DEPARTMENT OF PHYSICS
ATMOSPHERIC, OCEANIC AND PLANETARY PHYSICS

Prof. Philip Stier
Professor of Atmospheric Physics
Climate Processes Group

Clarendon Laboratory, Parks Road, Oxford OX1 3PU
Tel: +44(0)1865 272887 Fax: +44(0)1865 272923
philip.stier@physics.ox.ac.uk http://www.physics.ox.ac.uk/research/climate-processes

Dr Graham Feingold
Editor
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Dear Graham,

Subject: Revised manuscript

Thank you for extending the deadline for the resubmission of my manuscript "Limitations of passive remote sensing to constrain global cloud condensation nuclei". I am attaching a revised manuscript for your consideration.

I believe that the performed relatively minor changes improved the manuscript while at the same time not altering any of the main conclusions.

With best regards,

Philip

Philip Stier

Atmos. Chem. Phys. Discuss., 15, C13599–C13609, 2016
www.atmos-chem-phys-discuss.net/15/C13599/2016/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Limitations of passive satellite remote sensing to constrain global cloud condensation nuclei" *by* P. Stier

P. Stier

philip.stier@physics.ox.ac.uk

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I would like to thank the reviewer for the helpful comments that substantially improved the manuscript. I very much appreciated the detailed remarks and hope to have addressed all raised issues.

Specific comments

1. Title: "Limitations of passive satellite remote sensing to constrain global cloud condensation nuclei". This paper presents a theoretical study whose results suggest certain uncertainty in satellite data interpretation, assuming the numerical simulation well represents the Earth System. For the sake of accuracy, I recommend leaving titles in that spirit to studies based on observed data,

technical instrumentation limitations etc.

I do not fully understand to what extend this request is based on "accuracy". This work demonstrates that a key assumption used in many papers based on passive remote sensing (including our own work!) has significant limitations. As such, the title seems quite appropriate to me.

However, I have removed the word "satellite" in the revised manuscript as the same limitations apply to ground based passive remote sensing, e.g. from AERONET.

2. P. 32611 lines 26-29: "Therefore, use of this model allows to consistently assess the relationship between aerosol radiative properties and CCN as biases in the simulated fields are expected to be consistent". Please elaborate on the reasons for expected consistent biases in the simulated fields. What perturbations or errors are experienced in such simulation?

All aerosol models (even the ones used in the forward models of satellite retrievals) are subject to uncertainties in terms of the representation of aerosol amount, composition, size, mixing-state and radiative properties. In ECHAM-HAM, the diagnostics of CCN at various supersaturations is calculated from the prognostic size-distribution, mixing-state and composition. Aerosol radiative properties are calculated via Mie theory from the same prognostic size-distribution, mixing state and composition. Therefore any bias in e.g. the size or composition of a mode is consistent between the calculation of CCN and the corresponding radiative properties.

3. P. 32612 lines 1-2: "Nonetheless, it should be noted that the ability of models to mimic the spatial (in particular vertical) and temporal (co-)variability of aerosol and humidity fields introduces some uncertainty". Please give the reader some quantitative sense of the model uncertainties, in respect to CCN and aerosol optical depth, as required when comparing to other datasets.

The meteorology of the ECHAM base model has been extensively evaluated in Stevens

et al. (2013). ECHAM-HAM2 has been extensively evaluated against observations in Zhang et al., ACP, (2012) and its aerosol representation analysed in detail in Schutgens and Stier (2014). Unfortunately, currently no datasets exist to satisfactory evaluate CCN in global models. While we have compared our model against the most comprehensive published compilation of CCN datasets (Spracklen et al., 2011), our recent work on the importance of spatio-temporal sampling errors (Schutgens et al., ACP, 2016; Schutgens et al., ACPD, 2016) highlights the significance of sampling errors that can easily dwarf measurement errors or even model errors. I therefore refrain from publishing error estimates for which we cannot attribute the differences to model errors.

To overcome this unsatisfactory situation, we are working with partners in the Global Aerosol Synthesis and Science project on creating the largest consistent database of CCN and CCN related measurements (http://gassp.org.uk). We expect to be able to significantly advance the evaluation of CCN in global aerosol models.

4. P. 32615 lines 9-13: "We further investigate the role of the vertical aerosol distribution using the local (model layer) aerosol extinction coefficient (AEC) as well as the extinction aerosol index (AIAEC), defined here as local aerosol extinction coefficient times the local Ångström parameter". Please add more details regarding the "extinction aerosol index", which is presented for the first time. What's the nature of this metric and in what units (e.g. is it normalized by mass or not)? Besides the better correlation we see later in the paper – what is the physical logic behind the choice of that product?

The extinction aerosol index is defined as the local (not column integrated) equivalent of the commonly used Aerosol Index (AI). This is now properly defined in the introduction:

"We further investigate the role of the vertical aerosol distribution using the local (model layer) aerosol extinction coefficient (AEC) as well as the extinction aerosol index (AI$_{AEC}$), defined here as local aerosol extinction coefficient times the local Ångström

parameter: AI$_{AEC}$=AEC$\times\alpha_{AEC}$, where $\alpha_{AEC} = -\frac{\ln(AEC_{550nm}/AEC_{865nm})}{\ln(\lambda_{550nm}/\lambda_{865nm})}$ is evaluated from the local aerosol extinction coefficients, instead of from the column integrated aerosol optical depths used in AI. "

The motivation for the use of the AI$_{AEC}$ instead of AEC is the same as for AI instead of AOD: multiplication by the Angström parameter gives (generally) lower weight to larger particles to account for the fact that CCN numbers (in particular at higher supersaturation) are often dominated by Aitken mode sized particles.

5. P. 32616 line 5: "The ECHAM-HAM simulated annual-mean surface CCN concentrations (Fig. 1) show distinct land–sea contrast, with maxima over the main aerosol source areas". The colour scale of Fig. 1 does not ease the "distinct" observation of land- continent contrast. Please modify the colour scale (i.e by using logarithmic scale to focus on variance in low concentrations), or alternatively add calculated values, indicating that contrast.

The main purpose of Fig. 1 and Fig. 2 is to highlight the difference in the geographical spread of CCN at different supersaturations vs. the spread of aerosol radiative properties. The colorbar has been designed to be consistent across the different aerosol properties. None of the (many) colourbars I tried is perfect but the one in the revised manuscript is probably a bit better than the original one.

6. P. 32617 lines 13-16: "Note that maps of global correlations for alternative aerosol radiative properties proposed as superior proxies of CCN (Fig. 7), specifically (a) fine mode aerosol optical depth, (b) dry aerosol optical depth and (c) aerosol index do not show significantly improved correlations.". In spite claim (c), it seems the Fig. 7(c) has the best correlation in the panel. Global regional mean correlation values of those maps (over continents) would better make the point.

This is a good point. I have now included global mean correlation coefficients in the title of each figure and discuss its variation quantitatively:

"A number of alternative aerosol radiative properties have been proposed to provide superior proxies of CCN. Note that maps of their correlations and the corresponding global mean values (Fig. 7), specifically of (b) fine mode aerosol optical depth ($\overline{r} = 0.50$), (c) dry aerosol optical depth ($\overline{r} = 0.45$) and (d) aerosol index ($\overline{r} = 0.53$) do not show significantly improved correlations as compared to (a) aerosol optical depth ($\overline{r} = 0.44$). Usage of the non-parametric Spearman's rank correlation coefficient (e) gives very similar correlations ($\overline{\rho} = 0.41$). Sampling $CCN_{0.2\%}$ at the model simulated lowest cloud base gives slightly reduced ($\overline{r} = 0.36$) but spatially very similar correlations with AOD (f). "

7. P. 32618 lines 15-20: "This is likely due to the fact that not only aerosol water uptake but also aerosol removal via scavenging is positively correlated to relative humidity (via clouds and precipitation). This hypothesis is supported by the drop off of this correlation around and below cloud base (green line). However, correlations of column integrated AOD and surface CCN are consistently high for this region as well as for the northern high-latitude oceans.". Having ECHAM6 fully running, it should be simple to add precipitation maps (or values) and easily support this hypothesis. Such comparison would also strengthen the reliability of ECHAM6 model for this study.

It would be easy to add precipitation maps or values but these would be very similar to the quite detailed evaluation of ECHAM6 precipitation published in the ECHAM6 evaluation paper (e.g. Fig. 5,7,8,9 in Stevens et al., JAMES, 2013). However, evaluation of the base state provides only limited insight into the covariability of relative humidity, precipitation and aerosol extinction for which no suitable observations with sufficient coverage exist. In principle, this could have been investigated using dedicated sensitivity studies with ECHAM-HAM but that would have required to change the model aerosol radiation code to compute and output 3D fields of dry aerosol extinction. In the light of the minor relevance of this statement for the overall conclusions of the paper I have not taken on this fairly substantial task.

8. P. 32619 lines 7-11: "Note that also correlations between surface layer CCN and AIAEC deteriorate for higher supersaturations (sampling smaller particles of the aerosol size distribution), as expected from Mie theory, as the smaller particles contribute less to total extinction (Fig. 10). This is particularly evident over the continents with significant primary fine mode aerosol emissions.". This statement is inaccurate. In many cases, as expected from Mie theory, particle populations of smaller sizes contribute more to total extinction. Please see Fig. 1 below for example, showing simulated extinction coefficients for black carbon aerosol as a function of the population's mass concentration and mean radius, simulated using SHDOM (Evans, 1998). For this calculation, aerosol size distribution was log-normal (σ=0.7), refractive index of 1.87-0.71i and density of 1.8 g/cm3, at wavelength of 550nm.

This statement holds as the Mie scattering efficiency in the relevant $r = [0.05, 0.5]\mu$m size range (CCN at higher supersaturations are primarily determined by Aitken mode aerosol) decreases monotonically (see e.g. Fig. 5.7 in Liou, 2002). Scattering coefficients are additionally weighted with a factor of r^2 through the scattering cross-section. As a consequence, Aitken mode particles contribute only marginally to aerosol optical depth but significantly to CCN. This is nicely illustrated in Fig. 2 of Schutgens and Stier (2014), showing the contributions of each ECHAM-HAM mode to total AOD and $CCN_{1\%}$. The two Aitken modes (HAM modes 2 and 5 in green colors) are very important for global $CCN_{1\%}$ but do not significantly contribute to AOD (reproduced in Fig. 1).

