optical depth (AOD), derived globally at the wavelength of 550 nm, while the other parameters accounting for the aerosol size distribution, such as the Ångström exponent (AE) or fine-mode aerosol optical depth, are only derived over ocean (Levy et al., 2013). Additionally, the aerosol index (AI) can be derived by multiplying AOD by AE. The MODIS aerosol products have been extensively validated using highly-accurate observations made by the Aerosol Robotic Network (AERONET) (Sayer et al., 2014) showing good agreement with in-situ measurements. The uncertainty in MODIS retrievals of AOD from validation studies (Levy et al., 2007) was quantified at $0.03 + 0.05 \times \tau_A$ over ocean and $0.05 + 0.15 \times \tau_A$ over land, where τ_A is the reference AOD value from AERONET. In this study we primarily focus on the analysis of liquid cloud properties. However, MODIS aerosol data (Levy et al., 2013) is needed to assess aerosol-cloud interactions.

The Level-2 MODIS physical and optical cloud properties are derived trough a combination of infrared emission and shortwave reflectance techniques at a spatial resolution varying from 1 km to 5 km, depending on the parameter (Platnick et al., 2017). Collection 6.1, which is used in this work, provides cloud optical parameters divided into different products accordingly to the cloud phase and retrieved at wavelengths of 2.1 μm, at 1.6 μm and 3.7 μm (Hubanks et al., 2016; Platnick et al., 2017). As the COSP simulator simulates cloud properties at 2.1 μm, the same wavelength is selected in the MODIS observations for both ice and liquid clouds. MODIS offers two scientific L3 cloud fractions datasets, namely the cloud mask cloud fraction and the cloud optical properties cloud fraction (datasets with prefix 'Cloud Fraction' and 'Cloud Retrieval Fraction', respectively). From now on we refer to the cloud mask cloud fraction as CF, and to the cloud optical properties cloud fraction as COP CF. While the CF counts the proportion of the pixels classified by the cloud mask as cloudy or partly cloudy, the COP CF counts the proportion of the pixels for which cloud optical properties have been successfully derived. The main difference between these two definitions roots in the approach of handling partly cloudy pixels. As the task of the cloud mask is to identify fully clear pixels, partly cloudy pixels are counted as cloudy in CF, while in the COP CF they are counted as clear because the retrieval algorithm aims to include only fully cloudy pixels. The different treatment of partly cloudy pixels directly impacts the number of cloud pixels, and consequently many other retrieved cloud properties. Therefore differences are expected in our results and as already reported by Pincus et al. (2012). MODIS observations are here used as a reference dataset. However, MODIS data contains its own errors and limitations. Many studies compared MODIS liquid cloud microphysical properties with in-situ and airborne campaign measurements finding strong correlations for COT but a systematic significant overestimation of MODIS cloud-top droplet effective radius (CER) for marine stratus and stratus cumulus clouds due to possible instrument limitation and algorithm retrieval assumptions (e.g. Noble and Hudson, 2015; Painemal and Zuidema, 2011; Min et al., 2012). A good CER correlation between MODIS and in-situ data was however observed by e.g. Preißler et al. (2016) for marine warm stratiform clouds at higher latitudes. A bias in MODIS CER is propagated into the derivation of MODIS LWP, which also shows a positive bias with respect to the observations (e.g. King et al., 2013; Noble and Hudson, 2015; Painemal and Zuidema, 2011; Min et al., 2012). Overestimated MODIS LWP were also found over a high-latitude measurement land site (e.g. Sporre et al., 2016) for clouds from all altitudes in the atmosphere. Marchant et al. (2016) showed that the C6 cloud phase discrimination algorithm is significantly improved over C5 but some situations continue to be problematic over regions located at higher latitudes (i.e., polar areas, Greenland, and large desert areas).

In this study, we derive CDNC following the method presented in Bennartz (2007) and this additional cloud parameter is used in the computation of ACI. More information is provided in Sect. 3.2.

2.2 COSP - The CFMIP Observation Software Package

The simulator COSP (Bodas-Salcedo et al., 2011) is a publicly available software package (https://www.earthsystemcog.org/projects/cfmip/) developed by the CMIP community (Webb et al., 2017). It consists of a module coded in FORTRAN90, which simulates cloud properties and can be implement in any model.

The simulator's working principle is based on using climate model fields to mimic radiances to which a retrieval algorithm is applied to obtain satellite-like fields for the comparison with satellite observations.

This process is summed up in three main phases. As model grids are very coarse ($\sim$100 km), the model fields are first down-scaled: each model gridbox mean profile is broken into subcolumns, whose size is more representative of a satellite retrieval area ($\sim$10 km). Details of this downscaling process and corresponding assumptions are provided in Pincus et al. (2012). Next, each sub-column profile is processed by a forward radiative transfer model to create synthetic radiances at the satellite retrieval area-level. The last step aggregates the simulator outputs to produce diagnostics (for example temporal averages and histograms) statistically comparable to the real satellite observations. A comprehensive explanation about the methodology and results of the COSP MODIS simulator is presented in Pincus et al. (2012).

2.3 Models

2.3.1 ECHAM-HAM

ECHAM-HAMMOZ (echam6.3-ham2.3-moz1.0) is a global aerosol-chemistry climate model (Schultz et al., 2018; Kokkola et al., 2018; Tegen et al., 2019; Neubauer et al., 2019) where ECHAM refers to the atmospheric model of the model configuration, HAM to the aerosol model, and MOZ to the chemistry model. In this study only the global aerosol-climate model part of ECHAM-HAMMOZ is used. Instead of the comprehensive MOZ chemistry model, sulphate chemistry is calculated in HAM for which the details have been given by Zhang et al. (2012) and references therein.

ECHAM-HAMMOZ, referred to here as ECHAM-HAM, consists of the general circulation model ECHAM (Stevens et al., 2013) coupled to the latest version of the aerosol module HAM (Tegen et al., 2019) and uses a two-moment cloud microphysics scheme that includes prognostic equations for the cloud droplet and ice crystal number concentrations as well as cloud water and cloud ice (Lohmann and Diehl, 2006; Lohmann et al., 2007, 2008; Lohmann and Hoose, 2009). Aerosol microphysical processes such as nucleation, coagulation, condensational growth are computed by the modal scheme M7 (Vignati et al., 2004). HAM computes further processes such as emissions, sulfur chemistry (Feichter et al., 1996), dry depositon, wet deposition, sedimentation, aerosol optical properties, aerosol-radiation and aerosol-cloud interactions.

Next to the two-moment cloud microphysics scheme the stratiform cloud scheme includes an empirical cloud cover scheme (Sundqvist et al., 1989).

The cirrus scheme is based on Kärcher and Lohmann (2002) and described in Lohmann et al. (2008), cloud droplet activation uses the Abdul-Razzak and Ghan (2000) parameterization, the autoconversion of cloud droplets to rain follows the method from Khairoutdinov and Kogan (2000), immersion and contact freezing in mixed-phase clouds follows the scheme from Lohmann and Diehl (2006), and cumulus convection is represented by the parameterization of Tiedtke (1989) with modifications developed by Nordeng (1994) for deep convection.

Simulations were performed at T63 ($1.9° \times 1.9°$) spatial resolution using 31 vertical levels (L31) and COSP v1.4. Horizontal winds and surface pressure were nudged towards the ERA-Interim (Dee et al., 2011) reanalysis for 2008, and observed sea surface temperatures and sea ice cover for 2008 were used (Taylor et al., 2000). Three-hourly instantaneous output was used. The output of the COSP satellite simulator is also three-hourly. The implementation of the COSP satellite simulator in ECHAM-

HAM does not allow instantaneous output. The COSP satellite simulator is called every radiation time step (i.e. every two hours) and the output of the COSP satellite simlator is averaged over the three hourly output period. This means that on average 50% of the values in the output of the COSP satellite simulator are instantenous values (i.e. from only one time step) and 50% of the values are an average over two radiation time steps (i.e. an average over two instantaneous values which are two hours apart).

