We are grateful to the evaluations from the reviewers, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in italic and our response in bold. (The autoconversion and accretion data from HadGEM-UKCA are now available and Figure 6 is updated accordingly in the revised manuscript.)

Anonymous Referee #1

Received and published: 23 October 2015

The article investigates aerosol-cloud interactions and aerosol indirect radiative effects across a range of different climate models. The novel and very interesting aspect of the paper is that the large-scale dynamical settings are taken into account for the evaluation of aerosol-cloud interactions. The authors find a different sensitivity of the liquid water content to cloud condensation nuclei under different large-scale conditions, where regions of subsidence and strong monthly mean updraft are most sensitive. The comparison of different climate models indicates that models particularly strongly diverge in exactly these regimes. A further interesting finding is that the model predictions of the aerosol effects varies much more if different large-scale conditions are taken into account than for global results.

Promoting the idea of binning aerosol-cloud interactions into different dynamical settings is very helpful and an important aspect of advancing our understanding of aerosol-cloud interactions. The impact of large-scale dynamics on cloud-aerosol interactions has received only recently appropriate attention and this paper strongly contributes to highlight the importance of the large-scale dynamics. The large-scale (time and spatial) perspective taken in the paper allows to assess and compare a large number of large-scale dynamical regimes and helps to argue for the importance of large-scale dynamics. However, certainly more detailed work is required to understand the relation between large-scale dynamics, aerosols and cloud processes. The following points should be addressed before final publication.

Specific issues:

1. Please specify how monthly-mean GCM data for PI and PD is extracted from the models, i.e., time-slice experiments or average over specific time period (dates). If averages over a specific time period are used, it should be discussed what other changes in the state of the atmosphere could lead to changes in LWP coinciding with changes in aerosol number density. By just applying the eq. (1) these changes can project on the aerosol susceptibility although they are not physically related to aerosol-cloud interactions.

The monthly-mean GCM data for PI and PD is obtained by averaging over specific time period of 5 years. Natural variability of the simulations might

have some influence on the result. That's why all simulations were nudged toward reanalysis winds from operational forecast centers (some were also nudged toward analyzed temperature). For example, Figure S1 shows the vertical pressure velocity at 500hPa for PD, PI and the difference between them. It can be seen that the difference is quite minor. Nudging can significantly limits natural variability (Kooperman et al., 2012). Meanwhile, eq. (1) also allows some feedbacks, for example cloud feedback on CCN. An explanation about this has been added in the revised manuscript and now it reads (P. 9, l. 182-183): "Note that this metric allows some feedbacks, for example cloud effects on CCN." and (P. 11, l. 234-238): "Only ω 500 in PD runs is used to derive dynamical regimes and then these dynamical regimes are applied to PI simulations as well, with the assumption that ω 500 does not change much from PI to PD. This assumption is reasonable because both PD and PI runs were nudged toward the reanalysis data here, which ensures ω 500 is very similar between PD and PI.".

2. It would help to clarify the definition of "dynamical regime", if the characteristic spatial and temporal scales of the dynamic processes depicted by the chosen definition would be specified. It should also be discussed how relevant such a coarse definition is for aerosol-cloud interactions, particularly in regions with very transient dynamic systems as for instance in the extra tropics. It would be also helpful to include an additional figure showing the typical distribution of dynamical regimes as used in this study over the globe eventually for different seasons.

Since vertical pressure velocity is used as a criterion here, dynamic regimes generally follow the features of vertical pressure velocity distributions. Figure S1.A shows the distribution of annual mean distribution of ω500. Descending regimes are mostly located at subtropical regions and western coasts of continents, while ascending regimes locate around ITCZ and northern Pacific where storm tracks prevail. As for temporal change, the seasonal evolution of dynamic regimes follows seasonal changes in the major meteorological systems. For example, ascending regimes move north as ITCZ move north and descending regimes move accompanying with the movement of subtropical high. The characteristics of dynamic and thermodynamic regimes were discussed in detail in Bony et al. (2004). For some more specific dynamic regimes, such as stratocumulus, transitional clouds and trade wind cumulus, Figure S2 to S4 show the spatial distribution and temporal features of them. The spatial distribution in different seasons is similar to annual mean result (Fig. 4). As season changes, the spatial patterns do not change much and only the change of cloud fraction is evident. These clarifications of dynamic regimes with spatial and temporal patterns have now been added to the methodology part of revised manuscript and now it reads (P. 10, l. **202-210):** "Since vertical pressure velocity is used as a major criterion here, dynamic regimes generally follow the features of vertical pressure velocity distributions. Descending regimes are mostly located at subtropical regions and western coasts of continents, while ascending regimes locates around ITCZ and northern Pacific where storm tracks prevail. The seasonal evolution of dynamic regimes follows seasonal changes in the major meteorological systems. For example, ascending regimes move north/south as ITCZ move north/south and descending regimes move accompanying with subtropical high move. The characteristics of dynamic and thermodynamic regimes were discussed in detail in Bony et al. (2004)."

As is already stated in the text (P. 10. l. 199-P. 201): "Note however that the use of monthly means may obscure some details in the microphysical relationships, especially where the variability of cloud properties is high.". we acknowledge that the definition could be a little bit coarse. However, it is simple and it provides an effective way to separate different dynamic regimes. Bony and Dufresne (2005) adopted this definition of dynamic regimes and found evident subtropical cloud feedbacks uncertainties among climate models. More importantly, through this definition we do see different features of aerosol-cloud interactions within different dynamic regimes and find strong spread among different models, which could in turn suggest that this definition is effective and useful to understand the uncertainties associated with aerosol-cloud interactions in global climate models.

3. It should be specified how changes in LWP and CCN are computed: Are the values first binned according to ω 500 in PD and PI runs and then subtracted or are the grid point differences binned according to ω 500 from either PD or PI runs? If the latter is used some justification is required, as the spatial pattern of ω 500 may be different between PD and PI runs.

The latter one is used. Only $\omega 500$ in PD runs is used to derive dynamical regimes and then these dynamical regimes are applied to PI simulations as well, with the assumption that $\omega 500$ does not change much from PI to PD. This assumption is reasonable because both PD and PI runs were nudged toward the reanalysis data in this study, which ensures $\omega 500$ is very similar between PD and PI. Figure S1.C shows the difference of $\omega 500$ between PD and PI in CAM5-CLUBB as an example. It can be seen that the difference is indeed very small.

Deriving dynamical regimes from PD and PI runs separately could be another choice as the reviewer has pointed out. Actually, we did test this method at the very beginning of our work. But with this approach, the grid points can be different between PD and PI under each dynamical regime, which might introduce other differences than those from aerosols. In order to avoid this complexity and further considering the fact that the distribution of $\omega 500$ are very similar between PD and PI, we finally decided to choose the latter approach (i.e., gird-point differences are binned according to $\omega 500$ from PD runs).

This is now clarified in the revised manuscript: (P. 11, l. 234-238): "Only ω 500 in PD runs is used to derive dynamical regimes and then these dynamical regimes are applied to PI simulations as well, with the assumption that ω 500 does not change much from PI to PD. This assumption is reasonable because both PD and PI runs were nudged toward the reanalysis data here, which ensures ω 500 is very similar between PD and PI.".

4. Are the LWP and CCN values for different ω 500 arithmetic means for the values in each bin?

Yes. We first sort these 12-month global grid values into 20 dynamical regimes according to their ω 500 values, keeping the number of grids in each bin equal. Mean values of LWP, CCN and other fields for each bin are calculated from averaging the values of all grids belonging to that particular bin. Now this explanation has been added to the first paragraph of Section 3.2a (P. 16, l. 328-333): "Figure 1 shows LWP and CCN as a function of vertical pressure velocity at 500 hPa (ω 500) derived from PD simulations. To derive Figure 1, the 12-month monthly global grid values are first sorted into 20 dynamical regimes according to their ω 500 values, keeping the number of samples in each bin equal. LWP, CCN and values of other fields for each bin are then calculated from averaging the values of all samples in that particular bin. ".

5. The summary is a bit fuzzy and hard to read, particularly the 3rd to 5th paragraph. Please try to reformulate these. The comparison to findings from previous studies should be more clearly described and potential reasons for discrepancies summarized. Furthermore a short statement on the impact of neglecting mixed phase and ice-phase processes on the results should be included.

The summary is reformulated now. Generally it has been shortened to present the most important results more clearly and concisely. The second paragraph and the 3rd paragraph are now combined into one paragraph. Now the text of 2nd to 4th paragraph in the summary reads:

"The response of liquid water path (LWP) to aerosol perturbations, λ =dlnLWP/dlnCCN, a metric to quantify cloud lifetime effect of aerosols (Wang et al., 2012), shows a large spread within dynamical regimes among GCMs, although the global means are close. This diversity indicates that the aerosol cloud lifetime effect is regime-dependent. It is in strong ascending regimes and subsidence regimes that λ differs most between GCMs (Fig. 2a). Stratocumulus regimes have traditionally been the focus for studying aerosol indirect effects because of their significant cooling effect in climate system (e.g., Ackerman et al., 2004; Bretherton et al., 2007;

Gettelman et al., 2013). However, our results highlight that regimes with strong large-scale ascent should be another important regime to focus on in the future. Our results indicate that aerosol indirect forcing in regimes of vertical ascent is close to, or even larger than that in low cloud regimes (Fig. 7). Note however that these GCMs do not treat aerosol effects in their representations of deep convection that dominates clouds and LWP in regimes with strong ascent, while new versions of CAM exist where a version of the MG microphysics has been embedded in the deep convective parameterization (Song and Zhang, 2011).

By adding LTS as another criterion, we further separated different low cloud types under large-scale subsidence and revealed some further differences in cloud lifetime effect of aerosols on different types of low clouds. For example, the large λ in subsidence regimes in CAM5-CLUBB and ECHAM6-HAM2 comes from both stratocumulus and trade wind cumulus, while in CAM5-CLUBB-MG2 it mostly comes from trade wind cumulus (Fig. 5). It is also interesting to note that the distribution of λ in SPRINTARS and SPRINTATSKK is more likely to depend on LTS rather than vertical pressure velocity."

A discussion about mixed phase and ice phase process has been added to the end the summary (P. 32, l. 687-693):" It is our future plan to carry in-depth analysis to further understand some of the findings documented here, such as the large spread in λ in regimes of vertical ascent in different models. For example, LWP response to aerosol perturbation documented in this study may include contributions from mixed-phase and ice clouds. In- depth analysis of cloud macrophysics and microphysics processes will help to improve the understanding of the model uncertainty."

Minor issues Introduction

1. p. 23686, l. 15ff: Add a sentence with some references on the influence of aerosols on clouds by their potential to modify latent heating and cooling profiles.

Done. The sentence "It is worth noting that delaying the onset of precipitation may further modify latent heating profiles, which could lead to the invigoration of convective clouds (Andreae et al., 2004; Rosenfeld et al., 2008)." **has been added to the second paragraph of the introduction now (P. 4, l. 71-73).**

2. p. 23686, l. 17: Give references to articles considering mixed-phase and ice phase clouds.

Done. Now the text reads (P. 4, l. 74-77): "There are also adjustments on mixed-phase and ice clouds (e.g., Storelvmo et al., 2008; Lohmann and Hoose, 2009; Liu et al. 2012; Storelvmo et al., 2008; Wang et al., 2014). The focus of this study is on liquid cloud response to aerosol perturbation, primarily from large-scale clouds."

3. p. 23687, l. 8ff: Repeating the information from two sentences earlier. Also the

next sentence is very long, please reformulate.

The sentence "Ackerman et al. (2004) also demonstrated that the reduced cloud droplet size due to increases in aerosol reduces cloud droplet sedimentation." is **deleted now. The following sentence is reformulated into two sentences, which are** "They showed that the entrainment rate was reduced by decreasing available boundary-layer turbulence kinetic energy (TKE). However, Bretherton et al. (2007) found that TKE remained unchanged and changes in entrainment rate is mainly caused by reduced evaporative cooling from removing out liquid water" (P5, I 98-101).

4. p. 23692, l.9: "that the frequency of the following sorted dynamic regimes": unclear please reformulated.

We have changed this. Now the text reads (P. 13, l 263-265): "The similar patterns of ω 500 (due to nudging) in these simulations ensure that dynamic regimes defined by ω 500 do not vary much between models."

5. p. 23695, l. 18: replace "largest λ " with "largest global λ "

Done.

6. p. 23696, l. 14ff: sentence starting with "A major improvement ..." is unclear. Please reformulated.

We apology for this mistake made in the typesetting of the paper. It is an incomplete sentence. The full sentence should be "A major improvement of CAM-CLUBB is the better simulation of the transition of stratocumulus to trade wind cumulus over subtropical oceans (Bogenschutz et al., 2013). Fig. 2a shows that ... ". It has now been fixed in the revised manuscript (P. 19, l. 404-407).

7. p. 23697, l. 2: remove "where storm tracks prevail". This is not really required here and makes sentence hard to read.

This is now removed.

8. p. 23697, l. 7: add "spatial" before "pattern"

Done.

9. p. 23700, l. 19: sentence starting with "By sorting into ..." is unclear. Please reformulate.

We have changed this. Now it is reformulated to "It is in moderate regimes (-20

 $hPa/d < \omega 500 < 10$ hPa/d) where the result is consistent with Gettelman et al. (2015), which shows larger AUTO/PRECL in CAM5 than CAM5-MG2." (P. 25, l. 531-533).

10. p. 23703, l. 18: "Despite the closer global means ..." unclear, please reformulate.

Done. Now it is changed to "The response of liquid water path (LWP) to aerosol perturbations, λ =dlnLWP/dlnCCN, a metric to quantify cloud lifetime effect of aerosols (Wang et al., 2012), shows a large spread within dynamical regimes among GCMs, although the global means are close. "(P. 30, line 632-635)

11. p. 23704, l. 24: "Results derived from large eddy ..." unclear, please reformulate. **Done. The sentence is reformulated to** "Results derived from large eddy simulation (LES) and single column model (SCM) (e.g., Ackerman et al., 2004; Guo et al., 2011) have shown that λ could be negative under low precipitation situations, which indicates that λ is expected to be smaller under low precipitation situations." (P. 31, l. 661-664).

12. p. 23705, l. 7: replace "can reduce" by "reduces", remove "only"

Done.

13. p. 23705, l. 9: replace by "total SCRE decreases in models with prognostic rain scheme compared to those with a diagnostic rain scheme"

Done.

14. p. 23705, l. 20: Monthly mean ω 500 also does not necessarily represent the same conditions as also dynamical conditions may vary quite significantly on sub-monthly timescales.

Yes, we agree that monthly mean $\omega 500$ may not represent the variation in dynamical regimes on sub-monthly timescales. However, as we stated in our response to the reviewer's specific comment #2, sorting model data according to $\omega 500$ is an effective way to reveal uncertainties in aerosol indirect effects among different models. As is already stated in the text (P. 10. l. 199-201): "Note however that the use of monthly means may obscure some details in the microphysical relationships, especially where the variability of cloud properties is high.", we acknowledge the limitation of using monthly data, and we do plan to carry further analysis with hourly data in the future (see our discussion in the last paragraph.

15. p. 23706, l. 1: remove "A" from "Appendix A" since there is only one appendix.

Done.

16. caption of Tab. 4: replace "global regimes" by "all dynamical regimes".

Done.

Anonymous Referee #2

Received and published: 5 November 2015

This paper presents an analysis of the regime dependence of the susceptibility of LWP to changes in CCN, from 10 GCMs. The main goal of this analysis is show the importance of examing aerosol-cloud interactions different cloud and dynamical regimes, focusing only on warm clouds. The paper shows that lambda differs most between GCMs in regions of strong ascending regimes and subsidence regimes. Interestingly, the analysis shows that the sensitivity of LWP to changes in aerosol in regions of vertical ascent are equal to or even larger than that in low cloud regions. To the best of my knowledge this is the first paper that assesses aerosol-cloud interactions by dynamic regime, using GCMs. This is an important step to understanding aerosol-cloud interactions, so it is good to see this. In general, I think the paper and the overall results will be of interest to a broad community, but I think there needs to be some more detail about the method and some more analysis to understand the significance of the results. For this reason, I am recommending the paper should be accepted for publication once the following changes have been undertaken.

General comments:

1. It is not completely clear from the paper how the presented LWP and CCN are calculated. From the description in Table 2, I have to assume that the presented is averaged LWP and CCN are spatial averages for the present day, where the space can be the globe or the dynamic regime. The relative change in LWP and CCN from the GCM is the relative change in the spatial average over time (PI to PD). Is this correct? If so, it would be very useful if this could be explicitly stated in the text. At present, I feel that I am having to piece together the method from snippets throughout the entire text (including figure and table captions).

Yes, as described in the caption of Table 2, LWP and CCN are annual spatial averages over ocean from PD simulations, for the purpose of showing annual mean state of each model. As for LWP and CCN of each dynamical regime, they are both spatial and temporal averages from 12-month monthly data. As for how to get LWP and CCN for each bin, please see also our answer to specific comment #4 from reviewer #1. This is now added in the methodology section to further clarify how these fields are calculated (P. 8, l. 171-175): "It is directly calculated as the relative change of monthly mean LWP from pre-industrial (PI) to present day (PD) divided by the relative change of

CCN. Here dlnLWP=(LWP_{PD}-LWP_{PI})/LWP_{PI} and dlnCCN=(CCN_{PD}-CCN_{PI})/CCN_{PI}, where LWP_{PD} and LWP_{PI} are LWP in PD and PI, respectively, while CCN_{PD} and CCN_{PI} are CCN in PD and PI, respectively.".

2. Equation 1 defines the susceptibility of LWP to changes in CCN, but it is not clear in this paper how this is calculated. Given past work on precipitation susceptibility, I assume that LWP susceptibility is calculated by binning LWP and the associated in CCN from PI to PD into dynamic regime bins. Then, within a bin, a linear regression is applied to the lnLWP and lnCCN, to obtain lambda. Is this correct? If so this should be stated, so that others can perform the same analysis. Further, this work, particularly figure 2 and table 1 only present a single value for each dynamic bin. It would be very useful and would add to the paper if the authors could present error bars on this figure, or state the correlation for each regression, so that the reader can understand the significance of the trend in lambda with dynamic regime. Past work, e.g. Jiang et al, Terai et al, Hill et al, all presented error bars or correlations coefficients with their work, which helps the reader to understand significance. Is the correlation of LWP to CCN good in the GCMs tested?