The figure presented in the review shows scattering coefficients for aerosol distributions of varying geometric mean radii. Interpretation in terms of particle size can only be made along horizontal lines of constant mass. In this case, the large increase in particle numbers with decreasing radii ($N \propto r^{-3}$) while keeping mass constant overcomes the decrease in scattering efficiency per particle. However, this situation does not apply to the criticised statement in the manuscript "as expected from Mie theory, as the smaller

particles contribute less to total extinction", which simply states that for a fixed aerosol distribution, smaller particles in the Aitken mode range, which increasingly contribute to CCN at higher supersaturation, contribute less to total extinction, which is consistent with theory and nicely illustrated in of Schutgens and Stier (2014) reproduced as Fig. 1 in this reply.

To avoid any ambiguity I have slightly modified this statement to "sampling the smaller Aitken mode range of the aerosol size distribution".

9. P. 32619 line 18: "This study overcomes this limitation...". I still find it hard to understand how a climate model could "overcome" instrument sampling and retrieval limitations. If the author means it overcomes difficulties in interpreting satellite data, it should be demonstrated and generalized to more than a year-long simulation, and proven to be robust to variation in all related parameters in the model (e.g. relative humidity, precipitation, sea surface temperature etc.). Otherwise, the boundaries of this statement should be clarified.

The full statement cited reads "However, the underlying assumptions cannot be robustly tested with the small number of measurements available so that no reliable global estimate of cloud condensation nuclei exists. This study overcomes this limitation using a fully self-consistent global model (ECHAM-HAM) of aerosol radiative properties and cloud condensation nuclei."

"Overcome" in this sentence explicitly refers the small number of available CCN measurements. It is not claimed that this study overcomes instrument sampling errors (which are very important - our recent work on this is now specifically acknowledged in the introduction (Schutgens et al., ACP, 2016a,b)). We also explicitly state that our correlations are not affected by retrieval errors (as the model calculates CCN and AOD/AI directly from the same aerosol population, without having to retrieve AOD/AI from radiances).

The use of "a yearlong simulation" provides actually very robust statistics (for each of

192x96=18432 grid-columns 1460 6-hourly data-pairs) so that the results are robust with respect to longer simulation periods.

10. P. 32620 line 10: "... and aerosol index do not show significant improvements.". As mentioned above in comment 6, to the naked eye it seems that AI shows the best correlation in that panel.

Good point. See response and updated text in response to comment 6.

11. P. 32620 lines 16-18: "...Satellite retrievals based on visible wavelengths are most sensitive to larger particles...". Please see comment 8 above and Fig 1 below. Satellites may be more sensitive to smaller particles in many cases. Especially when aerosol mean radii are below 0.2 micron (which is typical for various combustion by-products).

As outlined in response to comment 8 above, aerosol extinction can of course increase if the particle size decreased while holding the total mass constant as this implies a huge increase in particle numbers (analogue to the Twomey effect). However, this situation is not relevant here: assuming a fixed size-distribution and mass this statement generally holds (although the non-monotonic nature of the Mie scattering efficiency is noted).

12. P. 32620 line 27: "... it should be noted that this approach is free from retrieval errors...". For supporting this claim, I suggest expanding the description of the model's inputemission maps (e.g. AEROCOM), to clarify they are "free from retrieval errors" as well.

This comment feels like an overcompliation of matters. Obviously, some components in any global model will have been constrained by satellite retrievals - this is a crucially important part of model development.

However, "free from retrieval errors" in this context refers to the self-consistent calculation of CCN and aerosol radiative properties, as outlined in the introduction:

" *self-consistent* in this context refers to the fact that the calculation of the aerosol radiative properties (based on Mie theory) and CCN (based on Köhler theory) are fully consistent in terms of the size-distribution, composition and mixing state, unaffected by any independent assumptions or errors common to remote sensing retrievals."

To make this clear, I have changed the statement to " it should be noted that this self-consistent approach is free from retrieval errors".

13. Figure 3: There is a notable 'crossed out' region over India and the Indian Ocean. Please mention in the figure caption and as well in the article itself whether this region was neglected in any analysis and why. Also, I suspect that extensive desert dust loads in that area may impact the simulated correlations between CCN and aerosol optical parameters.

This is a misunderstanding: this region is not crossed out, as indicated by legend hatching is used for this region as it partly overlaps with the region "South-East Asia". I have revised this in the legend to "India (hatched)".

Desert dust could contribute to the simulated low correlations. However, the simulated contributions of dust to the total extinction is relatively low for the southern part of the "India" region (around 10-15%, not shown) that still shows distinct anti-correlation between AOD and CCN.

Technical corrections

1. P. 32617 line 2: Fig 4d does not exist. Please correct.

Thank you. This has been corrected to "Fig. 4a".

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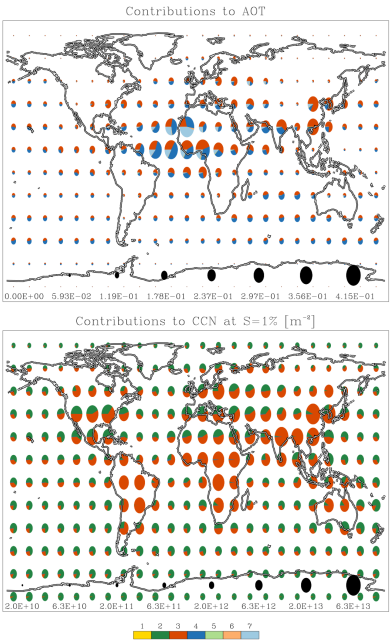

Figure 2. Contributions by different modes to AOT and CCN for the baseline experiment. The pie chart colors show contribution by mode (see legend below lowest panel); the pie chart's size shows the overall magnitude (legend at the bottom of each panel). From top to bottom: AOT at 550 nm (linear scale); column-integrated CCN at $S = 1\,\%$ (logarithmic scale).

Fig. 1.

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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Limitations of passive satellite remote sensing to constrain global cloud condensation nuclei" *by* P. Stier

P. Stier

philip.stier@physics.ox.ac.uk

Received and published: 2 May 2016

I would like to thank the reviewer for the helpful comments that substantially improved the manuscript. I very much appreciated the detailed remarks and hope to have addressed all raised issues.

General comments

(1) A key issue for the credibility of the results is the methodology. The author relies on a model (ECHAM-HAM) that is referred to as "self-consistent". It needs to be clarified to what extent this model is unique, i.e. it would seem that there are several other models out there with the same capability. Secondly, given the model's coarse spatial resolution (1.8 degrees horizontally), meaning that rela-

tive humidity distributions and vertical velocities are not resolved, the term "fully self-consistent" sounds rather excessive. I suggest "self-consistent" instead of "fully self-consistent".

There exist of course a number of (global) aerosol models (c.f. Myhre et al., 2013). A smaller subset of these models deals explicitly with aerosol microphysics (c.f. Mann et al., 2014). Of these models, only few explicitly diagnose CCN at various supersaturations from the prognostic size-distribution and composition and only few calculate aerosol radiative properties via Mie theory from the same prognostic size-distribution and composition. So while it is nowhere claimed that this model is unique, there exist only few models with comparable self-consistent diagnostics. To overcome this unsatisfactory situation I have proposed explicit CCN as diagnostic for future experiments of the AeroCom intercomparison project.

The proposed distinction of between "fully self-consistent" and "self-consistent" seems fairly arbitrary. The fact that some processes cannot be explicitly resolved in global models means that global models (as models in general) are not perfect but that does not change the definition of self-consistency introduced in the paper as quoted above.

In any case, I have removed the "fully" in the usage of "self-consistent".

(2) The figures need to be improved, in particular: a) In Figure 1, the color scale must be changed to better highlight the signals. As it is now, both panels look almost universally blue, with little information to the reader. b) The panels in Figure 6 are too small, so it's almost impossible for the reader to extract any information out of them. c) The panels in Figures 4 and 5 are too small. It is imperative that the reader can easily read the labels (e.g. "South America r=0.50", etc.), but currently this is very difficult.

Figure 1: the main purpose of Fig. 1 and Fig. 2 is to highlight the difference in the geographical spread of CCN at different supersaturations vs. the spread of aerosol radiative properties. The colourbar has been designed to be constant across the different

properties. None of the (many) colourbars I tried is perfect but the one in the revised manuscript is probably a bit better than the original one.

Figure 4,5,6: I agree that the figures appear too small in the ACPD layout (which is a common problem with ACPD) and will liaise with the production team to ensure good reproduction in the final ACP format.

(3) The logical thread of the paper could be improved. As it is now, the reader quite early on becomes convinced that AOD is an inadequate proxy for CCN at the surface. Yet, one has to wait until top of page 32619 and Figures 9-10 before a good alternative is proposed. And, that part of the paper – i.e. lines 1-11 on page 32619 – is very brief compared to the more lengthy discussion of the less successful attempts described on pages 32617-32618.

As indicated by the title, the focus of this study is to highlight limitations in commonly used proxies for cloud condensation nuclei, which is the underlying logic of this order.

I have slightly extended on the discussion of the correlations with aerosol extinction coefficients and extinction aerosol index in the revised manuscript.

Specific comments

(a) Page 32609, line 16: "and humidity" is redundant and should be removed, because the discussion is "at fixed supersaturation"

The idea is that if humidity as well as size, shape and composition are constant also the water uptake is constrained, so that aerosol extinction (at this humidity) is linear in CCN concentration.

(b) Page 32609, lines 16-17: An equation needs to be provided for the claim that "CCN concentrations at fixed supersaturation are linearly related to aerosol light extinction".

The full statement reads "Assuming identical size, shape, composition and humidity,

CCN concentrations at fixed supersaturation are linearly related to aerosol light extinction, so that AOD, the column integrated aerosol extinction, could be expected to provide a first order proxy for CCN. "

For aerosols with identical size, shape and composition the CCN concentration is well defined for each supersaturation. The additional constraint of constant humidity determines the water uptake per particle, constraining the composition and the wet radius of the particles. The aerosol extinction is simply the sum of the extinction of each aerosol particle at ambient radius and composition and therefore linear in the number of CCN.

(c) Page 32610, line 14: "Not clear what "also" refers to.

"Also" here refers to AI vs. the prior discussion of AOD and extinction.

(d) Page 32610, lines 24-28: Past tense should be used when referring to the Liu and Li (2014) study.

Good point. I have cleaned up the use of tenses in the introduction.