2.3.2 ECHAM-HAM-SALSA

ECHAM-HAM-SALSA is identical to the ECHAM-HAM setup (echam6.3-ham2.3-moz1.0), with the difference that the sectional aerosol module SALSA (Kokkola et al., 2008, 2018) is used instead of the modal model M7 used in the ECHAM-HAM setup. SALSA calculates the aerosol microphysical processes: nucleation, coagulation, condensation, and hydration. In this setup, the aerosol model HAM applies also the sectional scheme for the rest of the aerosol processes, i.e. emissions, removal, aerosol radiative properties, and aerosol-cloud interactions. In addition to differences in the aerosol size distribution scheme, also the wet deposition schemes differ between the ECHAM-HAM and ECHAM-HAM-SALSA setups. In addition, while ECHAM-HAM uses the cloud activation parameterization for modal models (Abdul-Razzak and Ghan, 2000), SALSA uses the activation parameterization for the sectional representation (Abdul-Razzak and Ghan, 2002). Along with the details of these differences, the implementation and the evaluation of SALSA with the ECHAM-HAMMOZ model version which is used in this study has been presented by Kokkola et al. (2018).

Similarly to ECHAM-HAM, simulations were performed at T63 ($1.9° \times 1.9°$) spatial resolution using 47 vertical levels (L47) and COSP v1.4. Horizontal winds and surface pressure were nudged towards the ERA-Interim (Dee et al., 2011) reanalysis for 2008, and observed sea surface temperatures and sea ice cover for 2008 were used (http://www-pcmdi.llnl.gov/projects/amip/). Three-hourly instantaneous output was used.

2.3.3 NorESM

The Norwegian Earth System Model (NorESM) (Kirkevåg et al., 2013; Bentsen et al., 2013; Iversen et al., 2013) is largely based on the Community Earth System Model (CESM) model (http://www.cesm.ucar.edu) but uses a different ocean model and a different aerosol scheme in the Community Atmospheric Model (CAM) (Neale et al., 2010).

The aerosol microphysics scheme in the NorESM version of CAM, called CAM-Oslo, consists of 12 log-normally shaped background modes which are tagged according to emission source and chemical composition (Kirkevåg et al., 2018). The shape of these modes can change due to condensation and coagulation.

In the current simulations, the NorESM model was run with the CAM-Oslo version 5.3 (Kirkevåg et al., 2018), which is configured with the microphysical two moment scheme MG1.5 (Morrison and Gettelman, 2008; Gettelman et al., 2015) for stratiform clouds. The scheme includes prognostic equations for liquid (mass and number) and ice (mass and number) and a version of the Khairoutdinov and Kogan (2000) autoconversion scheme where subgrid variability of cloud water (Morrison and Gettelman, 2008) has been included. The aerosol activation into cloud droplets is based on Abdul-Razzak and Ghan (2000) and the heterogeneous freezing in CAM5.3-Oslo is based on Wang et al. (2014) with a correction applied to the contact angle model (Kirkevåg et al., 2018). Moreover, CAM5.3-Oslo has a shallow convection scheme (Park and Bretherton, 2009) and a deep convection scheme (Zhang and McFarlane, 1995). The simulation was run with the Community Land Model (CLM) version 4.5 (Oleson et al., 2010) with satellite phenology. Included in CLM is the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012), which interactively calculates the emissions of biogenic volatile organic vapors. Both isoprene and monoterpenes take part in the formation of secondary organic aerosol in CAM5.3-Oslo. The sea surface temperatures and sea ice in the simulation were prescribed monthly averages for the years 1982-2001.

The resolution for the simulation was $0.9° \times 1.25°$ and the surface pressures as well as horizontal winds were nudged against ERA-Interim reanalysis data (Berrisford et al., 2011) from 2008. CAM-Oslo was run with COSP version 1.4 producing three-hourly instantaneous outputs.

3 Methods

3.1 Post-processing of the datasets

The comparison of satellite retrievals and model variables is not always straightforward. Satellite-retrieved physical quantities may be derived slightly differently than the corresponding parameters in the model, and differences can be attributed to discrepancies in the retrieved quantities viewed from space versus model fields (i.e. retrieval assumptions, sensor limitations, spatial resolution) (Bodas-Salcedo et al., 2011). In this study we aim at highlighting the differences between observations and models, which stem from different aerosol and cloud physical parametrization by using the COSP satellite simulator. Satellite simulators, such as COSP, represent a compromise between model fields and retrieved fields. Simulators use model fields to reproduce what the satellite sensor would see if the atmosphere had the clouds of a climate model. By taking the characteristics of the MODIS instrument into account COSP generates simulated fields of cloud parameters, which can be quantitatively compared to MODIS observations. The COSP diagnostics are then successively aggregated to the simulator outputs and are provided at the original model resolution. Prior to their intercomparison, post-processing of the COSP diagnostics and satellite data is necessary for obtaining a robust evaluation. COSP-derived parameters are in the original model resolution and represent grid-averaged values. As MODIS observations are grid values representative only of in-cloud pixels, the COSP grid-averaged values are divided by the corresponding cloud fractions. The three-hour outputs from the models were aggregated to daily averages and successively re-gridded and co-located by linear interpolation onto the finer satellite regular grid of $1° \times 1°$. Each grid point of cloud variables from MODIS observations and MODIS diagnostics was screened using a minimum threshold of 30% of cloud fraction to minimize the source of errors introduced by the retrieval algorithm and to ensure the existence of large-scale clouds. The screening does not introduce a significant loss in the data pool and provide grounds for a robust intercomparison as also shown in Bennartz (2007) and Ban-Weiss et al. (2014). For each time step, only grid points having a valid observation simultaneously in each one of the four datasets were included in the final dataset for the statistical analysis.

The MODIS algorithm retrieves cloud properties in the proximity of the top of a cloud while the direct model outputs provide values through the entire vertical structure of a simulated atmospheric column. To overcome this issue, when comparing the direct model output CDNC and satellite-derived CDNC, for each grid box we selected the column maximum value of the CDNC direct output of the models. Additionally, we selected only gridpoints with temperature T > 273° K to exclude mixed-phase and ice clouds.

Note that all discussed cloud parameter are diagnosed using satellite simulators and are compared to the corresponding MODIS satellite observations. However, we use two direct model diagnostics in the study:

– AOD, which is used to derive the AI, a proxy for cloud condensation nuclei for the computation of ACI

– $\mathrm{CDNC}_{\mathrm{direct}}$, which is compared with COSP-simulated and MODIS-derived estimates

3.2 Cloud droplet number concentration (CDNC)

The CDNC were derived from CER and COT from MODIS observations and COSP simulations by combining Eqs. (6) and (9) from Bennartz and Rausch (2017) in the following equation:

$$\mathrm{CDNC} = \gamma \cdot \mathrm{COT}^{0.5} \cdot \mathrm{CER}^{-2.5}, \tag{1}$$

where COT is cloud optical thickness, CER is the cloud droplet effective radius and $\gamma = 1.37 \cdot 10^{-5}$ $\mathrm{m}^{0.5}$ (Quaas et al., 2006). The assumption of not accounting for temperature effect and setting γ as a bulk costant applies rather well to the stratiform clouds in the marine boundary layer but less so for convective clouds (Bennartz, 2007; Rausch et al., 2010). The assumption of not accounting for temperature effect and setting γ as a bulk costant applies rather well to the warm stratiform clouds in the marine boundary layer but less for convective clouds (Bennartz, 2007; Rausch et al., 2010; Grosvenor et al., 2018). The equation represents the "Idealized Stratiform Boundary Layer Cloud" (ISBLC) model (Bennartz and Rausch, 2017) which is based on the following assumptions:

– the cloud is horizontally homogeneous;

– the LWC increases linearly from the cloud base to the cloud top;

– the CDNC is constant throughout the vertical extent of the cloud.

While the ISBLC model describes important aspects of stratiform boundary layer clouds, its assumption will never be fully valid for a real cloud. Issues related to the ISBLC model assumptions are extensively elaborated in Bennartz (2007) and Bennartz and Rausch (2017) and references therein. Despite the above mentioned approach for deriving CDNC is less reliable over land ares, we will provide MODIS CDNC values globally (land and ocean). These estimates will be used as a reference dataset, rather than "truth" data, for enabling the comparison of COSP-derived CDNC and the model direct output of CDNC.

