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

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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On the characteristics of aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

Aerosol-cloud interactions continue to constitute a major source of uncertainty for the estimate of climate radiative forcing. The variation of aerosol indirect effects (AIE) in climate models is investigated across different dynamical regimes, determined by monthly mean 500 hPa vertical pressure velocity ( $\omega_{500}$ ), lower-tropospheric stability (LTS) and large-scale surface precipitation rate derived from several global climate models (GCMs), with a focus on liquid water path (LWP) response to cloud condensation nuclei (CCN) concentrations. The LWP sensitivity to aerosol perturbation within dynamic regimes is found to exhibit a large spread among these GCMs. It is in regimes of strong large-scale ascend ( $\omega_{500} < -25 \text{ hPa d}^{-1}$ ) and low clouds (stratocumulus and trade wind cumulus) where the models differ most. Shortwave aerosol indirect forcing is also found to differ significantly among different regimes. Shortwave aerosol indirect forcing in ascending regimes is as large as that in stratocumulus regimes, which indicates that regimes with strong large-scale ascend are as important 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 ( $> 0.1 \text{ mm d}^{-1}$ ) 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 than that globally, pointing to the need to reduce the uncertainty in AIE in different dynamical regimes.

## 1 Introduction

By scattering and absorbing sunlight, aerosol particles can modify the solar radiation reaching the earth system, which is termed the direct effect. The direct radiative effect of anthropogenic aerosols combined with subsequent rapid adjustments of the surface energy budget, atmospheric state variables, and cloudiness to aerosol radiative effects is referred as Effective Radiative Forcing from aerosol-radiation interactions (ERFari;

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### On the characteristics of aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Boucher et al., 2013). Apart from ERFari, aerosols can also alter the Earth's radiation balance via interactions with clouds, such as effects on cloud albedo and subsequent changes to the cloud lifetime and thermodynamics as rapid adjustments, known as the aerosol indirect effect(s) (AIE). These radiative effects are called Effective Radiative Forcing from aerosol-cloud 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) concentration from anthropogenic aerosols leads to smaller cloud droplet sizes assuming constant liquid water content. The increased number but decreased droplet sizes in turn increase cloud albedo due to more efficient backscattering. This is called the cloud albedo effect or the first AIE, also known as the Twomey effect (Twomey, 1977). Moreover, the smaller cloud droplet sizes are hypothesized to lead to decreases in precipitation efficiency, which may further alter cloud liquid water path (LWP) and cloud lifetime (Albrecht, 1989). These adjustments are also referred to as the cloud lifetime effect or the second AIE. There are also adjustments on mixed-phase, ice and convective clouds but the focus of this study is on the large-scale warm clouds LWP response to CCN concentrations.

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

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Further modeling studies (e.g., Ackerman et al., 2004; Stevens and Feingold, 2009; Guo et al., 2011) suggest that cloud top entrainment plays a critical role as a dynamic feedback, to balance LWP and modify the lifetime of boundary layer clouds. Ackerman et al. (2004) found that an increase in droplet number concentration ( $N_d$ ) reduces cloud water sedimentation while accelerating the cloud-top entrainment rate, which makes the humidity of air overlying the boundary layer, wet or dry, critically important in determining the response of LWP. When surface precipitation is weak ( $< 0.1 \text{ mm day}^{-1}$ ) and the overlying air is dry, LWP decreases in response to increasing aerosol. Ackerman et al. (2004) also demonstrated that the reduced cloud droplet size due to increases in aerosol reduces cloud droplet sedimentation. They showed that the sedimentation effect reduces the entrainment rate by decreasing boundary-layer turbulence kinetic energy (TKE) available for entrainment while Bretherton et al. (2007) found that TKE remained unchanged but reduced evaporative cooling by removing out liquid water should be the main cause of reduced entrainment rate. LES studies (e.g., Wang and Feingold, 2009a) with a large model domain that is able to resolve mesoscale circulations (on the order of ten kilometers) in marine stratocumulus showed that aerosols can shift cloud regimes through their impact on precipitation and associated dynamical feedbacks. This can represent a more significant impact on cloud radiative forcing than the conventional AIE.

Many state-of-the-art global climate models (GCMs) appear to overestimate AIE when compared with satellite observations (e.g., Quaas et al., 2009; Wang et al., 2012), despite of some uncertainties in satellite derived estimates (e.g., Penner et al., 2011; Gryspeerd et al., 2014a; Gryspeerd et al., 2014b). The multi-scale interactions between clouds, aerosols and large-scale dynamics (Stevens and Feingold, 2009; Wang et al., 2011; Ma et al., 2015) and complex microphysical processes (e.g., Bretherton et al., 2007; Gettelman et al., 2013) cause uncertainties in estimating AIE by GCMs. One possible source of overestimation of AIE is their inability to reproduce negative LWP responses to aerosol perturbations, which are found in some observations and LES studies, partly because they do not explicitly simulate the droplet size effect on the

---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

5 entrainment process and on sub-grid cloud organizations associated with changes in precipitation. Guo et al. (2011) found that this effect could be captured through applying a parameterization based on multi-variate probability density functions with dynamics (MVD PDFs) in single-column simulations. They found decreased LWP in response to increasing aerosols concentration and suggested that the implementation of MVD PDFs in GCMs may help lower the magnitude of the simulated AIE. A negative correlation between LWP and aerosol loading was further found for clouds with weak precipitation and dry air above the PBL in a subsequent global model study (Guo et al., 2015).

10 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 autoconversion and accretion (Gettelman et al., 2013). Since autoconversion acts as a sink of LWP, it is crucial in the formation of precipitation, thus plays an important role in determining the cloud lifetime effect. The autoconversion rate is directly dependent on droplet number concentration ( $N_d$ ) while the accretion rate is only weakly dependent on  $N_d$  (Khairoutdinov and Kogan, 2000; Gettelman et al., 2013). Furthermore, the ratio of the autoconversion rate to the large-scale surface precipitation rate is found to be strongly correlated with the LWP response to anthropogenic aerosol perturbations (e.g., Wang et al., 2012). Posselt and Lohmann (2009) suggested this ratio is related to the rain scheme adopted in GCMs. They showed that the adoption of different rain schemes (prognostic vs. diagnostic) in a GCM leads to a different LWP response to aerosol perturbations. A prognostic rain scheme shifts the importance of (warm) rain production from autoconversion process to the accretion process and therefore reduces the AIE (Posselt and Lohmann, 2009; Gettelman et al., 2015).

25 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

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 dynamical regimes over global oceans (60° S–60° N), with a focus on cloud lifetime effects of aerosols (2nd AIE). We note that the term “cloud lifetime effects” can be somehow misleading, since aerosol effects on cloud liquid water may have little to do with cloud lifetime per se (e.g., Small et al., 2009). Nevertheless, this term is still used in some occasions in this paper for convenience. The paper is organized as follows. Methods and models are described in Sect. 2, and results and discussions are presented in Sect. 3. The paper concludes with the summary in Sect. 4.

## 2 Methodology and models

The susceptibility of LWP to aerosol perturbations is defined as

$$\lambda = d \ln LWP / d \ln CCN. \quad (1)$$

As simulated LWP and CCN can be quite different among GCMs, the logarithmic form of LWP and CCN is adopted in the  $\lambda$  formula.  $\lambda$  is a metric to quantitatively measure cloud lifetime effect of aerosols in models and is derived from pre-industrial (PI) to present day (PD) global monthly-mean GCM data in this study. This parameter was used by Wang et al. (2012) to constrain the cloud lifetime effects of aerosols over global oceans using precipitation frequency susceptibility ( $S_{pop}$ ) derived from A-Train satellite observations. Lebo and Feingold (2014) examined the relationship between  $\lambda$  and  $S_{pop}$  to aerosol perturbations for stratocumulus and trade-wind cumulus simulated

by LES and found that  $\lambda$  may increase in marine stratocumulus while decrease in the case of trade-wind cumulus in response to increasing  $S_{\text{pop}}$ , suggesting a cloud regime dependence of this relationship.