The susceptibility of LWP to changes in CCN is not calculated from the linear regression between lnLWP and lnCCN. This is directly calculated as the relative change of LWP from PI to PD divided by the relative change of CCN,i.e., λ =dlnLWP/dlnCCN=[(LWP_{PD}-LWP_{PI})/LWP_{PI}]/[(CCN_{PD}-CCN_{PI})/CCN_{PI}]. For this reason, we do not provide the error bars or correaltions in Figure 2 and Table 1. The same approach was also used by Wang et al. (2012) to constrain the cloud lifetime of aerosols. The detailed formula is added to methodology part now. We found that using the term 'susceptibility' might be somehow misleading, so now it has been changed to 'the response of LWP to changes in CCN' in the revised manuscript and the text reads (P. 8, 1. 171-175):".It is directly calculated as the relative change of monthly mean LWP from pre-industrial (PI) to present day (PD) divided by the relative change of CCN. Here dlnLWP=(LWP_{PD}-LWP_{PI})/LWP_{PI} and dlnCCN=(CCN_{PD}-CCN_{PI})/CCN_{PI}, where LWP_{PD} and LWP_{PI} are LWP in PD and PI, respectively, while CCN_{PD} and CCN_{PI} are CCN in PD and PI, respectively.".

3. The paper very clearly states that the focus of this work is warm phase clouds, so it focuses on LWP alone. This is fine, but given that all the GCMs include ice phase processes, it would be useful if the authors would discuss whether the GCMs are producing changes in the ice phase and mixed phase processes and whether these changes are influencing their results. For example, is the sensitivity of the LWP to aerosol in ascending regimes only the result of changes in warm phase rain processes or is there an impact resulting from change in the ice phase and mixed phase processes. I think this type of discussion would give some more insight into the results presented. We first would like to clarify that there was a mistake in the last sentence in second paragraph of introduction in the original manuscript. Actually all liquid clouds were sampled in this study, not only warm clouds, so cloud water melt from mixed and ice phase processes is also included. We have corrected this in the revised manuscript and it reads (P. 4, l. 74-77): "There are also adjustments on mixed-phase and ice clouds (e.g., Storelvmo et al., 2008; Lohmann and Hoose, 2009; Liu et al. 2012; Storelvmo et al., 2008; Wang et al., 2014). The focus of this study is on liquid cloud response to aerosol perturbation, primarily from large-scale clouds.".

It is possible that some of the changes in liquid water path and aerosol indirect forcing may come from changes in ice phase and mixed phase processes (e.g., Gettelman, 2015). Detailed and specific discussions on the role of mixed-phase and ice phase clouds required in-depth analysis and is beyond the scope of the current manuscript and we intend to leave this for a separate study in the future. This has been now stated in the end of summary (P. 32, l. 687-693): " It is our future plan to carry in-depth analysis to further understand some of the findings documented here, such as the large spread in λ in regimes of vertical ascent in different models. For example, LWP response to aerosol perturbation documented in this study may include contributions from mixed-phase and ice clouds. In-depth analysis of cloud macrophysics and microphysics processes will help to improve the understanding of the model uncertainty."

Specific Comments:

1. Page 23685, abstract - "with strong large scale ascend" should be changed to "strong large scale ascent"

Corrected.

2. Page 23688, paragraph 2 – I feel that the authors are inferring that autoconversion is a natural process in the warm rain formation. I would argue that autoconversion is modelling necessity only related to bulk microphysics schemes. For example, bin microphysics and superdroplet schemes do not include a specific parametrisation for autoconversion because it is dealt with the collection equations. The second sentence on paragraph 2 needs to be modified so it is explicitly stated that this relates to only bulk microphysics schemes.

Thanks and we now clarified this in the revised manuscript. Now the text reads (P. 6, l. 128-129): "In warm clouds, cloud microphysical processes are dominated by autoconversion and accretion in bulk microphysics schemes (Gettelman et al., 2013)."

3. Page 23688, paragraph 2, last sentence – The last sentence is correct, i.e, using a prognostic rain scheme enhances the dominance of accretion. However, it may be

useful to state that this alone might not be a panacea. For example, Hill et al 2015 showed that for an all-else-equal test, there is still significant differences in the precipitation susceptibility from single moment prognostic rain schemes.

Yes, thanks for the suggestion. We realized the last sentence might be kind of misleading, which could underline too much the effect of adding prognostic rain scheme. As being pointed out here, we agree that prognostic rain scheme might not be a panacea. This is also the motivation of comparing results derived from models with (e.g. CAM5-MG2) and without (e.g., CAM5-NCAR) prognostic rain-scheme in our work. It has been modified now. Now the statement (P. 7, l. 142-147): "However, Hill et al. (2015) shows that adding prognostic rain scheme alone still cannot reduce the spread precipitation among different of susceptibility of cloud microphysics parameterizations and further shows that increasing the complexity of the rain representation to double-moment significantly reduces the spread of precipitation sensitivity and improves overall consistency between bulk and bin schemes." has been added.

4. Page 23692, second paragraph, last 2 sentences – I found this a bit confusing. I think this is saying that the same LWP are not being presented because the models report different LWP, with some including LWP from mixed phase clouds, while others do not. Is this important? Does the impact of changing aerosol on mixed phase clouds impact the results and conclusions from the regime analysis? This point relates back the general comment (3).

Different models treat LWP differently. As summarized in Table 1, most versions of CAM5 sample LWP from stratiform clouds. CAM5-CLUBB and CAM5-CLUBB-MG2 also include shallow convective clouds because higher-order turbulence closure (CLUBB) unifies the treatment of boundary layer turbulence, stratiform clouds and shallow convection (Bogenschutz et al., 2013). SPRINTARS and HadGEM3-UKCA sample LWP from both stratiform clouds and convective clouds. This difference can contribute to the spread of λ in our study, and that's why we show Table 1 and also give some descriptions in the manuscript. This is now noted in the manuscript and it reads: (P. 16, l. 342-345): " The model spread of LWP response is larger in the ascending regimes than in the subsiding regimes. This may be partly related to the fact that the types of clouds included in LWP are not the same in different models (Table 1)."

However, all models sample LWP from liquid clouds, which also include cloud water melt from mixed phase clouds and ice phase clouds. So changes in LWP in the paper may include aerosol impact in mixed-phase or ice clouds. See our answer to general comment (3).

5. Page 23693, second paragraph, last sentence – I like that the authors have stated

that large differences CCN may not correspond to large differences in the Nd because treatment of cloud base updraft. However, it raises the question whether lambda should be defined as the change in LWP vs the change in Nd, not CCN? I am aware that this definition would be difficult to compare with observations, but given that LWP is dependent on Nd, not necessarily CCN, it would be useful to know whether the results presented are sensitive to this definition. Could the authors add some discussion to address this?

As the focus of this study is about aerosol indirect effects, the current definition of λ provides the direct measure of cloud response to anthropogenic aerosol perturbation and therefore serves our purpose well. As the reviewer noted, the alternative definition of λ as dlnLWP/dlnNd would be difficult to compare with observations, and this new definition also can not directly measure clouds response to anthropogenic aerosols. However, we agreed that the reviewer raised an important point. The interaction between clouds and anthropogenic aerosols arises through a chain of processes, from effects of the CCN on Nd to effects of Nd on cloud water. This chain of processes can be expressed as dlnLWP/dlnCCN=(dlnLWP/dlnNd)*(dlnNd/dlnCCN). It is highly interesting to examine this chain of processes to improve our understanding of aerosol-cloud interactions. In a separate study using the same set of model simulations, we did examine this chain of processes (Ghan et al., 2016). Further discussion about this has been added to Section 3.1 and the text reads (P. 15, l. 312-325): "We also should note that large differences in CCN shown in Table 2 do not necessarily correspond to equally large differences in droplet concentration (N_d), since N_d is primarily dependent on cloud base updraft that is an extremely uncertain parameter and may vary significantly between the GCMs. It therefore seems reasonable to define λ as the change in LWP vs. the change in cloud droplet number concentration (N_d) , which would provide a direct insight into how clouds response to N_d change since LWP directly depends on N_d , not necessarily on CCN. However, this alternative definition of λ as dlnLWP/dlnN_d would be difficult to compare with observations, and this also does not directly measure cloud response to anthropogenic aerosols. The interactions between clouds and anthropogenic aerosols arise through a chain of processes, from effects of the CCN on N_d to effects of N_d on cloud water, which be expressed can as dlnLWP/dlnCCN=(dlnLWP/dlnN_d)*(dlnN_d/dlnCCN). This chain of processes has now been examined in Ghan et al., (2016) based on the same set of model simulations documented in this study. "

6. Page 23696, second paragraph, sentence beginning "A major improvement of CAMCLUBB...", this sentence does not make sense. I think some words are missing

Sorry for the mistake here. This is now fixed and please see our answer to specific comment #6 from reviewer #1.

7. Page 23702, second paragraph, sentence beginning "Here we investigate the LWP response to aerosol perturbations under low precipitation...". Are the results sensitive to the precipitation threshold applied? Previous work has shown that precipitation susceptibility is sensitive to this threshold.

This is a good point. The results can be potentially sensitive to precipitation threshold. We now tested how our results are sensitive to different thresholds of precipitation (0.01, 0.05, 0.1 and 0.2 mm d⁻¹). Table S1 shows the fractional occurrences of low surface precipitation over global oceans for different thresholds. As we expect, the fractional occurrence of low precipitation increases as the threshold increases. However, the LWP response to aerosol perturbations under low and high precipitation does not change much as the threshold changes. For example, although the threshold has been changed to four times larger from 0.05 mm d⁻¹ to 0.2 mm d⁻¹, λ under high and low precipitation situations barely change. This indicates the results of LWP response to CCN changes under low and high precipitation are insensitive to the threshold we applied.

We also looked at aerosol indirect effects (dSCRE) under low and high precipitation with different thresholds (Figure S5-S7 and Figure 7). The aerosol indirect effects contributed by low and high precipitation do change with different thresholds. This is mainly caused by the changing fractional occurrences. However, the conclusion is still reliable no matter how the threshold changes. High precipitation situations contribute the most part of aerosol indirect effects.

As monthly data is used here, the results may not be as sensitive as it would be if instantaneous data is used. As we noted in the summary part, using instantaneous data may also produce different results and we intend to carry a similar analysis using the instantaneous data in a future study. We now added discussion in the end of Section 3 and it reads (P. 28, l. 608-623): " Our sensitive tests indicate that results in Table 4 and Figure 7 can be potentially sensitive to the precipitation threshold applied to separate high precipitation and low precipitation situations (not shown). The occurrence frequency of low precipitation situations increases with increasing threshold and the magnitude of increase can be different for different models. For example, when the precipitation threshold increases from 0.01 mm d^{-1} to 0.20 mm d^{-1} , the occurrence frequency of low precipitation situations increases from 2% to 37% in CAM5-PNNL while it increases from near 0% to 5% in CAM5-CLUBB. Increasing the precipitation threshold also increases the contribution of low precipitation situations to the total aerosol indirect forcing as the occurrence frequency of low precipitation situations increase. However, our results indicate that the LWP response to aerosol perturbations under low and high precipitation does not change much as the precipitation threshold changes and that high precipitation situations generally contribute more to the total aerosol indirect

forcing for precipitation threshold in the range of 0.01 to 0.20 mm d⁻¹. More work is needed to explore this further such as how results may be different when instantaneous precipitation data (e.g., 3-houly data) is used."

References:

Bogenschutz, P. A., Gettelman, A., Morrison, H., Larson, V. E., Craig, C., and Schanen, D. P.: Higher-Order Turbulence Closure and Its Impact on Climate Simulations in the Community Atmosphere Model, J. Clim., 26, 9655-9676, 10.1175/jcli-d-13-00075.1, 2013.

Bony, S., Dufresne, J. L., Le Treut, H., Morcrette, J. J., and Senior, C.: On dynamic and thermodynamic components of cloud changes, Clim. Dyn., 22, 71-86, 10.1007/s00382-003-0369-6, 2004.

Bony, S., and Dufresne, J. L.: Marine boundary layer clouds at the heart of tropical cloud feedback uncertainties in climate models, Geophys. Res. Lett., 32, 10.1029/2005gl023851, 2005.

Gettelman, A.: Putting the clouds back in aerosol-cloud interactions, Atmos. Chem. Phys., 15, 12397-12411, 10.5194/acp-15-12397-2015, 2015.

Ghan, S. J., Wang, M., Zhang, S., Ferrachat, S., Gettelman, A., Griesfeller, J., Kipling, Z., Lohmann, U., Morrison, H, Neubauer, D., Partridge, D., Stier, P., Takemura, T., Wang, H., Zhang, K.: Challenges in Constraining Anthropogenic Aerosol Effects on Cloud Radiative Forcing Using Present-day Spatiotemporal Variability, Proc. Natl. Acad. Sci. U. S. A., Accepted, 2016.

Kooperman, G. J., Pritchard, M. S., Ghan, S. J., Wang, M. H., Somerville, R. C. J., and Russell, L. M.: Constraining the influence of natural variability to improve estimates of global aerosol indirect effects in a nudged version of the Community Atmosphere Model 5, J. Geophys. Res.-Atmos., 117, 10.1029/2012jd018588, 2012.

Hill, A. A., B. J. Shipway, and I. A. Boutle (2015), How sensitive are aerosol-precipitation interactions to the warm rain representation?, J. Adv. Model. Earth Syst., 7, 987–1004, doi:10.1002/2014MS000422.

Jiang, H., Feingold, G., and Sorooshian, A.: Effect of Aerosol on the Susceptibility and Efficiency of Precipitation in Warm Trade Cumulus Clouds, J. Atmos. Sci., 67(11), 3525–3540, doi:10.1175/2010JAS3484.1, 2010. Penner, J. E., Xu, L., and Wang, M. H.: Satellite methods underestimate indirect climate forcing by aerosols, Proc. Natl. Acad. Sci. U. S. A., 108, 13404-13408, 10.1073/pnas.1018526108, 2011.

Stevens, B., and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461, 607-613, 10.1038/nature08281, 2009.

Terai, C. R., R. Wood, D. C. Leon, and P. Zuidema (2012), Does precipitation susceptibility vary with increasing cloud thickness in marine stratocumulus?, Atmos. Chem. Phys., 12, 4567–4583, doi:10.5194/acp-12-4567-2012.

Wang, M. H., Ghan, S., Liu, X. H., L'Ecuyer, T. S., Zhang, K., Morrison, H., Ovchinnikov, M., Easter, R., Marchand, R., Chand, D., Qian, Y., and Penner, J. E.: Constraining cloud lifetime effects of aerosols using A-Train satellite observations, Geophys. Res. Lett., 39, 10.1029/2012gl052204, 2012.

Table S1. The fractional occurrences of low surface precipitation in PD cases over global oceans for different precipitation thresholds. Low precipitation situations refer to monthly surface precipitation rate (PRECL) less than the threshold.

Model	f, low thre=0.01	f, low thre=0.05	f, low thre=0.1	f, low thre=0.2
CAM5	0.02	0.18	0.27	0.37
CAM5-MG2	0.23	0.32	0.39	0.48
CAM5-PNNL	0.02	0.19	0.28	0.37
CAM5-CLUBB	0.00	0.01	0.02	0.05
CAM5-CLUBB-MG2	0.03	0.08	0.16	0.30
ECHAM6-HAM2	0.04	0.12	0.18	0.25
SPARINTARS	0.00	0.02	0.03	0.06
SPARINTARS-KK	0.00	0.02	0.03	0.05
ModelE2-TOMAS	0.00	0.00	0.001	0.01
HadGEM3-UKCA	0.01	0.02	0.06	0.15

	thre=	=0.01	thre=	=0.05	thre	=0.1	thre	=0.2
	λ^{a}	λ^{b}	λ^{a}	λ^{b}	λ^{a}	λ^{b}	λ^{a}	λ^{b}
Model	low,	high,	low,	high,	low,	high,	low,	high,
	down	down	down	down	down	down	down	down
CAM5	0.16	0.18	0.20	0.18	0.21	0.19	0.21	0.19
CAM5-MG2	0.16	0.22	0.18	0.23	0.19	0.24	0.21	0.25
CAM5-PNNL	0.12	0.16	0.16	0.17	0.17	0.17	0.18	0.17
CAM5-CLUBB	Nan	0.30	0.34	0.30	0.33	0.30	0.34	0.31
CAM5-CLUBB-MG2	0.18	0.30	0.21	0.33	0.26	0.33	0.31	0.32
ECHAM6-HAM2	0.13	0.24	0.24	0.23	0.25	0.23	0.27	0.22
SPARINTARS	Nan	0.01	0.06	0.01	0.06	0.01	0.04	0.01
SPARINTARS-KK	Nan	0.04	0.25	0.04	0.24	0.04	0.20	0.04
ModelE2-TOMAS	Nan	0.00	Nan	0.00	-0.011	0.001	-0.01	0.00
HadGEM3-UKCA	0.03	0.03	0.04	0.03	0.04	0.03	0.04	0.03

Table S2. λ under low and high surface precipitation situations only over downdraft regimes for different thresholds.

 $^a\,\lambda$ under low PRECL for downdraft regimes

 $^{\rm b}\,\lambda$ under high PRECL for downdraft regimes

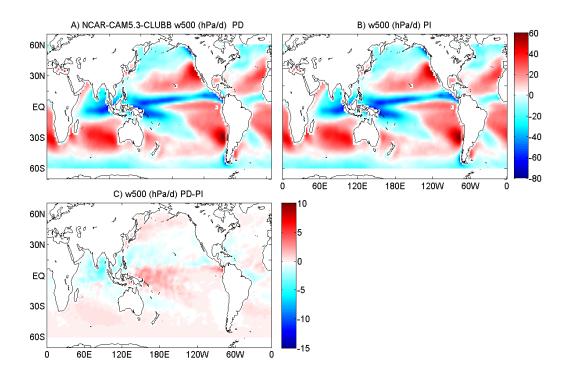


Figure S1. Annual average vertical pressure velocity at 500hPa level derived from A) present day simulation (PD), B) pre-industrial simulation (PI) and C) their difference PD-PI in CAM5-CLUBB.