3.3 Assessing the aerosol-cloud-interactions

The aerosol-cloud-interactions is quantified here trough the parameter ACI_{CDNC} defined as the change in the selected cloud property, CDNC, to a change in AI, which is used here as a proxy for cloud condensation nuclei (CCN):

$$ACI_{CDNC} = \frac{d\ln(CDNC)}{d\ln(AI)} \tag{2}$$

The CDNC was computed from the CER and COT from the COSP-MODIS simulations and MODIS retrievals. Additionally, AI was derived from ECHAM-HAM, ECHAM-HAM-SALSA, and NorESM MODIS-COSP diagnostics, and MODIS satellite observations following Feingold et al. (2001). The mean values and standard deviations of the parameters involved in the computation of ACI_{CDNC} are presented in Table 1. We discarded pixels retrieved when liquid cloud fraction is $\leq$0.3 to reduce noise-contamination and to focus on large-scale clouds. The screened parameters were used to derive CDNC. The ACI_{CDNC} parameter was calculated globally for each season. When calculating the ACI_{CDNC} for the 1° x 1° gridboxes Eq.2 is applied to data for the selected season and grid box. This methodology (Grandey and Stier, 2010) can be thought of as computing the linear regression slope of a scatter plot of ln(CDNC) versus ln(AI), where each point represents a day for which both aerosol and cloud data exist for the considered grid box. When computing ACI_{CDNC} for large areas, the ACI_{CDNC} of each gridbox needs to be weighted by the corresponding number of data points (Grandey and Stier, 2010). This step was included in the post-processing of the datasets.

4 Results

4.1 Global bias distributions

In this section we compare on a global scale aerosol and cloud properties from the three models by subtracting MODIS retrievals from the modeled COSP diagnostics. From now on we will refer to the difference between the simulated parameters and MODIS retrieved values using the term bias.

Overall, the spatial distributions of the biases (Fig. 1-5) show large discrepancies around the polar and ice-covered areas, such as Greenland and Antarctica. Over these areas large discrepancies are expected due to the inaccuracy of the MODIS retrieval algorithm due to viewing geometry (i.e. large zenith or viewing angles) and to correctly classify opaque clouds, snow/ice surfaces and optically thin clouds over really bright or warm surfaces (Marchant et al., 2016).

Figure 1 presents the differences between the MODIS-COSP cloud fraction diagnostics and COP CF for ice clouds CF_{ice} (Fig. 1b-d), and liquid clouds CF_{liq} (Fig. 1f-h), as well as the differences between MODIS total COP CF (Fig. 1j-l), and CF (Fig. 1n-p). Additionally, for each comparison the MODIS spatial distribution is presented as reference (Fig. 1a,e,i,m). It was already highlighted in section 2.1 that the cloud fraction retrieved from the optical properties (CF_{ice}, CF_{liq} and COP CF) excludes partly cloudy pixels, representing a limitation in the comparison of the data. Thus, lower values of MODIS COP cloud fractions are expected. A widespread positive bias is observed for CF_{ice} and CF_{liq}, indicating higher values of the COSP-simulated cloud fractions than the MODIS observations. Prevalent cloud regimes can be recognized in the bias distributions. ECHAM-HAM and ECHAM-HAM-SALSA well represent the amount of ice clouds which are generally found in the intertropical convergence zone (ITCZ) and the marine subtropical stratocumulus and stratus regions, whereas liquid clouds are better represented over land areas and in the subtropical stratocumulus region. NorESM shows positive biases for ice cloud amount over stratus clouds regions and around the ITCZ, but shows smaller biases for liquid stratus cloud regimes than ECHAM-HAM and ECHAM-HAM-SALSA

The total cloud fraction bias shows a positive bias between the MODIS-COSP CF simulated by the three models and MODIS COP CF (Fig. 1j-l) and a negative bias when MODIS CF is considered (Fig. 1n-p). Consequently, MODIS CF is higher than the MODIS COP CF product. This outcome is to be expected, and possibly originates from the different treatment in the MODIS algorithm of partly cloudy pixels in the computation of CF and COP CF, as discussed in section 2.1. Additionally, all models underestimate CF in marine subtropical stratocumulus regions.

The spatial distribution of the cloud physical and optical properties is remarkably similar among the model datasets with the exception of CER_{ice}, IWP (Fig. 2 d and l) and COT (Fig. 3g,k) for NorESM. These strong biases are explained by the fact that in the NorESM COSP 1.4 implementation code includes radiative active snow in the computation of the effective radius and optical thickness of ice clouds. However, this does not affect the properties of liquid clouds.

CER_{ice} and IWP are underestimated in ECHAM-HAM and ECHAM-HAM-SALSA. This is likely caused by the cirrus scheme which does not account for heterogenous nucleation or pre-existing ice crystals during formation of cirrus clouds (Neubauer et al., 2019; Lohmann and Neubauer, 2018). Interestingly, dissimilarities can also be observed between ECHAM-HAM and ECHAM-HAM-SALSA, despite the fact that the models share the same cloud module. ECHAM-HAM CER_{liq} is on average 5 μm smaller than in ECHAM-HAM-SALSA in the mid-latitude belt, and ECHAM-HAM-SALSA CER_{liq} is larger around the polar areas (Fig. 2g) and shows a large positive bias for LWP over ocean (Fig. 2o) in comparison to ECHAM-HAM. LWP is also overestimated by NorESM but over land areas (Fig. 2p), while ECHAM-HAM shows a good agreement with MODIS (Fig. 2n).

The evaluation of COT shows homogeneous results and comparable values of root mean square errors (Fig. 3) with the exception of NorESM COT biases for ice and liquid clouds, which are particularly high over land. It appears that some tuning parameters, for example the autoconversion parameter, are particularly low and affect the convection scheme by suppressing precipitation, thus creating thick clouds. The comparison of the differences between the biases of ECHAM-HAM and of ECHAM-HAM-SALSA shows localized differences over India, China and Russia for IWP (Fig. 2j,k) and over China for water cloud COT (Fig. 3e,f). These are also regions where aerosol microphysics has a fundamental role as shown in Kokkola et al. (2018). ECHAM-HAM and ECHAM-HAM-SALSA generally overestimate COT. The atmospheric model ECHAM shows a similar estimation when running without an aerosol model. This overestimation has been previouslt reported by Stevens et al. (2013).

Figure 4 shows global biases for CDNC derived from the MODIS retrievals and the COSP diagnostics following the method presented in Sect.3.2 (Fig. 4b-d), and the daily averages of the direct output of the models (Fig. 4e-g). The CDNC derived from MODIS (Fig. 4a) are shown globally (land and ocean) despite the assumptions described in 1 are less reliable over land areas, which are highlighted with contour lines. The differences between MODIS-COSP diagnostics and MODIS observations are very clear. Overall the MODIS derived CDNC is lower than that derived from COSP simulated values, but higher than the direct output values. Consequently, the CDNC from direct model output is lower than MODIS-COSP diagnostics. Possible explanations could be either related to the COSP method for deriving CER_{liq} and COT_{liq} or the approach used for deriving CDNC from CER_{liq} and COT_{liq} or related to the fact that the computation of the direct CDNC requires a minimum number of CDNC set to 40 cm^{-3}. This outcome is very similar to what has been found by Ban-Weiss et al. (2014), where the CDNC satellite-simulated values are higher than the standard CAM5 model output near clout top as well as the column maximum values. Therefore, this result represent an interesting topic that extends beyond the scope of this paper but it should further developed in future work. The biases between CDNC COSP-derived and modeled direct values are very different, but within each product the biases are similar, although local differences are observed. For example, the CDNC values from ECHAM-HAM-SALSA are lower in the polar regions and higher in the mid-latitude belt in comparison with the ECHAM-HAM and NorESM diagnostics. Local differences can also be observed in the direct output where ECHAM-HAM-SALSA shows higher values of CDNC over the oceans in the southern Hemisphere (Fig. 4f). A direct comparison of CDNC derived from MODIS-COSP simulated variable and the model CDNC direct outputs is shown in the supplementary material. ECHAM-HAM and ECHAM-HAM-SALSA were run with identical tuning parameter settings which were optimized for ECHAM-HAM. This choice was made to distinguish the differences in aerosol-cloud interactions coming from different aerosol microphysics modules. The differences in CDNC between these two model setups originates from the cloud activation schemes, i.e. for HAM the modal cloud activation scheme of Abdul-Razzak and Ghan (2000) and for HAM-SALSA the sectional cloud activation scheme (Abdul-Razzak and Ghan, 2002). The cloud activation scheme of ECHAM-HAM-SALSA produces a higher number of CDNC than ECHAM-HAM (Fig. 4c) because SALSA microphysics module simulates generally higher number of particles larger than 100 nm in diameter which act as cloud condensation nuclei. Despite the higher CDNC, $\mathrm{CER_{liq}}$ is larger in ECHAM-HAM-SALSA than in ECHAM-HAM. Although it is expected that $\mathrm{CER_{liq}}$ decreases with increasing CDNC, higher LWP in ECHAM-HAM-SALSA in turn results in larger $\mathrm{CER_{liq}}$ and outweighs the CDNC effect on $\mathrm{CER_{liq}}$. The causes for differences in LWP between the two model versions are very difficult to diagnose but candidates for causing them are differences in wet scavenging, convective detrainment or freezing.