Dynamical regimes can be defined by environment characteristics such as large-scale vertical pressure velocity (e.g., Bony and Dufresne, 2005) and lower-tropospheric stability (LTS, defined as the difference in potential temperature between 700hPa and the surface,  $\theta_{700\text{hPa}} - \theta_{\text{surface}}$ ; e.g., Medeiros and Stevens, 2011). Medeiros and Stevens (2011) noted that low clouds and deep convective clouds could be separated by  $\omega_{500}$  while different low cloud types under large-scale subsidence can only be depicted by using LTS. In this study the monthly-averaged vertical pressure velocity ( $\omega$ ) in the mid-troposphere (defined as at 500 hPa) is used as a proxy for large-scale motions (Bony and Dufresne, 2005). Note that  $\omega_{500}$  with positive (negative) value means descending (ascending) motions. We decompose global ( $60^\circ\text{S}$ – $60^\circ\text{N}$ ) large-scale circulations over ocean as a group of dynamical regimes (equally sampled) by  $\omega_{500}$  (and LTS). Ascending regimes and descending regimes are defined by  $\omega_{500}$  and descending regimes are further divided into stratocumulus, transitional clouds and trade wind cumulus regimes by LTS. This method is straight-forward to apply to GCM results and gives us a direct view of the relationship between clouds and their favorable large-scale environmental characteristics. 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.

As the perturbations in cloud radiative forcing from anthropogenic aerosols (indirect effect) are typically on the order of  $1\text{ W m}^{-2}$ , which is small compared to the cloud radiative forcing (shortwave radiative effect of  $\sim -47\text{ W m}^{-2}$  and longwave radiative effect of  $\sim 27\text{ W m}^{-2}$ ; Boucher et al., 2013), long integrations are required to produce statistically significant results. The Newtonian relaxation method (nudging) provides a way to estimate AIE within a relatively short integration time, while giving statistically significant results (Lohmann and Hoose, 2009; Kooperman et al., 2012). Nudging here refers to the method of adding a forcing to the prognostic model equations, determined by the

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





---

**On the  
characteristics of  
aerosol indirect effect**

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



5 difference between a model-computed value and a prescribed value at the same time and model grid-cell, to constrain the model results with prescribed atmospheric conditions. Kooperman et al. (2012) implemented nudging to constrain PD and PI simulations toward identical meteorological fields and found that the use of nudging provided  
 10 a more stable estimate of AIE in shorter simulations and increased the statistical significance of the anthropogenic aerosol perturbation signal. All simulations used in this study were nudged toward reanalysis winds provided by operational forecast centers. Some simulations 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 emissions for years  
 15 1850 and 2000 (Lamarque et al., 2010).

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 SPRINTARS. These models show large differences in their aerosol and cloud treatments. For example, while most models (CAM5, CAM5-PNNL, CAM5-MG2, CAM5-CLUBB, CAM5-CLUBB-MG2, ECHAM6-HAM2, and SPRINTARS-KK) use the autoconversion scheme from Khairoutdinov and Kogan (2000, hereafter KK), autoconversion rate in ModelE-TOMAS is independent of cloud droplet number concentration and the Berry scheme (Berry, 1967) is used for SPRINTARS. Most models use diagnostic rain schemes, while an updated Morrison and Gettelman microphysics scheme with a prognostic rain scheme (MG2; Gettelman et al., 2015) is adopted in CAM5-MG2 and CAM5-MG2-CLUBB. HadGEM3-UKCA also adopts a prognostic rain scheme (Abel and Boutle, 2012). While most models only account for aerosol effects on large-scale stratiform clouds, CAM5-CLUBB and CAM5-CLUBB-MG2 use a higher-order turbulence closure (CLUBB) to unify the treatment of boundary layer turbulence, stratiform clouds and shallow convection, and therefore include aerosol effects on shallow convection (Bogenschutz et al., 2013). A brief description of each model is provided in Appendix A.  
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### 3 Results

#### 3.1 Annual mean

We first examine the annual climatology in different simulations to get an overall picture of the general differences/similarities among these models (details within dynamic regimes are examined in Sect. 3.2). All of the simulations reproduce the general pattern of large-scale circulations ( $\omega_{500}$ ): strong ascending motions within the inter-tropical convergence zone (ITCZ) and subsidence dominating subtropical eastern ocean regions (not shown). The similar patterns of  $\omega_{500}$  (due to nudging) in these simulations promise that the frequency of following sorted dynamic regimes does not vary much between models.

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

There are large differences among global LWP annual means. CAM5-MG2 has the lowest LWP among these simulations ( $30.0 \text{ g m}^{-2}$ ). The LWP means over oceans are  $31.1$ ,  $39.4$  and  $35.2 \text{ g m}^{-2}$  in CAM5, CAM5-PNNL and CAM5-CLUBB, respectively. HadGEM3-UKCA simulates higher LWP ( $57.1 \text{ g m}^{-2}$ ) than all versions of CAM5. LWPs in ModelE2-TOMAS ( $80.4 \text{ g m}^{-2}$ ) and ECHAM6-HAM2 ( $84.6 \text{ g m}^{-2}$ ) are greater than the aforementioned GCMs, but less than in SPRINTARS and SPRINTARS-KK ( $139.1$  and  $98.9 \text{ g m}^{-2}$  respectively) which include convective clouds LWP. Even though CAM5-CLUBB simulates a higher LWP in storm track regions and ECHAM6-HAM2 produces

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



much more LWP associated with deep convection in the ITCZ, all models here display reasonable patterns of global LWP distributions (not shown).

The differences in CCN (at 0.1 % supersaturation) among these simulations are not as large as the differences in LWP (Table 2). The global annual mean CCN in CAM5-PNNL is slightly larger than the one in other versions of CAM5. CCN concentrations simulated by CAM5-PNNL, ECHAM6-HAM2 and ModelE2-TOMAS are largest among these simulations and are more than twice those simulated by SPRINTARS, SPRINTARS-KK and HadGEM3-UKCA, which are the lowest. Since these models are using same emissions, differences of CCN between the models are mainly due to different aerosol lifetime between models. We also should note that large differences in CCN do not necessarily correspond to equally large differences in droplet concentration ( $N_d$ ), since  $N_d$  is primarily dependent on cloud base updraft which is an extremely uncertain parameter and may vary significantly between the GCMs.

The LWP response to aerosol perturbations,  $\lambda$ , 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  $\lambda$  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 more sensitive to the aerosol perturbations. LWP is much less sensitive to the changes of CCN in SPRINTARS and SPRINTARS-KK with  $\lambda$  of 0.01 and 0.04 respectively.  $\lambda$  is also small in HadGEM3-UKCA (0.03) due to the large relative increase of CCN while small relative increase of LWP. Since the aerosol effect on precipitation formation is turned off in ModelE2-TOMAS (its autoconversion parameterization is not a function of  $N_d$ ), LWP barely responds to the increase of CCN ( $\lambda$  is  $-0.001$ ). The variation in  $\lambda$  closely follows that of the relative enhancement of LWP ( $d\ln LWP$ ), as the variation of the relative enhancement of CCN ( $d\ln CCN$ ) among the simulations is generally much smaller than that of  $d\ln LWP$ .