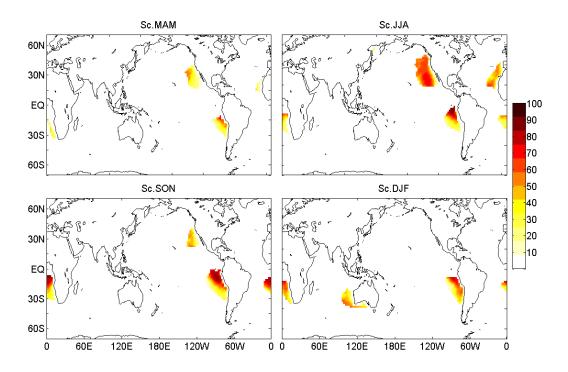


Figure S2. The seasonal mean (MAM, JJA, SON and DJF) cloud fraction of stratocumulus regime derived from PD monthly simulation in CAM5-CLUBB.

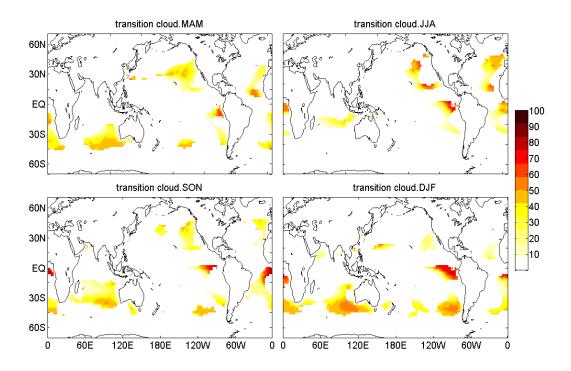


Figure S3. Same as Fig. S2, but for transitional clouds.

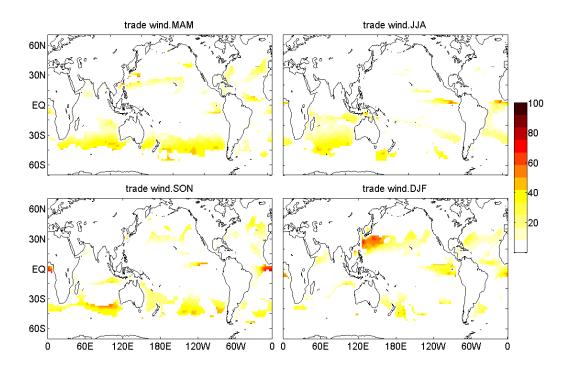


Figure S4. Same as Fig. S2, but for trade wind cumulus.

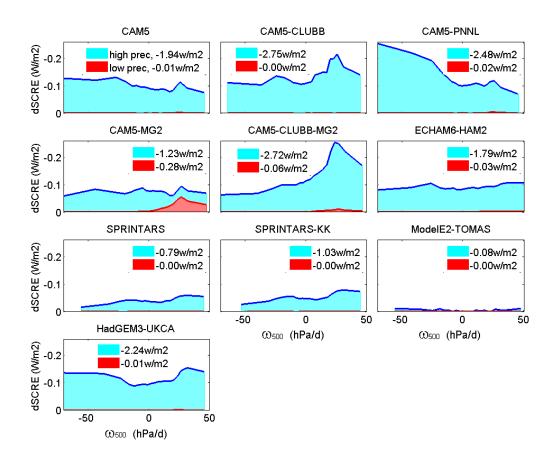


Fig S5. Same as Figure 7, but for threshold=0.01 mm d⁻¹.

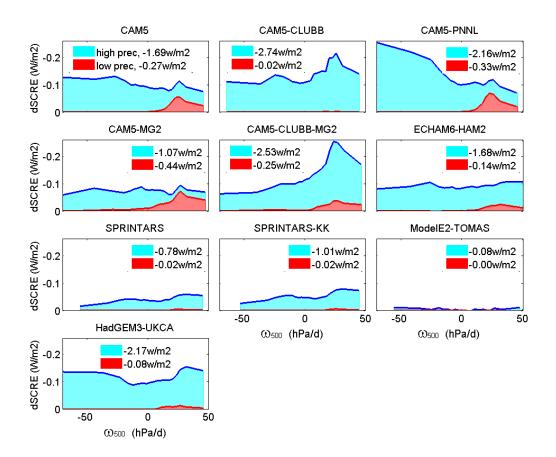


Fig S6. Same as Figure 7, but for threshold= 0.05 mm d^{-1} .

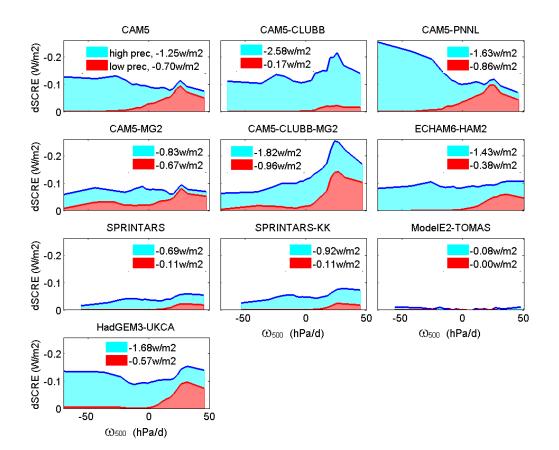


Fig S7. Same as Figure 7, but for threshold=0.2 mm d⁻¹.

On the characteristics of aerosol indirect effect based on dynamic regimes in global climate models

5	S. Zhang ^{1,2,3} , M. Wang ^{1,2,3} , S. J. Ghan ³ , A. Ding ^{1,2} , H. Wang ³ , K. Zhang ³ , D.
6	Neubauer ⁴ , U. Lohmann ⁴ , S. Ferrachat ⁴ , T. Takeamura ⁵ , A. Gettelman ⁶ , H. Morrison ⁶ ,
7	Y. H. Lee ⁷ , D. T. Shindell ⁷ , D. G. Partridge ^{8,9,10} , P. Stier ⁸ , Z. Kipling ⁸ , C. Fu ^{1,2}
8	
9	¹ Institute for Climate and Global Change Research and School of Atmospheric
10	Sciences, Nanjing University, Nanjing, China
11	² Collaborative Innovation Center of Climate Change, Jiangsu Province, China
12	³ Atmospheric Science and Global Change Division, Pacific Northwest National
13	Laboratory, Richland, Washington, USA
14	⁴ ETH Zurich, Institute for Atmospheric and Climate Science, Zurich, Switzerland
15	⁵ Research Institute for Applied Mechanic, Kyushu University, Fukuoka, Japan
16	⁶ National Center for Atmospheric Research, Boulder, Colorado, USA
17	⁷ Earth and Ocean Sciences, Nicholas School of the Environment, Duke University,
18	Durham, North Carolina, USA
19	⁸ Atmospheric, Oceanic and Planetary Physics, Department of Physics, University of
20	Oxford, Oxford, UK

21	⁹ Department of Environmental Science and Analytical Chemistry, Stockholm
22	University, Stockholm, Sweden
23	¹⁰ Bert Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden
24	To be submitted to Atmospheric Chemistry and Physics
25	
26	*Corresponding author: Minghuai Wang (minghuai.wang@nju.edu.cn)
27	

28 Abstract

Aerosol-cloud interactions continue to constitute a major source of uncertainty for 29 the estimate of climate radiative forcing. The variation of aerosol indirect effects (AIE) 30 in climate models is investigated across different dynamical regimes, determined by 31 monthly mean 500 hPa vertical pressure velocity (ω_{500}), lower-tropospheric stability 32 (LTS) and large-scale surface precipitation rate derived from several global climate 33 models (GCMs), with a focus on liquid water path (LWP) response to cloud 34 condensation nuclei (CCN) concentrations. The LWP sensitivity to aerosol 35 perturbation within dynamic regimes is found to exhibit a large spread among these 36 GCMs. It is in regimes of strong large-scale ascent ($\omega_{500} < -25$ hPa/d) and low clouds 37 (stratocumulus and trade wind cumulus) where the models differ most. Shortwave 38 aerosol indirect forcing is also found to differ significantly among different regimes. 39 Shortwave aerosol indirect forcing in ascending regimes is close to that in subsidence 40 regimes, which indicates that regimes with strong large-scale ascent are as important 41

as stratocumulus regimes in studying AIE. It is further shown that shortwave aerosol
indirect forcing over regions with high monthly large-scale surface precipitation rate (>
0.1 mm/d) contributes the most to the total aerosol indirect forcing (from 64% to
nearly 100%). Results show that the uncertainty in AIE is even larger within specific
dynamical regimes compared to the uncertainty in its global mean values, pointing to
the need to reduce the uncertainty in AIE in different dynamical regimes.
Key words: aerosol indirect effects, dynamic regimes, aerosol-cloud interactions

49

50 **1 Introduction**

By scattering and absorbing sunlight, aerosol particles can modify the solar 51 52 radiation reaching the earth system, which is termed the direct effect. The direct radiative effect of anthropogenic aerosols combined with subsequent rapid 53 adjustments of the surface energy budget, atmospheric state variables, and cloudiness 54 to aerosol radiative effects is referred as Effective Radiative Forcing from 55 aerosol-radiation interactions (ERFari) (Boucher et. al, 2013). Apart from ERFari, 56 aerosols can also alter the Earth's radiation balance via interactions with clouds, such 57 as effects on cloud albedo and subsequent changes to the cloud lifetime and 58 thermodynamics as rapid adjustments, known as the aerosol indirect effect(s) (AIE). 59 These radiative effects are called Effective Radiative Forcing from aerosol-cloud 60 61 interactions (ERFaci) (Boucher et. al, 2013).

For liquid clouds, there are two principal ways through which aerosols interact with them in AIE. First, an increase in cloud condensation nuclei (CCN)

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64	concentration from anthropogenic aerosols leads to smaller cloud droplet sizes
65	assuming constant liquid water content. The increased number but decreased droplet
66	sizes in turn increase cloud albedo due to more efficient backscattering. This is called
67	the cloud albedo effect or the first AIE, also known as the Twomey effect (Twomey,
68	1977). Moreover, the smaller cloud droplet sizes are hypothesized to lead to decreases
69	in precipitation efficiency, which may further alter cloud liquid water path (LWP) and
70	cloud lifetime (Albrecht, 1989). These adjustments are also referred to as the cloud
71	lifetime effect or the second AIE. It is worth noting that delaying the onset of
72	precipitation may further modify latent heating profiles, which could lead to the
73	invigoration of convective clouds (e.g., Andreae et al., 2004; Rosenfeld et al., 2008).
74	There are also adjustments on mixed-phase and ice clouds (e.g., Storelvmo et al.,
75	2008; Lohmann and Hoose, 2009; Liu et al. 2012; Storelvmo et al., 2008; Wang et al.,
76	2014). The focus of this study is on liquid cloud response to aerosol perturbation,
77	primarily from large-scale clouds.

AIE could be large enough to offset much of the global warming induced by anthropogenic greenhouse gases, yet its magnitude is still very uncertain (IPCC, 2013). The uncertainty in the cloud lifetime effect of aerosols is particularly large.

The complexity of microphysical-dynamical-radiative feedbacks involved in the cloud lifetime effect has been noted in previous studies. Conventional theory regarding the cloud lifetime effect suggests that higher CCN concentration slows down precipitation formation and hence leads to more LWP (Albrecht, 1989). However, this theory is inconsistent with some observations (Coakley and Walsh,

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86	2002; Kaufman et al., 2005; Matsui et al., 2006; Chen et al., 2014) and large eddy
87	simulations (LESs) (e.g., Ackerman et al., 2004; Lu and Seinfeld, 2005; Wang and
88	Feingold, 2009b) that found either increase or decrease in LWP in responses to
89	increases in CCN concentration.

Further modeling studies (e.g., Ackerman et al., 2004; Stevens and Feingold, 90 2009; Guo et al., 2011) suggest that cloud top entrainment plays a critical role as a 91 dynamic feedback, to balance LWP and modify the lifetime of boundary layer clouds. 92 Ackerman et al. (2004) found that an increase in droplet number concentration (N_d) 93 reduces cloud water sedimentation while accelerating the cloud-top entrainment rate, 94 which makes the humidity of air overlying the boundary layer, wet or dry, critically 95 important in determining the response of LWP. When surface precipitation is weak 96 $(<0.1 \text{ mm day}^{-1})$ and the overlying air is dry, LWP decreases in response to increasing 97 aerosol. They showed that the entrainment rate was reduced by decreasing available 98 boundary-layer turbulence kinetic energy (TKE). However, Bretherton et al. (2007) 99 found that TKE remained unchanged and changes in entrainment rate are mainly 100 caused by reduced evaporative cooling from removing out liquid water. LES studies 101 (e.g., Wang and Feingold, 2009a) with a large model domain that is able to resolve 102 mesoscale circulations (on the order of ten kilometers) in marine stratocumulus 103 showed that aerosols can shift cloud regimes through their impact on precipitation and 104 associated dynamical feedbacks. This can represent a more significant impact on 105 cloud radiative forcing than the conventional AIE. 106

Many state-of-the-art global climate models (GCMs) appear to overestimate AIE 107 when compared with satellite observations (e.g., Quaas et al., 2009; Wang et al., 108 2012), despite of some uncertainties in satellite derived estimates (e.g., Penner et al., 109 2011; Gryspeerdt et al., 2014a; Gryspeerdt et al., 2014b). The multi-scale interactions 110 between clouds, aerosols and large-scale dynamics (Stevens and Feingold, 2009; 111 Wang et al., 2011; Ma et al., 2015) and complex microphysical processes (e.g., 112 Bretherton et al., 2007; Gettelman et al., 2013) cause uncertainties in estimating AIE 113 by GCMs. One possible source of overestimation of AIE is their inability to reproduce 114 negative LWP responses to aerosol perturbations, which are found in some 115 observations and LES studies, partly because they do not explicitly simulate the 116 droplet size effect on the entrainment process and on sub-grid cloud organizations 117 associated with changes in precipitation. Guo et al. (2011) found that this effect could 118 be captured through applying a parameterization based on multi-variate probability 119 density functions with dynamics (MVD PDFs) in single-column simulations. They 120 found decreased LWP in response to increasing aerosols concentration and suggested 121 that the implementation of MVD PDFs in GCMs may help lower the magnitude of the 122 simulated AIE. A negative correlation between LWP and aerosol loading was further 123 found for clouds with weak precipitation and dry air above the PBL in a subsequent 124 global model study (Guo et al., 2015). 125

Another likely source for the overestimation of cloud lifetime effects in GCMs is
the treatment of cloud microphysics (Penner et al., 2006; Posselt and Lohmann, 2009;
Wang et al., 2012). In warm clouds, cloud microphysical processes are dominated by

129	autoconversion and accretion in bulk microphysics schemes (Gettelman et al., 2013).
130	Since autoconversion acts as a sink of LWP, it is crucial in the formation of
131	precipitation, thus plays an important role in determining the cloud lifetime effect.
132	The autoconversion rate is directly dependent on droplet number concentration $\left(N_{d}\right)$
133	while the accretion rate is only weakly dependent on N_{d} (Khairoutdinov and Kogan,
134	2000; Gettelman et al., 2013). Furthermore, the ratio of the autoconversion rate to the
135	large-scale surface precipitation rate is found to be strongly correlated with the LWP
136	response to anthropogenic aerosol perturbations (e.g., Wang et al., 2012). Posselt and
137	Lohmann (2009) suggested this ratio is related to the rain scheme adopted in GCMs.
138	They showed that the adoption of different rain schemes (prognostic vs. diagnostic) in
139	a GCM leads to a different LWP response to aerosol perturbations. A prognostic rain
140	scheme <u>can</u> shift the importance of (warm) rain production from autoconversion
141	process to the accretion process and therefore reduces the AIE (Posselt and Lohmann,
142	2009; Gettelman et al., 2015). However, Hill et al. (2015) shows that adding
143	prognostic rain scheme alone still cannot reduce the spread of susceptibility of
144	precipitation among different cloud microphysics parameterizations and further shows
145	that increasing the complexity of the rain representation to double-moment
146	significantly reduces the spread of precipitation sensitivity and improves overall
147	consistency between bulk and bin schemes.

Previous studies are mostly confined to global averages (e.g Quaas et al., 2009;
Wang et al., 2012) or a specific dynamic environment (e.g., Bretherton et al., 2007;
Guo et al., 2011). However, aerosols, clouds, precipitation distributions and

dynamical feedbacks are all related to the prevailing meteorological environment (Stevens and Feingold, 2009). Clouds are sensitive to changes in dynamical regimes, which can be defined by large-scale circulations, thermodynamic structure and meteorological backgrounds (Bony et al., 2004). Gryspeerdt and Stier (2012) and Gryspeerdt et al. (2014c) used satellite data and found that the characteristics of aerosol cloud-albedo effect (droplet number sensitivity) vary with cloud regimes and pointed out the importance of regime-based studies of aerosol-cloud interactions.