Figure 5 presents AOD and AI biases. The biases between values of AI from direct model output and MODIS observations are quite close among the model as their average is about of +0.2. The main divergence is observed in the ECHAM-HAM bias where higher AI values are simulated around the mid-latitude belt. Tegen et al. (2019) found indications that the particle size of mineral dust and sea salt aerosol particles may be too small in ECHAM-HAM. More discrepancies can be observed in the AOD bias: ECHAM-HAM-SALSA AOD values are higher over ocean, and NorESM AOD are much higher over deserts and other bright surfaces (such as Africa and Australia).

Other localized distinctions in aerosol loading distribution can be observed over regions which are typically strongly affected by primary emissions (such as the Sahara, India, Southeast Asia, Russia, Canada, central Africa, and South America). The different representation of size distribution, microphysical processing of aerosols and sink processes has a significant effect on the modelled AOD as shown for the aerosol module SALSA2.0 by Kokkola et al. (2018). The overestimation of AOD in the tropical oceans and underestimation of AOD at higher latitudes and over land in ECHAM-HAM has also been found by Tegen et al. (2019).

4.2 Joint histogram

The analysis of the CTP-COT joint histogram enables to determine how well the data sources represent the vertical cloud structures and regimes. Figure 6 shows the comparison of the simulated and observed global mean cloud fraction as a function of cloud top pressure and cloud optical thickness. ECHAM-HAM and ECHAM-HAM-SALSA (Fig. 6a,b) show a nearly identical result by concentrating a large fraction of clouds at low level (CTP $\leq$ 680 hPa) and in the interval 3.6 $\leq$ COT $\leq$ 23. NorESM (Fig. 6c) also concentrates its largest amount of clouds at low levels in the same COT interval as in Fig. 6a and Fig. 6b, but detects also a higher fraction (about 2-2.5%) of optically thick clouds 9.4 $\leq$ COT $\leq$ 60 throughout the atmosphere. A second cloud fraction peak is observed for optically thin clouds (COT $\leq$ 1.3) at very high levels (180 $\leq$ CTP $\leq$ 310) for NorESM. This bimodal distribution resembles the vertical distribution of the MODIS cloud fraction shown in Fig.6d. The MODIS observations are mostly in the category of high-level clouds (CTP $\leq$ 440 hPa) and low-level clouds (680 hPa $\leq$ CTP). MODIS shows on average more mid-level clouds than NorESM and a higher fraction at low-level for 3.6 $\leq$ COT $\leq$ 23 similarly to ECHAM-HAM and ECHAM-HAM-SALSA. Figure 6e shows the differences in cloud vertical distribution where MODIS is generally having the highest cloud fraction except for mid-level. MODIS also presents the highest percentage of clouds for COT $\geq$ 3.6. NorESM and MODIS detects nearly the same amount of clouds for 1.3 $\leq$ COT $\leq$ 3.6, while for optically very thin clouds (COT $\leq$ 1.3) a good agreement is obtained between all datasets and NorESM shows the highest percentage of cloud fractions.

4.3 Aerosol-cloud interactions

The daily mean values of CDNC and AI were used to assess how clouds are affected by the changes of the CCN proxy. Uncertainties were computed as the 95% confidence intervals using daily averages. Positive estimates of $\mathrm{ACI_{CDNC}}$ indicate an increase of CDNC as a function of AI, which could be an indication of the aerosol indirect effects. The potential limitations to this approach are discussed in Sect.refsummary.

Figure 7 shows estimates of $\mathrm{ACI_{CDNC}}$ on a global scale, although over ocean areas only, for each season and, separately, for the entire period under study as 'All'. The same analysis is iterated on a regional scale, including both land and ocean areas, and presented in the supplementary material (Fig.S4) Error bars are representative of the boundaries of the 95% confidence interval. The positive $\mathrm{ACI_{CDNC}}$ values resulting from MODIS and the three models suggest that changes in AI are connected with an increase of CDNC and the trend seems to be independent of the time of the year. The modeling $\mathrm{ACI_{CDNC}}$ estimates are similar in the models; however, the results are statistically indistinguishable owing to fully overlapping confidence bars (Cumming et al., 2007). MODIS $\mathrm{ACI_{CDNC}}$ estimates show negative values for the winter months (DJF), especially over the Northern Hemisphere (Fig.S4). As these negative values are derived over land regions, it could be indicative of retrieval biases over bright surfaces (i.e. snow or ice). Furthermore, it is important to inform the readers that MODIS aerosol size parameters over land (i.e. AE or fine-AOD) are no longer official products directly provided by the MODIS aerosol team. The publication of these variable were discontinued due to low quantitative MODIS skill (Mielonen et al., 2011; Levy et al., 2013). Using spectral AOD, we derived AE over land and derived AI on a global scale to allow estimates of $\mathrm{ACI_{CDNC}}$ on a global scale (Fig.S4). However, the AE values over land were not evaluated. However, negative $\mathrm{ACI_{CDNC}}$ values may be associated with the presence of different types of aerosol (i.e. hydrophobic aerosol such as dust, black carbon) and their proximity to clouds, which may affect or inhibits the growth of cloud droplets (Chen et al., 2018; Jiang et al., 2018; Costantino and Bréon, 2013). Over ocean negative $\mathrm{ACI_{CDNC}}$ values from MODIS observations have been systematically found over subtropical marine stratuscumulus regions (i.e. N. Atlantic Ocean, N.America, S.Atlantic Ocean). In these regions Chen et al. (2014) found a decrease in LWP with increasing AI for non-precipitating scenes. Additionally, negative $\mathrm{ACI_{CDNC}}$ values were suggested owing to wet scavenging or mixing of environmental air by entrainment (Ackerman et al., 2004). While both processes affect LWP, CDNC is not necessarily changing. This indicates limits in the derivation of CDNC from retrieved quantities for MODIS. Also water uptake by aerosol particles and effects of meteorology can have a significant impact on the estimation of $\mathrm{ACI_{CDNC}}$ derived from the relationship between CDNC and AI (Neubauer et al., 2017).

Cloud properties (Fig. 2 and Fig. 3) are more similar for ECHAM-HAM and ECHAM-HAM-SALSA, which share the same atmospheric model, rather than between the two and NorESM. Nevertheless, the $\mathrm{ACI_{CDNC}}$ estimates show good agreement between the three models and, even more important, with $\mathrm{ACI_{CDNC}}$ derived from MODIS observations.

5 Summary, discussion and conclusions

The differences between observed and modeled aerosol and cloud properties can be related to many factors, among which are the different parametrizations of aerosol and cloud physical processes in the models, or differences in observation characteristics by satellite, as well as meteorological influences on aerosol-cloud interactions. In this study we focus on the differences due to the physical parametrization of aerosol and cloud properties, and minimize the impact of the other factors. This objective was achieved by using a satellite simulator, which resolves the issue related to the incongruities between model and satellite views, and by nudging modeled winds to meteorological observation, solving the discrepancies between observed and modeled meteorology.

The results show that the aerosol module in a climate model, in our case ECHAM-HAM and ECHAM-HAM-SALSA, has a smaller effect on the simulation of cloud properties than switching to another atmospheric model, NorESM. However, the three models differ from each other in the spatial and vertical representation of clouds. The COSP cloud fraction diagnostics are comparable to MODIS products but the difference between the two MODIS products of total cloud fractions is significant. Despite having identical cloud modules, ECHAM-HAM and ECHAM-HAM-SALSA diverge when comparing liquid water cloud properties yet both fail to represent high level clouds. The discrepancies between ECHAM-HAM and ECHAM-HAM-SALSA may originate from different amounts of activated droplets and different ice nucleation rates. While the NorESM cloud vertical distribution is closer to MODIS, large biases are found globally for cloud droplet size and water content in ice clouds due to the contribution of radiatively active snow (Kay et al., 2012). The inclusion of radiatively active snow in the physical model and the COSP module mitigates the underestimation of model mid-level and high clouds but heavily impacts the magnitude of the global values of the cloud properties in ice clouds.

The differences observed in the simulation of cloud properties are reflected in the estimations of ACI_{CDNC}. ACI_{CDNC} is generally larger for ECHAM-HAM-SALSA, while similarly lower values are found for ECHAM-HAM, NorESM and MODIS.