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## 3.2 Regime dependence

### 3.2.1 LWP, CCN and $\lambda$

Figure 1 shows LWP and CCN as a function of vertical pressure velocity at 500 hPa ( $\omega_{500}$ ) derived from PD simulations. In general, SPRINTARS (default and KK) simulates much higher LWP in all dynamic regimes and ECHAM6-HAM2/ModelE2-TOMAS in most regimes than different versions of CAM5 runs (default, PNNL, CLUBB and MG2; Fig. 1a), which is consistent with global means in Table 2. A peak of LWP is found around  $\omega_{500} = 0 \text{ hPa d}^{-1}$  in CAM5, ModelE2-TOMAS and ECHAM6-HAM2. For SPRINTARS, LWP decreases from 190 to  $100 \text{ g m}^{-2}$  as  $\omega_{500}$  increases from  $-60$  to  $40 \text{ hPa d}^{-1}$ . In all simulations LWP is low in regimes where  $\omega_{500}$  is larger than  $10 \text{ hPa d}^{-1}$ , i.e., regimes dominated by low clouds. HadGEM3-UKCA simulates larger LWP than CAM5 especially in ascending regimes. Figure 1b shows that CCN concentrations peak at around  $25 \text{ hPa d}^{-1}$  among all the models. This peak is partly caused by little precipitation (and therefore low wet scavenging rate) in subsidence regimes as well as by the fact that these dynamic regimes are located near continents where the sources of anthropogenic aerosols are strong. Furthermore, CCN concentrations are low at around  $0 \text{ hPa d}^{-1}$ , which could be explained by the fact that most regimes around  $0 \text{ hPa d}^{-1}$  are located over the oceans far away from continents (i.e. remote marine aerosols) and anthropogenic aerosol source regions (figures not shown). Generally, CCN in two versions of SPRINTARS and HadGEM3-UKCA is less than other models in most regimes, consistent with Table 2.

All the simulations show positive  $\lambda$  within all dynamical regimes (Fig. 2a), which is consistent with the theory proposed by Albrecht (1989) that an increase in aerosols leads to more liquid cloud water. However, for each configuration,  $\lambda$  can vary significantly between regimes in CAM5 and ECHAM6-HAM2 (Fig. 2a), which indicates that changes in LWP in response to aerosol perturbations are regime-dependent in these GCMs. For example,  $\lambda$  in CAM5-PNNL ranges from 0.35 in strong ascending

---

### On the characteristics of aerosol indirect effect

S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

regions to 0.11 in strong subsidence regimes, which means that LWP in strong ascending regimes is more sensitive to aerosol perturbations than in strong subsidence regimes. Exceptions are ModelE2-TOMAS, SPRINTARS (default and SPRINTARS-KK) and HadGEM3-UKCA, in which  $\lambda$  is low in magnitude. (i.e., LWP changes little in response to the changes of CCN, consistent with the global annual means shown in Table 2).

By comparing these configurations, we note that although the global means of  $\lambda$  in CAM5 and ECHAM6-HAM2 are close, from 0.19 in ECHAM6-HAM2 to 0.25 in CAM5-CLUBB,  $\lambda$  in the different dynamical regimes can differ significantly among these simulations (Fig. 2). For example, LWP in CAM5-PNNL is much more sensitive to CCN perturbations than in ECHAM6-HAM2 in strong ascending regimes; and in strong subsidence regimes, LWP in CAM5-CLUBB and ECHAM6-HAM2 is more sensitive than in CAM5-PNNL and CAM5. Models that use the MG2 with prognostic rain scheme (i.e. CAM5-MG2 and CAM5-CLUBB-MG2) simulate larger  $\lambda$  than the models that use the default MG scheme in most regimes, only except for strong subsidence regimes. However, generally the shapes of the  $\lambda$  distribution are very similar.  $\lambda$  in CAM5-CLUBB-MG2 is large in both ascending and subsidence regimes, which explains the largest  $\lambda$  in CAM5-CLUBB-MG2 among all configurations (Table 2). Except for the models producing very low values of  $\lambda$  (SPRINTARS, SPRINTARS-KK, ModelE2-TOMAS and HadGEM3-UKCA),  $\lambda$  from the other models converges around  $0 \text{ hPa d}^{-1}$  and then diverges greatly in strong 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 agree most, while it is in strong ascending and descending regimes where models differ most. The diversity of  $\lambda$  within dynamical regimes in different GCMs highlights the need to distinguish different dynamical regimes in studying AIE.

When analyzing the numerator and denominator of  $\lambda$  separately, we found that this large spread in  $\lambda$  is mainly contributed by the numerator,  $\text{dlnLWP}$ .  $\text{dlnLWP}$  ranges from about 0 to 0.22 among the models (Fig. 2a) while the denominator  $\text{dlnCCN}$ , is more stable than  $\text{dlnLWP}$  within dynamical regimes and fluctuates around 0.45, except for

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



larger  $\text{dlnCCN}$  in HadGEM3-UKCA (Fig. 2b). In summary, the ratio of  $\text{dlnLWP}$  to  $\text{dlnCCN}$  ( $\lambda$ ) therefore changes more consistently with  $\text{dlnLWP}$  within dynamical regimes.

The decreasing trends of  $\lambda$  with increasing  $\omega$  in CAM5, CAM5-MG2, and CAM5-PNNL are similar, which is opposite to the increasing trends derived from ECHAM6-HAM2, CAM5-CLUBB and CAM5-CLUBB-MG2. It is interesting that the regime-dependence of  $\lambda$  simulated by CAM5-CLUBB and CAM5-CLUBB-MG2 is quite different from that simulated by CAM5, CAM5-MG2, and CAM5-PNNL even though all these 5 model versions are originally from CAM5 and share many similarities. In CAM5, CAM5-MG2 and CAM5-PNNL, three separate parameterization schemes are used to treat planetary boundary layer (PBL) turbulence, stratiform cloud microphysics and shallow convection. In CAM5-CLUBB and CAM5-CLUBB-MG2, instead, a higher-order turbulence closure, Cloud Layers Unified by Binormals (CLUBB), is adopted to replace these three separate schemes to provide a unified treatment of these processes (Bogenschutz et al., 2013). A major improvement of CAM-CLUBB is the better simulation of the transition of stratocumulus to *ture 2a* shows that  $\lambda$  in CAM5-CLUBB and CAM5-CLUBB-MG2 is quite different from that in CAM5 simulations without CLUBB (i.e., CAM5, CAM5-MG2 and CAM5-PNNL) in regimes where  $\omega_{500}$  is larger than  $10 \text{ hPa d}^{-1}$ . Under such suppressed conditions, low clouds such as trade wind cumulus and stratocumulus are typically formed. This higher  $\lambda$  might be expected because CAM5-CLUBB 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  $\text{dlnLWP}$  differs most in Fig. 2b, we plot the global distribution of annual averaged  $\text{dlnLWP}$  in different simulations, shown in Fig. 3. The ascending regimes where

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ECHAM6-HAM2 differs significantly from the two CAM5 configurations (CAM5, CAM5-PNNL) are located over the North Pacific Ocean (from 30 to 60° N), where storm tracks prevail, for weak ascending motions and the Southern coast of Asia for strong ascending motions. The spatial patterns in ECHAM6-HAM2, CAM5-CLUBB, CAM5-CLUBB-MG2 and HadGEM3-UKCA share some similarities over Northern Pacific Ocean, but the magnitude in CAM5-CLUBB and CAM5-CLUBB-MG2 is larger than in ECHAM6-HAM2 and HadGEM3-UKCA. Moreover, not only the pattern but also the magnitude of  $\text{dlnLWP}$  in ECHAM6-HAM2 differ significantly from those in CAM5, CAM5-MG2 and CAM5-PNNL. For the Southern coast of Asia where strong ascending motions dominate, all simulations show a relative increase of LWP. However,  $\text{dlnLWP}$  in ECHAM6-HAM2 in this region is much smaller than in all CAM5 simulations. This makes  $\text{dlnLWP}$ , and thus  $\lambda$ , in ECHAM6-HAM2 much less than in the five CAM5 models (CAM5, CAM5-MG2, CAM5-PNNL, CAM5-CLUBB and CAM5-CLUBB-MG2) in ascending regimes, as shown in Fig. 2b and a.