(e) Page 32611, lines 27-28: To say that the biases are "consistent" sounds strange. How about replacing "to be consistent" by "affect the two of them similarly" or something like that?

Replaced by "are expected to affect both parameters similarly".

(f) Page 32613, line 22: Insert "by" before "Kazil"

Changed to "... by Kazil et al. (2010)"

(g) Page 32614, line 2: "empirical estimation" is rather cryptic. Can you provide some insight into the physics involved?

Activation schemes are generally based on approximations of the supersaturation balance equation in which the updrafts provide the source term for supersaturation and the condensation on the growing droplet spectrum the sink term. No analytical solu-

tions exist for this equation so the widely used Abdul-Razzak & Ghan (2000) scheme is based on a fit of parcel model simulations. I would refer the interested reader to the literature cited in the manuscript.

(h) Page 32614, lines 3-4: Recently, a significant sensitivity to the activation scheme has been found in several studies, e.g. Gantt et al. (2014, ACP). How might the results of this study be affected by the choice of activation scheme?

Aerosol activation schemes estimate the maximum supersaturation for a given updraft velocity and particle distribution. As our results focus on CCN at fixed supersaturations the results are not directly dependent on the choice of the aerosol activation scheme. However, it should be noted that adsorption activation on insoluble aerosol would increase CCN for the insoluble HAM modes, which currently do not contribute to CCN in HAM (as outlined in the model description).

(i) Page 32614, line 14: "wet- able" should be 'wettable'

Corrected.

(j) Page 32616, lines 11-12: It sounds strange that Saharan dust isn't explicitly mentioned here (as an example of aerosols downstream of source regions), because it is the most striking feature in the figure.

Good point. I have added "such as the Saharan dust outflow" to this sentence.

(k) Page 32617, lines 15-16: How do you define "significantly improved"? Clearly, Figure 7a shows some improvement.

Good point. See response to reviewer 1. I have now included global mean correlation coefficients in the title of each figure and discuss its variation quantitatively.

(l) Page 32618, line 12: Something wrong with "particularly than over". Please rephrase.

Rephrased to "particularly higher than over South America"

(m) Page 32619, line 5: "significantly improved", compared to what?

This directly links to the previous sentence "These results suggest that vertically integrated aerosol radiative properties, as retrieved from satellite imagers, are of limited suitability as proxy for global surface or cloud base CCN"

(n) Page 32619, line 6: "surface extinction aerosol index" needs to be defined.

Agreed. This is now properly defined in the introduction.

"We further investigate the role of the vertical aerosol distribution using the local (model layer) aerosol extinction coefficient (AEC) as well as the extinction aerosol index (AI$_{AEC}$), defined here as local aerosol extinction coefficient times the local Ångström parameter: AI$_{AEC}$=AEC$\times\alpha_{AEC}$, where $\alpha_{AEC} = -\frac{\ln(AEC_{550nm}/AEC_{865nm})}{\ln(\lambda_{550nm}/\lambda_{865nm})}$ is evaluated from the local aerosol extinction coefficients, instead of from the column integrated aerosol optical depths used in AI. "

(o) Page 32619, line 9: "as the smaller particles contribute less to total extinction" is not a full explanation. Something is missing.

I am not entirely sure what is meant by this comment but have clarified this statement as follows:

"as expected from Mie theory, as the smaller particles selected by higher supersaturations contribute less to total extinction"

(p) Page 32619, line 22: How large is "large" and how long is "long"?

Clarified to " large (continental) spatial scales and long (monthly) averaging periods"

(q) Page 32620, line 2: A verb is missing. I suggest resolving that by replacing "an analysis" by "according to our analysis".

Corrected to "according to this analysis, the temporal correlation..."

(r) Page 32620, line 2: "local (grid)": Need to remind the reader here what the

model resolution actually is, i.e. we are not dealing with a cloud-resolving model.

Which would not be strictly local either... Clarified to “local (global-model grid) scale”.

(s) Page 32620, lines 6-7: “This suggests particularly limited constraints” is cryptic. Please rephrase.

Changed to “This suggests that constraints from passive satellite remote sensing are particularly limited in areas key for radiative forcing due to aerosol–cloud interactions.”

(t) Page 32621, lines 12- 15: The parentheses should be removed, because this is highly relevant information.

I do not believe that the parentheses make this statement less relevant.

(u) Page 32628, Figure 1: The caption must explicitly state that the figures are from simulations with ECHAM-HAM.

Good point. I have updated all captions accordingly.

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Limitations of passive ~~satellite~~ remote sensing to constrain global cloud condensation nuclei

P. Stier

Atmospheric, Oceanic and Planetary Physics, Department of Physics, University of Oxford, Oxford, UK

Correspondence to: P. Stier (philip.stier@physics.ox.ac.uk)

Abstract. Aerosol–cloud interactions are considered a key uncertainty in our understanding of climate change (Boucher et al., 2013). Knowledge of the global abundance of aerosols suitable to act as cloud condensation nuclei (CCN) is fundamental to determine the strength of the anthropogenic climate perturbation. Direct measurements are limited and sample only a very small fraction of the globe so that remote sensing from satellites and ground based instruments is widely used as a proxy for cloud condensation nuclei (Nakajima et al., 2001; Andreae, 2009; Clarke and Kapustin, 2010; Boucher et al., 2013). However, the underlying assumptions cannot be robustly tested with the small number of measurements available so that no reliable global estimate of cloud condensation nuclei exists. This study overcomes this limitation using a ~~fully~~ self-consistent global model (ECHAM-HAM) of aerosol radiative properties and cloud condensation nuclei. An analysis of the correlation of simulated aerosol radiative properties and cloud condensation nuclei reveals that common assumptions about their relationships are violated for a significant fraction of the globe: 71 % of the area of the globe shows correlation coefficients between $CCN_{0.2\%}$ at cloud base and aerosol optical depth (AOD) below 0.5, i.e. AOD variability explains only 25 % of the CCN variance. This has significant implications for satellite based studies of aerosol–cloud interactions. The findings also suggest that vertically resolved remote sensing techniques, such as satellite-based high spectral resolution lidars, have a large potential for global monitoring of cloud condensation nuclei.

1 Introduction

Aerosol–cloud interactions play an important role in the global climate system through modification of aerosol and cloud properties and abundance (Boucher et al., 2013; Twomey, 1974; Albrecht, 1989; Lohmann and Feichter, 2005). The activation of suitable aerosols (cloud condensation nuclei, CCN) to cloud droplets is the primary aerosol effect on warm clouds (and ice or mixed-phase clouds initiated from the liquid phase). Knowledge of the global abundance of aerosols suitable to act as cloud condensation nuclei is fundamental to determine the strength of the anthropogenic perturbation causing the radiative effect of aerosol–cloud interactions. Most estimates of the effect of aerosol–cloud interactions on the global radiation balance rely on global aerosol models. However, large uncertainties associated with the representation of clouds and aerosol effects on cloud microphysics and dynamics in current climate models (Boucher et al., 2013; Stevens and Feingold, 2009) demand for independent observational constraints. Unfortunately, direct observations of CCN are spatio-temporally sparse (Andreae, 2009; Spracklen et al., 2011) and provide insufficient constraints on their global distribution. Consequently, satellite retrieved aerosol radiative properties, such as aerosol optical depth (AOD), have been widely used as proxy for CCN in satellite based studies of aerosol–cloud interactions (Kaufman and Nakajima, 1993; Kaufman et al., 2005; Rosenfeld et al., 2008; Grandey and Stier, 2010; Boucher et al., 2013; Gryspeerdt et al., 2014).

Assuming identical size, shape, composition and humidity, CCN concentrations at fixed supersaturation are linearly related to aerosol light extinction, so that AOD, the column integrated aerosol extinction, could be expected to provide a first order proxy for CCN. However, for realistic aerosol distributions extinction and CCN concentrations are non-linearly related to size, complicating the retrieval of CCN based on extinction measurements (Ghan and Collins, 2004; Kapustin et al., 2006). It has been suggested from theory and an analysis of satellite retrievals (Nakajima et al., 2001) that

aerosol index (Deuze et al., 2001)

$$\mathrm{AI} = \mathrm{AOD} \times \alpha \quad (1)$$

where the Ångström parameter

$$\alpha = -\frac{\ln(\mathrm{AOD}_{\lambda_1}/\mathrm{AOD}_{\lambda_2})}{\ln(\lambda_1/\lambda_2)} \quad (2)$$

provides a superior proxy of CCN, as it gives lower weight to large (low α) aerosol and reduces the impact of large but low number-concentration sea salt and dust particles.

A significant body of prior work has evaluated the suitability of aerosol radiative properties as proxy for CCN based on local in-situ data and local remote sensing using sun-photometers or lidars. Ghan and Collins (2004) devised a method for estimating CCN at cloud base from lidar retrievals and surface CCN measurements, which they evaluated in (Ghan et al., 2006) using aircraft, surface in-situ, and surface remote sensing measurements. This study highlighted deteriorating retrieval quality for higher supersaturations and for scenes with vertical inhomogeneity. A continental-scale compilation of co-located observations of AERONET sun-photometer (Holben et al., 1998) retrieved AOD and ground-based CCN measurements revealed a statistically robust power–law relationship between AOD and CCN for continental scales and (month) long averaging periods (Andreae, 2009). Limited-scale in-situ observations (Kapustin et al., 2006) ~~show~~ showed that also the relationship of AI to aerosol number and CCN is strongly affected by relative humidity (increasing particle size and extinction but not aerosol number) and complex aerosol size distributions. An analysis of aircraft measurements for the ARCTAS measurement campaign over Canada ~~shows~~ showed reasonable temporal correlations between CCN and AOD ($r^2 = 0.59$) and a significant improvement in correlation when using in-situ dry extinction instead of vertically integrated ambient AOD (Shinozuka et al., 2015). An analysis of a large compilation of aircraft measurements over the Pacific ~~reveals~~ revealed that regional campaign-average vertical profiles of extinction and CCN proxies show generally a strong correlation (Clarke and Kapustin, 2010) but it is unclear how representative this is for the temporal correlation of extinction and CCN at cloud base. Liu and Li (2014) ~~investigate~~ investigated the correlation of CCN and aerosol radiative properties using data from five Atmospheric Radiation Measurement (ARM) Climate Research Facility sites. They ~~find~~ found variable correlations between surface CCN and AERONET retrieved AOD for the different sites, with lower correlations for the Azores and Niger sites and attribute this to the dominance of large particles. They generally ~~find~~ found improved correlations using AI as compared to AOD and best correlations between in-situ surface scattering/extinctions coefficients and in-situ measured scattering aerosol index. The importance of many of the above factors has also been realised in the context of deriving surface aerosol mass from AOD retrievals for air pollution applications over the continental United States (van Donkelaar et al., 2010), employing a chemical transport model to derive local linear conversion factors from AOD to surface mass.