Although satellite simulators allow robust comparisons, their reliability is flawed when the observational data is not well explained or the simulator itself fails to address specific characteristics. Therefore, their strengths and weaknesses need to be accounted for as to successfully use simulation diagnostics in model-observation comparisons as illustrated in details by Pincus et al. (2012), and Kay et al. (2012) to successfully use simulator diagnostics in model-observation comparisons. For example, simulators have limitations in depicting horizontally heterogeneous cloud regimes as they do not account for sub-pixel clouds, which may explain the differences in the detection of small cloud fractions between observations and models. However, simulator and observational errors are here neglected because we considered them to be less important in the explanation of the model biases. The observed biases in the modeled clouds could originate from errors in the model calculation as well from the cloud parametrization; the identification of the specific reasons for these discrepancies is beyond the scope of this study.

The results presented here indicate that the cloud droplet number concentration appears to be more sensitive to changes in aerosols in models than observations and these results are in agreement with many previous studies found in the literature (e.g. Ban-Weiss et al., 2014; Quaas et al., 2004; McComiskey and Feingold, 2012; Penner et al., 2011). Some of the challenges and limitations in assessing ACI_{CDNC} are now highlighted. AOD retrievals are limited to cloud-free conditions, which creates challenges to studying the ACI_{CDNC} where the intention is to study collocated aerosol and cloud observations. Unless height-resolving instruments (i.e. lidars) are considered, the vertical location of the AOD level is unknown. Aerosol and cloud measurements may contain retrieval errors, which are further propagated to ACI_{CDNC} estimates, as well as they reciprocally may bias the respective retrievals (Jia et al., 2019) . The interpretation of the observed aerosol-cloud relationships is complicated by the effect of meteorology (Quaas et al., 2010; Gryspeerdt et al., 2014, 2016; Brenguier et al., 2003). As cloud formation happens in high humidity conditions, aerosol humidification can severely affect the assessment of ACI by causing positive correlation between AOD and cloud properties (Myhre et al., 2007; Quaas et al., 2010; Grandey et al., 2013; Gryspeerdt et al., 2014). Additionally to aerosol particles, water vapour also affects precipitation (Boucher et al., 2013), obviously linked to the presence of clouds, and consequently causes spurious correlations between aerosols and clouds (Koren et al., 2012). Therefore, some of the differences in the $\mathrm{ACI}_{\mathrm{CDNC}}$ estimates from satellites and models could be associated with limitations in satellite measurements. For example, the estimates of $\mathrm{ACI}_{\mathrm{CDNC}}$ might suffer from an averaging effect due to the large spatial averages of satellite aerosol and cloud properties. L3 data can introduce spurious relationships between aerosols and cloud properties (e.g. McComiskey and Feingold, 2012; Christensen et al., 2017), and provide a rather limited pool of data samples enabling the analysis only over large regions. This was not explored in this study because we used the same spatial resolution for both the true model estimate and for the satellite-based model estimate for the $\mathrm{ACI}_{\mathrm{CDNC}}$. Neubauer et al. (2017) performed a detailed study on the impact of meteorology, cloud regimes, aerosol swelling, and wet scavenging on microphysical cloud properties using ECHAM-HAM. The results highlight that a minimum distance between cloud and aerosol gridded data should be taken into account in the anaylsis of satellite data, and that dry aerosols should be selected to reduce the influence of aerosol growth due to humidity for model simulations when comparing satellite-based and model estimates for $\mathrm{ACI}_{\mathrm{CDNC}}$. Similarly to our results, Neubauer et al. (2017) find a systematical overestimation of the sensitivity of modeled LWP and CDNC compared to MODIS observations, and often a disagreement in sign in the comparison of cloud parameters. The results suggest that the derivation of CDNC from satellite observations may be limited by entrainment mixing of environmental air or precipitation. Furthermore, the models can not resolve the entrainment mixing at the top of stratocumulus clouds, which puts the LWP sensitivity to aerosol change in the models into question. In conclusion, this study identified limitations and deficiencies in the models, and their acknowledgment is important for the model development process and the correct interpretation of modelling diagnostics. We highlighted many discrepancies in cloud spatial and vertical representations and the results showed that the three models overall similarly underestimate cloud fraction for the stratocumulus cloud regime being when compared to MODIS. We discovered that IWC is systematically lower in ECHAM-HAM-SALSA than in ECHAM-HAM due to a higher cloud droplet freezing rate which consecutively triggers a reduced sedimentation of ice clouds. This outcome explains the contradictory result in ECHAM-HAM-SALSA that shows the largest global averages for CER among the models despite having the highest number of CDNC. Further investigation is needed to explain the differences in ice cloud properties between ECHAM-HAM and ECHAM-HAM-SALSA. The clouds simulated by NorESM are too thick over land and this issue is not only seen in COSP-variables but also in the default model output due to a very low autoconversion parameter which caused the suppression of precipitation over land, thus thicker clouds. Additionally, in support to Ban-Weiss et al. (2014), the study revealed that the direct model CDNC is systematically larger than the values derived from COSP-diagnostics and MODIS observation. This discrepant results from the comparison of COSP-derived CDNC and the modeled direct output of CDNC, where the MODIS data represents solely the reference dataset rather than "truth" data, stem from the aerosol model set-up and this discrepancy has potentially important implications for the modelling community, using satellite observations to evaluate standard (not satellite-simulated) model output.

Finally, we point out that the model deficiencies identified here may lead to an improvement of model parametrization and to more robust results. As future work, a regional-based analysis would enable a better understanding of the physical processes responsible for the model biases. Additional research should be conducted to evaluate the aerosol-cloud-interaction following the approach suggested by Neubauer et al. (2017). These further steps would potentially benefit the modeling community interested in climate applications.

Data availability. The MODIS satellite data used in this study are publicly available at https://ladsweb.nascom.nasa.gov. ECHAM-HAM data are available from David Neubauer (david.neubauer@env.ethz.ch), ECHAM-HAM-SALSA data are available from Harri Kokkola (harri.kokkola@fmi.fi) and NorESM data from Moa K. Sporre (moa.sporre@nuclear.lu.se).

Author contributions. ECHAM-HAM-SALSA, ECHAM-HAM, and NorESM data (and corresponding descriptive text in Sect. 2.3 Data) were provided by DN, HK, and MS, respectively. IH assisted in setting up the NorESM simulations. GS conducted the data analysis, and wrote the majority of manuscript, except for the sections describing the models. PK, HK, AA participated in reading the results. GL, UL, PS helped to set up the concept idea of the paper. MS, DV, HK, PK, UL and GL contributed to review the manuscript.

Competing interests. No competing interests to declare.