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 perturbations, the spatial patterns of  $\text{dlnLWP}$  in these four simulations shown in Fig. 3 are indeed different from each other. HadGEM3-UKCA simulates larger  $\text{dlnLWP}$  in middle northern subtropical oceans, which is similar to CAM5-CLUBB and ECHAM6-HAM2 but with smaller magnitude. However, the pattern in SPRINTARS is unlike any models discussed above. SPRINTARS simulates larger  $\text{dlnLWP}$  over the North Pacific Ocean, the North Atlantic Ocean and the western coasts of continents than other parts of the global ocean. SPRINTARS-KK simulates the same pattern as SPRINTARS only with larger values. Meanwhile,  $\text{dlnLWP}$  in ModelE2-TOMAS shows no special global pattern and the values are all near zero, which indicates LWP in ModelE2-TOMAS has indeed little response to aerosol perturbations as autoconversion rate in ModelE2-TOMAS is not influenced by cloud droplet number concentrations.

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



On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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  $\text{dlnLWP}$  in ECHAM6-HAM2 and HadGEM3-UKCA is not as large as that in CAM5-CLUBB and CAM5-CLUBB-MG2. Another difference among these models is in regions dominated by more intensive subsidence, over Western coasts of North America, South America and Africa. In these regions  $\text{dlnLWP}$  in ECHAM6-HAM2 and the two CAM5 models with CLUBB is large while it is small in the three CAM5 models without CLUBB.

To examine the cloud lifetime effect in different cloud regimes more specifically, another criterion, lower-tropospheric stability ( $\text{LTS} = \theta_{700\text{hPa}} - \theta_{\text{surface}}$ ), is added to distinguish stratocumulus from trade wind cumulus regimes, following Medeiros and Stevens (2011). Table 3 lists the criteria of different low cloud types conditionally sampled by  $\omega_{500}$  and LTS. The annual mean cloud fractions of each low cloud type in CAM5-CLUBB are shown in Fig. 4; the distributions in other simulations are generally similar to CAM5-CLUBB (figures not shown). The cloud type distribution is consistent with satellite observations that stratocumuli occurs over subtropical oceans near western continents while trade wind cumuli dominate over oceans further away from continents (Medeiros and Stevens, 2011). Figure 4 shows that some differences in  $\text{dlnLWP}$  between models shown in Fig. 3 are located at regions dominated by low clouds (i.e., stratocumulus and trade wind cumulus).

The joint distributions of LTS and  $\omega_{500}$  over global oceans between  $60^\circ\text{S}$  and  $60^\circ\text{N}$  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  $\omega$  intervals. LTS ranges from 8 to 24 K while  $\omega$  ranges from  $-100$  to  $60\text{ hPa d}^{-1}$ . Instances with slight downward vertical motions and moderate LTS are most frequent.

Figure 5 shows that, though  $\omega_{500}$  plays the primary role in determining the  $\text{dlnLWP}/\text{dlnCCN}$  distribution, LTS can reveal further details of the differences among





tion rate are used in calculating AUTO/PRECL. Generally the curves of AUTO/PRECL are smoother than  $\lambda$  (Fig. 6a and 2a). The ratio from different simulations shows large diversity in ascending regimes and subsidence regimes. In all versions of CAM5 and SPRINTARS the ratio decreases with increasing  $\omega_{500}$  in ascending regimes and then increases in descending regimes. However, the ratio in ECHAM6-HAM2 remains unchanged in ascending regimes and then increases under subsidence. As discussed above,  $\lambda$  was shown to be highly correlated with this ratio from global average results (Wang et al., 2012). According to our results, the correlation also applies well for individual dynamical regimes in ECHAM6-HAM2 and CAM-CLUBB, in which the correlation coefficients between  $\lambda$  and AUTO/PRECL are 0.98 and 0.86 respectively. However, these high correlation coefficients are not found in other simulations, in which the correlation coefficients are lower than 0.7, which indicates that the relationship of AUTO/PRECL and  $\lambda$  in these models is changing from regime to regime, in other word, this relationship is regime-dependent.

Wang et al. (2012) and Gettelman et al. (2013) found that the diagnostic rain scheme used in the CAM configurations might overestimate the role of autoconversion over accretion. Gettelman et al. (2015) found using instantaneous microphysical process rates that adding the new microphysics with prognostic precipitation to cloud scheme (MG2) decreases the ratio of autoconversion to accretion. By sorting into dynamical regimes, resulted in moderate regimes ( $-20 \text{ hPa d}^{-1} < \omega_{500} < 10 \text{ hPa d}^{-1}$ ) in CAM5 is consistent with Gettelman et al. (2015), which show larger AUTO/PRECL in CAM5 than CAM5-MG2. However, in other regimes of CAM5 and all regimes of CAM5-CLUBB, adding the prognostic precipitation (MG2) increases the ratio of AUTO/PRECL. The result of larger AUTO/PRECL in some regimes of models with MG2 seems different from the results of Gettelman and Morrison (2015) in idealized tests of MG2 and of Gettelman et al. (2015) in CAM simulations with MG2. We have verified using the same model output from Gettelman et al. (2015) that the difference is not due to the simulations performed. The difference is likely due to (a) the use of instantaneous output in Gettelman et al. (2015) for process rate comparisons while monthly data is

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



used here. (b) Microphysics variables and precipitation are sorted by  $\omega_{500}$  here while Gettelman et al. (2015) sorted them by LWP that the microphysics sees, which includes contributions from deep convection. (c) Vertical integrals of autoconversion rate are used here while Gettelman et al. (2015) were using vertical averages.

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

Note that low precipitation situations are only found in subsidence regimes ( $\omega_{500} > 0 \text{ hPa d}^{-1}$ ). 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  $\lambda$  and the fractional occurrences of each precipitation situation in descending regimes. The fractional occurrence of low precipitation increases evidently in subsidence regimes, compared with that over global ocean. We find that the averages of  $\lambda$  under low precipitation are larger than those under high precipitation in most models (CAM5, CAM5-PNNL, CAM5-CLUBB, SPRINTARS, SPRINTARS-KK and HadGEM3-UKCA) (Table 4). This result is different from some LES and single column model (SCM) results showing that smaller  $\lambda$  values are found for low surface precipitation rather than

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



(Fig. 7), consistent with the argument that prognostic precipitation schemes reduce aerosol indirect forcing (Posselt and Lohmann, 2009; Wang et al., 2012; Gettelman and Morrison, 2015). However, adopting a prognostic precipitation scheme is found to increase dSCRE under low precipitation situations. This is partly from the increase in the occurrence frequency of low precipitation instances when MG2 is adopted (Table 4).

## 4 Summary

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

The susceptibility of liquid water path (LWP) to aerosol perturbations,  $\lambda = \text{dlnLWP}/\text{dlnCCN}$ , is a metric that quantitatively measures cloud lifetime effect of aerosols (Wang et al., 2012). The diversity of  $\lambda$  within dynamical regimes indicates that the aerosol cloud lifetime effect is regime-dependent. Despite the closer global means,  $\lambda$  shows a large spread within dynamical regimes among GCMs. The changes of LWP in subsidence regimes are more sensitive to CCN perturbations than those in ascending regimes in CAM5-CLUBB, ECHAM6-HAM2 and SPRINTARS while the opposite is true in CAM5, CAM5-MG2 and CAM5-PNNL. This large spread indicates that the aerosol indirect effect associated with cloud regimes is not well represented in current climate models.