In summary, previous work on the relationship between aerosol radiative properties and CCN has been based on in-situ CCN data in combination with aerosol radiative properties from in-situ measurements or remote sensing. Studies have found: a variable degree of correlations for different regions and aerosol regimes; generally improved correlations between AI and CCN as compared to AOD and CCN; a degradation of correlations in regions of high relative humidity; an impact of vertical layering on the correlation of surface CCN and aerosol radiative properties. However, the limited availability of direct measurements as well as their limited representativeness in the light of sampling errors (Schutgens et al., 2016b, a) has made it impossible to provide a global, statistically robust, assessment of the suitability of aerosol radiative properties as constraint for CCN. Consequently, no reliable global observational dataset of cloud condensation nuclei exists and a large body of literature uses AOD/AI almost synonymously for CCN.

This work provides a global assessment of the link between aerosol radiative properties and CCN, overcoming the insufficient global coverage of direct observations through use of a ~~fully~~ self-consistent global model (ECHAM-HAM, Stier et al., 2005, 2007; Zhang et al., 2012) of aerosol radiative properties and CCN. It is clear that no perfect global model of aerosol radiative properties or CCN exists (e.g. Myhre et al., 2013; Mann et al., 2014) so *self-consistent* in this context refers to the fact that the calculation of the aerosol radiative properties (based on Mie theory) and CCN (based on Köhler theory) are fully consistent in terms of the size-distribution, composition and mixing state, unaffected by any independent assumptions or errors common to remote sensing retrievals. Therefore, use of this model allows to consistently assess the relationship between aerosol radiative properties and CCN as biases in the simulated fields are expected to ~~be consistent~~affect both parameters similarly. Nonetheless, it should be noted that the ability of models to mimic the spatial (in particular vertical) and temporal (co-)variability of aerosol and humidity fields introduces some uncertainty.

While the introduced methodology would lend itself to the derivation of CCN retrieval from satellite retrieved aerosol radiative properties, this is not the focus of this study. Likewise, it should be pointed out that this work does not investigate the link between aerosol radiative properties and the number of activated cloud droplets, which additionally requires the knowledge of (highly uncertain) updraft velocities at cloud base or the point of activation. Instead, this work aims to provide the first consistent global analysis of the suitability of aerosol radiative properties as observational constraint for CCN.

2 Methods

In this study we employ the aerosol-climate model ECHAM-HAM, version ECHAM-6.1_HAM-2.2, with a prognostic representation of the composition, size distribution, and mixing state of the major global aerosol components: sulfate, black carbon, particulate organic matter, sea salt, and mineral dust. More details and an extensive evaluation of this base model can be found in (Stier et al., 2005, 2007; Zhang et al., 2012; Schutgens and Stier, 2014) as well as part of the AeroCom intercomparison (Myhre et al., 2013; Stier et al., 2013; Mann et al., 2014).

2.1 The atmospheric general circulation model ECHAM6

The atmospheric general circulation model (GCM) ECHAM6 (Stevens et al., 2013) is the sixth-generation climate model developed at the Max Planck Institute for Meteorology. ECHAM6 solves prognostic equations for vorticity, divergence, surface pressure, and temperature, expressed in terms of spherical harmonics with a triangular truncation. Non linear processes and the physical parameterisations are solved on a corresponding Gaussian grid. Water vapour, cloud liquid water, cloud ice, and trace components are transported in grid-point space with a flux form semi-Lagrangian transport scheme (Lin and Rood, 1996). ECHAM6 contains a microphysical cloud scheme (Lohmann and Roeckner, 1996; Lohmann et al., 2007) with prognostic equations for cloud liquid water and ice. Cloud cover is represented using an assumed humidity distribution function (Sundqvist et al., 1989). Convective clouds and convective transport are based on the mass-flux scheme of Tiedtke (1989) with modifications by (Nordeng, 1994) and a modified triggering related to a prognostic treatment of the temperature variance in the planetary boundary layer (Stevens et al., 2013). Radiative transfer is represented using the rapid radiation transfer suite of models optimised for general circulation modeling (Iacono et al., 2008) with 16 and 14 bands in the longwave and shortwave parts of the spectrum, respectively.

2.2 The aerosol module HAM

The microphysical aerosol module HAM (Stier et al., 2005, 2007; Zhang et al., 2012) predicts the evolution of an ensemble of seven interacting internally- and externally-mixed log-normal aerosol modes. In the current setup, the components comprise: sulfate, black carbon, particulate organic matter, sea salt, and mineral dust. The microphysical core M7 (Vignati et al., 2004) calculates coagulation among the modes and the condensation of gas-phase sulfuric acid on the existing aerosol population. In the revised version HAM-2.0, the equilibrium water update is based on κ-Köhler theory (Petters and Kreidenweis, 2007) and a range of aerosol nucleation parameterisations have been introduced ~~(Kazil et al., 2010)~~ ~~by~~ Kazil et al. (2010) in addition to the original binary nucleation scheme. In this study we are employing a parameterisation of neutral and charged nucleation Kazil and Lovejoy (2007) as described in Kazil et al. (2010). Aerosol radiative properties, as well as the sink processes of dry deposition, sedimentation, and wet deposition, are parameterised in dependence on the prognostic aerosol size distribution, composition, and mixing state and coupled to the ECHAM meteorology. Emissions of mineral dust, sea salt and DMS from seawater are calculated online. For all other compounds, emission strength, distribution, and height are based on the AEROCOM aerosol model inter-comparison (http://aerocom.met.no) Phase II ACCMIP-MACCity emission inventory (Lamarque et al., 2010) for the year 2000. We implement an explicit Köhler theory based activation scheme with empirical estimation of maximum supersaturation in updrafts derived from explicit parcel model calculations (Abdul-Razzak and Ghan, 2000). The total number of activated particles is calculated as sum of the integrated log-normal aerosol number distribution from the radius of activation for each mode. Köhler theory requires detailed information about the aerosol composition. While the composition is relatively well defined for some aerosol components, such as sea salt, the detailed composition of other components, such as particulate organic matter is insufficiently understood. In many measurements of aerosol chemical composition, a non-negligible fraction of the aerosol mass cannot be identified and is often attributed to organics (Jimenez et al., 2009). Given the large uncertainties in the identification and simulation of organics, as well as low measured κ values (Petters and Kreidenweis, 2007), we ignore ionic contributions of organics to the solute and treat organics as well as dust in the hydrophilic modes as ~~wet-able~~ wettable and in the hydrophobic modes as entirely hydrophobic.

2.2.1 Cloud Condensation Nuclei

In addition to the application of Köhler theory in the activation scheme of HAM, this scheme is also used for a consistent diagnostics of cloud condensation nuclei at fixed, prescribed supersaturations.

2.2.2 Aerosol radiative properties

Aerosol radiative properties are calculated in the framework of Mie theory. For each aerosol mode, effective refractive indices are calculated by volume-averaging the refractive indices of all components, including aerosol water, which is parameterised in terms of ambient relative humidity. The effective complex refractive indices and the Mie size-parameters for each mode serve as input to look-up tables for the aerosol radiative properties, providing extinction cross-section, single scattering albedo, and asymmetry parameter to the ECHAM radiation scheme. Long-wave

(LW) radiative properties have been introduced and coupled to the ECHAM LW radiation scheme and black carbon refractive indices have been revised for version HAM-2.0 (Stier et al., 2007). We additionally introduce diagnostics of: Aerosol Index (AI) calculated online from the aerosol optical depth at wavelengths comparable to the MODIS AI product: $\mathrm{AI} = \mathrm{AOD} \times \alpha$, where the Ångström parameter $\alpha = -\frac{\ln(\mathrm{AOD}_{550\mathrm{nm}}/\mathrm{AOD}_{865\mathrm{nm}})}{\ln(\lambda_{550\mathrm{nm}}/\lambda_{865\mathrm{nm}})}$; fine mode AOD calculated as sum of Aitken and Accumulation mode AOD; dry AOD approximated from total AOD minus the AOD times the volume fraction of aerosol water.

We further investigate the role of the vertical aerosol distribution using the local (model layer) aerosol extinction coefficient (AEC) as well as the extinction aerosol index ($\mathrm{AI}_{\mathrm{AEC}}$), defined here as local aerosol extinction coefficient times the local Ångström parameter: $\mathrm{AI}_{\mathrm{AEC}}=\mathrm{AEC}\times\alpha_{AEC}$, where $\alpha_{AEC} = -\frac{\ln(\mathrm{AEC}_{550\mathrm{nm}}/\mathrm{AEC}_{865\mathrm{nm}})}{\ln(\lambda_{550\mathrm{nm}}/\lambda_{865\mathrm{nm}})}$ is evaluated from the local aerosol extinction coefficients, instead of from the column integrated aerosol optical depths used in AI.

2.3 Simulation setup

All simulations were performed from October 1999 to December 2000 and constrain the large-scale meteorology to the year 2000 by nudging (Jeuken et al., 1996) the model to the ECMWF ERA40 reanalysis data (Simmons and Gibson, 2000). Only the year 2000 data are analysed. We employ a horizontal resolution of T63 in spectral space with a corresponding resolution of $1.8° \times 1.8°$ on a Gaussian grid. The vertical resolution is set to 31 levels, extending from the surface up to 10 hPa.

2.4 Statistical analysis

The statistical analysis is performed on 6 hourly instantaneous model output, unless longer averaging periods are described. Correlations are reported as linear Pearson correlation coefficient of log-transformed parameters, providing consistency with the majority of prior work. Fits are derived from linear regression of the log-transformed parameters to derive power-law expressions. Note that the results remain largely unchanged when using the non-parametric Spearman's rank correlation coefficient (Fig. 7e).

3 Results

The ECHAM-HAM simulated annual-mean surface CCN concentrations (Fig. 1) show distinct land–sea contrast, with maxima over the main aerosol source areas. CCN concentrations at the lower 0.2 % supersaturation (activating only the larger particles of the CCN spectrum into cloud droplets) are lower than at the higher supersaturation of 1.5 % (activating also smaller particles of the CCN spectrum).