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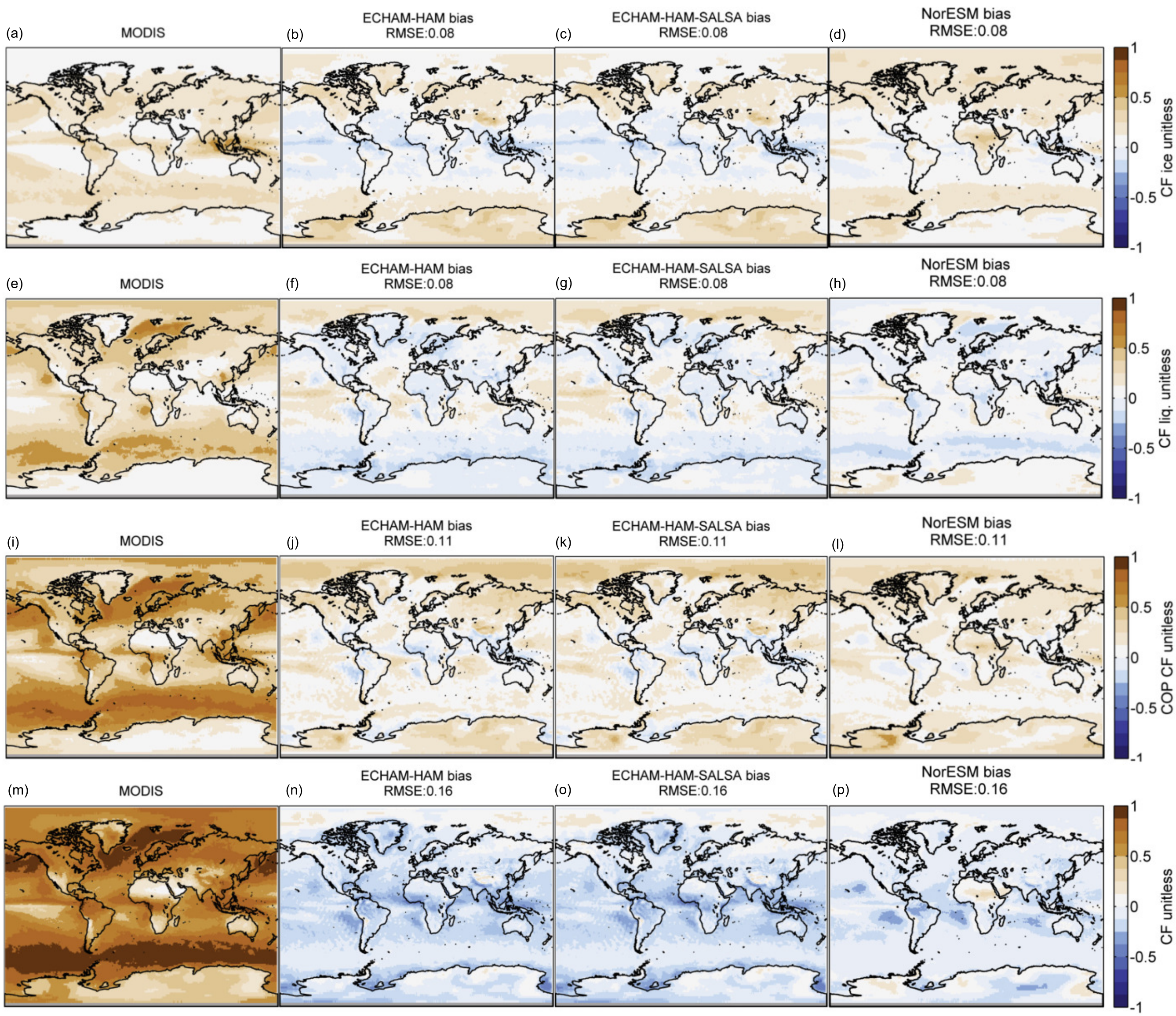

Figure 1. Annual global mean bias in cloud fraction. The bias represents the difference between MODIS-COSP diagnostics from ECHAM-HAM, ECHAM-HAM-SALSA, NorESM and MODIS observations. COSP-simulated total ice and liquid cloud fractions are compared with MODIS retrieval ice fraction (b-d), and with MODIS retrieval liquid cloud fraction (f-h), respectively. COSP-simulated total cloud fraction is compared with MODIS retrieval total cloud fraction (COP CF) (j-l), and cloud mask cloud fraction (CF) (n-p). Pixels with liquid cloud fraction $\leq 30\%$ are screened.The averages represent in-cloud values. High latitudes (Lat $> 60°$ N or Lat $> 60°$ S) are excluded in the computation of the root mean square error (RMSE). MODIS spatial distribution is presented as reference (a,e,i,m).

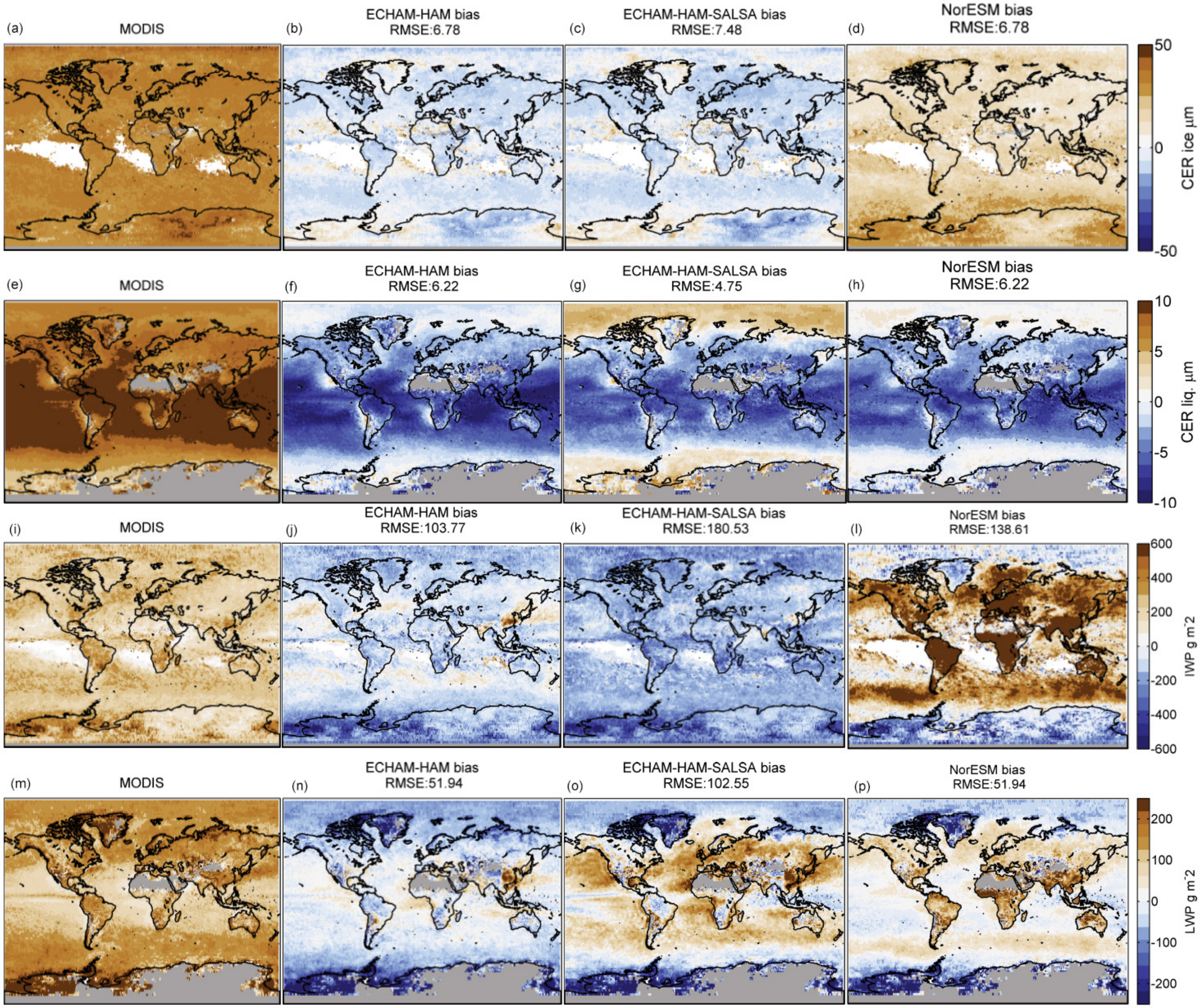

Figure 2. Annual global mean bias in cloud effective radius and water path. The bias represents the difference calculated subtracting MODIS observation to MODIS-COSP diagnostics from ECHAM-HAM, ECHAM-HAM-SALSA, and NorESM. Ice cloud effective radius (CER_{ice}) from MODIS-COSP is compared with MODIS observations in (b)-(d) and liquid cloud effective radius (CER_{liq}) in (f)-(h). The biases related to the comparison of COSP-simulated ice water path (IWP) are showed in (j)-(l) and for liquid water path (LWP) in (n)-(p). Pixels with liquid cloud fraction $\leq$ 30% are screened. The averages represent in-cloud values. Pixels with liquid cloud fraction $\leq$ 30% are screened. Values are in-cloud concentrations. High latitudes (Lat $>$ 60° N or Lat $>$ 60° S) are excluded in the computation of the root mean square error (RMSE). MODIS spatial distribution is presented as reference (a,e,i,m).