It is in strong ascending regimes and subsidence regimes that  $\lambda$  differs most between GCMs. Our results further show that aerosol indirect forcing (change in short-wave cloud radiative effect from PI to PD, dSCRE) in regimes of vertical ascent is close

### On the characteristics of aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

to, or even larger than in low cloud regimes (Fig. 7). While recent studies on aerosol indirect effects mostly focus on the stratocumulus regimes due to their significant cooling effect, our result highlights that regimes of vertical ascent should be another important regime to focus on in the future to reduce the uncertainties in the estimate of AIE in climate models. Note 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 separate different low cloud types under large-scale subsidence. It is found that the large  $\lambda$  in subsidence regimes in CAM5-CLUBB and ECHAM6-HAM2 is caused by both stratocumulus and trade wind cumulus. Nevertheless, in ascending regimes,  $\lambda$  in CAM5-CLUBB is large and is more similar to CAM5 and CAM5-PNNL than to ECHAM6-HAM2. We also found that in ascending regimes simulations with the MG2 scheme (CAM5-MG2 and CAM5-CLUBB-MG2) exhibit stronger sensitivity of LWP in response to aerosol perturbation than simulations without the MG2 scheme (CAM5 and CAM5-CLUBB). As for low cloud regimes,  $\lambda$  is larger in transitional cloud regimes than stratocumulus cloud regimes and trade wind cloud regimes in CAM5-CLUBB-MG2, different from CAM5-CLUBB. LWP in HadGEM3-UKCA is more sensitive to aerosol perturbation in transitional cloud regime and stratocumulus regime than trade wind cloud regime. The distribution of  $\lambda$  in SPRINTARS and SPRINTATSKK is more likely to depend on LTS rather than vertical pressure velocity.

Our results show that  $\lambda$  under low precipitation situations (monthly-mean surface precipitation rate less than  $0.1 \text{ mm d}^{-1}$ ) is larger than under high precipitation situations (monthly-mean surface precipitation rate larger than  $0.1 \text{ mm d}^{-1}$ ) in all models except for models with prognostic rain scheme (MG2). Results derived from large eddy simulation (LES) and single column model (SCM; e.g., Ackerman et al., 2004; Guo et al., 2011) where  $\lambda$  could be negative under low precipitation situations and thus is expected to be smaller than under high precipitation situations. Further efforts are

---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

needed to understand the differences among different models and the difference between global model results and results from process-level studies. It is further found that high precipitation situations constitute most of dSCRE (from 64 % in CAM5-MG2 to nearly 100 % in CAM5-CLUBB, Fig. 7) and the contributions from low precipitation situations are relatively small, ranging from 0 to 36 %.

Adding prognostic precipitation (MG2) to cloud microphysical scheme for global climate model can reduce the shortwave cloud radiative effect (SCRE) only for high precipitation situations. Considering low precipitation situations are much less prevalent than high precipitation situations, total SCRE appears to be decreased in models with prognostic rain scheme than with diagnostic rain scheme.

The regime categorization used in this study is derived from monthly mean data. Giving the high variability of precipitation and microphysics processes on short time scales, we acknowledge that instantaneous data (e.g. 3 hourly) might provide more reliable information. For example, instantaneous data may help to reconcile some of discrepancies between our studies and that of Gettelman et al. (2015) regarding the prognostic rain scheme noted in Sect. 3.2.2. However, it is challenging to calculate  $\lambda$  and aerosol indirect forcing using instantaneous data. Here  $\lambda$  and aerosol indirect forcing are derived from the difference between present day (PD) and pre-industrial (PI) simulations. Using instantaneous data will not guarantee that the sorted bins of dynamical regimes include the same instances from PI to PD, giving the high variability of instantaneous data. Since the main goal in this manuscript is to demonstrate the importance of examining aerosol indirect effects in different cloud and dynamical regimes, the use of monthly-mean data serves this goal well. 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  $\lambda$  in regimes of vertical ascend in different models.



## Appendix A: Global aerosol-climate models

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

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

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).

### On the characteristics of aerosol indirect effect

S. Zhang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



## On the characteristics of aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 aerosol-chemistry climate model. In this study only the global aerosol-climate model part of ECHAM-HAMMOZ is used and for the sake of brevity referred to as 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 HAM2 (Stier et al., 2005; Zhang et al., 2012) and uses a two-moment cloud microphysics scheme that includes prognostic equations for the cloud droplet and ice crystal number concentrations as well as cloud water and cloud ice (Lohmann et al., 2007; Lohmann and Hoose, 2009). The activation of aerosol particles into cloud droplets is parameterized by Lin and Leitch (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 transport-climate model based on a general circulation model, MIROC (Watanabe et al. 2010). In this study, the horizontal and vertical resolutions are T106 ( $1.125^\circ \times$  approx.  $1.125^\circ$ ) and 56 layers, respectively. SPRINTARS is coupled with the radiation and cloud microphysics schemes in MIROC to calculate the aerosol-radiation and aerosol-cloud

interactions. A prognostic scheme for determining the cloud droplet and ice crystal number concentrations is introduced (Takemura et al. 2009). The default autoconversion scheme in MIROC-SPRINTARS is based on Berry (1967), and the activation of aerosol particles into cloud droplet is based on the ARG scheme.

5   SPRINTARS-KK: This is the same as SPRINTARS, but the default autoconversion scheme in SPRINTARS is replaced with the KK autoconversion 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 sigma layers from the surface to 0.1 hPa (80 km). In the model, clouds are distinguished into convective and large-scale stratiform clouds. The clouds parameterizations are similar to Del Genio (1993) and Del Genio (1996) but have been improved in several respects (see details in Schmidt, 2014; Schmidt, 2006). Using a prognostic treatment of cloud droplet number concentration (CDNC) from Morrison and Gettleman (2008), ModelE2-TOMAS represents the first aerosol indirect effects only on large-scale stratiform clouds (Menon et al., 2010). In ModelE2-TOMAS, CDNC and a critical supersaturation are computed using a physical-based activation parameterization from Nenes and Seinfeld (2002) with a model updraft velocity that is computed based on a large-scale vertical velocity and sub-grid velocity.

HadGEM3-UKCA: HadGEM3-UKCA is a global composition climate model (<http://www.ukca.ac.uk>). It consists of the third generation of the Hadley Centre Global Environmental Model (Hewitt et al, 2011) developed at the UK Met Office. This general circulation model is non-hydrostatic and uses a semi-Lagrangian transport scheme. We are using the atmospheric configuration: General Atmosphere (GA) 4.0 as documented in Walters et al. (2014), except for the addition of the UKCA aerosol and chemistry scheme which is fully coupled with the radiation scheme of HadGEM3 (Belouin et al., 2013). UKCA is a two-moment pseudo-modal scheme which carries both

---

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

aerosol number concentration and component mass as prognostic tracers. It calculates the evolution of five aerosol species, sulfate, particulate organic matter, black carbon, sea salt and dust, in both internally and externally mixed particles. The aerosol scheme in UKCA is based on the Global Model of Aerosol Processes (GLOMAP-mode, Mann et al., 2010). The main exception is that dust is calculated separately using 6 size bins. UKCA hence only considers 5 modes. The tropospheric chemistry part of UKCA is described in 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 cloud scheme (PC2; Wilson et al., 2008), in which the cloud droplet number concentration is diagnosed from the expected number of aerosols that are available to activate at each timestep (West et al., 2014). Cumulus convection is represented by a mass flux convection scheme based on Gregory and Rowntree (1990) with various extensions (Walters et al., 2014). Simulations were performed at N96L85 resolution, a regular  $1.25^\circ$  latitude  $\times$   $1.875^\circ$  longitude grid in the horizontal, with 85 hybrid-height vertical levels.