In this study, we investigate how AIE in several GCMs varies under different 158 dynamical regimes over global oceans (60°S-60°N), with a focus on cloud lifetime 159 effects of aerosols (2nd AIE). We note that the term "cloud lifetime effects" can be 160 somehow misleading, since aerosol effects on cloud liquid water may have little to do 161 with cloud lifetime per se (e.g., Small et al., 2009). Nevertheless, this term is still used 162 in some occasions in this paper for convenience. The paper is organized as follows. 163 Methods and models are described in Section 2, and results and discussions are 164 presented in Section 3. The paper concludes with the summary in Section 4. 165

166

2 Methodology and models

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The <u>response</u> of LWP to aerosol perturbations is defined as

$$\lambda = d \ln LWP / d \ln CCN$$

As simulated LWP and CCN can be quite different among GCMs, the logarithmic form of LWP and CCN is adopted in the λ formula. λ is a metric to quantitatively measure cloud lifetime effect of aerosols in models. It is directly calculated as the

relative change of monthly mean LWP from pre-industrial (PI) to present day (PD) 172 divided by the relative change of CCN. Here dlnLWP=(LWPPD-LWPPI)/LWPPI and 173 dlnCCN=(CCN_{PD}-CCN_{PI})/CCN_{PI}, where LWP_{PD} and LWP_{PI} are LWP in PD and PI, 174 respectively, while CCN_{PD} and CCN_{PI} are CCN in PD and PI, respectively. This 175 parameter was used by Wang et al. (2012) to constrain the cloud lifetime effects of 176 aerosols over global oceans using precipitation frequency susceptibility (S_{pop}) derived 177 from A-Train satellite observations. Lebo and Feingold (2014) examined the 178 relationship between λ and S_{pop} to aerosol perturbations for stratocumulus and 179 trade-wind cumulus simulated by LES and found that λ may increase in marine 180 stratocumulus while decrease in the case of trade-wind cumulus in response to 181 increasing S_{pop} , suggesting a cloud regime dependence of this relationship. Note that λ 182 allows some feedbacks, for example cloud effects on CCN. 183

Dynamical regimes can be defined by environment characteristics such as 184 large-scale vertical pressure velocity (e.g., Bony and Dufresne, 2005) and 185 lower-tropospheric stability (LTS, defined as the difference in potential temperature 186 between 700hPa and the surface, θ_{700hPa} - $\theta_{surface}$) (e.g., Medeiros and Stevens, 2011). 187 Medeiros and Stevens (2011) noted that low clouds and deep convective clouds could 188 be separated by ω_{500} while different low cloud types under large-scale subsidence can 189 only be depicted by using LTS. In this study the monthly-averaged vertical pressure 190 velocity (ω) in the mid-troposphere (defined as at 500hPa) is used as a proxy for 191 large-scale motions (Bony and Dufresne, 2005). Note that ω_{500} with positive (negative) 192 value means descending (ascending) motions. We decompose global (60°S~60°N) 193

194	large-scale circulations over ocean as a group of dynamical regimes (equally sampled)
195	by ω_{500} (and LTS). Ascending regimes and descending regimes are defined by ω_{500}
196	and descending regimes are further divided into stratocumulus, transitional clouds and
197	trade wind cumulus regimes by LTS. This method is straight-forward to apply to
198	GCM results and gives us a direct view of the relationship between clouds and their
199	favorable large-scale environmental characteristics. Note however that the use of
200	monthly means may obscure some details in the microphysical relationships,
201	especially where the variability of cloud properties is high.
202	Since vertical pressure velocity is used as a major criterion here, dynamic regimes
203	generally follow the features of vertical pressure velocity distributions. Descending
204	regimes are mostly located at subtropical regions and western coasts of continents,
205	while ascending regimes locates around ITCZ and northern Pacific where storm tracks
206	prevail. The seasonal evolution of dynamic regimes follows seasonal changes in the
207	major meteorological systems. For example, ascending regimes move north/south as
208	ITCZ move north/south and descending regimes move accompanying with subtropical
209	high move. The characteristics of dynamic and thermodynamic regimes were
210	discussed in detail in Bony et al. (2004).
211	As the perturbations in cloud radiative forcing from anthropogenic aerosols

As the perturbations in cloud radiative forcing from antiropogenic aerosois (indirect effect) are typically on the order of 1 W m⁻², which is small compared to the cloud radiative forcing (shortwave radiative effect of ~-47 W m⁻² and longwave radiative effect of ~27 W m⁻²) (Boucher et al., 2013), long integrations are required to produce statistically significant results. The Newtonian relaxation method (nudging)

216	provides a way to estimate AIE within a relatively short integration time, while giving
217	statistically significant results (Lohmann and Hoose, 2009; Kooperman et al., 2012).
218	Nudging here refers to the method of adding a forcing to the prognostic model
219	equations, determined by the difference between a model-computed value and a
220	prescribed value at the same time and model grid-cell, to constrain the model results
221	with prescribed atmospheric conditions. Kooperman et al. (2012) implemented
222	nudging to constrain PD and PI simulations toward identical meteorological fields and
223	found that the use of nudging provided a more stable estimate of AIE in shorter
224	simulations and increased the statistical significance of the anthropogenic aerosol
225	perturbation signal. All simulations used in this study were nudged toward reanalysis
226	winds (year 2006 to 2010) provided by operational forecast centers. Some simulations
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227	were further nudged toward reanalysis temperature, but this was discouraged because
227	were further nudged toward reanalysis temperature, but this was discouraged because
227 228	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al.,
227 228 229	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850
227 228 229 230	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850 and 2000 (Lamarque et al., 2010) and 5-year simulations were performed in each case
227 228 229 230 231	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850 and 2000 (Lamarque et al., 2010) and 5-year simulations were performed in each case (PI and PD). Sea surface temperature, sea-ice extent and greenhouse gas
227 228 229 230 231 232	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850 and 2000 (Lamarque et al., 2010) and 5-year simulations were performed in each case (PI and PD). Sea surface temperature, sea-ice extent and greenhouse gas concentrations are prescribed to climatological values in all simulations. Monthly data
227 228 229 230 231 232 233	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850 and 2000 (Lamarque et al., 2010) and 5-year simulations were performed in each case (PI and PD). Sea surface temperature, sea-ice extent and greenhouse gas concentrations are prescribed to climatological values in all simulations. Monthly data were then obtained by averaging over the 5-year integration period.
227 228 229 230 231 232 233 233	were further nudged toward reanalysis temperature, but this was discouraged because it might affect the moist convection activities simulated in the model (Zhang et al., 2014). All models were driven by the same IPCC <u>aerosol</u> emissions for years 1850 and 2000 (Lamarque et al., 2010) and 5-year simulations were performed in each case (PI and PD). Sea surface temperature, sea-ice extent and greenhouse gas concentrations are prescribed to climatological values in all simulations. Monthly data were then obtained by averaging over the 5-year integration period. Only ω_{500} in PD runs is used to derive dynamical regimes and then these

237 PI runs were nudged toward the reanalysis data here, which ensures ω_{500} is very 238 similar between PD and PI.

239 A total of ten aerosol-climate models participated in this study. This includes five versions of Community Atmosphere Model (CAM) 5.3, and two versions of 240 SPRINTARS. These models show large differences in their aerosol and cloud 241 treatments. For example, while most models (CAM5, CAM5-PNNL, CAM5-MG2, 242 CAM5-CLUBB, CAM5-CLUBB-MG2, ECHAM6-HAM2, and SPRINTARS-KK) 243 use the autoconversion scheme from Khairoutdinov and Kogan (2000, hereafter KK), 244 autoconversion rate in ModelE-TOMAS is independent of cloud droplet number 245 concentration and the Berry scheme (Berry, 1967) is used for SPRINTARS. Most 246 models use diagnostic rain schemes, while an updated Morrison and Gettelman 247 microphysics scheme with a prognostic rain scheme (MG2) (Gettelman et al., 2015) is 248 adopted in CAM5-MG2 and CAM5-MG2-CLUBB. HadGEM3-UKCA also adopts a 249 prognostic rain scheme (Abel and Boutle, 2012). While most models only account for 250 aerosol effects large-scale stratiform clouds. CAM5-CLUBB 251 on and CAM5-CLUBB-MG2 use a higher-order turbulence closure (CLUBB) to unify the 252 treatment of boundary layer turbulence, stratiform clouds and shallow convection, and 253 therefore include aerosol effects on shallow convection (Bogenschutz et al., 2013). A 254 brief description of each model is provided in the Appendix. 255

256 **3 Results**

257 **3.1 Annual mean**

258 We first examine the annual climatology in different simulations to get an overall

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picture of the general differences/similarities among these models (details within dynamic regimes are examined in section 3.2). All of the simulations reproduce the general pattern of large-scale circulations (ω_{500}): strong ascending motions within the inter-tropical convergence zone (ITCZ) and subsidence dominating subtropical eastern ocean regions (not shown). The similar patterns of ω_{500} (due to nudging) in these simulations <u>ensure</u> that dynamic regimes <u>defined by ω_{500} </u> do not vary much between models.

Table 1 lists the types of clouds included in LWP and rain analyzed in this study 266 and the different rain scheme (prognostic or diagnostic) in these 10 GCM simulations. 267 Table 2 lists global annual means of aerosol, precipitation and cloud parameters in PD 268 simulations and λ for each model. Note that all versions of CAM5 calculate LWP only 269 for large-scale clouds while SPRINTARS, SPRINTARS-KK and HadGEM3-UKCA 270 also count LWP from convective clouds. As for ModelE2-TOMAS, LWP includes 271 stratiform anvil clouds that formed from convective detrainment of water vapor and 272 ice. ECHAM6-HAM2 also includes the contribution of convective detrainment of 273 liquid water and ice to stratiform clouds Also note that CAM5 models with CLUBB 274 include LWP in the shallow convective regimes, which partly explains why these 275 models produce more LWP than their corresponding CAM5 models without CLUBB 276 (Table 2). 277

There are large differences among global LWP annual means. CAM5-MG2 has the lowest LWP among these simulations (30.0 g m⁻²). The LWP means over oceans are 31.1 g m⁻², 39.4 g m⁻² and 35.2 g m⁻² in CAM5, CAM5-PNNL and

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CAM5-CLUBB, respectively. HadGEM3-UKCA simulates higher LWP (57.1 g m⁻²) 281 than all versions of CAM5. LWPs in ModelE2-TOMAS (80.4 g m⁻²) and 282 ECHAM6-HAM2 (84.6 g m⁻²) are greater than the aforementioned GCMs, but less 283 than in SPRINTARS and SPRINTARS-KK (139.1 g m⁻² and 98.9 g m⁻² respectively) 284 which include LWP from convective clouds. Even though CAM5-CLUBB simulates a 285 higher LWP in storm track regions and ECHAM6-HAM2 produces much more LWP 286 associated with deep convection in the ITCZ, all models here display reasonable 287 patterns of global LWP distributions (not shown). 288

The differences in CCN (at 0.1% supersaturation) among these simulations are 289 not as large as the differences in LWP (Table 2). The global annual mean CCN in 290 CAM5-PNNL, which has a different treatment of wet scavenging processes (Wang et 291 al., 2013), is slightly larger than the one in other versions of CAM5. CCN 292 concentrations simulated by CAM5-PNNL, ECHAM6-HAM2 and ModelE2-TOMAS 293 are largest among these simulations and are more than twice those simulated by 294 SPRINTARS, SPRINTARS-KK and HadGEM3-UKCA, which are the lowest. Since 295 these models are using same emissions, differences of CCN between the models are 296 mainly due to different aerosol lifetime between models. 297

The LWP response to aerosol perturbations, λ , in ECHAM6-HAM2 (0.19) is close to those derived from three CAM5 configurations (0.20 in CAM5, 0.19 in CAM5-PNNL and 0.25 in CAM5-CLUBB). Notice that λ in CAM5-MG2 and CAM5-CLUBB-MG2 is larger than that in CAM5 and CAM5-CLUBB, respectively, which indicates that the changes of LWP in the models, using the MG2 scheme, are

303	more sensitive to the aerosol perturbations. LWP is much less sensitive to the changes
304	of CCN in SPRINTARS and SPRINTARS-KK with λ of 0.01 and 0.04 respectively. λ
305	is also small in HadGEM3-UKCA (0.03) due to the large relative increase of CCN
306	while small relative increase of LWP. Since the aerosol effect on precipitation
307	formation is turned off in ModelE2-TOMAS (its autoconversion parameterization is
308	not a function of $N_{\text{d}}),$ LWP barely responds to the increase of CCN (λ is -0.001). The
309	variation in λ closely follows that of the relative enhancement of LWP (dlnLWP), as
310	the variation of the relative enhancement of CCN (dlnCCN) among the simulations is
311	generally much smaller than that of dlnLWP.
312	We should note that large differences in CCN shown in Table 2 do not
313	necessarily correspond to equally large differences in droplet concentration (N _d), since
314	$N_{\underline{d}}$ is primarily dependent on cloud base updraft that is an extremely uncertain
315	parameter and may vary significantly between the GCMs. It therefore seems
316	reasonable to define λ as the change in LWP vs. the change in cloud droplet number
317	concentration ($N_{\underline{d}}$), which would provide a direct insight into how clouds response to
318	$N_{\underline{d}}$ change since LWP directly depends on $N_{\underline{d}}$, not necessarily on CCN. However, this
319	alternative definition of λ as dlnLWP/dlnN _d would be difficult to compare with
320	observations, and this also does not directly measure cloud response to anthropogenic
321	aerosols. The interactions between clouds and anthropogenic aerosols arise through a
322	chain of processes, from effects of the CCN on N _d to effects of N _d on cloud water,
323	which can be expressed as dlnLWP/dlnCCN=(dlnLWP/dlnN _d)*(dlnN _d /dlnCCN). This
324	chain of processes has now been examined in Ghan et al., (2016) based on the same
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325 set of model simulations documented in this study.

326 **3.2 Regime dependence**

327 a. LWP, CCN and λ

Figure 1 shows LWP and CCN as a function of vertical pressure velocity at 500 hPa (ω_{500}) derived from PD simulations. To derive Figure 1, the 12-month monthly global grid values are first sorted into 20 dynamical regimes according to their ω 500 values, keeping the number of samples in each bin equal. LWP, CCN and values of other fields for each bin are then calculated from averaging the values of all samples in that particular bin.

In general, SPRINTARS (default and KK) simulates much higher LWP in all 334 dynamic regimes and ECHAM6-HAM2/ModelE2-TOMAS in most regimes than 335 different versions of CAM5 runs (default, PNNL, CLUBB and MG2) (Figure 1a), 336 which is consistent with global means in Table 2. A peak of LWP is found around 337 $\omega_{500} = 0$ hPa/d in CAM5, ModelE2-TOMAS and ECHAM6-HAM2. For SPRINTARS, 338 LWP decreases from 190 g m $^{-2}$ to 100 g m $^{-2}$ as ω_{500} increases from -60 hPa/d to 40 339 340 hPa/d. In all simulations LWP is low in regimes where ω_{500} is larger than 10 hPa/d, i.e., regimes dominated by low clouds. HadGEM3-UKCA simulates larger LWP than 341 CAM5 especially in ascending regimes. The model spread of LWP response is larger 342 in the ascending regimes than in the subsiding regimes. This may be partly related to 343 the fact that the types of clouds included in LWP are not the same in different models 344 (Table 1). Figure 1b shows that CCN concentrations peak at around 25 hPa/d among 345

346	all the models. This peak is partly caused by little precipitation (and therefore low wet
347	scavenging rate) in subsidence regimes as well as by the fact that these dynamic
348	regimes are located near continents where the sources of anthropogenic aerosols are
349	strong. Furthermore, CCN concentrations are low at around 0 hPa/d, which could be
350	explained by the fact that most regimes around 0 hPa/d are located over the oceans far
351	away from continents (i.e. remote marine aerosols) and anthropogenic aerosol source
352	regions (figures not shown). Generally, CCN in two versions of SPRINTARS and
353	HadGEM3-UKCA is less than other models in most regimes, consistent with Table 2.
354	All the simulations show positive λ within all dynamical regimes (Figure 2a),
355	which is consistent with the theory proposed by Albrecht (1989) that an increase in
356	aerosols leads to more liquid cloud water. However, λ can vary significantly between
357	regimes in CAM5 and ECHAM6-HAM2 (Figure 2a), which indicates that changes in
358	LWP in response to aerosol perturbations are regime-dependent in these GCMs. For
359	example, λ in CAM5-PNNL ranges from 0.35 in strong ascending regions to 0.11 in
360	strong subsidence regions, which means that LWP in strong ascending regimes is
361	more sensitive to aerosol perturbations than in strong subsidence regimes. Exceptions
362	are ModelE2-TOMAS, SPRINTARS (default and SPRINTARS-KK) and
363	HadGEM3-UKCA, in which $\boldsymbol{\lambda}$ is low in magnitude.(i.e., LWP changes little in
364	response to the changes of CCN, consistent with the global annual means shown in
365	Table 2).

366 We note that although the global means of λ in all CAM5 configurations and 367 ECHAM6-HAM2 are close, from 0.19 in ECHAM6-HAM2 to 0.25 in

CAM5-CLUBB, λ in the different dynamical regimes can differ significantly among 368 these simulations (Figure 2). For example, LWP in CAM5-PNNL is much more 369 sensitive to CCN perturbations than in ECHAM6-HAM2 in strong ascending regimes; 370 and in strong subsidence regimes, LWP in CAM5-CLUBB and ECHAM6-HAM2 is 371 more sensitive than in CAM5-PNNL and CAM5. Models that use the MG2 with 372 prognostic rain scheme (i.e. CAM5-MG2 and CAM5-CLUBB-MG2) simulate larger 373 λ than the models that use the default MG scheme in most regimes, only except for 374 strong subsidence regimes. However, generally the shapes of the λ distribution are 375 very similar. λ in CAM5-CLUBB-MG2 is large in both ascending and subsidence 376 regimes, which explains the largest global λ in CAM5-CLUBB-MG2 among all 377 configurations (Table 2). Except for the models producing very low values of λ 378 (SPRINTARS, SPRINTARS-KK, ModelE2-TOMAS and HadGEM3-UKCA), λ from 379 the other models converges around 0 hPa/d and then diverges greatly in strong 380 381 ascending regimes (from 0.10 to 0.46) and, to a less extent, in strong subsidence regimes. This indicates that it is in regimes with weak vertical velocity where models 382 agree most, while it is in strong ascending and descending regimes where models 383 differ most. The diversity of λ within dynamical regimes in different GCMs highlights 384 the need to distinguish different dynamical regimes in studying AIE. 385

When analyzing the numerator and denominator of λ separately, we found that this large spread in λ is mainly contributed by the numerator, dlnLWP. dlnLWP ranges from about 0 to 0.22 among the models (Figure 2a) while the denominator dlnCCN, is more stable than dlnLWP within dynamical regimes and fluctuates around 390 0.45, except for larger dlnCCN in HadGEM3-UKCA (Figure 2b). In summary, the 391 ratio of dlnLWP to dlnCCN (λ) therefore changes more consistently with dlnLWP 392 within dynamical regimes.

