RESPONSE TO REVIEWS AND COMMENTS OF: *SIMPLE PLUMES: A PARAMETERIZATION OF ANTHROPOGENIC AEROSOL OPTICAL PROPERTIES AND AN ASSOCIATED TWOMEY EFFECT FOR CLIMATE STUDIES*

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General Response

The reviewers are thanked for their careful reading of our manuscript and their thoughtful and constructive suggestions. We are pleased to note that the reviewers recognize the value of the ideas, and all recommend publication, even if reviewer 3 is somewhat more conditional in his/her judgement.

The most substantial issues that arose in the reviews can be broadly grouped as follows: (i) the need to compactly summarize the assumptions and limitations of our approach (Reviewer 1 and 2); (ii) somewhat more exploration of the sensitivity of our approach to different assumptions, particularly as relates to the apparently large radiative efficiency (Reviewer 2); (iii) the need to provide some additional justification for our assumptions, including an apparent reliance on an unpublished climatology. These have been addressed as follows:

- We have reformulated the Conclusions by adding two additional paragraphs to highlight the major assumptions that have gone into the development of MACv2-SP and their potential implications. In stating the underlying assumption of the climatology, and outlining the main simplifications adopted to arrive at it, we hopefully better emphasize that MACv2-SP is a designed to induce a reasonable aerosol forcing, not a climatology of the aerosol.
- In response to the reviewer's concerns we have looked more in depth at the radiative efficiency in MACv2-SP. Our initial analysis was inadvertently based on the full eleven years of the simulations, but a shock in the first year is associated with a large ($-0.15\,\mathrm{W m^{-2}}$) clear-sky adjustment in the southern hemisphere and this lead to large discrepancies between clear-sky ERF and clear-sky IRF that is unphysical. Discarding the first year in our analysis and only analyzing the last ten-years of the ensembles gives a much better agreement between clear-sky ERF and clear-sky IRF, and results in the estimates of radiative efficiency and cloud masking matching the AeroCom mean to two significant digits. In addition to correcting this analysis we also performed two additional simulations with a different single-scattering albedo for the industrial plumes. The analysis of the clear-sky IRF of these runs is also included in the revised manuscript thus

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demonstrating the sensitivity of the forcing to assumptions adopted within the climatology.

- Throughout we provide additional justification for the assumptions made, following the guidance of the reviews (especially Reviewer 2). Most critically, we address the appearance that MACv2-SP is disproportionately dependent on MACv2 which is not yet published (Reviewer 3). MACv2 is mostly an update of MACv1, and though it does not differ fundamentally from MACv1 it does address deficiencies in that data set, most importantly the lack of remote observations over the ocean. However in the end MACv2 only provides a guide, the real measure of MACv2-SP is in the patterns of aerosol forcing it produces, and these, when compared to past work, e.g., AeroCOM, show that MACv2-SP is not only a good encapsulation of MACv2, but understanding of aerosol forcing more broadly.

In addition to the above, our interaction with the community and our ongoing analysis of the climatology revealed some minor bugs, in particular in the implementation of the treatment of the seasonal cycle of the tropical biomass plumes. This was responsible for what appeared to be a curious and strong tropical cloud adjustment near the Atlantic ITCZ. We corrected this bug in the climatology and reran all the simulations. Doing so removed the curious, and seemingly un-realistic feature near the Atlantic ITCZ. As a result of the new runs and the discarding of one year of spin-up in our analysis the forcing numbers have changes and, as detailed above, are now much more consistent with the AeroCom results.

All of the reviewer comments are addressed specifically below. In addressing these comments, large and small, we generally followed the rule that comments merit a change in the manuscript.

Specific Response (Editor)

The editor asked that we alter the title to conform with the journal policy.

To address the need to specify the name of the climatology and its relation to CMIP6, yet avoid it being lost in a sea of acronyms we propose:

> *MACv2-SP: A parameterization of anthropogenic aerosol optical properties and an associated Twomey effect for use in CMIP6*

Note that this is a model of the aerosol forcing that is needed for many CMIP6 MIPs, and more broadly. It is not a MIP in its own right, hence it was not clear how to adjust the name to follow exactly the convention suggested by the editor, but we are open to further suggestions.

Specific Response (Anonymous Referee #1)

The reviewer identified and recommended the removal of a text fragment.

We thank the reviewer for noticing this, and wonder how we could have missed it after reading it so many time. The text fragment will be removed in the revised manuscript.

Specific Response (Anonymous Referee #2)

The reviewer raised a number of points related to the need to better justify particular assumptions, and discuss their potential impact on the radiative forcing

The reviewer's comments are very helpful in addressing those areas where further clarification or justification of our assumptions would be helpful. In the revised manuscript we address all of these. Specifically:

(1) Difference in MACv1 and MACv2 globally averaged AOD: The annual average total AOD from MACv1 to MACv2 did change only very little (maybe 0.003 now smaller in MACv2) and is still between 0.12 and 0.13. The anthropogenic AOD at 550nm, however, changed from about 0.038 to ca 0.030 in from MACv1 to MACv2 due to the use of ACCMIP (rather than AeroCom) emissions to define the scaling fraction for the fine-mode AOD (based on fine-mode AOD simulation differences for present day and pre-industrial times with global model (ensemble)s). The ACCMIP simulations have a lower reduction (mainly due to organics) back in time and the now higher pre-industrial fine-mode background leave less fine-mode AOD for anthropogenic purposes. As a result the anthropogenic AOD of MACv2 and MACv2-SP is now much closer to the AeroCom phase II median value of 0.276. This is noted in the revised text.