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On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**On the  
characteristics of  
aerosol indirect effect**

---

S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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---

**On the  
characteristics of  
aerosol indirect effect**

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**On the  
characteristics of  
aerosol indirect effect**

---

S. Zhang et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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---

**On the  
characteristics of  
aerosol indirect effect**

S. Zhang et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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---

**On the  
characteristics of  
aerosol indirect effect**S. Zhang et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**On the  
characteristics of  
aerosol indirect effect**

S. Zhang et al.

**Table 1.** The types of clouds included in liquid water path (LWP) and surface rain rate and different rain schemes in 10 participating models.

Model	LWP	Rain	Rain scheme
CAM5	$S^a$	$S$	$d^c$
CAM5-MG2	$S$	$S$	$p^d$
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	$p$
ECHAM6-HAM2	$S$	$S$	$d$
SPRINTARS	$S + C^b$	$S + C$	$d$
SPRINTATRS-KK	$S + C$	$S + C$	$d$
ModelE2-TOMAS	$S$ +anvil clouds	$S$ +anvil clouds	$d$
HadGEM3-UKCA	$S + C$	$S$	$p$

<sup>a</sup>  $S$  in LWP and Rain stands for stratiform clouds. <sup>b</sup>  $C$  in LWP and Rain stands for convective clouds. <sup>c</sup>  $d$  in Rain schemes represents diagnostic rain scheme. <sup>d</sup>  $p$  in Rain schemes represents prognostic rain scheme.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## On the characteristics of aerosol indirect effect

S. Zhang et al.

**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 ( $\lambda$ , dlnLWP/dlnCCN) of the 10 GCM simulations.

Model	$\lambda$	LWP ( $\text{g m}^{-2}$ )	CCN ( $10^{11} \text{ m}^{-2}$ )	dlnLWP	dlnCCN	PRECL ( $\text{mm d}^{-1}$ )	SCRE ( $\text{W m}^{-2}$ )
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

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)

[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


## On the characteristics of aerosol indirect effect

S. Zhang et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Table 3.** Criteria used to conditional sampling stratocumulus, transitional clouds and trade wind cumulus regimes (adopted from Medeiros and Stevens, 2011).

	Stratocumulus	Transitional clouds	Trade wind cumulus
LTS (K)	$LTS \geq 18.5$	$18.5 > LTS \geq 15.4$	$15.4 > LTS \geq 11.3$
$\omega_{500 \text{ hPa}}$ ( $\text{hPa d}^{-1}$ )	$\omega_{500 \text{ hPa}} > 10$	$\omega_{500 \text{ hPa}} > 10$	$\omega_{500 \text{ hPa}} > 10$

## On the characteristics of aerosol indirect effect

S. Zhang et al.

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

Model	$\lambda^a$	$\lambda^b$	$f^c$	$f^d$	$f^e$	$f^f$
	low, descend	high, descend	low, descend	high, descend	low, glb	high, 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

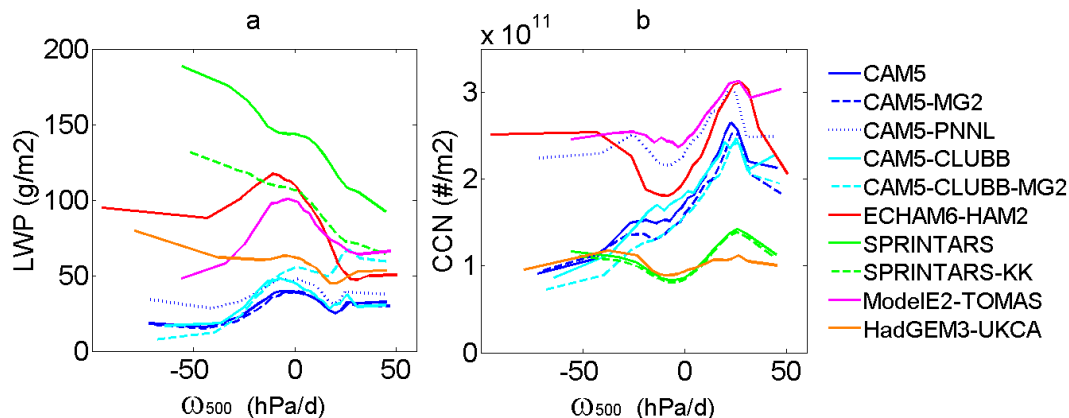
<sup>a</sup>  $\lambda$  under low PRECL for descend regimes. <sup>b</sup>  $\lambda$  under high PRECL for descend regimes. <sup>c</sup> Fractional occurrence of low PRECL for descend regimes. <sup>d</sup> Fractional occurrence of high PRECL for descend regimes. <sup>e</sup> Fractional occurrence of low PRECL over global regimes. <sup>f</sup> Fractional occurrence of high PRECL over global regimes.





## On the characteristics of aerosol indirect effect

S. Zhang et al.



**Figure 1.** (a) LWP and (b) column-integrated CCN (at 0.1% supersaturation) as a function of 500 hPa vertical pressure velocity ( $\omega_{500}$ ) 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).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