The decreasing trends of λ with increasing ω in CAM5, CAM5-MG2, and 393 CAM5-PNNL are similar, which is opposite to the increasing trends derived from 394 ECHAM6-HAM2, CAM5-CLUBB and CAM5-CLUBB-MG2. It is interesting that 395 the regime-dependence of λ simulated by CAM5-CLUBB and CAM5-CLUBB-MG2 396 is quite different from that simulated by CAM5, CAM5-MG2, and CAM5-PNNL 397 even though all these 5 model versions are originally from CAM5 and share many 398 CAM5, CAM5-MG2 and CAM5-PNNL, 399 similarities. In three separate parameterization schemes are used to treat planetary boundary layer (PBL) turbulence, 400 stratiform cloud macrophysics and shallow convection. In CAM5-CLUBB and 401 CAM5-CLUBB-MG2, instead, a higher-order turbulence closure, Cloud Layers 402 Unified by Binormals (CLUBB), is adopted to replace these three separate schemes to 403 provide a unified treatment of these processes (Bogenschutz et al., 2013). A major 404 improvement of CAM-CLUBB is the better simulation of the transition of 405 stratocumulus to trade wind cumulus over subtropical oceans (Bogenschutz et al., 406 2013). Fig. 2a shows that λ in CAM5-CLUBB and CAM5-CLUBB-MG2 is quite 407 different from that in CAM5 simulations without CLUBB (i.e., CAM5, CAM5-MG2 408 and CAM5-PNNL) in regimes where ω_{500} is larger than 10 hPa/d. Under such 409 suppressed conditions, low clouds such as trade wind cumulus and stratocumulus are 410 typically formed. This higher λ might be expected because CAM5-CLUBB 411

formulations apply the MG microphysics (and effects of aerosols on cloud microphysics) to shallow convective regimes. The better representation of low clouds in CAM5-CLUBB, and the representation of double-moment microphysics and AIE in shallow convective regimes from the unified parameterization may help to explain the different behaviors between CAM5 runs with CLUBB (CAM5-CLUBB and CAM5-CLUBB-MG2) and CAM5 runs without CLUBB (CAM5, CAM5-MG2 and CAM5-PNNL) in subsidence regimes._

In order to find out the crucial geographic locations of dynamic regimes where 419 dlnLWP differs most in Fig. 2b, we plot the global distribution of annual averaged 420 dlnLWP in different simulations, shown in Fig. 3. The ascending regimes where 421 ECHAM6-HAM2 differs significantly from the two CAM5 configurations (CAM5, 422 CAM5-PNNL) are located over the North Pacific Ocean (from 30°N to 60°N), for 423 weak ascending motions and the Southern coast of Asia for strong ascending motions. 424 The spatial patterns in ECHAM6-HAM2, CAM5-CLUBB, CAM5-CLUBB-MG2 and 425 HadGEM3-UKCA share some similarities over Northern Pacific Ocean, but the 426 magnitude in CAM5-CLUBB and CAM5-CLUBB-MG2 is larger than in 427 ECHAM6-HAM2 and HadGEM3-UKCA. Moreover, not only the spatial pattern but 428 also the magnitude of dlnLWP in ECHAM6-HAM2 differ significantly from those in 429 CAM5, CAM5-MG2 and CAM5-PNNL. For the Southern coast of Asia where strong 430 ascending motions dominate, all simulations show a relative increase of LWP. 431 However, dlnLWP in ECHAM6-HAM2 in this region is much smaller than in all 432 433 CAM5 simulations. This makes dlnLWP, and thus λ , in ECHAM6-HAM2 much less

434 than ir	n the five CAM5	models (CAM5	, CAM5-MG2,	, CAM5-PNNL,	, CAM5-CLUBB
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and CAM5-CLUBB-MG2) in ascending regimes, as shown in Fig. 2b and Fig. 2a.

436 Despite the fact that SPRINTARS (default and KK), ModelE2-TOMAS and HadGEM3-UKCA all show almost no relative change of LWP in response to aerosol 437 perturbations, the spatial patterns of dlnLWP in these four simulations shown in Fig. 3 438 are indeed different from each other. HadGEM3-UKCA simulates larger dlnLWP in 439 middle northern subtropical oceans, which is similar to CAM5-CLUBB and 440 ECHAM6-HAM2 but with smaller magnitude. However, the pattern in SPRINTARS 441 is unlike any models discussed above. SPRINTARS simulates larger dlnLWP over the 442 North Pacific Ocean, the North Atlantic Ocean and the western coasts of continents 443 than other parts of the global ocean. SPRINTARS-KK simulates the same pattern as 444 SPRINTARS only with larger values. Meanwhile, dlnLWP in ModelE2-TOMAS 445 shows no special global pattern and the values are all near zero, which indicates LWP 446 in ModelE2-TOMAS has indeed little response to aerosol perturbations as 447 autoconversion rate in ModelE2-TOMAS is not influenced by cloud droplet number 448 concentrations. 449

Figure 3 shows that the differences in subsidence regimes in Fig. 2b are mainly contributed by middle northern subtropical oceans and western coasts of continents. In middle northern subtropical oceans, the relative changes of LWP in ECHAM6-HAM2, HadGEM3-UKCA and the two CAM5 models with CLUBB (CAM5-CLUBB and CAM5-CLUBB-MG2) are much more sensitive to the aerosol perturbations than in the three CAM5 models without CLUBB (CAM5,

CAM5-PNNL and CAM5-MG2), even though dlnLWP in ECHAM6-HAM2 and 456 HadGEM3-UKCA is not as large as that in CAM5-CLUBB and 457 CAM5-CLUBB-MG2. Another difference among these models is in regions 458 dominated by more intensive subsidence, over Western coasts of North America. 459 South America and Africa. In these regions dlnLWP in ECHAM6-HAM2 and the two 460 461 CAM5 models with CLUBB is large while it is small in the three CAM5 models without CLUBB. 462

To examine the cloud lifetime effect in different cloud regimes more specifically, 463 another criterion, lower-tropospheric stability (LTS= θ_{700hPa} - $\theta_{surface}$), is added to 464 distinguish stratocumulus from trade wind cumulus regimes, following Medeiros and 465 Stevens (2011). Table 3 lists the criteria of different low cloud types conditionally 466 sampled by ω_{so} and LTS. The annual mean cloud fractions of each low cloud type in 467 CAM5-CLUBB are shown in Fig. 4; the distributions in other simulations are 468 generally similar to CAM5-CLUBB (figures not shown). The cloud type distribution 469 is consistent with satellite observations that stratocumuli occurs over subtropical 470 oceans near western continents while trade wind cumuli dominate over oceans further 471 away from continents (Medeiros and Stevens, 2011). Fig. 4 shows that some 472 differences in dlnLWP between models shown in Fig. 3 are located at regions 473 dominated by low clouds (i.e., stratocumulus and trade wind cumulus). 474

The joint distributions of LTS and ω_{500} over global oceans between 60°S and 60°N derived from the models are shown in Fig. 5. Note that the bins here are not equally sampled as in previous figures but divided into equal LTS and ω intervals. 478 LTS ranges from 8K to 24K while ω ranges from -100 hPa/d to 60 hPa/d. Instances
479 with slight downward vertical motions and moderate LTS are most frequent.

480 Fig. 5 shows that, though ω_{500} plays the primary role in determining the dlnLWP/dlnCCN distribution, LTS can reveal further details of the differences among 481 various low cloud types in subsidence regimes. The large λ in strong subsidence 482 regimes in ECHAM6-HAM2 and CAM5-CLUBB is mainly caused by stratocumulus 483 and trade wind cumulus. As for regions of ascending motions, LTS is confined 484 between 12K and 14K. λ in CAM5, CAM5-PNNL and CAM5-CLUBB in ascending 485 regimes is larger than in regimes with weak large-scale vertical velocity (ω_{500} around 486 487 0 hPa/d) and larger than in ECHAM6-HAM2 in ascending regimes. In ascending regimes, LWP is more sensitive to the change of CCN in the two CAM5 models with 488 the MG2 scheme (CAM5-MG2 and CAM5-CLUBB-MG2) than in the two 489 corresponding CAM5 models without the MG2 scheme (CAM5 and CAM5-CLUBB), 490 which is consistent with Fig. 2a. In CAM5-CLUBB-MG2, λ is larger in transitional 491 cloud regimes than in stratocumulus cloud regimes and trade wind cloud regimes, 492 which is evidently different from the low cloud regimes in CAM5-CLUBB. 493 HadGEM3-UKCA simulates higher LWP response in transitional clouds and 494 stratocumulus regimes than trade wind cloud regime. It is also interesting to note that 495 λ in SPRINTARS and SPRINTARS-KK shows stronger dependence on LTS than on 496 497 ω₅₀₀.

b. Microphysics process rates and precipitation

The balance between autoconversion and accretion is found to be critical in 23

500	determining cloud lifetime effect in climate models (Posselt and Lohmann, 2009;
501	Wang et al., 2012). Autoconversion rate is sensitive to cloud droplet concentration
502	while accretion has little dependence of droplet number. If the role of accretion
503	dominates over autoconversion (with all other effects equal), the effect of aerosols on
504	clouds is expected to be weakened in GCMs (Posselt and Lohmann, 2009; Gettelman
505	et al 2013). Wang et al. (2012) found that the cloud lifetime effect is highly correlated
506	with the ratio of autoconversion rate to large-scale surface precipitation rate
507	(AUTO/PRECL, where PRECL also includes ice and snow) over global oceans in
508	climate models. AUTO/PRECL for different dynamical regimes is shown in Fig. 6a.
509	Here <u>PD</u> monthly-averaged autoconversion rate and surface precipitation rate are used
510	in calculating AUTO/PRECL. Generally the curves of AUTO/PRECL are smoother
511	than λ (Fig. 6a and Fig. 2a). The ratio from different simulations shows large diversity
512	in ascending regimes and subsidence regimes. In all versions of CAM5 and
513	SPRINTARS the ratio decreases with increasing ω_{500} in ascending regimes and then
514	increases in descending regimes. The ratio is especially large in CAM5-CLUBB-MG2
515	and HadGEM3-UKCA in descending regimes. However, the ratio in
516	ECHAM6-HAM2 remains unchanged in ascending regimes and then increases under
517	subsidence. As discussed above, λ was shown to be highly correlated with this ratio
518	from global average results (Wang et al., 2012). According to our results, the
519	correlation also applies well for individual dynamical regimes in ECHAM6-HAM2.
520	HadGEM3-UKCA and CAM-CLUBB, in which the correlation coefficients between
521	λ and AUTO/PRECL are 0.98, 0.92 and 0.86 respectively. However, these high

522 correlation coefficients are not found in other simulations, in which the correlation 523 coefficients are lower than 0.7, which indicates that the relationship of AUTO/PRECL 524 and λ in these models is changing from regime to regime (i.e., this relationship is 525 regime-dependent).

Wang et al. (2012) and Gettelman et al. (2013) found that the diagnostic rain 526 scheme used in the CAM configurations might overestimate the role of 527 autoconversion over accretion. Using instantaneous microphysical process rates, 528 Gettelman et al. (2015) found that adding the new microphysics with prognostic 529 precipitation to cloud scheme (MG2) decreases the ratio of autoconversion to 530 accretion. It is in moderate regimes (-20 hPa/d $\leq \omega_{500} \leq 10$ hPa/d) where the result is 531 consistent with Gettelman et al. (2015), which shows larger AUTO/PRECL in CAM5 532 than CAM5-MG2. However, in other regimes of CAM5 and all regimes of 533 CAM5-CLUBB, adding the prognostic precipitation (MG2) increases the ratio of 534 AUTO/PRECL. The result of larger AUTO/PRECL in some regimes from models 535 with MG2 seems different from the results of Gettelman and Morrison (2015) in 536 idealized tests of MG2 and of Gettelman et al. (2015) in CAM simulations with MG2. 537 We have verified using the same model output from Gettelman et al. (2015) that the 538 difference is not due to the simulations performed. The difference is likely due to: (a) 539 the use of instantaneous output in Gettelman et al. (2015) for process rate 540 comparisons while monthly data is used here; (b) Microphysics variables and 541 precipitation are sorted by ω_{500} here while Gettelman et al. (2015) sorted them by 542 LWP that the microphysics sees, which includes contributions from deep convection; 543

(c) Vertical integrals of autoconversion rate are used here while <u>vertical mean values</u>
are used in Gettelman et al. (2015).

As discussed in Section 1, precipitation is a key process in interactions between 546 aerosols and clouds. A decrease in surface precipitation increases cloud water while a 547 decrease in cloud-top sedimentation increases the entrainment rate and thus dries out 548 LWP when the free troposphere air is dry (Ackerman et al., 2004). Here we 549 investigate the LWP response to aerosol perturbations under low precipitation 550 (monthly-averaged surface precipitation rate less than $0.1 \text{ mm } d^{-1}$) and high 551 precipitation (monthly-averaged surface precipitation rate larger than 0.1 mm d⁻¹). 552 Table 4 lists the occurrence frequency of each situation in different simulations. It 553 shows that instances with low PRECL occurs much less often (from 2.2% in 554 CAM5-CLUBB to 38.8% in CAM5-MG2) than those with high PRECL. The 555 occurrence frequency of low precipitation situations is increased with the MG2 556 scheme (CAM5-MG2 and CAM5-CLUBB-MG2), compared with simulations without 557 MG2. This increase is especially evident in CAM5-CLUBB (from 0.02 in 558 CAM5-CLUBB to 0.16 in CAM5-CLUBB-MG2). This is consistent with Gettelman 559 et al. (2015), who showed surface precipitation decreases slightly in GCMs with 560 MG2. 561

Note that low precipitation situations are only found in subsidence regimes ($\omega_{500} > 0$ hPa/d). Thus, the sensitivity of the LWP response to aerosol change under low and high precipitation is compared only in subsidence regimes. Table 4 also shows λ and the fractional occurrences of each precipitation situation in descending regimes. The

566	fractional occurrence of low precipitation increases evidently in subsidence regimes,
567	compared with that over global ocean. We find that the averages of $\boldsymbol{\lambda}$ under low
568	precipitation are larger than those under high precipitation in most models (CAM5,
569	CAM5-PNNL, CAM5-CLUBB, <u>ECHAM6-HAM2,</u> SPRINTARS, SPRINTARS-KK
570	and HadGEM3-UKCA) (Table4). This result is different from some LES and single
571	column model (SCM) results showing that smaller λ values are found for low surface
572	precipitation rather than high precipitation due to a decrease of LWP in response to
573	increasing CCN (Ackerman et al., 2004; Guo et al., 2011). The decrease in LWP in
574	these previous studies is found to come from the entrainment drying due to increased
575	entrainment from increasing aerosol loading (e.g., Bretherton et al., 2007) and this
576	effect has not been explicitly included in most GCMs. Exceptions are CAM5 runs
577	with the prognostic precipitation scheme MG2 (CAM5-MG2, CAM5-CLUBB-MG2).
578	It can be seen from Table 4 that λ under low surface precipitation is smaller than
579	under high precipitation only when MG2 scheme is used. It is still unclear what might
580	cause this difference. It is interesting to note that λ under low surface precipitation is
581	still higher for HadGEM3-UKCA though a prognostic precipitation scheme is applied
582	in HadGEM3-UKCA.

583 c. Shortwave cloud radiative effect

The shortwave cloud radiative effect (SCRE) is defined as the difference between all-sky and clear sky shortwave radiative fluxes at the top of atmosphere. <u>Here SCRE</u> is adjusted to the "clean-sky" SCRE, which is estimated as a diagnostic with aerosol optical depth set to zero (Ghan, 2013). Recent studies on aerosol indirect effects mostly focus on stratocumulus clouds due to their significant cooling effect (e.g., Lu
and Seinfeld, 2005; Bretherton et al., 2007). However, by sorting the change of SCRE
(dSCRE) from PI to PD into dynamical regimes, our results suggest that the regimes
of ascending motions are as important as the <u>subsidence</u> regimes and in some
simulations dSCRE in ascending regimes is even larger than under <u>subsidence</u>
regimes (e.g., CAM5-PNNL) (Fig. 7). This suggests that ascending regimes are
crucial regimes in studying aerosol climate effect.

We also examined dSCRE contributed by low and high precipitation situations 595 (note that the total dSCRE is the sum of dSCRE under low and high precipitation 596 situation). It is found that high precipitation situations constitute most of dSCRE 597 (from 64% in CAM5-MG2 to nearly 100% in CAM5-CLUBB, Fig. 7) and the 598 contributions from clouds with low precipitation rates are generally small, ranging 599 from 0% to 36%, due to their low occurrence frequency. dSCRE is reduced by 33% 600 for high precipitation situations from CAM5 to CAM5-MG2, and 15% from 601 CAM5-CLUBB to CAM5-CLUBB-MG2 (Fig. 7), consistent with the argument that 602 prognostic precipitation schemes reduce aerosol indirect forcing (Posselt and 603 Lohmann, 2009; Wang et al., 2012; Gettelman and Morrison, 2015). However, 604 adopting a prognostic precipitation scheme is found to increase dSCRE under low 605 precipitation situations. This is partly from the increase in the occurrence frequency of 606 low precipitation instances when MG2 is adopted (Table 4). 607

608Our sensitive tests indicate that results in Table 4 and Figure 7 can be potentially609sensitive to the precipitation threshold applied to separate high precipitation and low

610	precipitation situations (not shown). The occurrence frequency of low precipitation
611	situations increases with increasing threshold and the magnitude of increase can be
612	different for different models. For example, when the precipitation threshold increases
613	from 0.01 mm d ⁻¹ to 0.20 mm d ⁻¹ , the occurrence frequency of low precipitation
614	situations increases from 2% to 37% in CAM5-PNNL while it increases from near 0%
615	to 5% in CAM5-CLUBB. Increasing the precipitation threshold also increases the
616	contribution of low precipitation situations to the total aerosol indirect forcing as the
617	occurrence frequency of low precipitation situations increases. However, our results
618	indicate that the LWP response to aerosol perturbations under low and high
619	precipitation does not change much as the precipitation threshold changes and that
620	high precipitation situations generally contribute more to the total aerosol indirect
621	forcing for precipitation threshold in the range of 0.01 to 0.20 mm d ⁻¹ . More work is
622	needed to explore this further such as how results may be different when
623	instantaneous precipitation data (e.g., 3-houly data) is used.