The corresponding annual-mean AOD (Fig. 2a) shows similar maxima in the main aerosol source areas. However, it also shows high values over the sea-salt aerosol dominated storm track regions, dust source regions, such as the Saharan dust outflow, and generally higher levels downwind of the source areas (mainly because AOD is a column integrated quantity while (Fig. 1) shows surface CCN values). AI, giving lower weight to large particles, is in better spatial agreement with the annual mean CCN distribution than AOD (Fig. 2b).

These results confirm the common understanding that CCN are related to AOD and (better) AI for large spatial scales and long averaging periods. This is also confirmed in the fit of regional annual mean AOD and CCN pairs (Fig. 4a) for the main continents defined in Fig. 3.

The derived fit of the mean values ($y = 0.0002x^{1.074}$ for $\mathrm{CCN}_{0.5\%}$) is statistically robust ($r^2 = 0.70$) and compares to a fit ($y = 0.0027x^{0.640}$ for $\mathrm{CCN}_{0.4\%}$, $r^2 = 0.88$) of campaign mean co-located surface based CCN measurements and AERONET sun-photometer retrievals of AOD (Andreae, 2009). The inclusion of oceanic regions dominated by large sea salt particles with high extinction per particle, deteriorates the relationship of AOD and $\mathrm{CCN}_{0.2\%}$ (Fig. 4b) and r^2 decreases from 0.65 to 0.47. The fit for land and ocean regions combined (Fig. 4c) improves using AI instead of AOD ($r^2 = 0.84$).

Despite the good fit of the regional annual mean CCN and AI, the fit of individual monthly means within each region (colour coded scatter and fits in Fig. 4~~d~~a) is variable, with r ranging from 0.41 to 0.93 for individual regions.

The mean goodness of fit deteriorates from $r^2 = 0.57$ to $r^2 = 0.46$ and $r^2 = 0.41$ varying the averaging periods of CCN and AI pairs from monthly via daily to 6 h instantaneous data (Fig. 5).

The global distribution of (temporal) Pearson's correlation coefficients between surface $\mathrm{CCN}_{0.2\%}$ and vertically integrated aerosol optical depth (map in Fig. 6) reveals variable suitability of vertically integrated AOD as proxy for surface CCN. While correlations are generally positive and exceed $r = 0.6$ for large parts of the high latitudes and the tropical oceans, significant areas of the continents and subtropical subsidence regions show low or even negative correlations.

~~Note that maps of global correlations for~~ A number of alternative aerosol radiative properties ~~proposed as~~ have been proposed to provide superior proxies of CCN. Note that maps of their correlations and the corresponding global mean values (Fig. 7), specifically ~~(a~~of (b) fine mode aerosol optical depth ~~,~~(b$\bar{r} = 0.50$), (c) dry aerosol optical depth ~~and~~ (e$\bar{r} = 0.45$) and (d) aerosol index ($\bar{r} = 0.53$) do not show significantly improved correlations ~~.~~as compared to (a) aerosol optical depth ($\bar{r} = 0.44$). Usage of the non-parametric Spearman's rank correlation coefficient (e) gives very similar correlations ($\bar{\rho} = 0.41$). Sampling $\mathrm{CCN}_{0.2\%}$ at the model simulated lowest cloud base gives slightly reduced ($\bar{r} = 0.36$) but spatially very similar correlations with AOD (~~d). Usage of the non-parametric Spearman's rank correlation~~

~~coefficient (e) gives slightly enhanced but spatially very similar correlations.~~f).

The analysis of the vertical structure of aerosol extinction and CCN reveals the reasons for the large spatial variability in these correlations (Fig. 6). In agreement with observations (Clarke and Kapustin, 2010), the correlation between the annual mean vertical profiles of the aerosol extinction coefficient (AEC) and CCN is robust (r ranges from 0.84 to 0.98 for the selected regions), even in areas for which the temporal correlation of column integrated AOD and surface CCN is poor, such as the South-East Atlantic. This can be explained by vertical decoupling: while the temporal correlation of surface CCN with vertically integrated AOD is low ($r \approx 0.2$) in this region, the correlation of surface CCN with the surface extinction coefficient is robust ($r > 0.6$). Generally, the temporal correlation of extinction coefficients with CCN at the same layer (orange) is significantly stronger than the correlation of CCN at each layer with vertically integrated AOD (red). The extinction coefficient is temporally well correlated with relative humidity throughout the troposphere (pink). The correlation of vertically integrated AOD with CCN at cloud base is generally comparable to the correlation with surface CCN. For the Indian region, surface level CCN are even anti-correlated with vertically integrated AOD. Here, surface level extinction is strongly correlated with relative humidity (pink), while surface level CCN are efficiently removed by scavenging during high relative humidity events associated with strong precipitation. Over Europe and North America, correlations of column integrated AOD and surface CCN are generally intermediate to high, in particular north of the sub-tropical subsidence areas. Correlations of extinction coefficients with relative humidity are higher over Europe than over North American and particularly higher than over South America, where the predominant carbonaceous aerosols take up relatively little water. Interestingly, correlations of extinction coefficients with relative humidity are also low for the Southern Ocean region, despite the fact that the dominant (by extinction) sea salt aerosol is highly hygroscopic. This is likely due to the fact that not only aerosol water uptake but also aerosol removal via scavenging is positively correlated to relative humidity (via clouds and precipitation). This hypothesis is supported by the drop off of this correlation around and below cloud base (green line). However, correlations of column integrated AOD and surface CCN are consistently high for this region as well as for the northern high-latitude oceans.

These results suggest that vertically integrated aerosol radiative properties, as retrieved from satellite imagers, are of limited suitability as proxy for global surface or cloud base CCN: 71 % of the area of the globe shows correlation coefficients between $CCN_{0.2\%}$ at cloud base and AOD below 0.5 (i.e. AOD variability explains only 25 % of the CCN variance). The fractional area of $r < 0.5$ increases to 83 and 96 % for $CCN_{0.5\%}$ and $CCN_{1.5\%}$, respectively. Corresponding areas for $r < 0.5$ between CCN and aerosol index are somewhat lower (52, 66, 91 % for $CCN_{0.2\%}$, $CCN_{0.5\%}$, $CCN_{1.5\%}$, respectively, Fig. 8).

Could vertically resolved aerosol radiative properties, e.g. from space-born lidars, provide stronger constraints on CCN and ultimately the radiative effect of aerosol cloud interactions?

The correlation of surface CCN with surface aerosol extinction coefficients (AEC) (Fig. 9a) is significantly improved for most of the globe. This highlights the important role of the aerosol vertical distribution for determining CCN at specific altitudes.Correlations further improve for surface extinction aerosol index AI_{AEC} (Fig. 9b) with $r > 0.8$ for most of the globe. This can be attributed to the lower weight AI gives to large aerosols, reducing the impact of low number-concentration sea salt and dust particles.

Note that also correlations between surface layer CCN and AI_{AEC} deteriorate for higher supersaturations (sampling ~~smaller particles~~ the smaller Aitken mode range of the aerosol size distribution), as expected from Mie theory, as the smaller particles selected by higher supersaturations contribute less to total extinction (Fig. 10). This is particularly evident over the continents with significant primary fine mode aerosol emissions.

4 Conclusions

Direct measurements of cloud condensation nuclei are limited and sample only a very small fraction of the globe so that remote sensing from satellites and ground based instruments is widely used as a proxy for cloud condensation nuclei. However, the underlying assumptions cannot be robustly tested with the small number of measurements available so that no reliable global estimate of cloud condensation nuclei exists.

This study overcomes this limitation using a ~~fully~~ self-consistent global model (ECHAM-HAM) of aerosol radiative properties and cloud condensation nuclei.

An analysis of the correlation of simulated aerosol radiative properties and cloud condensation nuclei confirms findings from earlier work that continental mean CCN are related to AOD ($r^2 = 0.65$) for large (continental) spatial scales and long (monthly) averaging periods but r^2 drops to 0.47 when oceanic regions are included. Use of AI improves the goodness of fit, including oceanic regions, to $r^2 = 0.84$.

The mean goodness of fit for CCN and AI pairs over continental and oceanic regions deteriorates from $r^2 = 0.57$ to $r^2 = 0.46$ and $r^2 = 0.41$ varying the averaging period from monthly via daily to 6 h instantaneous data.

However, aerosol–cloud interactions occur locally (McComiskey and Feingold, 2012): ~~an analysisof~~ according to this analysis, the temporal correlation on the local (global-model grid) scale 71 % of the area of the globe shows correlation coefficients between $CCN_{0.2\%}$ at cloud base and AOD below 0.5 (i.e. AOD variability explains only 25 %

of the CCN variance). The areas with low correlations include the main marine stratocumulus decks, considered most susceptible to aerosol perturbations (Boucher et al., 2013). This suggests ~~particularly limited~~ that constraints from passive satellite remote sensing are particularly limited in areas key for radiative forcing due to aerosol–cloud interactions. Note that correlations for alternative aerosol radiative properties proposed as superior proxies of CCN such as fine mode aerosol optical depth, dry aerosol optical depth and aerosol index do not show significant improvements.

A number of reasons contribute to the low correlations between aerosol radiative properties and CCN, in particular over sub-tropical subsidence areas: aerosol extinction is heavily affected by humidity, in particular at cloud base, and often the local correlation of relative humidity with aerosol extinction coefficients is larger than the correlation of local CCN with column integrated AOD. Satellite retrievals based on visible wavelengths are most sensitive to larger particles, corresponding to CCN at small supersaturations. Correlations between CCN and AI decrease with increasing supersaturations, in particular over the continents with significant primary fine mode aerosol emissions. Additionally, surface or cloud-base CCN and column AOD are often decoupled: and correlations of CCN with local aerosol extinction coefficients throughout the troposphere significantly exceed the correlations with column AOD. Consequently, correlations of surface CCN with surface AEC are significantly larger than with column AOD and are further improved for surface AI_{AEC} for which $r > 0.8$ for most of the globe.

While the ability of this global model to mimic the spatial (in particular vertical) and temporal (co-)variability of aerosol and humidity fields introduces some uncertainty, it should be noted that this self-consistent approach is free from retrieval errors, which would add additional uncertainty when using real satellite data. Advances in computational capabilities now make high-resolution, large-domain simulations of aerosols, clouds and their interactions possible. Such simulations should be increasingly used to test common assumptions in the assessment of aerosol cloud interactions from space (e.g. Gryspeerdt et al., 2015).