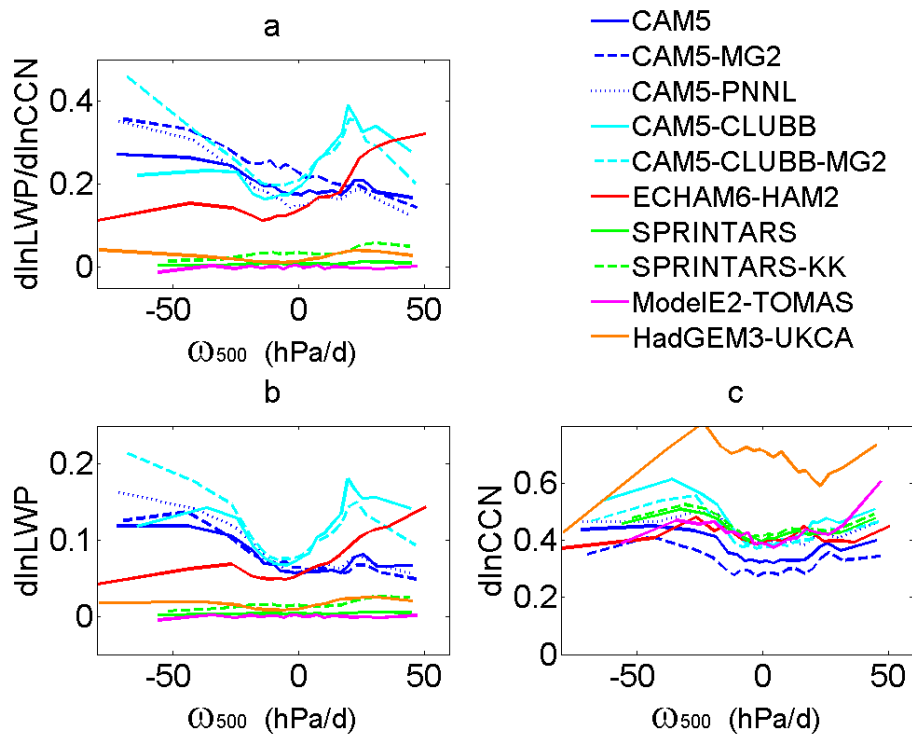
Printer-friendly Version

Interactive Discussion



## On the characteristics of aerosol indirect effect

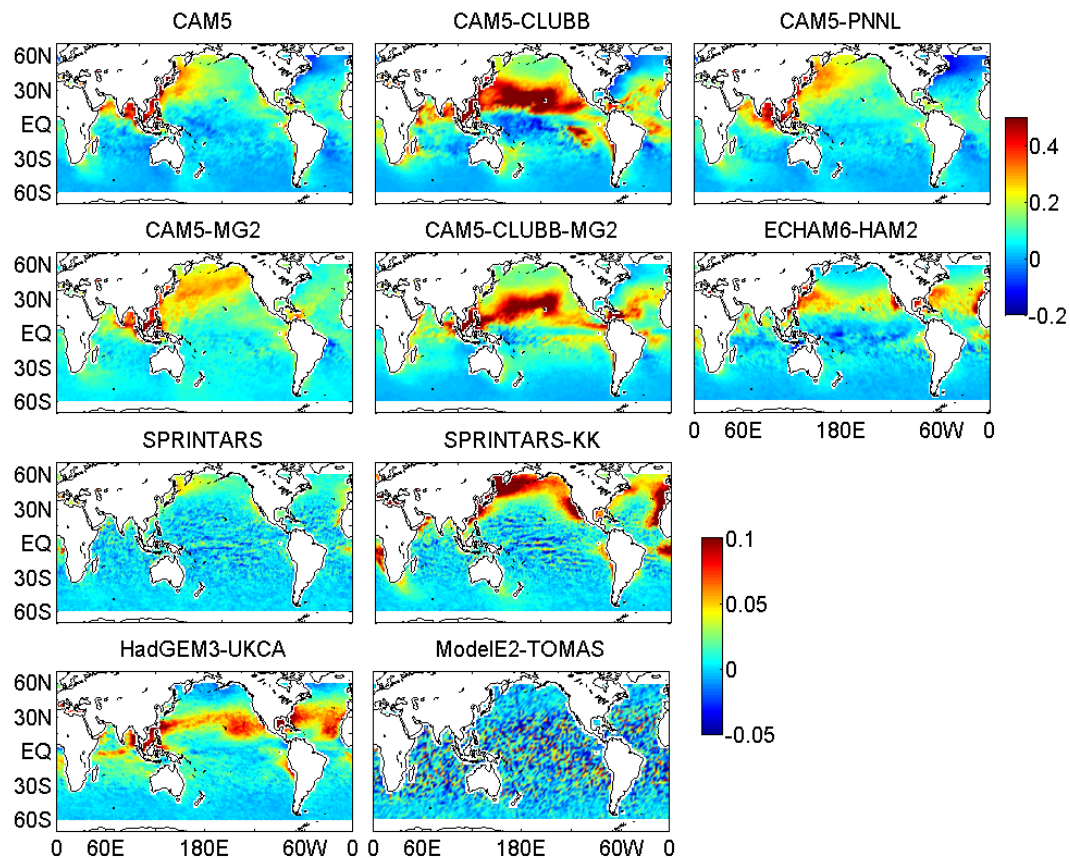
S. Zhang et al.



**Figure 2.** Same as Fig. 1a, but for **(a)** the sensitivity of LWP to the change of CCN ( $\lambda$ ), **(b)** relative enhancement of liquid water path ( $d\ln LWP$ ) and **(c)** relative enhancement of cloud condensation nuclei ( $d\ln CCN$ ) from pre-industrial (PI) to present day (PD).

## On the characteristics of aerosol indirect effect

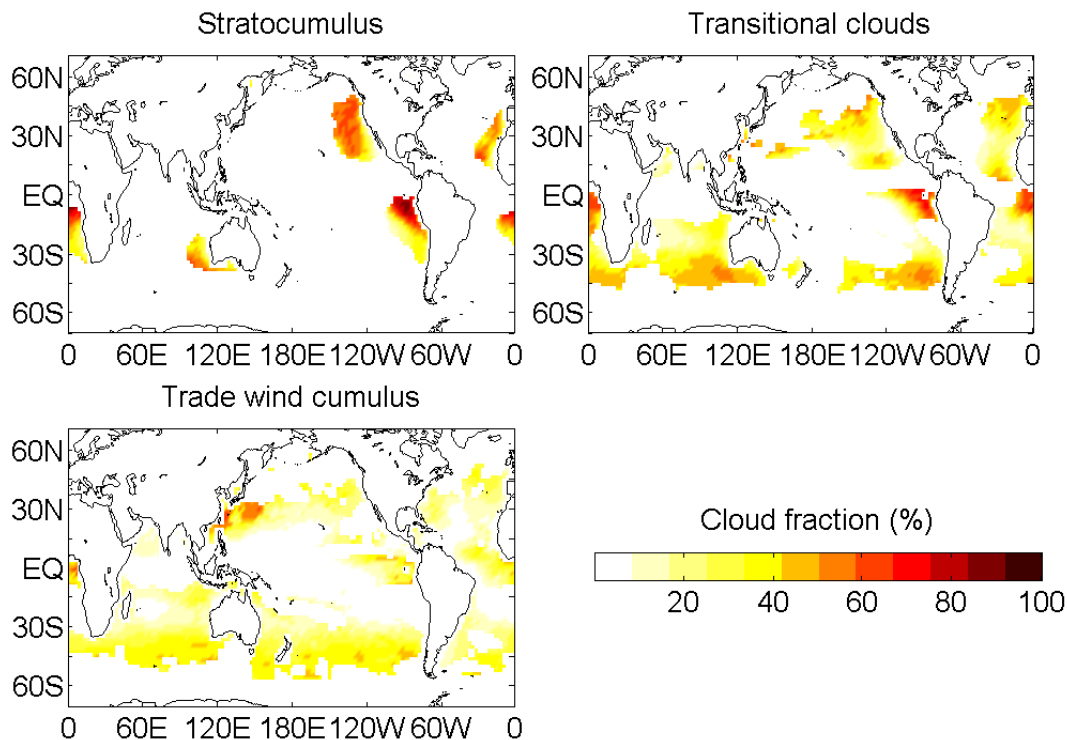
S. Zhang et al.



**Figure 3.** Relative change of annual averaged LWP from PI to PD ( $d\ln LWP$ ) simulations derived from the 10 GCM simulations.