624 **4 Summary**

We have examined the regime-dependence of aerosol indirect effects (AIE) over 625 global oceans (from 60° S to 60° N) in several GCMs (CAM5, CAM5-MG2, 626 CAM5-PNNL, CAM5-CLUBB, CAM5-CLUBB-MG2, ECHAM6-HAM2, 627 SPRINTARS, SPRINTARS-KK, ModelE2-TOMAS and HadGEM3-UKCA). Model 628 results are sorted into different dynamical regimes, characterized by the 629 monthly-mean mid-tropospheric 500hPa vertical pressure velocity (ω_{500}), 630 lower-tropospheric stability (LTS, θ_{700hPa} - $\theta_{surface}$) and surface precipitation rate. 631

632	The <u>response</u> of liquid water path (LWP) to aerosol perturbations,
633	λ =dlnLWP/dlnCCN, a metric to quantify cloud lifetime effect of aerosols (Wang et al.,
634	2012), shows a large spread within dynamical regimes among GCMs, although the
635	global means are close. This diversity indicates that the aerosol cloud lifetime effect is
636	regime-dependent. It is in strong ascending regimes and subsidence regimes that $\boldsymbol{\lambda}$
637	differs most between GCMs (Fig. 2a). Stratocumulus regimes have traditionally been
638	the focus for studying aerosol indirect effects because of their significant cooling
639	effect in climate system (e.g., Ackerman et al., 2004; Bretherton et al., 2007;
640	Gettelman et al., 2013). However, our results highlight that regimes with strong
641	large-scale ascent should be another important regime to focus on in the future. Our
642	results indicate that aerosol indirect forcing in regimes of vertical ascent is close to, or
643	even larger than that in low cloud regimes (Fig. 7). Note however that these GCMs do
644	not treat aerosol effects in their representations of deep convection that dominates
645	clouds and LWP in regimes with strong ascent, while new versions of CAM exist
646	where a version of the MG microphysics has been embedded in the deep convective
647	parameterization (Song and Zhang, 2011).
648	By adding LTS as another criterion, we further separated different low cloud

649 types under large-scale subsidence and revealed some further differences in cloud 650 lifetime effect of aerosols on different types of low clouds. For example, the large λ in 651 subsidence regimes in CAM5-CLUBB and ECHAM6-HAM2 comes from both 652 stratocumulus and trade wind cumulus, while in CAM5-CLUBB-MG2 it mostly 653 comes from trade wind cumulus (Fig. 5). It is also interesting to note that the distribution of λ in SPRINTARS and SPRINTATSKK is more likely to depend on
LTS rather than vertical pressure velocity.

Precipitation is another important factor in understanding simulated aerosol 656 indirect forcing and its spread across models. LWP is more sensitive to CCN change 657 under low precipitation situations (monthly-mean surface precipitation rate less than 658 0.1 mm d^{-1}) than under high precipitation situations (monthly-mean surface 659 precipitation rate larger than 0.1 mm d⁻¹) in all models except for CAM5 simulations 660 with prognostic rain scheme (MG2) (Table 4). Results derived from large eddy 661 simulation (LES) and single column model (SCM) (e.g., Ackerman et al., 2004; Guo 662 et al., 2011) have shown that λ could be negative under low precipitation situations, 663 which indicates that λ is expected to be smaller under low precipitation situations. 664 Further efforts are needed to understand the differences among different models and 665 the difference between global model results and results from process-level studies. 666 Our results indicate that grids with high precipitation contribute most to aerosol 667 indirect forcing (from 64% in CAM5-MG2 to nearly 100% in CAM5-CLUBB, Fig. 7) 668 and the contributions from model grids with low precipitation are relatively small, 669 ranging from 0% to 36%. Adding prognostic precipitation scheme (MG2) reduces the 670 shortwave cloud radiative effect (SCRE) for high precipitation situations. As low 671 precipitation situations are much less prevalent than high precipitation situations, total 672 673 SCRE decreases in models with prognostic rain scheme compared to those with a

674 diagnostic rain scheme.

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The regime categorization used in this study is derived from monthly mean 31

data. Giving the high variability of precipitation and microphysics processes on short 676 time scales, we acknowledge that instantaneous data (e.g. 3 hourly) might provide 677 more reliable information. For example, instantaneous data may help to reconcile 678 some of discrepancies between our studies and that of Gettelman et al. (2015) 679 regarding the prognostic rain scheme noted in Section 3.2b. However, it is challenging 680 to calculate λ and aerosol indirect forcing using instantaneous data. Here λ and aerosol 681 indirect forcing are derived from the difference between present day (PD) and 682 pre-industrial (PI) simulations. Using instantaneous data will not guarantee that the 683 sorted bins of dynamical regimes include the same instances from PI to PD, giving the 684 high variability of instantaneous data. Since the main goal in this manuscript is to 685 demonstrate the importance of examining aerosol indirect effects in different cloud 686 and dynamical regimes, the use of monthly-mean data serves this goal well. It is our 687 future plan to carry in-depth analysis to further understand some of the findings 688 documented here, such as the large spread in λ in regimes of vertical ascent in 689 different models. For example, LWP response to aerosol perturbation documented in 690 this study may include contributions from mixed-phase and ice clouds. In- depth 691 analysis of cloud macrophysics and microphysics processes will help to improve the 692 understanding of the model uncertainty. 693

694 Appendix. Global aerosol-climate models

695 CAM5: This is the default version of CAM5.3. The moist turbulence scheme is 696 based on Bretherton and Park (2009), which explicitly simulates stratus-697 radiation-turbulence interactions. The shallow convection scheme is from Park and

Bretherton (2009) and the deep convection parameterization is retained from CAM4.0 698 (Neale et al., 2008). The two-moment cloud microphysics scheme from Morrison and 699 Gettelman (2008) (MG) is used to predict both the mass and number mixing ratios for 700 cloud water and cloud ice with a diagnostic formula for rain and snow. The cloud ice 701 microphysics was further modified to allow ice supersaturation and aerosol effects on 702 ice clouds (Gettelman et al., 2010). The activation of aerosol particles into cloud 703 droplets is parameterized by Abdul-Razzak and Ghan (2000, hereafter ARG) and the 704 autoconversion scheme is based on Khairoutdinov and Kogan (2000) (KK). A modal 705 approach is used to treat aerosols in CAM5 (Liu et al., 2012; Ghan et al., 2012). 706 Aerosol size distribution can be represented by using either 3 modes or 7 modes, and 707 the default 3-mode treatment is used in this study. Simulations were performed at 1.9° 708 $\times 2.5^{\circ}$ horizontal resolution with finite volume dynamical core, using 30 vertical 709 710 levels.

CAM5-PNNL: This is the same as CAM5, but a new unified treatment of vertical 711 transport and in-cloud wet removal processes in convective clouds developed by 712 Wang et al. (2013) is applied. It has a more detailed treatment of aerosol activation in 713 convective updrafts and a mechanism is added for laterally entrained aerosols to be 714 activated and then removed. In addition, a few other changes have been introduced to 715 stratiform cloud wet scavenging processes in CAM5-PNNL to improve the fidelity of 716 717 the aerosol simulation, including the vertical distribution of aerosols and their transport to remote regions (Wang et al., 2013). 718

CAM5-MG2: This is the same as CAM5, but the original two-moment MG scheme with diagnostic treatment for rain and snow in CAM5 is replaced by the updated MG scheme (MG2) with prognostic scheme for rain and snow (Gettelman et al., 2015).

CAM5-CLUBB: This is the same as CAM5, but the separate treatments of boundary layer turbulence, large-scale cloud macrophysics and shallow convection in CAM5 is replaced by CLUBB, a higher-order turbulence closure that unifies these different treatments (Bogenschutz et al., 2013). This therefore includes aerosol effects on shallow convection.

CAM5-CLUBB-MG2: This is the same as CAM5-CLUBB, but the MG2 scheme
with prognostic rain and snow treatment replaces the original MG scheme with
diagnostic rain and snow treatment (Gettelman et al., 2015). This also includes aerosol
effects on shallow convection.

ECHAM6-HAM2: ECHAM-HAMMOZ (echam6.1-ham2.2-moz0.9) is a global 732 aerosol-chemistry climate model. In this study only the global aerosol-climate model 733 part of ECHAM-HAMMOZ is used and for the sake of brevity referred to as 734 735 ECHAM6-HAM2 (Neubauer et al., 2014). It consists of the general circulation model ECHAM6 (Stevens et al., 2013) coupled to the latest version of the aerosol module 736 HAM2 (Stier et al., 2005; Zhang et al., 2012) and uses a two-moment cloud 737 microphysics scheme that includes prognostic equations for the cloud droplet and ice 738 crystal number concentrations as well as cloud water and cloud ice (Lohmann et al., 739 2007; Lohmann and Hoose, 2009). The activation of aerosol articles into cloud 740 34

droplets is parameterized by Lin and Leaitch (1997) and the autoconversion scheme is based on the KK scheme. Cumulus convection is represented by the parameterization of Tiedtke (1989) with modifications by Nordeng (1994) for deep convection. Aerosol effects on convective clouds are not included, but there is a dependence of cloud droplets detrained from convective clouds on aerosol. Simulations were performed at T63 $(1.9^{\circ} \times 1.9^{\circ})$ spectral resolution using 31 vertical levels (L31).

SPRINTARS: SPRINTARS (Takemura et al. 2005) is a global aerosol 747 transport-climate model based on a general circulation model, MIROC (Watanabe et 748 al. 2010). In this study, the horizontal and vertical resolutions are T106 (1.125° x 749 approx. 1.125°) and 56 layers, respectively. SPRINTARS is coupled with the radiation 750 and cloud microphysics schemes in MIROC to calculate the aerosol-radiation and 751 aerosol-cloud interactions. A prognostic scheme for determining the cloud droplet and 752 ice crystal number concentrations is introduced (Takemura et al. 2009). The default 753 atuoconversion scheme in MIROC-SPRINTARS is based on Berry (1967), and the 754 activation of aerosol particles into cloud droplet is based on the ARG scheme. 755

756 SPRINTARS-KK: This is the same as SPRINTARS, but the default 757 autoconversion scheme in SPRINTARS is replaced with the KK auconversion 758 scheme.

ModelE2-TOMAS: ModelE2-TOMAS is a global-scale atmospheric chemistry-climate model, which consists of the state-of-the-art NASA GISS ModelE2 general circulation model (Schmidt, 2014) coupled to the TwO-Moment Aerosol Sectional (TOMAS) microphysics model (Lee and Adams, 2012; Lee et al., 2015).

ModelE2-TOMAS has 2° latitude by 2.5° longitude resolution, with 40 vertical hybrid 763 sigma layers from the surface to 0.1 hPa (80 km). In the model, clouds are 764 distinguished into convective and large-scale stratiform clouds. The clouds 765 parameterizations are similar to Del Genio (1993) and Del Genio (1996) but have 766 been improved in several respects (see details in Schmidt, 2014; Schmidt, 2006). 767 Using a prognostic treatment of cloud droplet number concentration (CDNC) from 768 Morrison and Gettleman (2008), ModelE2-TOMAS represents the first aerosol 769 indirect effects only on large-scale stratiform clouds (Menon et al., 2010). In 770 ModelE2-TOMAS, CDNC and a critical supersaturation are computed using a 771 physical-based activation parameterization from Nenes and Seinfeld (2002) with a 772 model updraft velocity that is computed based on a large-scale vertical velocity and 773 sub-grid velocity. 774

HadGEM3-UKCA: HadGEM3-UKCA is a global composition climate model 775 (http://www.ukca.ac.uk). It consists of the third generation of the Hadley Centre 776 Global Environmental Model (Hewitt et al, 2011) developed at the UK Met Office. 777 778 This general circulation model is non-hydrostatic and uses a semi-Lagrangian transport scheme. We are using the atmospheric configuration: General Atmosphere 779 (GA) 4.0 as documented in Walters et al., (2014), except for the addition of the 780 UKCA aerosol and chemistry scheme which is fully coupled with the radiation 781 scheme of HadGEM3 (Bellouin et al., 2013). UKCA is a two-moment pseudo-modal 782 scheme which carries both aerosol number concentration and component mass as 783 prognostic tracers. It calculates the evolution of five aerosol species, sulfate, 784

particulate organic matter, black carbon, sea salt and dust, in both internally and 785 externally mixed particles. The aerosol scheme in UKCA is based on the Global 786 Model of Aerosol Processes (GLOMAP-mode, Mann et al., 2010). The main 787 exception is that dust is calculated separately using 6 size bins. UKCA hence only 788 considers 5 modes. The tropospheric chemistry part of UKCA is described in 789 790 O'Connor et al. (2014). HadGEM3 uses a prognostic treatment of rain formulation (Abel and Boutle, 2012) and employs a prognostic cloud fraction and condensation 791 cloud scheme (PC2) (Wilson et al., 2008), in which the cloud droplet number 792 concentration is diagnosed from the expected number of aerosols that are available to 793 activate at each timestep (West et al., 2014). Cumulus convection is represented by a 794 mass flux convection scheme based on Gregory and Rowntree (1990) with various 795 extensions (Walters et al., 2014). Simulations were performed at N96L85 resolution, 796 a regular 1.25° latitude \times 1.875° longitude grid in the horizontal, with 85 797 hybrid-height vertical levels. 798

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References

830 831 832	Abel, S. J. and Boutle, I. A.: An improved representation of the raindrop size distribution for single-moment microphysics schemes, Q. J. Roy. Meteorol. Soc., 138, 2151–2162, doi:10.1002/qj.1949, 2012.
833 834	Abdul-Razzak, H. and Ghan, S. J.: A parameterization of aerosol activation 2. Multiple aerosol types, J. Geophys. Res., 105, 6837–6844, 2000.
835 836 837	Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, Nature, 432, 1014-1017, 10.1038/nature03174, 2004.
838 839	Albrecht, B. A.: AEROSOLS, CLOUD MICROPHYSICS, AND FRACTIONAL CLOUDINESS, Science, 245, 1227-1230, 10.1126/science.245.4923.1227, 1989.
840 841 842	Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M., and Silva-Dias, M. A. F. :Smoking rain clouds over the Amazon. Science, 303, 1337– 1342, 10.1126/science.1092779,2004.
843 844 845 846	Bellouin, N., Mann, G. W., Woodhouse, M. T., Johnson, C., Carslaw, K. S., and Dalvi, M.: Impact of the modal aerosol scheme GLOMAP-mode on aerosol forcing in the Hadley Centre Global Environmental Model, Atmos. Chem. Phys., 13, 3027-3044, 10.5194/acp-13-3027-2013, 2013.
847 848 849	Berry, E. X.: Modification of the warm rain process, Proc. First Natl. Conf. Weather Modification, Ed. American Meteorological Society, State University of New York, Albany, 81–88, 1968
850 851 852 853	Bogenschutz, P. A., Gettelman, A., Morrison, H., Larson, V. E., Craig, C., and Schanen, D. P.: Higher-Order Turbulence Closure and Its Impact on Climate Simulations in the Community Atmosphere Model, J. Clim., 26, 9655-9676, 10.1175/jcli-d-13-00075.1, 2013.
854 855 856	Bony, S., Dufresne, J. L., Le Treut, H., Morcrette, J. J., and Senior, C.: On dynamic and thermodynamic components of cloud changes, Clim. Dyn., 22, 71-86, 10.1007/s00382-003-0369-6, 2004.
857 858 859	Bony, S., and Dufresne, J. L.: Marine boundary layer clouds at the heart of tropical cloud feedback uncertainties in climate models, Geophys. Res. Lett., 32, 10.1029/2005gl023851, 2005.
860 861 862 863 864	Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, VM., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B., and Zhang, X. Y.: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. edited

- by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J.,
- Nauels, A., Xia, Y., Bex V., and Midgley, P. M., Cambridge University Press,
- 867 Cambridge, United Kingdom and New York, NY, USA, 2013.
- Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation,
- entrainment efficiency, and subtropical stratocumulus albedo, Geophys. Res. Lett., 34,
 10.1029/2006gl027648, 2007.
- 871 Bretherton, C. S., and Park, S.: A New Moist Turbulence Parameterization in the
- 872 Community Atmosphere Model, J. Clim., 22, 3422-3448, 10.1175/2008jcli2556.1,
 873 2009.
- Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based
- estimate of global aerosol-cloud radiative forcing by marine warm clouds, Nature
- 876 Geosci, 7, 643-646, 10.1038/ngeo2214, 2014.Coakley, J. A., and Walsh, C. D.: Limits
- to the aerosol indirect radiative effect derived from observations of ship tracks, J.
- Atmos. Sci., 59, 668-680, 10.1175/1520-0469(2002)059<0668:lttair>2.0.co;2, 2002.
- 879 Del Genio, A. D., and M.-S. Yao, Efficient cumulus parameterization for long-term
- climate studies: The GISS scheme, in The Representation of Cumulus Convection in
- 881 Numerical Models, AMS Meteorol. Monogr., vol. 46, edited by K. A. Emanuel and D.
- A. Raymond, pp. 181–184, Am. Meteorol. Soc., Washington D.C., 1993.
- Bel Genio, A. D., M. S. Yao, W. Kovari, and K. K. Lo, A prognostic cloud water
 parameterization for general circulation models, J. Clim., 9, 270–304, 1996.
- 885 Gettelman, A.: Putting the clouds back in aerosol-cloud interactions, Atmos. Chem.
 886 Phys., 15, 12397-12411, 10.5194/acp-15-12397-2015, 2015.
- 687 Gettelman, A., Liu, X., Ghan, S. J., Morrison, H., Park, S., Conley, A. J., Klein, S. A.,
- 888 Boyle, J., Mitchell, D. L., and Li, J. L. F.: Global simulations of ice nucleation and ice
- supersaturation with an improved cloud scheme in the Community Atmosphere
- 890 Model, J. Geophys. Res.-Atmos., 115, 10.1029/2009jd013797, 2010.
- Gettelman, A., Morrison, H., Terai, C. R., and Wood, R.: Microphysical process rates
 and global aerosol-cloud interactions, Atmos. Chem. Phys., 13, 9855-9867,
 10.5194/acp-13-9855-2013, 2013.
- 6894 Gettelman, A., and Morrison, H.: Advanced Two-Moment Bulk Microphysics for
- Global Models. Part I: Off-Line Tests and Comparison with Other Schemes, J. Clim.,
- 896 28, 1268-1287, 10.1175/JCLI-D-14-00102.1, 2015.
- 897 Gettelman, A., Morrison, H., Santos, S., Bogenschutz, P., and Caldwell, P. M.:
- 898 Advanced Two-Moment Bulk Microphysics for Global Models. Part II: Global Model
- 899 Solutions and Aerosol–Cloud Interactions, J. Clim., 28, 1288-1307,
- 900 10.1175/JCLI-D-14-00103.1, 2015.

