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

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.

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".

Bibliography

Liou, K.-N. (2002), An introduction to atmospheric radiation, 2nd ed., xiv, 583 p. pp., Academic Press, Amsterdam ; Boston.

Schutgens, N. A. J., and P. Stier (2014), A pathway analysis of global aerosol pro-

cesses, Atmos. Chem. Phys., 14, 11657-11686 doi: 10.5194/acp-14-11657-2014.

Schutgens, N. A. J., E. Gryspeerdt, N. Weigum, S. Tsyro, D. Goto, M. Schulz, and P. Stier (2016), Will a perfect model agree with perfect observations? The impact of spatial sampling, Atmos. Chem. Phys. Discuss., 2016 doi: 10.5194/acp-2015-973.

Schutgens, N. A. J., D. G. Partridge, and P. Stier (2016), The importance of temporal collocation for the evaluation of aerosol models with observations Atmos. Chem. Phys., 16, 1065–1079 doi: 10.5194/acp-16-1065-2016.

Spracklen, D. V., K. S. Carslaw, U. Poschl, A. Rap, and P. M. Forster (2011), Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, Atmos Chem Phys, 11(17), 9067-9087 doi: Doi 10.5194/Acp-11-9067-2011.

Stevens, B., M. Giorgetta, M. Esch, T. Mauritsen, T. Crueger, S. Rast, M. Salzmann, H. Schmidt, J. Bader, K. Block, R. Brokopf, I. Fast, S. Kinne, L. Kornblueh, U. Lohmann, R. Pincus, T. Reichler, and E. Roeckner (2013), Atmospheric component of the MPI-M Earth System Model: ECHAM6, J Adv Model Earth Sy, 5(2), 146-172 doi: Doi 10.1002/Jame.20015.

Zhang, K., D. O'Donnell, J. Kazil, P. Stier, S. Kinne, U. Lohmann, S. Ferrachat, B. Croft, J. Quaas, H. Wan, S. Rast, and J. Feichter (2012), The global aerosol-climate model ECHAM-HAM, version 2: sensitivity to improvements in process representations, Atmos. Chem. Phys., 12(19), 8911-8949 doi: Doi 10.5194/Acp-12-8911-2012.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 32607, 2015.

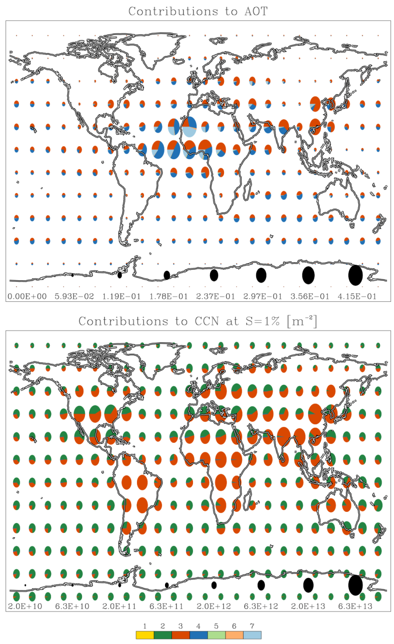

Figure 2. Contributions by different modes to AOT and CCN for the baseline experiment. The pie chart colors show contribution by mode (see legend below lowest panel); the pie chart's size shows the overall magnitude (legend at the bottom of each panel). From top to bottom: AOT at 550 nm (linear scale);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

Fig. 1.