The findings in this work have important implications for satellite based studies of aerosol–cloud interactions. They suggest that vertically resolved remote sensing techniques, such as satellite-based high-spectral resolution lidars as ATLID on the ESA/JAXA EarthCare satellite, have a large potential for global monitoring of cloud condensation nuclei. The additional improvement in correlations using the dual-wavelength extinction measurements in AI, suggests that multi-wavelength high-spectral resolution lidars, such as the NASA airborne HSRL (McPherson et al., 2010), could further advance observational constraints on CCN from space.

While the sparse sampling of lidars from space (the CALIOP space-born lidar, Winker et al., 2009, samples the globe sparsely in 16 days, in comparison to e-folding aerosol lifetimes ranging from about $1/2$ day for sea salt to 7 days for black carbon, Textor et al., 2006) may introduce sampling errors, these could be potentially mitigated through synergistic retrievals with co-located imaging radiometers. Ultimately, the assimilation into global aerosol models may provide the best observationally constrained dataset of global cloud condensation nuclei.

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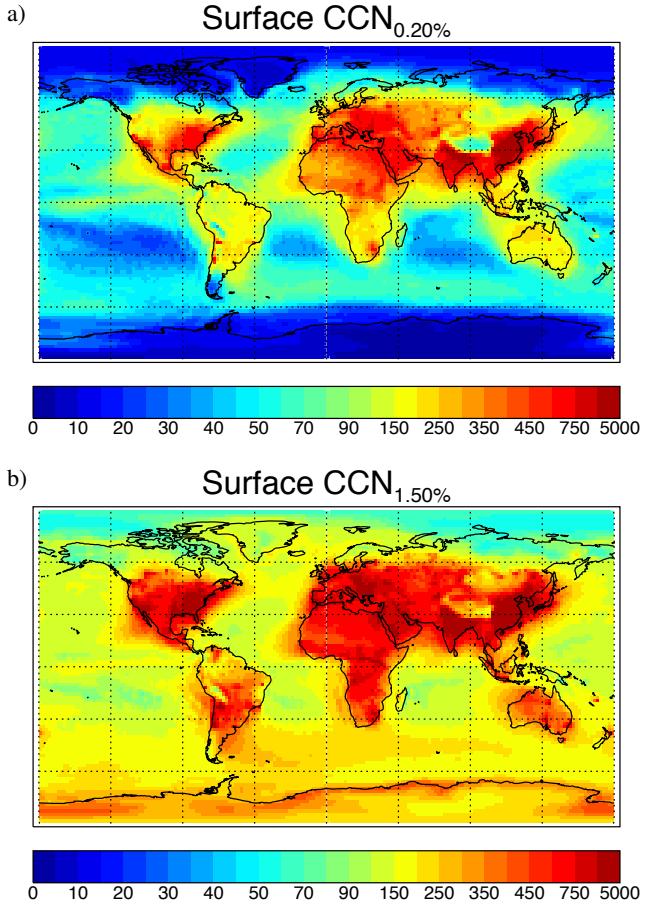

Figure 1. Annual-mean ECHAM-HAM simulated surface cloud condensation nuclei concentrations [cm^{-3}] at **(a)** 0.2 % and **(b)** 1.5 % supersaturation.

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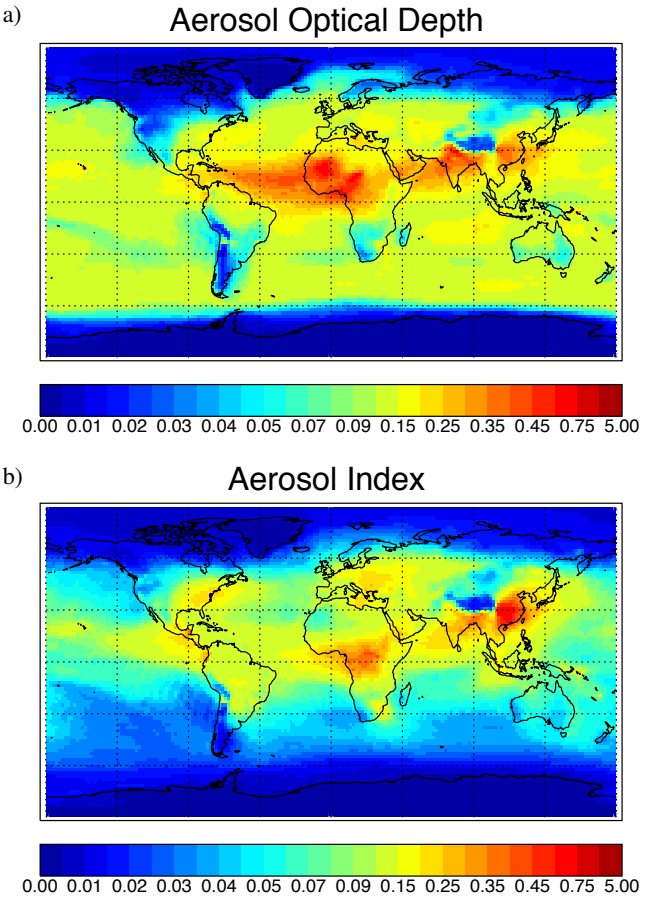

Figure 2. Annual-mean ECHAM-HAM simulated **(a)** aerosol optical depth at 550nm and **(b)** aerosol index between wavelengths of 550 and 865nm. Note the non-linear scale, comparable to Fig. 1.

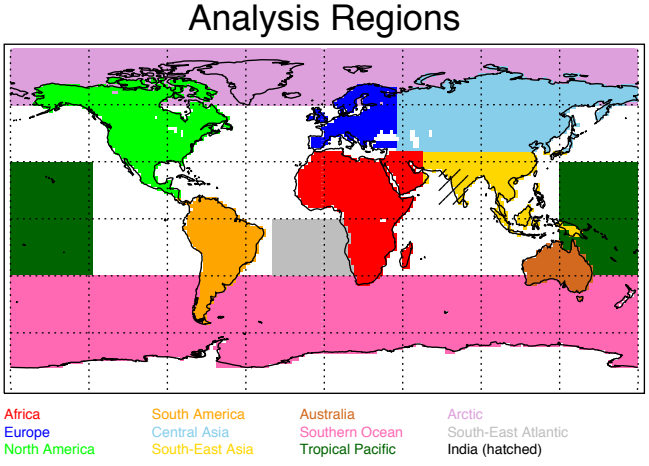

Figure 3. Map of regions used in the analysis.

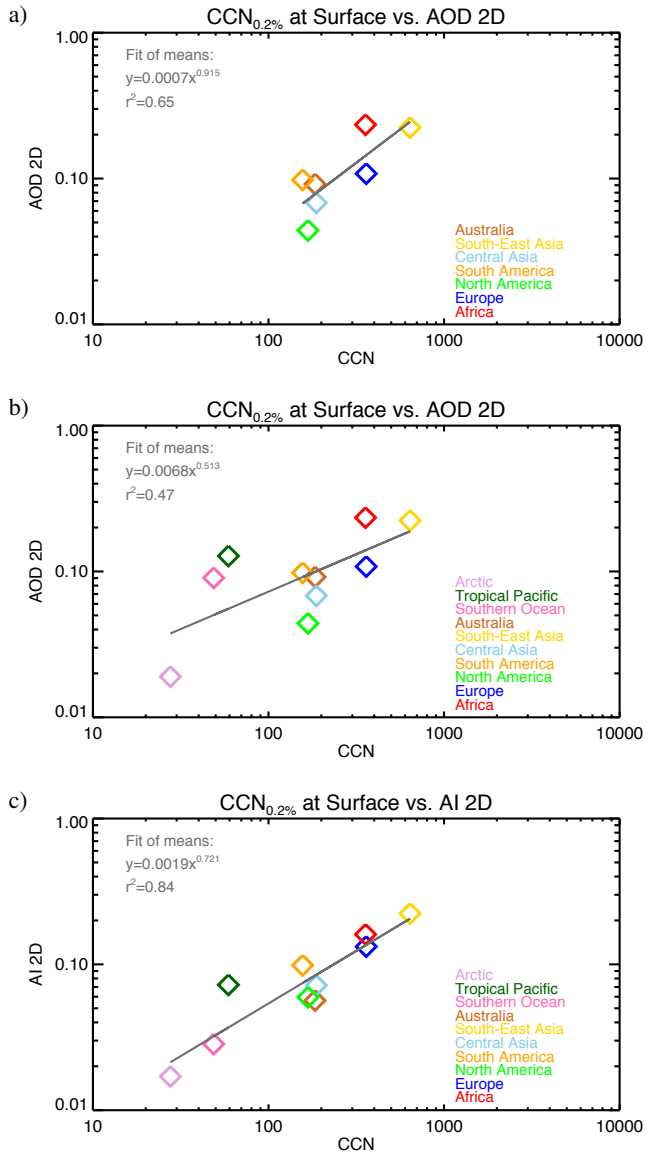

Figure 4. **(a)** Annual continental mean ECHAM-HAM simulated AOD [1] as function of $CCN_{0.2\%}$ [cm^{-3}] and their fit derived from linear regression (gray), **(b)** as **(a)** but including three ocean regions, **(c)** annual continental mean simulated AI as function of $CCN_{0.2\%}$ for continental and ocean regions as in **(b)**; Regional colour coding as in Fig. 3.