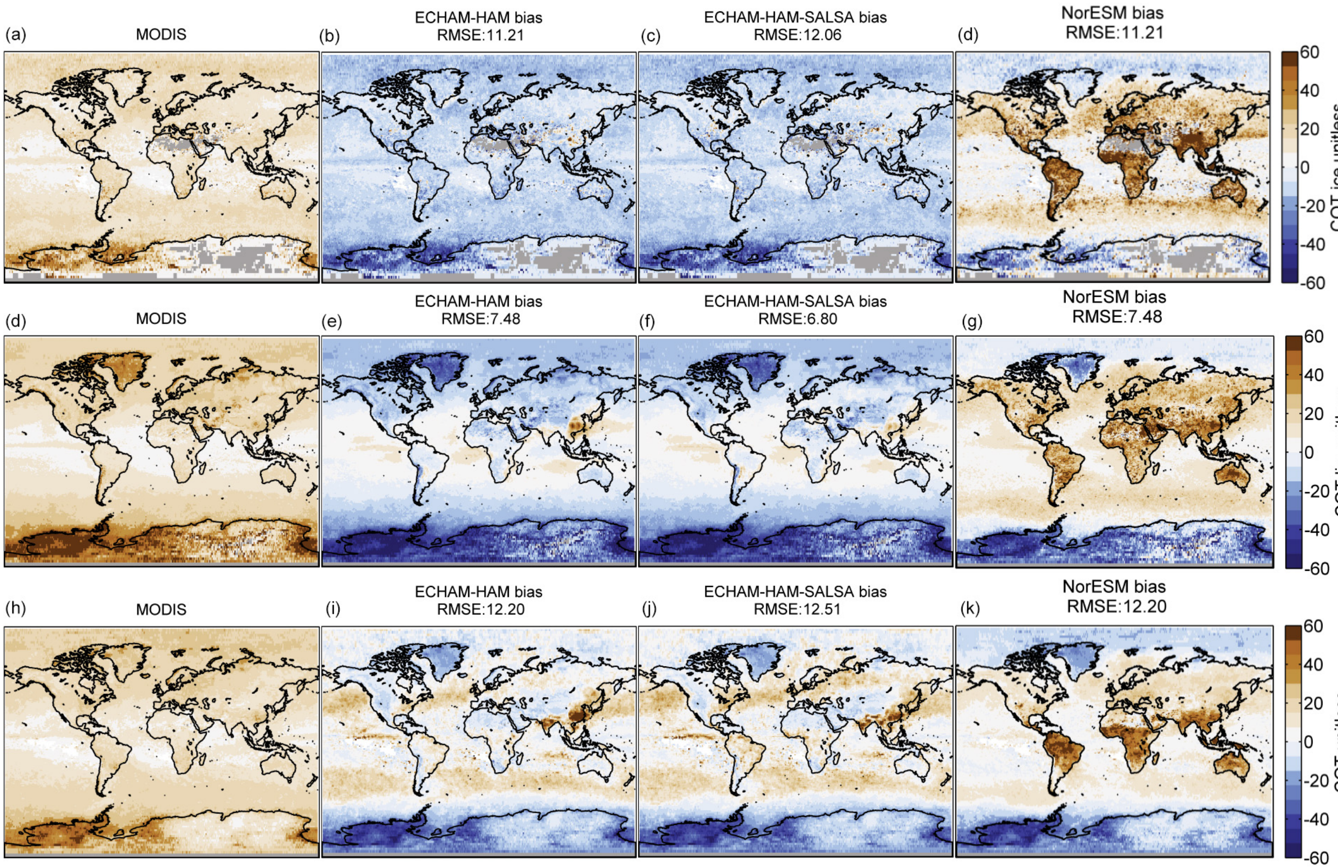

Figure 3. Annual global mean bias in cloud optical thickness for ice clouds (b-d), liquid water clouds (e-g) and total (combined ice and water clouds) COT (i-k) between MODIS and ECHAM-HAM, ECHAM-HAM-SALSA, and NorESM. The bias represents the difference between MODIS-COSP diagnostics and MODIS observations. Pixels with liquid cloud fraction $\leq 30\%$ are screened. The averages represent in-cloud values. High latitudes (Lat $> 60^\circ$ N or Lat $> 60^\circ$ S) are excluded in the computation of the root mean square error (RMSE). MODIS spatial distribution is presented as reference (a,d,h).

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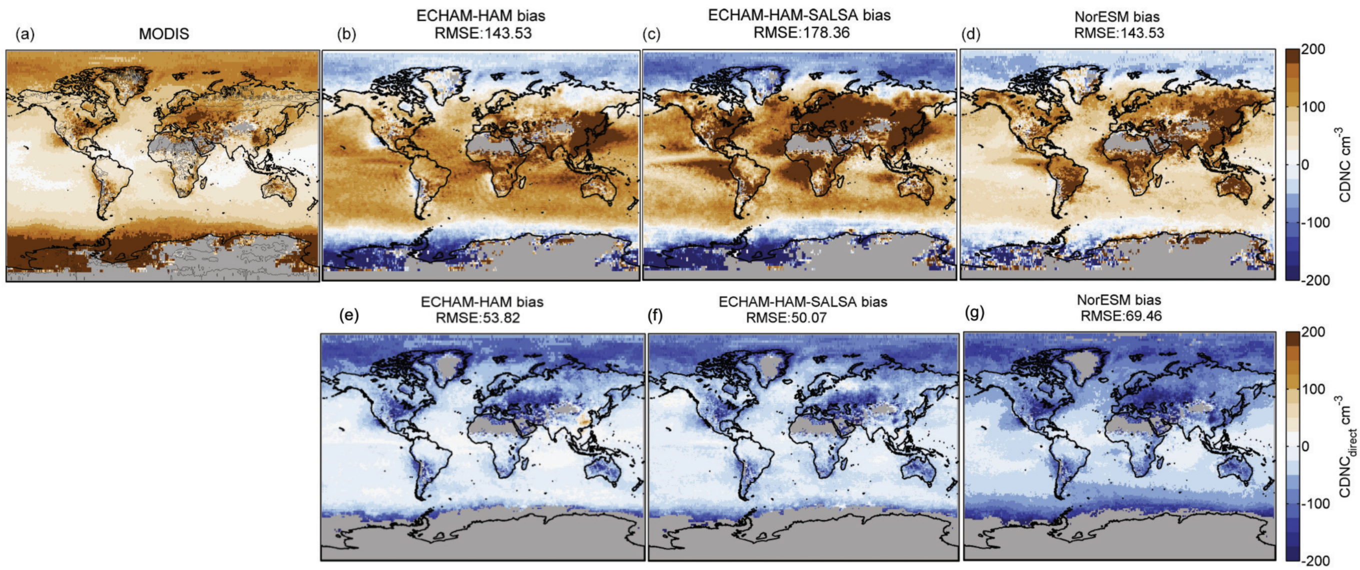

Figure 4. Cloud droplet number concentration (CDNC) annual mean bias.The bias represents the difference between CDNC derived from MODIS-COSP diagnostics and MODIS observations(b-d), and the model direct outputs and MODIS observations (f-h). Pixels with liquid cloud fraction $\leq 30\%$ are screened. The averages represent in-cloud values. High latitudes (Lat $> 60^\circ$ N or Lat $> 60^\circ$ S) are excluded in the computation of the root mean square error (RMSE). MODIS spatial distribution is presented as reference (a). Contour lines highlight the land areas where the assumptions for deriving MODIS CDNC are less reliable.