901 902	<u>Ghan, S. J.: Technical Note: Estimating aerosol effects on cloud radiative forcing,</u> <u>Atmos. Chem. Phys., 13, 9971–9974, doi:10.5194/acp-13-9971-2013, 2013.</u>
903 904 905 906	Ghan, S. J., Liu, X., Easter, R. C., Zaveri, R., Rasch, P. J., Yoon, J. H., and Eaton, B.: Toward a Minimal Representation of Aerosols in Climate Models: Comparative Decomposition of Aerosol Direct, Semidirect, and Indirect Radiative Forcing, J. Clim., 25, 6461-6476, 10.1175/jcli-d-11-00650.1, 2012.
907 908 909 910 911	Ghan, S. J., Wang, M., Zhang, S., Ferrachat, S., Gettelman, A., Griesfeller, J., Kipling, Z., Lohmann, U., Morrison, H, Neubauer, D., Partridge, D., Stier, P., Takemura, T., Wang, H., Zhang, K.: Challenges in Constraining Anthropogenic Aerosol Effects on Cloud Radiative Forcing Using Present-day Spatiotemporal Variability, Proc. Natl. Acad. Sci. U. S. A., Accepted, 2016.
912 913 914 915	Gregory, D. and Rowntree, P. R.: A massflux convection scheme with representation of cloud ensemble characteristics and stability dependent closure, Mon. Weather Rev., 118, 1483–1506, doi:10.1175/1520-0493(1990)118<1483:AMFCSW>2.0.CO;2, 1990.
916 917	Gryspeerdt, E., and Stier, P.: Regime-based analysis of aerosol-cloud interactions, Geophys. Res. Lett., 39, 10.1029/2012gl053221, 2012.
918 919 920	Gryspeerdt, E., Stier, P., and Grandey, B. S.: Cloud fraction mediates the aerosol optical depth-cloud top height relationship, Geophys. Res. Lett., 41, 3622-3627, 10.1002/2014g1059524, 2014a.
921 922 923 924 925	Gryspeerdt, E., Stier, P., and Partridge, D. G.: Links between satellite-retrieved aerosol and precipitation, Atmos. Chem. Phys., 14, 9677-9694, 10.5194/acp-14-9677-2014, 2014b.Gryspeerdt, E., Stier, P., and Partridge, D. G.: Satellite observations of cloud regime development: the role of aerosol processes, Atmos. Chem. Phys., 14, 1141-1158, 10.5194/acp-14-1141-2014, 2014c.
926 927 928	Guo, H., Golaz, J. C., and Donner, L. J.: Aerosol effects on stratocumulus water paths in a PDF-based parameterization, Geophys. Res. Lett., 38, 10.1029/2011gl048611, 2011.
929 930 931	Guo, H., Golaz, J. C., Donner, L. J., Wyman, B., Zhao, M., and Ginoux, P.: CLUBB as a unified cloud parameterization: Opportunities and challenges, Geophys. Res. Lett., 42, 4540-4547, 10.1002/2015GL063672, 2015.
932 933 934	Guo, Z., Wang, M., Qian, Y., Larson, V., Ghan, S., Bogenschutz, P., Gettelman, A.: Parametric behaviors of CLUBB in simulation of low clouds in the Community Atmosphere Model CAM5, under revision, J. Adv. Model. Earth Syst., 2015.
935 936	Hewitt, H. T., Copsey, D., Culverwell, I. D., Harris, C. M., Hill, R. S. R., Keen, A. B., McLaren, A. J., and Hunke, E. C.: Design and implementation of the infrastructure of

HadGEM3: the nextgeneration Met Office climate modelling system, Geosci. Model
Dev., 4, 223–253, doi:10.5194/gmd-4-223-2011, 2011.

939 IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of

940 Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on

941 Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J.

Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge

- University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp.,
- 944 2013

Kaufman, Y. J., Koren, I., Remer, L. A., Rosenfeld, D., and Rudich, Y.: The effect of
smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic
Ocean, Proc. Natl. Acad. Sci. U. S. A., 102, 11207-11212, 10.1073/pnas.0505191102,
2005.

Khairoutdinov, M., and Kogan, Y.: A new cloud physics parameterization in a
large-eddy simulation model of marine stratocumulus, Mon. Weather Rev., 128,
229-243, 10.1175/1520-0493(2000)128<0229:ancppi>2.0.co;2, 2000.

952 Kooperman, G. J., Pritchard, M. S., Ghan, S. J., Wang, M. H., Somerville, R. C. J.,

and Russell, L. M.: Constraining the influence of natural variability to improve

estimates of global aerosol indirect effects in a nudged version of the Community

Atmosphere Model 5, J. Geophys. Res.-Atmos., 117, 10.1029/2012jd018588, 2012.

Lamarque, J. F., et al. (2010), Historical (1850-2000) gridded anthropogenic and

biomass burning emissions of reactive gases and aerosols: methodology and

958 application, *Atmos Chem Phys*, 10(15), 7017-7039, doi:10.5194/Acp-10-7017-2010.

Lebo, Z. J., and Feingold, G.: On the relationship between responses in cloud water
and precipitation to changes in aerosol, Atmos. Chem. Phys., 14, 11817-11831,
10.5194/acp-14-11817-2014, 2014.

Lee, Y.-H., and P. J. Adams, A fast and efficient version of the TwO-Moment Aerosol
Sectional (TOMAS) global aerosol microphysics model, Aerosol. Sci. Technol., 46,
678–689, doi:10.1080/02786826.2011.643259, 2012.

Lee, Y. H., et al., Evaluation of the global aerosol microphysical ModelE2-TOMAS
model against satellite and ground-based observations, Geosci. Model Dev., 8(3),

- 967 631–667, doi:10.5194/gmd-8-631-2015, 2015.
- Lin, H. and Leaitch, W. R.: Development of an in-cloud aerosol activation

parameterization for climate modelling, in: Proceedings of the WMO Workshop on

970 Measurement of Cloud Properties for Forecasts of Weather, Air Quality and Climate,

971 World Meteorol. Organ., Geneva, pp. 328–335, 1997.

- 972 Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J. F.,
- 973 Gettelman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C.,

- Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M. J., Bretherton, C.
- 975 S., Flanner, M. G., and Mitchell, D.: Toward a minimal representation of aerosols in
- climate models: description and evaluation in the Community Atmosphere Model
- 977 CAM5, Geosci. Model Dev., 5, 709-739, 10.5194/gmd-5-709-2012, 2012.
- 978 Liu, X., Shi, X., Zhang, K., Jensen, E. J., Gettelman, A., Barahona, D., Nenes, A., and
- 279 Lawson, P.: Sensitivity studies of dust ice nuclei effect on cirrus clouds with the
- 980 Community Atmosphere Model CAM5, Atmos Chem Phys, 12(24), 12061-12079,
- 981 doi:Doi 10.5194/Acp-12-12061-2012, 2012
- 982 Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang,
- 983 J.: Cloud microphysics and aerosol indirect effects in the global climate model
- ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425–3446, doi:10.5194/acp-7-3425-2007,
 2007.
- Lohmann, U., and Hoose, C.: Sensitivity studies of different aerosol indirect effects in
 mixed-phase clouds, Atmos. Chem. Phys., 9, 8917-8934, 2009.
- Lu, M. L., and Seinfeld, J. H.: Study of the aerosol indirect effect by large-eddy
 simulation of marine stratocumulus, J. Atmos. Sci., 62, 3909-3932, 10.1175/jas3584.1,
 2005.
- 991 Ma, P.-L., Rasch, P. J., Wang, M., Wang, H., Ghan, S. J., Easter, R. C., Gustafson, W.
- 992 I., Liu, X., Zhang, Y., and Ma, H.-Y.: How does increasing horizontal resolution in a
- global climate model improve the simulation of aerosol-cloud interactions?, Geophys.
 Res. Lett., n/a-n/a, 10.1002/2015GL064183, 2015.
- 995 Matsui, T., Masunaga, H., Kreidenweis, S. M., Pielke, R. A., Tao, W. K., Chin, M.,
- and Kaufman, Y. J.: Satellite-based assessment of marine low cloud variability
- associated with aerosol, atmospheric stability, and the diurnal cycle, J. Geophys.
- 998 Res.-Atmos., 111, 10.1029/2005jd006097, 2006.
- Medeiros, B., and Stevens, B.: Revealing differences in GCM representations of low
 clouds, Clim. Dyn., 36, 385-399, 10.1007/s00382-009-0694-5, 2011.
- Menon, S., D. Koch, G. Beig, S. Sahu, J. Fasullo, and D. Orlikowski, Black carbon
 aerosols and the third polar ice cap, Atmos. Chem. Phys., 10, 4559–4571, 2010.
- 1003 Morrison, H., and A. Gettelman, A new two-moment bulk stratiform cloud
- 1004 microphysics scheme in the Community Atmosphere Model, version 3 (CAM3). Part
- I: Description and numerical tests, J. Clim., 21, 3642–3659, 2008. Nenes, A., and
- 1006 Seinfeld, J. H.: Parameterization of cloud droplet formation in global climate models,
- 1007 J. Geophys. Res.-Atmos., 108, 10.1029/2002jd002911, 2003.
- Neale, R. B., Richter, J. H., and Jochum, M.: The Impact of Convection on ENSO:
 From a Delayed Oscillator to a Series of Events, J. Clim., 21, 5904-5924,
- 1010 10.1175/2008jcli2244.1, 2008.

- 1011 Neubauer, D., Lohmann, U., Hoose, C., & Frontoso, M. G. Impact of the
- 1012 representation of marine stratocumulus clouds on the anthropogenic aerosol effect.
- 1013 Atmos. Chem. Phys. Disc., 14, 13681–13729, 2014.
- 1014 Nordeng, T. E.: Extended versions of the convective parametrization scheme at
- ECMWF and their impact on the mean and transient activity of the model in the
- 1016 tropics, ECMWF Research Department, Technical Momorandum 206, European
- 1017 Centre for Medium-range Weather Forecast, Reading, UK, 1994.
- 1018 O'Connor, F. M., Johnson, C. E., Morgenstern, O., Abraham, N. L., Braesicke, P.,
- 1019 Dalvi, M., Folberth, G. A., Sanderson, M. G., Telford, P. J., Voulgarakis, A., Young,
- 1020 P. J., Zeng, G., Collins, W. J., and Pyle, J. A.: Evaluation of the new UKCA
- 1021 climate-composition model Part 2: The Troposphere, Geosci. Model Dev., 7, 41–91,
- 1022 doi:10.5194/gmd-7-41-2014, 2014
- Park, S., and Bretherton, C. S.: The University of Washington Shallow Convection
 and Moist Turbulence Schemes and Their Impact on Climate Simulations with the
 Community Atmosphere Model, J. Clim., 22, 3449-3469, 10.1175/2008jcli2557.1,
 2009.
- Penner, J. E., Quaas, J., Storelvmo, T., Takemura, T., Boucher, O., Guo, H., Kirkevag,
 A., Kristjansson, J. E., and Seland, O.: Model intercomparison of indirect aerosol
 effects, Atmos. Chem. Phys., 6, 3391-3405, 2006.
- 1030 Penner, J. E., Xu, L., and Wang, M. H.: Satellite methods underestimate indirect
- climate forcing by aerosols, Proc. Natl. Acad. Sci. U. S. A., 108, 13404-13408,
 10.1073/pnas.1018526108, 2011.
- Posselt, R., and Lohmann, U.: Sensitivity of the total anthropogenic aerosol effect to
 the treatment of rain in a global climate model, Geophys. Res. Lett., 36,
 1035 | 10.1029/2008gl035796, 2009.
- 1036 Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S.,
 1037 Reissell, A., and Andreae, M. O.: Flood or drought: How do aerosols affect
- **1038** precipitation?, Science, 321, 1309-1313, 10.1126/science.1160606, 2008.
- 1039 Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A.,
- 1040 Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey,
- 1041 A., Feingold, G., Hoose, C., Kristjansson, J. E., Liu, X., Balkanski, Y., Donner, L. J.,
- 1042 Ginoux, P. A., Stier, P., Grandey, B., Feichter, J., Sednev, I., Bauer, S. E., Koch, D.,
- 1043 Grainger, R. G., Kirkevag, A., Iversen, T., Seland, O., Easter, R., Ghan, S. J., Rasch,
- 1044 P. J., Morrison, H., Lamarque, J. F., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol
- indirect effects general circulation model intercomparison and evaluation with
- satellite data, Atmos. Chem. Phys., 9, 8697-8717, 2009.

- 1047 Schmidt, G. A., Ruedy, R., Hansen, J. E., Aleinov, I., Bell, N., Bauer, M., Bauer, S.,
- 1048 Cairns, B., Canuto, V., Cheng, Y., Del Genio, A., Faluvegi, G., Friend, A. D., Hall, T.
- 1049 M., Hu, Y. Y., Kelley, M., Kiang, N. Y., Koch, D., Lacis, A. A., Lerner, J., Lo, K. K.,
- 1050 Miller, R. L., Nazarenko, L., Oinas, V., Perlwitz, J., Perlwitz, J., Rind, D., Romanou,
- 1051 A., Russell, G. L., Sato, M., Shindell, D. T., Stone, P. H., Sun, S., Tausnev, N.,
- 1052 Thresher, D., and Yao, M. S.: Present-day atmospheric simulations using GISS
- 1053 ModelE: Comparison to in situ, satellite, and reanalysis data, J. Clim., 19, 153-192,
- 1054 10.1175/jcli3612.1, 2006.
- 1055 Schmidt, G. A., Kelley, M., Nazarenko, L., Ruedy, R., Russell, G. L., Aleinov, I.,
- 1056 Bauer, M., Bauer, S. E., Bhat, M. K., Bleck, R., Canuto, V., Chen, Y. H., Cheng, Y.,
- 1057 Clune, T. L., Del Genio, A., de Fainchtein, R., Faluvegi, G., Hansen, J. E., Healy, R.
- 1058 J., Kiang, N. Y., Koch, D., Lacis, A. A., LeGrande, A. N., Lerner, J., Lo, K. K.,
- 1059 Matthews, E. E., Menon, S., Miller, R. L., Oinas, V., Oloso, A. O., Perlwitz, J. P.,
- 1060 Puma, M. J., Putman, W. M., Rind, D., Romanou, A., Sato, M., Shindell, D. T., Sun,
- 1061 S., Syed, R. A., Tausnev, N., Tsigaridis, K., Unger, N., Voulgarakis, A., Yao, M. S.,
- and Zhang, J. L.: Configuration and assessment of the GISS ModelE2 contributions to
- the CMIP5 archive, J. Adv. Model. Earth Syst., 6, 141-184, 10.1002/2013ms000265,
 2014.
- 1065 Small, J. D., Chuang, P. Y., Feingold, G., and Jiang, H. L.: Can aerosol decrease
- 1066 cloud lifetime?, Geophys. Res. Lett., 36, L16806, 10.1029/2009GL038888,
- 1067 2009.Song, X. L., and Zhang, G. J.: Microphysics parameterization for convective
- 1068 clouds in a global climate model: Description and single-column model tests, J.
- 1069 Geophys. Res.-Atmos., 116, 10.1029/2010jd014833, 2011.
- Stevens, B., and Feingold, G.: Untangling aerosol effects on clouds and precipitation
 in a buffered system, Nature, 461, 607-613, 10.1038/nature08281, 2009.
- 1072 Stevens, B., Giorgetta, M., Esch, M., Mauritsen, T., Crueger, T., Rast, S., Salzmann,
- 1073 M., Schmidt, H., Bader, J., Block, K., Brokopf, R., Fast, I., Kinne, S., Kornblueh, L.,
- 1074 Lohmann, U., Pincus, R., Reichler, T., and Roeckner, E.: Atmospheric component of
- the MPI-M Earth System Model: ECHAM6, J. Adv. Model. Earth Syst., 5, 146–172,
 doi:10.1002/jame.20015, 2013.
- 1077 Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L.,
- 1078 Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and
- 1079 Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5,
- 1080 1125–1156, doi:10.5194/acp-5-1125-2005, 2005.
- 1081 Storelvmo, T., Kristjansson, J. E., and Lohmann, U.: Aerosol influence on
- 1082 mixed-phase clouds in CAM-Oslo, J Atmos Sci, 65(10), 3214-3230,
- 1083 doi:doi:10.1175/2008jas2430.1, 2008.

Storelvmo, T., Kristjansson, J. E., Muri, H., Pfeffer, M., Barahona, D., and Nenes, A.:
Cirrus cloud seeding has potential to cool climate, Geophys Res Lett, 40(1), 178-182,
doi:Doi 10.1029/2012gl054201, 2013.