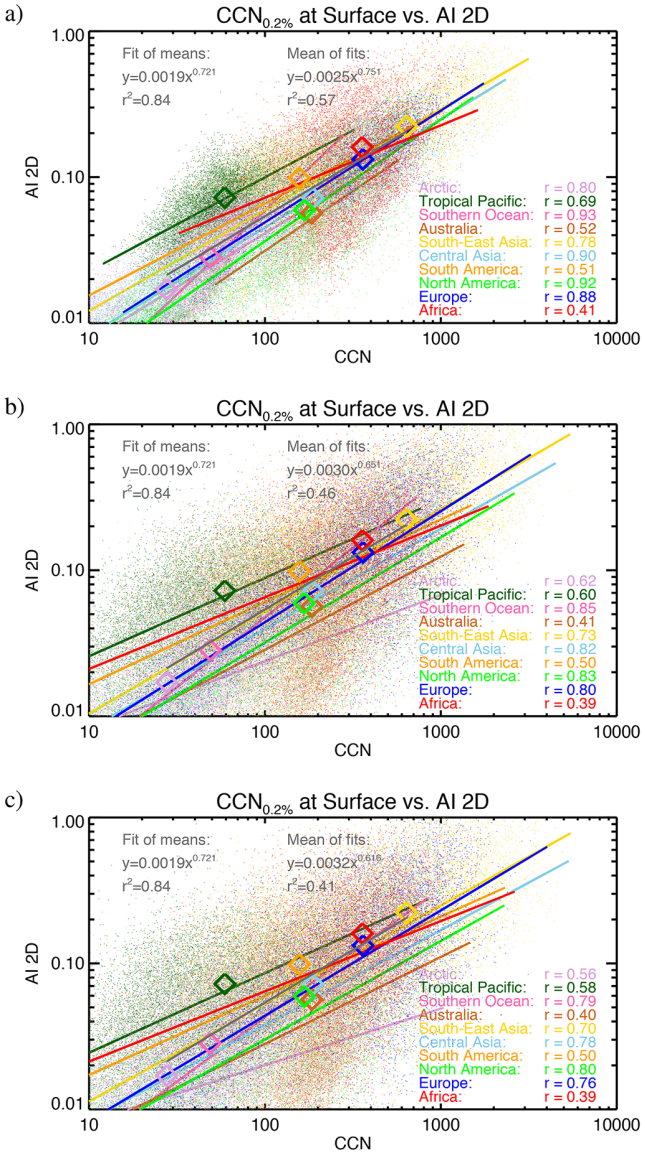

Figure 5. Annual continental mean ECHAM-HAM simulated AI [1] as function of $CCN_{0.2\%}$ [cm^{-3}] (symbols) and their fit derived from linear regression (gray); overlay of **(a)** montly mean, **(b)** daily mean and **(c)** instantaneous 6 hourly pairs of AI and $CCN_{0.2\%}$ (scatter) and their fit derived from linear regression. For visualisation, data in scatterplot randomly sub-sampled to 10 000 pairs. Regional colour coding as in Fig. 3.

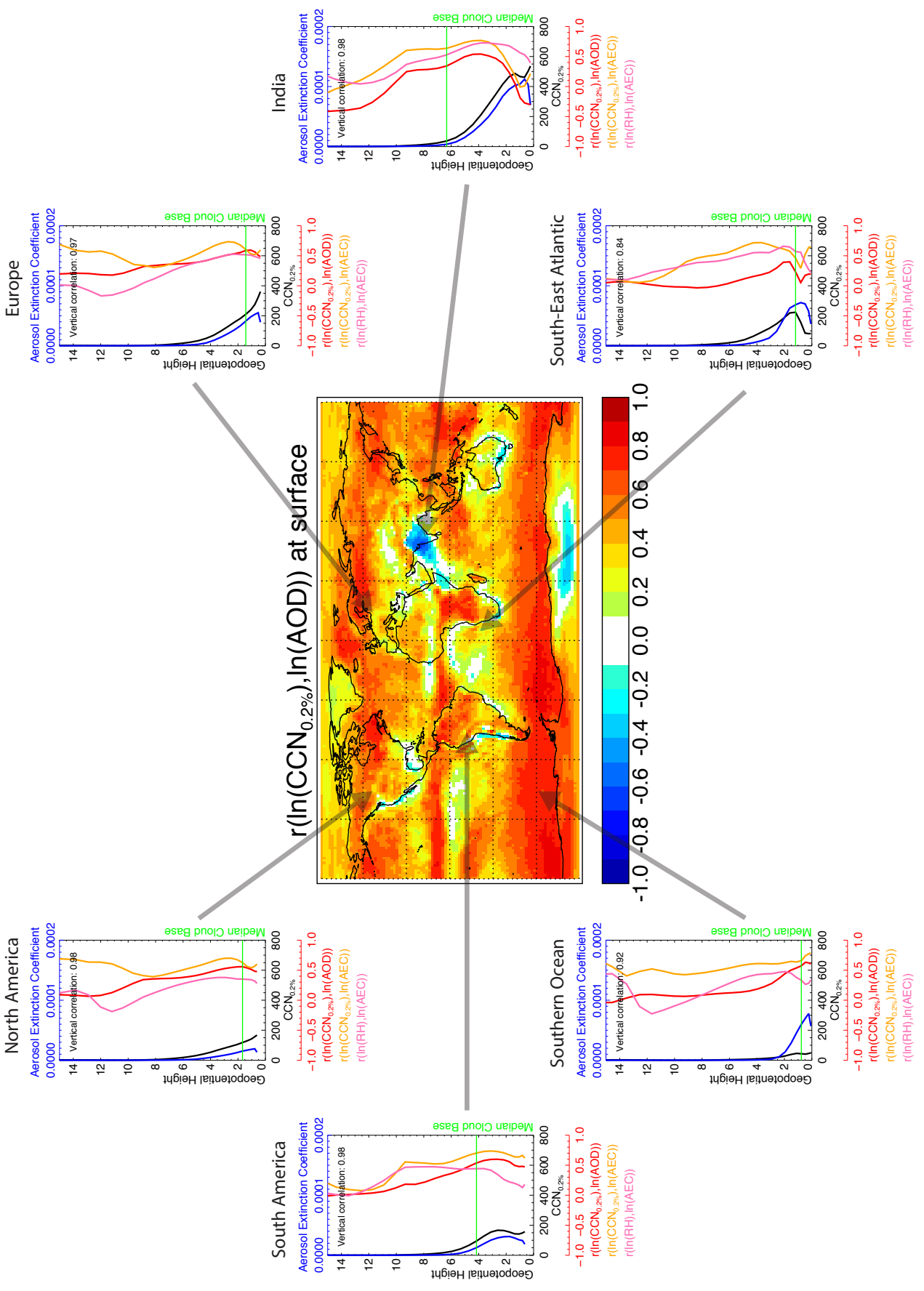

Figure 6. Map of Pearson's correlation coefficient r of ECHAM-HAM simulated surface $\ln(CCN_{0.2\%})$ with column integrated aerosol optical depth $\ln(AOD)$ calculated for each model grid box from one year of 6 hourly pairs. Annual-mean vertical profiles of $CCN_{0.2\%}$ [cm^{-3}] (black), aerosol extinction coefficient (AEC) [m^{-1}] (blue), profile of temporal correlation of $\ln(CCN_{0.2\%})$ with column integrated $\ln(AOD)$ (red), profile of temporal correlation of$\ln(CCN_{0.2\%})$ with vertically resolved extinction coefficient $\ln(AEC)$ orange) and temporal correlation of $\ln(RH)$ with vertically resolved extinction coefficient $\ln(AEC)$ (pink). Also shown is the model median stratiform cloud base for each region (green) – note that this corresponds to the lowest detrainment level in regions dominated by convection, such as India. Regions defined as in Fig. 3.

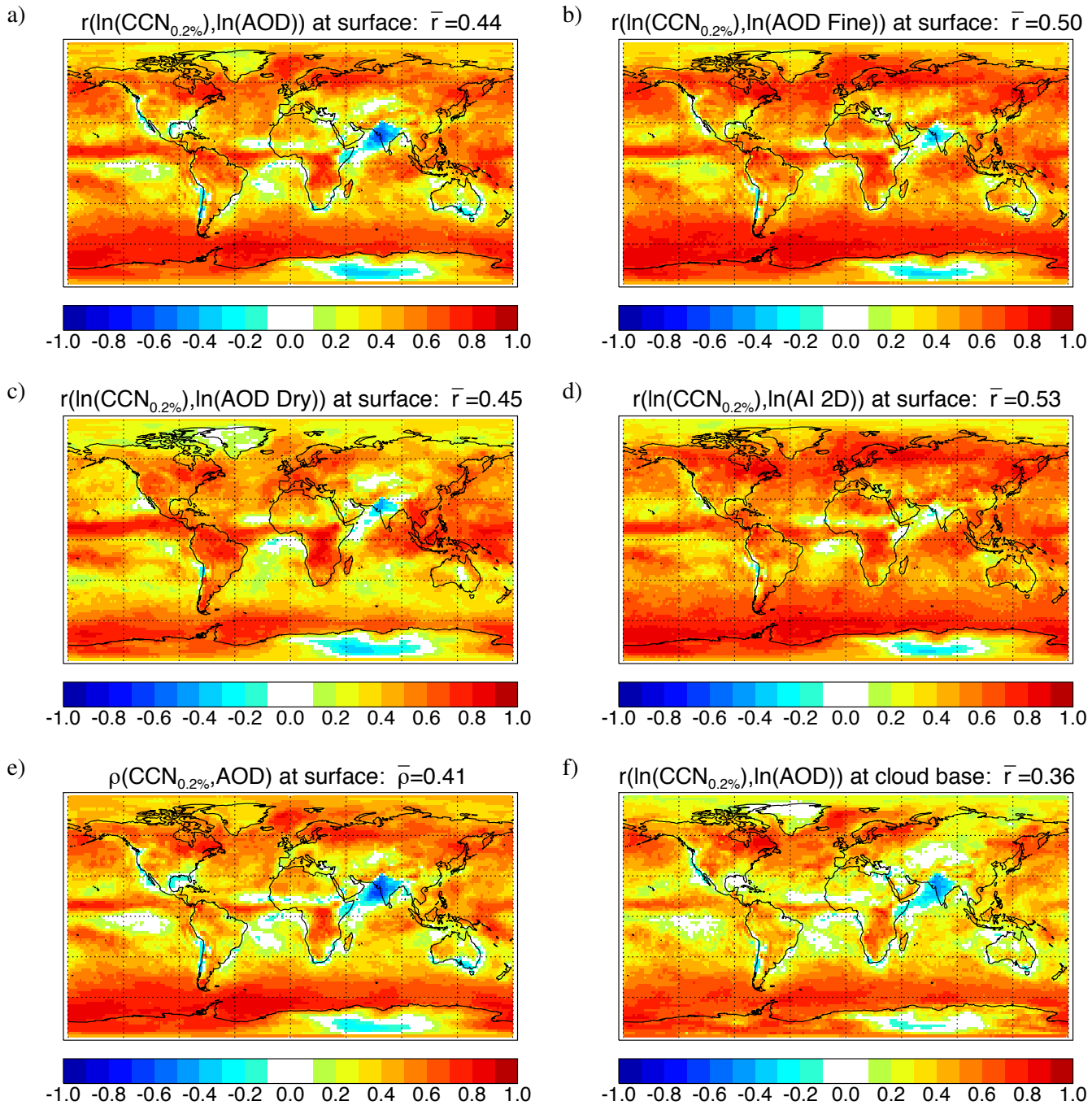

Figure 7. Map of Pearson's correlation coefficient of ECHAM-HAM simulated $CCN_{0.2\%}$ with aerosol radiative properties for **(a)** surface $CCN_{0.2\%}$ with vertically integrated ~~fine mode~~ aerosol optical depth, **(b)** surface $CCN_{0.2\%}$ with vertically integrated ~~dry~~ fine mode aerosol optical depth, **(c)** surface $CCN_{0.2\%}$ with vertically integrated ~~AI~~dry aerosol optical depth, **(d)** surface $CCN_{0.2\%}$ ~~sampled at cloud base~~ with vertically integrated ~~AOD and~~ AI, **(e)** ~~map of~~ Spearman's rank correlation coefficient for surface $CCN_{0.2\%}$ with vertically integrated AOD and **(f)** Pearson's correlation coefficient of $CCN_{0.2\%}$ sampled at cloud base with vertically integrated AOD. Global-mean correlation coefficients are given in the title of each plot.