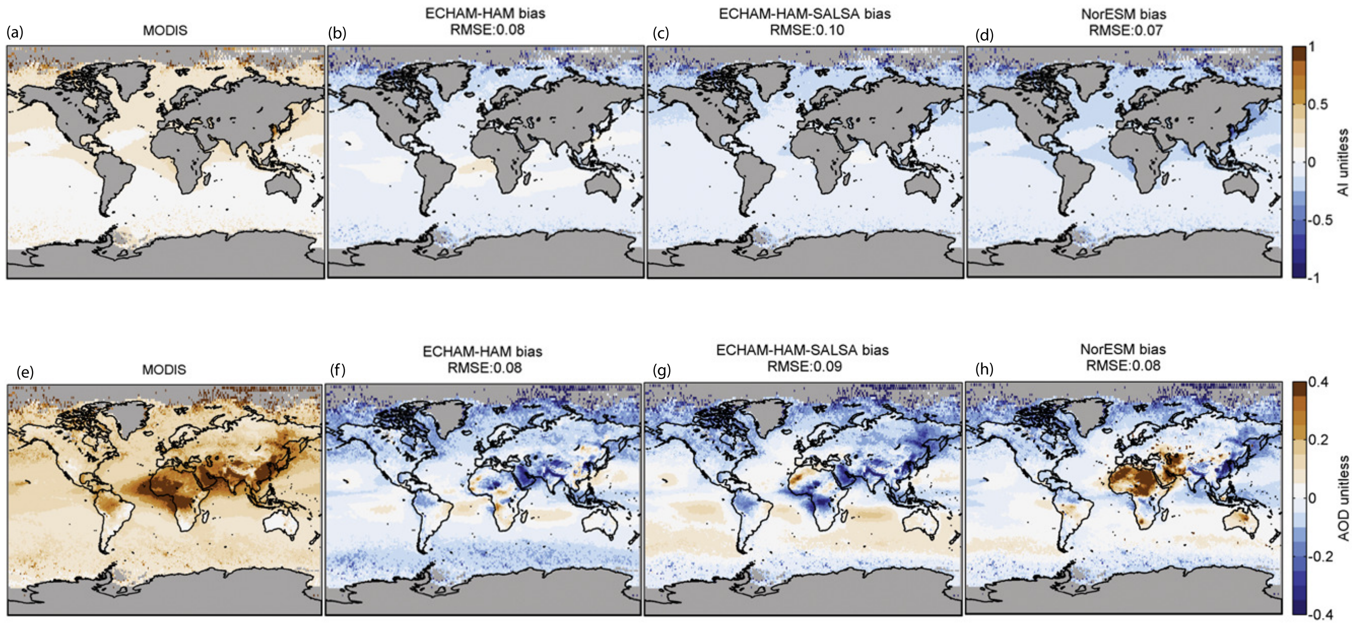

Figure 5. Aerosol Index (AI) (b-d) and Aerosol Optical Depth (f-h) annual mean bias. AI are representative of ocean values only. The bias represents the difference between the model direct outputs and MODIS observations. High latitudes (Lat $>$ 60° N or Lat $>$ 60° S) are excluded in the computation of the root mean square error (RMSE). MODIS spatial distribution is presented as reference (a,e).

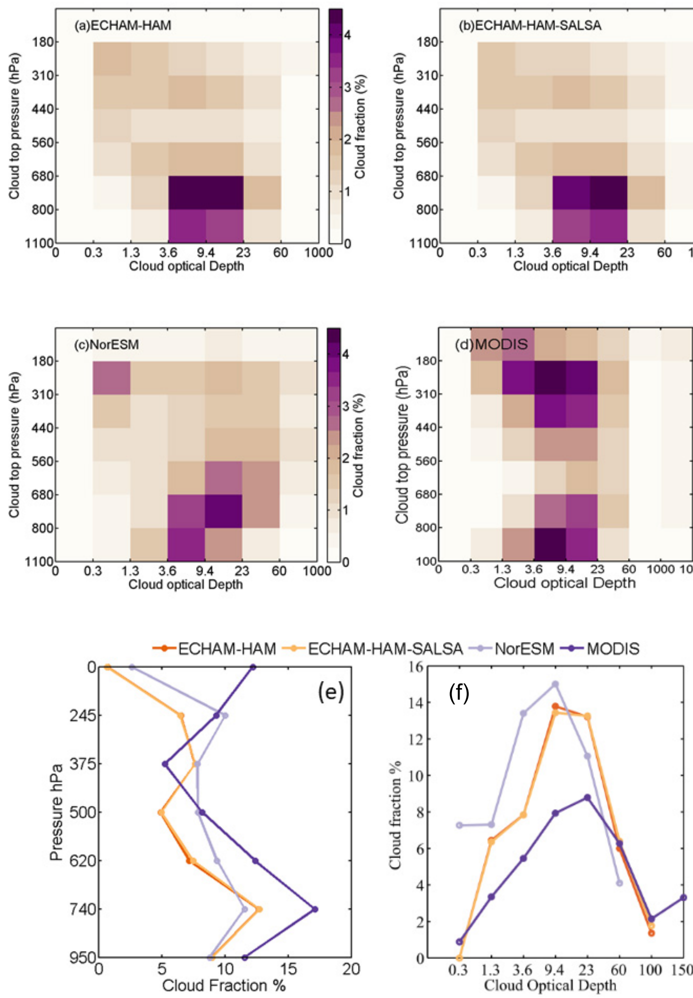

Figure 6. Vertical distribution analysis. Cloud fraction as a function of cloud top pressure and optical thickness for (a) ECHAM-HAM, (b) ECHAM-HAM-SALSA, (c) NorESM and (d) MODIS. The color scale represents the cloud fraction percentage. (e) Cloud fraction as a function of CTP (sum of all optical depth $\geq$0.3, and (f) cloud fraction as a function of COT (sum of all CTP layers for each COD-bin).

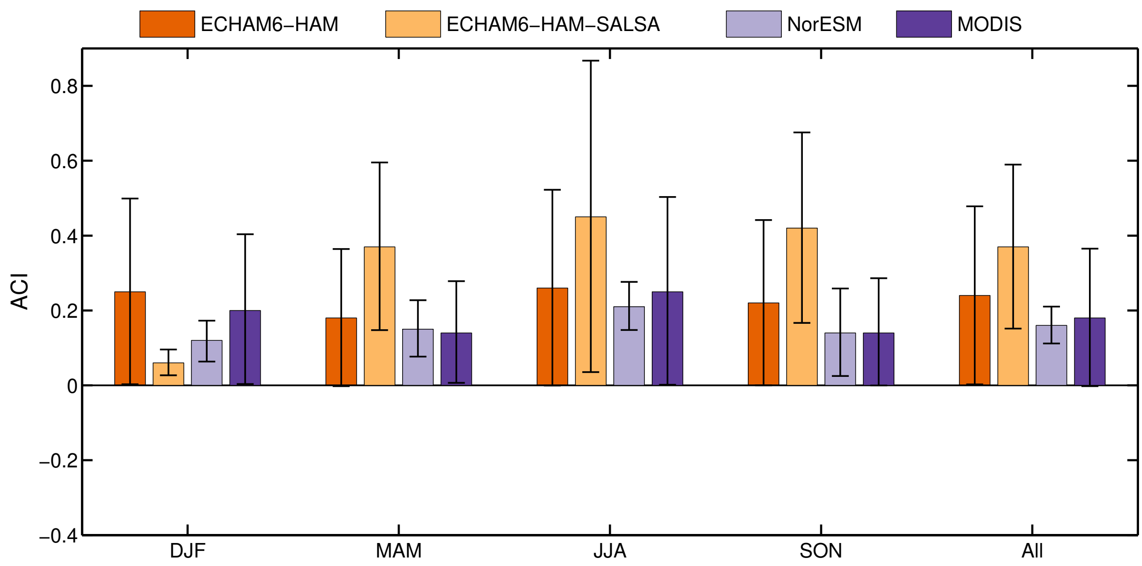

Figure 7. Global (over ocean only) estimates of the ACI_{CDNC} computed as the changes of ln(CDNC) to ln(AI). CDNC are derived from corresponding daily grid points of LWP and COT from MODIS observations and COSP-MODIS outputs following Bennartz (2007). ACI_{CDNC} values are calculated by season and for the entire period (1 January 2008 – 31 December 2008). Uncertainties estimates are calculated as 95% confidence interval from the daily values.

Table 1. Annual global in-cloud mean value ± standard deviation for the parameters used in the study. If a grid point has CF $\leq$ 30%, the point is set to fill values in all the datasets. The process leads to a reduction of 35% of datapoints in each dataset. 'CF all' is not screened for CF $\leq$ 30%. AI are representative of ocean values only.

Source	CF all	CF	LWP gm^{-2}	CER μm	COT	CDNC cm^{-3}	AI
MODIS	0.68± 0.35	0.83 ± 0.21	140 ± 142	15.3 ± 4.7	18.5±18.7	82 ± 82.12	0.15±0.20
ECHAM-HAM	0.56 ± 0.36	0.70 ± 0.21	106 ± 83	11 ± 1.9	9.6 ± 11.9	168 ± 122	0.14±0.20
ECHAM-HAM-SALSA	0.56 ± 0.36	0.70 ± 0.20	168 ± 159	12.5 ± 3.5	9.9± 11.9	177 ± 183	0.11±0.18
NorESM	0.63 ± 0.34	0.42 ± 0.28	161 ± 133	11.9 ± 2.7	28.3 ± 53.6	167 ± 124	0.17 ± 0.26

Table 2. Summary of the the models used in the study.

Model	Reference	Resolution	Aerosol scheme	Cloud microphysics
ECHAM-HAM	Tegen et al. (2019)	1.9° lat ×1.9° lon, 31 levels	HAM 2.3-M7	2-moment scheme
ECHAM-HAM-SALSA	Kokkola et al. (2018)	1.9° lat ×1.9° lon, 31 levels	HAM2.3-SALSA	2-moment scheme
NorESM	Kirkevåg et al. (2018)	0.9 ° lat × 1.25 ° lon, 30 levels	OsloAero	2-moment scheme MG1.5

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