On the  
characteristics of  
aerosol indirect effect

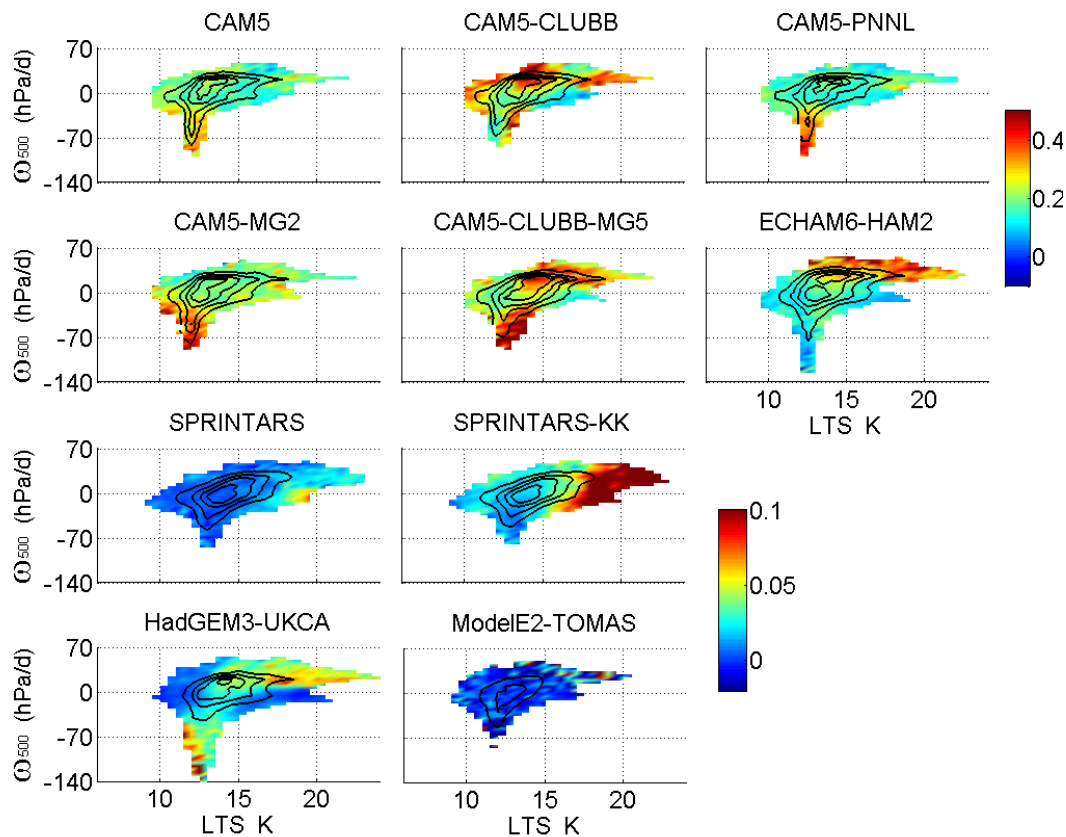
S. Zhang et al.



**Figure 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.

On the  
characteristics of  
aerosol indirect effect

S. Zhang et al.



**Figure 5.**  $d\ln LWP/d\ln CCN$  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.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

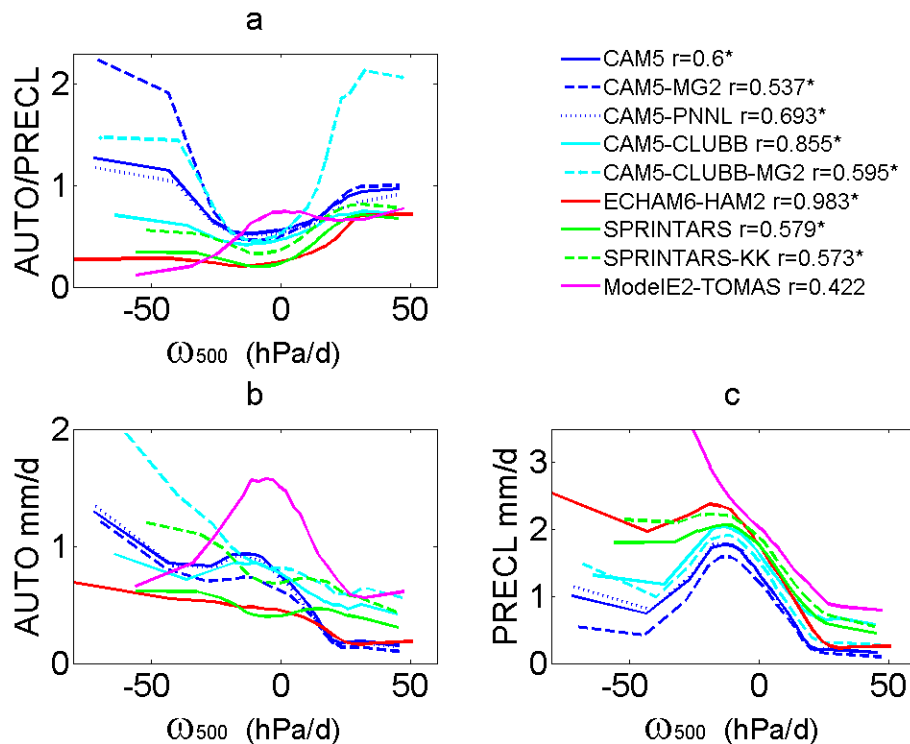
Printer-friendly Version

Interactive Discussion

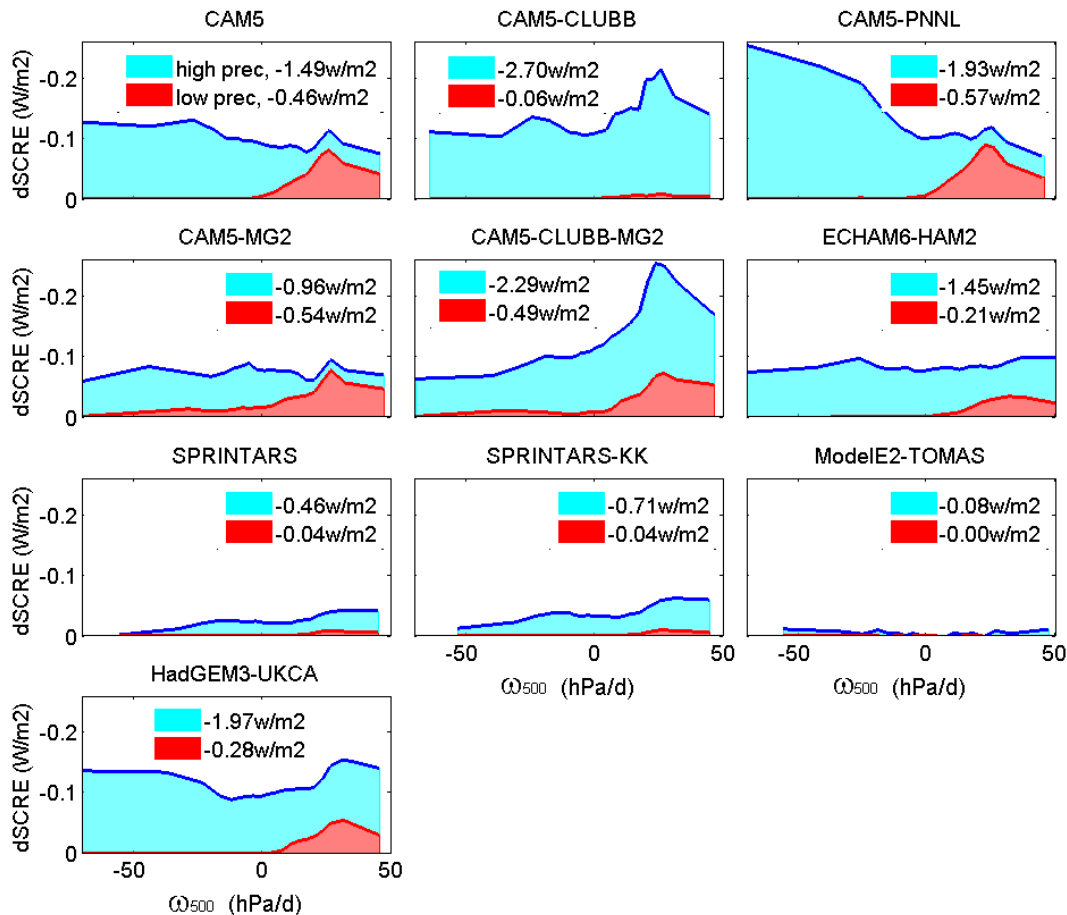


## On the characteristics of aerosol indirect effect

S. Zhang et al.



**Figure 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 (autoconversion rate data is not available in HadGEM3-UKCA, so not included here). The number marked in each simulation is the corresponding correlation coefficient between AUTO/PRECL and  $\lambda$  and number with mark “\*” indicates the correlation is significant (at 95% confidence).



**Figure 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.