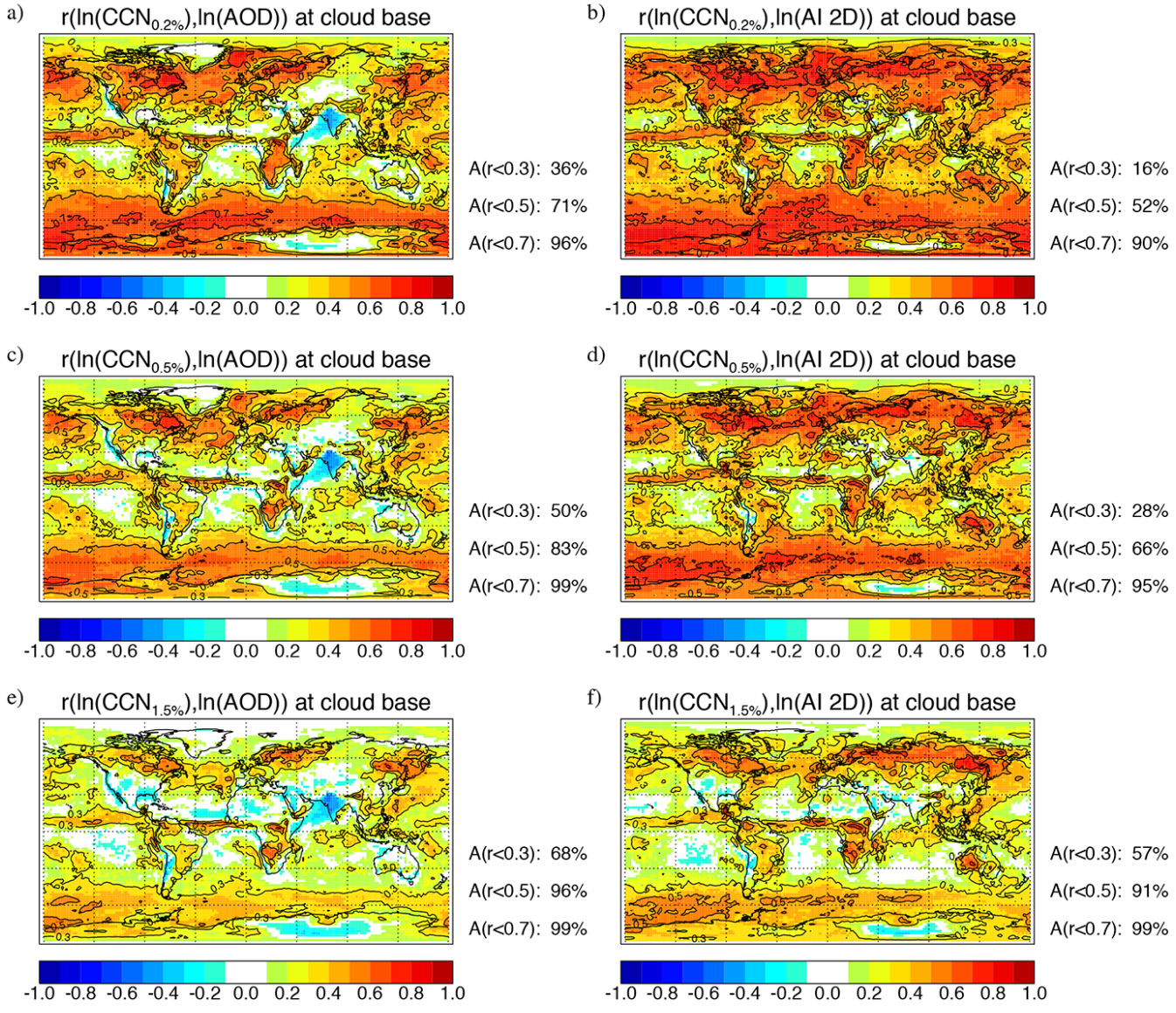

Figure 8. Map of Pearson's correlation coefficient of ECHAM-HAM simulated cloud base CCN with aerosol radiative properties for **(a)** $CCN_{0.2\%}$ with vertically integrated aerosol optical depth, **(b)** $CCN_{0.2\%}$ with vertically integrated aerosol index, **(c)** $CCN_{0.5\%}$ with vertically integrated aerosol optical depth, **(d)** $CCN_{0.5\%}$ with vertically integrated aerosol index, **(e)** $CCN_{1.5\%}$ with vertically integrated aerosol optical depth and **(f)** $CCN_{1.5\%}$ with vertically integrated aerosol index. Fractional area (A) of the globe with $r < 0.3, 0.5, 0.7$.

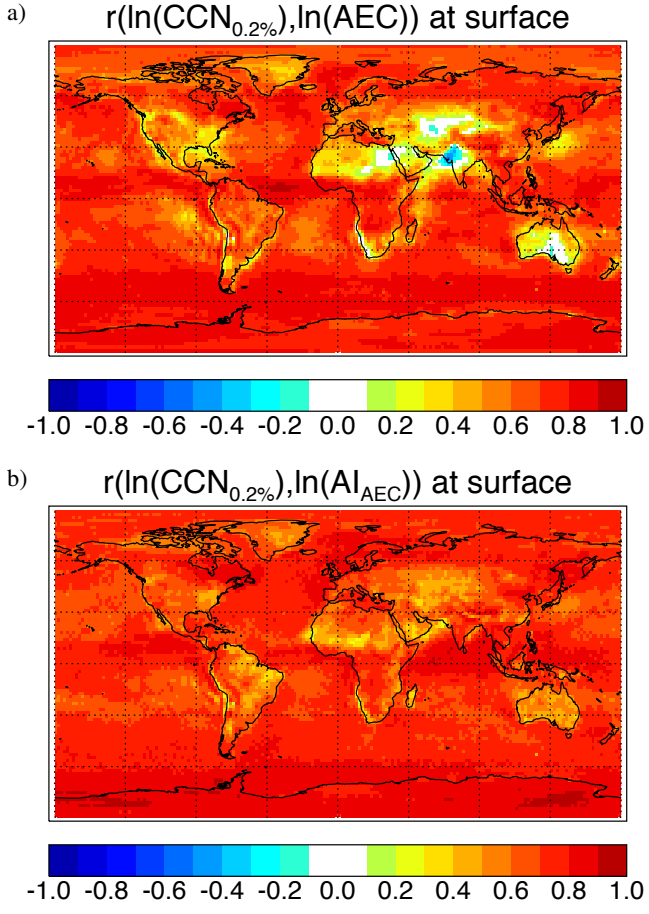

Figure 9. Map of Pearson's correlation coefficient of ECHAM-HAM simulated CCN with vertically resolved aerosol radiative properties: **(a)** surface $\ln(\mathrm{CCN}_{0.2\%})$ with surface $\ln(\mathrm{AEC})$ and **(b)** surface $\ln(\mathrm{CCN}_{0.2\%})$ with surface $\ln(\mathrm{AEC\text{-}AI})$ calculated for each model grid box from one year of 6 hourly pairs.

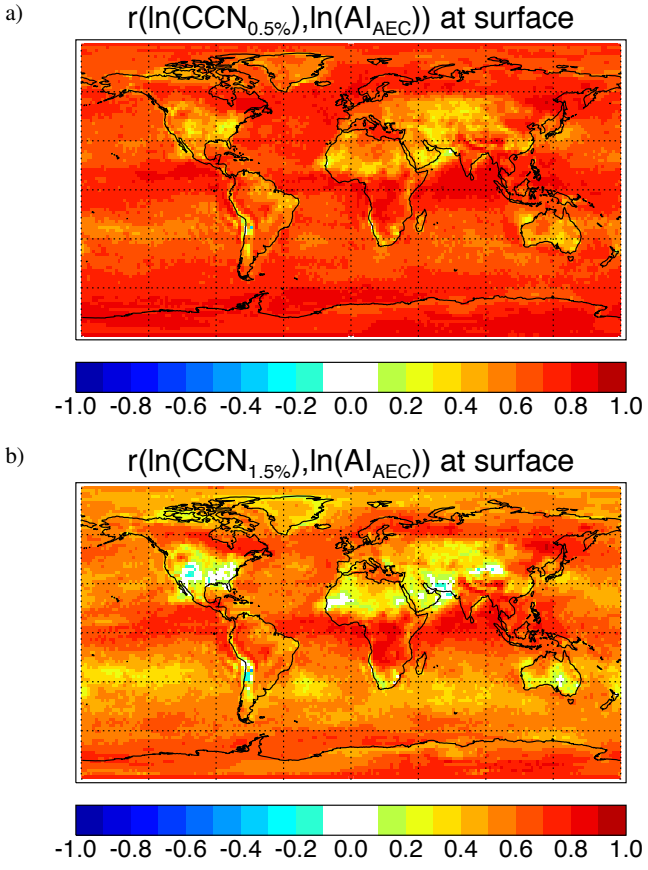

Figure 10. Map of Pearson's correlation coefficient of ECHAM-HAM simulated surface layer $\ln(\mathrm{AI}_{\mathrm{AEC}})$ with $\ln(\mathrm{CCN})$ at higher supersaturations: **(a)** $\mathrm{CCN}_{0.5\%}$, **(b)** $\mathrm{CCN}_{1.5\%}$.