1087 Takemura, T., Nozawa, T., Emori, S., Nakajima, T. Y., and Nakajima, T.: Simulation

- 1088 of climate response to aerosol direct and indirect effects with aerosol
- transport-radiation model, J. Geophys. Res.-Atmos., 110, 10.1029/2004jd005029,
 2005.
- Takemura, T., Egashira, M., Matsuzawa, K., Ichijo, H., O'Ishi, R., and Abe-Ouchi, A.:
 A simulation of the global distribution and radiative forcing of soil dust aerosols at the
- Last Glacial Maximum, Atmos. Chem. Phys., 9, 3061-3073, 2009.
- 1094 Tiedtke, M.: A comprehensive mass flux scheme for cumulus parameterization in
- 1095 large-scale models, Mon. Weather Rev., 117, 1779–1800, doi:10.1175/1520-
- 1096 0493(1989)117<1779:ACMFSF> 2.0.CO;2, 1989.
- 1097 Twomey, S.: INFLUENCE OF POLLUTION ON SHORTWAVE ALBEDO OF
- 1098 CLOUDS, J. Atmos. Sci., 34, 1149-1152,
- 1099 10.1175/1520-0469(1977)034<1149:tiopot>2.0.co;2, 1977.
- 1100 Walters, D. N., Williams, K. D., Boutle, I. A., Bushell, A. C., Edwards, J. M., Field, P.
- 1101 R., Lock, A. P., Morcrette, C. J., Stratton, R. A., Wilkinson, J. M., Willett, M. R.,
- 1102 Bellouin, N., Bodas-Salcedo, A., Brooks, M. E., Copsey, D., Earnshaw, P. D.,
- 1103 Hardiman, S. C., Harris, C. M., Levine, R. C., MacLachlan, C., Manners, J. C., Martin,
- 1104 G. M., Milton, S. F., Palmer, M. D., Roberts, M. J., Rodríguez, J. M., Tennant, W. J.,
- and Vidale, P. L.: The Met Office Unified Model Global Atmosphere 4.0 and JULES
- 1106 Global Land 4.0 configurations, Geosci. Model Dev., 7, 361-386,
- 1107 10.5194/gmd-7-361-2014, 2014.
- 1108 Wang, H. L., and Feingold, G.: Modeling Mesoscale Cellular Structures and Drizzle
- in Marine Stratocumulus. Part I: Impact of Drizzle on the Formation and Evolution of
- 1110 Open Cells, J. Atmos. Sci., 66, 3237-3256, 10.1175/2009jas3022.1, 2009a.
- 1111 Wang, H. L., and Feingold, G.: Modeling Mesoscale Cellular Structures and Drizzle
- in Marine Stratocumulus. Part II: The Microphysics and Dynamics of the Boundary
- 1113 Region between Open and Closed Cells, J. Atmos. Sci., 66, 3257-3275,
- 1114 10.1175/2009jas3120.1, 2009b.
- 1115 Wang, H., Easter, R. C., Rasch, P. J., Wang, M., Liu, X., Ghan, S. J., Qian, Y., Yoon,
- 1116 J. H., Ma, P. L., and Vinoj, V.: Sensitivity of remote aerosol distributions to
- 1117 representation of cloud-aerosol interactions in a global climate model, Geosci. Model
- 1118 Dev., 6, 765-782, 10.5194/gmd-6-765-2013, 2013.

- 1119 Wang, M., Ghan, S., Ovchinnikov, M., Liu, X., Easter, R., Kassianov, E., Qian, Y.,
- and Morrison, H.: Aerosol indirect effects in a multi-scale aerosol-climate model
- 1121 PNNL-MMF, Atmos. Chem. Phys., 11, 5431-5455, 10.5194/acp-11-5431-2011, 2011.
- 1122 Wang, M. H., Ghan, S., Liu, X. H., L'Ecuyer, T. S., Zhang, K., Morrison, H.,
- 1123 Ovchinnikov, M., Easter, R., Marchand, R., Chand, D., Qian, Y., and Penner, J. E.:
- 1124 Constraining cloud lifetime effects of aerosols using A-Train satellite observations,
- 1125 Geophys. Res. Lett., 39, 10.1029/2012gl052204, 2012.
- 1126 Wang, M., Liu, X., Zhang, K. and Comstock, J.: Aerosol indirect effects on cirrus
- throughice nucleation in CAM5 with a statistical cirrus cloud scheme, in press, J. Adv.
- 1128 Model. Earth Syst. 6, doi:10.1002/2014MS000339, 2014.
- 1129 Watanabe, M., Suzuki, T., O'Ishi, R., Komuro, Y., Watanabe, S., Emori, S., Takemura,
- 1130 T., Chikira, M., Ogura, T., Sekiguchi, M., Takata, K., Yamazaki, D., Yokohata, T.,
- 1131 Nozawa, T., Hasumi, H., Tatebe, H., and Kimoto, M.: Improved Climate Simulation
- by MIROC5. Mean States, Variability, and Climate Sensitivity, J. Clim., 23,
- 1133 6312-6335, 10.1175/2010jcli3679.1, 2010.
- 1134 West, R. E. L., Stier, P., Jones, A., Johnson, C. E., Mann, G. W., Bellouin, N.,
- 1135 Partridge, D. G., and Kipling, Z.: The importance of vertical velocity variability for
- estimates of the indirect aerosol effects, Atmos. Chem. Phys., 14, 6369-6393,
- 1137 10.5194/acp-14-6369-2014, 2014.
- 1138 Wilson, D. R., Bushell, A. C., Kerr-Munslow, A. M., Price, J. D., and Morcrette, C. J.:
- 1139 PC2: A prognostic cloud fraction and condensation scheme. I: Scheme description, Q.
- 1140 J. R. Meteorol. Soc., 134, 2093–2107, doi:10.1002/qj.333, 2008
- 1141 Zhang, K., O'Donnel, D., Kazil, J., Stier., P., Kinne, S., Lohmann, U., Ferrachat, S.,
- 1142 Croft, B., Quaas, J., Wan, H., Rast, S. and Feichter, J.: The global aerosol-climate
- 1143 model ECHAM-HAM, version 2: sensitivity to improvements in process
- 1144 representations, Atmos. Chem. Phys., 12, 8911–8949, doi:10.5194/acp-12-8911-2012,
 1145 2012.
- 1146 Zhang, K., Wan, H., Liu, X., Ghan, S. J., Kooperman, G. J., Ma, P.-L., Rasch, P. J.,
- 1147 Neubauer, D., and Lohmann, U.: Technical Note: On the use of nudging for aerosol-
- climate model intercomparison studies, Atmos. Chem. Phys., 14, 8631-8645,
- 1149 doi:10.5194/acp-14-8631-2014, 2014.
- 1150

1151 Tabel 1. The types of clouds included in liquid water path (LWP) and surface rain

Model	LWP	Rain	Rain scheme
CAM5	\mathbf{S}^{*}	S	d&
CAM5-MG2	S	S	$\mathbf{p}^{@}$
CAM5-PNNL	S	S	d
CAM5-CLUBB	S+shallow convective clouds	S+shallow convective clouds	d
CAM5-CLUBB-MG2	S+shallow convective clouds	S+shallow convective clouds	р
ECHAM6-HAM2	S+convective detrainment	S	d
SPRINTARS	$S+C^{\#}$	S+C	d
SPRINTATRS-KK	S+C	S+C	d
ModelE2-TOMAS	S+anvil clouds	S+anvil clouds	d
HadGEM3-UKCA	S+C	S	р

rate and different rain schemes in 10 participating models

1153 * S in LWP and Rain stands for stratiform clouds.

1154 # C in LWP and Rain stands for convective clouds.

1155 & d in Rain schemes represents diagnostic rain scheme.

1156 *(a)* p in Rain schemes represents prognostic rain scheme.

1157

Table 2.Global ocean (60°S-60°N) averages of LWP, column-integrated cloud condensation nuclei (CCN, at 0.1% supersaturation) concentration, precipitation rate (PRECL), shortwave cloud radiative effect (SCRE) derived from the present day (PD) cases and the relative change from pre-industrial (PI) to PD of LWP and CCN (dlnLWP and dlnCCN) and the sensitivity of LWP to CCN concentration change (λ , dlnLWP/dlnCCN) of the 10 GCM simulations.

		LWP	CCN			PRECL	SCRE
Model	λ	(g	(10^{11})	dlnLWP	dlnCCN	(mm	$(W m^{-2})$
		m ⁻²)	m^{-2})			d^{-1})	(" ")
CAM5	0.20	31.1	1.86	0.07	0.36	0.90	-61.9
CAM5-MG2	0.23	30.0	1.73	0.07	0.32	0.76	-67.9
CAM5-PNNL	0.19	39.4	2.51	0.08	0.42	0.91	-64.6
CAM5-CLUBB	0.25	35.2	1.88	0.11	0.45	1.26	-57.7
CAM5-CLUBB-MG2	0.27	47.1	1.66	0.11	0.42	1.08	-70.6
ECHAM6-HAM2	0.19	84.6	2.39	0.07	0.41	1.35	-54.5
SPRINTARS	0.01	139.1	1.07	0.00	0.43	1.42	-62.6
SPRINTATRS-KK	0.04	98.9	1.04	0.02	0.45	1.59	-57.0
ModelE2-TOMAS	0.00	80.4	2.66	0.00	0.43	2.17	-68.1
HadGEM3-UKCA	0.03	57.1	1.01	0.01	0.67	0.87	-58.9

1168	Table 3. Criteria used to conditional sampling stratocumulus, transitional clouds and

trade wind cumulus regimes (adopted from Medeiros and Steve	ns (2011))
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		Stratocumulus	Transitional	Trade wind	
			clouds	cumulus	
	LTS (K)	LTS≥18.5	18.5>LTS≥15.4	15.4>LTS≥11.3	
0	ω_{500hPa} (hPa d ⁻¹)	ω _{500hPa} >10	ω _{500hPa} >10	ω _{500hPa} >10	

1174	Table 4. The fractional occurrences of low and high surface precipitation in PD cases
1175	over downdraft regimes ($\omega_{500} > 0$ hPa/d) and global oceans and λ under these low and
1176	high surface precipitation situations only over downdraft regimes. Low precipitation
1177	situations refer to monthly surface precipitation rate (PRECL) less than 0.1 mm d^{-1}
1178	while high precipitation situations refer to PRECL larger than 0.1mm d ⁻¹ .

1179

	λ^{a}	λ^{b}	f^c	f^d	f^e	f^{f}
Model	low,	high,	low,	high,	low,	high,
	down	down	down	down	glb	glb
CAM5	0.21	0.19	0.47	0.54	0.27	0.73
CAM5-MG2	0.19	0.24	0.57	0.43	0.39	0.61
CAM5-PNNL	0.17	0.17	0.48	0.52	0.28	0.72
CAM5-CLUBB	0.33	0.30	0.04	0.96	0.02	0.98
CAM5-CLUBB-MG2	0.26	0.33	0.22	0.78	0.16	0.84
ECHAM6-HAM2	0.25	0.23	0.31	0.69	0.18	0.82
SPARINTARS	0.06	0.01	0.06	0.94	0.03	0.97
SPARINTARS-KK	0.24	0.04	0.05	0.95	0.03	0.97
ModelE2-TOMAS	-0.011	0.001	0.002	0.998	0.001	0.999
HadGEM3-UKCA	0.04	0.03	0.11	0.89	0.06	0.94

- 1181 ^a λ under low PRECL for downdraft regimes
- 1182 ^b λ under high PRECL for downdraft regimes
- ^c Fractional occurence of low PRECL for downdraft regimes
- ^d Fractional occurence of high PRECL for downdraft regimes
- ^e Fractional occurence of low PRECL over <u>all dynamical</u> regimes
- ^f Fractional occurrence of high PRECL over <u>all dynamical</u> regimes

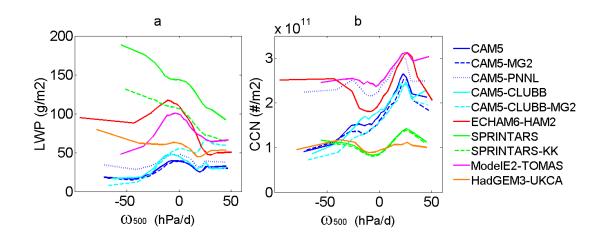
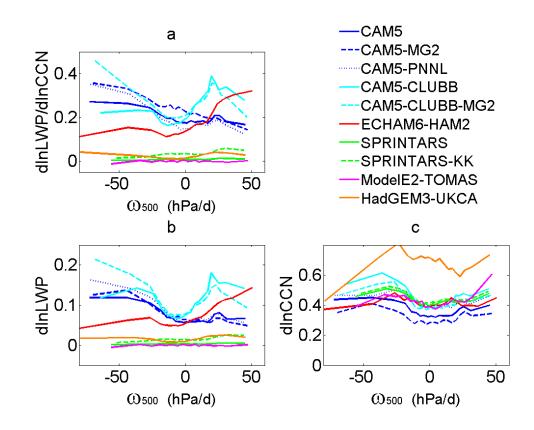


Fig 1. (a) LWP and (b) column-integrated CCN (at 0.1% supersaturation) as a function of 500 hPa vertical pressure velocity (ω_{500}) derived from different models: CAM5 (blue solid line), CAM5-MG2 (blue dashed line), CAM5-PNNL (blue dotted line), CAM5-CLUBB (cyan solid line), CAM5-CLUBB-MG2 (cyan dashed line), ECHAM6-HAM2 (red solid line), SPRINTARS (green solid line), SPRINTARS-KK (green dashed line), ModelE2-TOMAS (purple solid line) and HadGEM3-UKCA (orange solid line).

1196

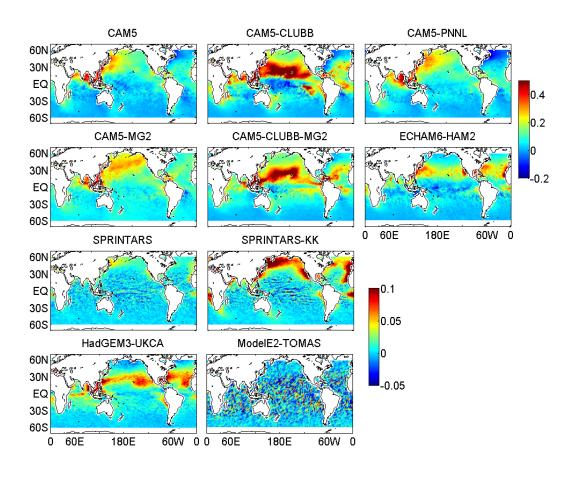


1200 Fig 2. Same as Fig. 1a), but for (a) the sensitivity of LWP to the change of CCN (λ),

1201 (b) relative enhancement of liquid water path (dlnLWP) and (c) relative enhancement

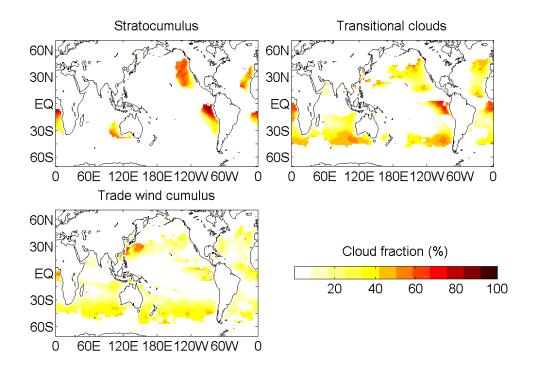
1202 of cloud condensation nuclei (dlnCCN) from pre-industrial (PI) to present day (PD).





1209 Fig 3. Relative change of annual averaged LWP from PI to PD (dlnLWP)

simulations derived from the 10 GCM simulations.



1214

Fig 4. The annual mean cloud fraction (averaged on the months when the regime occurs) of stratocumulus regime (top left), transitional clouds regime (top right) and trade wind cumulus regime (bottom left) derived from PD monthly simulation in CAM5-CLUBB. The definitions of different cloud types are listed in Table 3.



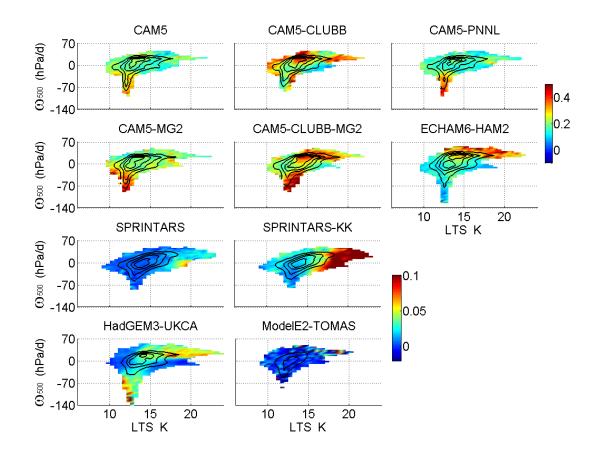


Fig 5. dlnLWP/dlnCCN conditioned on vertical motion and LTS derived from the 10
GCM simulations. Solid lines are contours of grid number distribution and each line
interval is 20% of the total counted data.

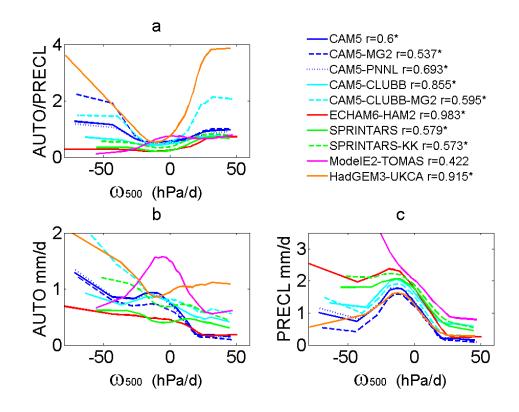


Fig 6. Same as Fig. 1, but for (b) column-integrated autoconversion rate (AUTO), (c) the large-scale surface precipitation rate (PRECL) and (a) their ratio AUTO/PRECL from the 9 GCM simulations. The number marked in each simulation is the corresponding correlation coefficient between AUTO/PRECL and λ and number with mark '*' indicates the correlation is significant (at 95% confidence).

1228

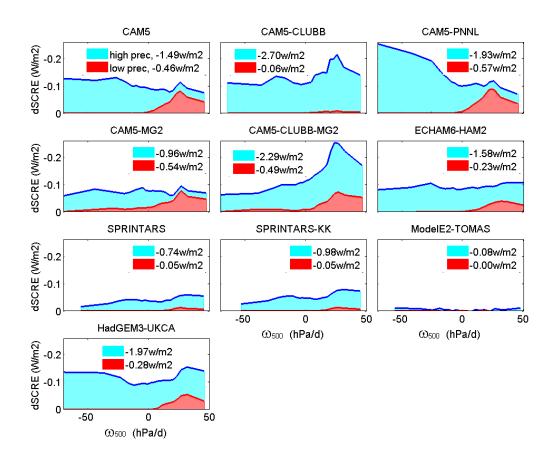


Fig 7. Change in shortwave cloud radiative effect (dSCRE, shown in blue line) from
PI to PD as a function of dynamic regimes. Red patches are dSCRE contributed by
low precipitation situations while blue patches are by high precipitation situations.