(2) Why AOD maxima do not align better with the perceived centers of emissions: The plume centers are defined at the location of the local AOD maximum in MAC. These need not coincide as the AOD maximum is also influenced by transport and deposition. This is now noted in the text.

(3) Differences in vertical profiles between MACv2 and MACv2-SP: The vertical structure of the MACv2 AOD is not well constrained by observations, and very dependent on modelling. Hence we do not have a lot of confidence that the very small secondary maxima near the tropopause in the mid-latitudes is a real feature. The vertical structure near the surface also depends on the representation of local topography, which explains why in some regions it is lower in MACv2-SP than in MACv2 (e.g., over China). Even if these features were being reliably represented by MACv2 we believe that any attempt to capture them in the framework of MACv2-SP would be overfitting. The whole idea of MACv2-SP is that such details are clearly much smaller than differences among different state-of-the-art aerosol models (cf Table 4). Additional evidence to support this point of view is provided by sensitivity studies described in Fiedler et al., (in review with JAMES). These points are now mentioned in the revised manuscript.

(4) Annual cycle in industrial plumes Even if emissions do not change over the annual cycle the meteorology and aerosol sinks will change, and thereby impose an annual cycle in AOD. This is now noted.

(5) Justification for optical properties: MACv2-SP assumes than the fine-mode has a typical effective radius of 0.14 μm. This value is based on AERONET statistics of detailed size-distributions data of aerosol size smaller than 0.5 μm in radius. For such sizes a mid-visible (550nm) Angstrom parameter of 2.0 and a mid-visible asymmetry-factor of 0.63 are representative choices. The assumed SSA seems low at initial glance, but remember that the size-distributions in Dubovik (2002)

have aside from the fine-mode AOD usually also a non-negligible coarse mode AOD, which in the mid-visible is less absorbing, so such low SSA values for just the fine-mode AOD fraction is (considered by us as) quite realistic. This view is substantiated with the good agreement with other studies in terms of clear-sky radiative efficiency, and our experiments exploring different values for SSA.

(6) Constant τ_{bg}: This has been revised. But the background was scaling in time just like the anthropogenic AOD, which did not make sense. So the revised code has a year-to-year constant, yet plume-dependent background AOD that only varies from month-to-month.

(7) Emission based scaling not physical: We want to keep it simple to enable experimentation and that the parameterization could be fit to more complex representations of the aerosol to address research questions once it has been better established. To make this clearer, the revised manuscript states explicitly that factors uncorrelated with anthropogenic NH_3 or SO_2 emissions may also influence the time history of regional pattens of anthropogenic aerosol optical depth. The simplification of scaling to emissions is also noted in the conclusion, to further highlight this simplification.

The reviewer is concerned about our use of AeroCom I, versus AeroCom II, models for a specification of cloud active properties:

We wish we could share the reviewers optimism regarding progress in modelling cloud-aerosol interactions on global scale. Recent studies such as the one by Wyant et al., comparing aerosol modelling to VOCALS data (*ACP*, **15**, 2015) are sobering. Because we have so little confidence in the modelling (we actually analyzed both AeroCom I and II models) priority is given to satellite retrievals in the derivation of the CDNC factor as function of AOD. We have revised the manuscript to make this point more clear. The figure showing that the AeroCom models show roughly similar behavior is based on AeroCom 1 to be consistent with the 1850 base year and because AeroCom2 did not look any more reasonable, and in some respects substantially less compelling. Our reservations regarding the modelling are however only touched upon as the manuscript did not seem to be a good place to belabor the challenges in modelling aerosol-cloud interactions. Still note, that AeroCom II model simulations were used to define the fine-mode AOD fraction that is anthropogenic fraction and also when comparing AODf–cloud droplet population density relationships in comparison to our applied relationship from satellite remote sensing.

The reviewer is surprised about the large radiative efficiency:

We were too, and the reviewers comments prompted us to look at this more deeply. After much analysis, additional experiments, and more detailed comparisons with runs using double radiation calls, we realized that the apparently large radiative efficiency was the result of an analysis error. Remove the first year from the analysis, during which there is a strong adjustment in the model, leads to much more sensible numbers and almost perfect alignment with the AeroCom Mean. The value is still larger than ECHAM5, but this is likely attributable to the much greater absorption in that model, as it used a radiative transfer model with a very coarse spectral treatment, four bands compared to 112 in PSRad (as used in the MPI-ESM1)

Specific (more minor) Comments

Please excuse the terseness of our responses, the reviewer's comments were thoughtful and helpful and almost all led to changes.

(1) Yes, look carefully at AR5, the modeling was not viewed as having quantitative skill and bounds came from top-down estimates. Indeed, modeling approaches typically tune the results to the expected observation.
(2) Not sure if we understand the reviewer's point. The result from a single model study should not be taken as a fact, and that they are controversial is documented by the cited references. More studies are coming out that call into question purported circulation responses to the aerosol. We agree that this is because robust circulation responses to forcing are difficult to establish, albeit important. That said the literature contains many strong claims about circulation responses to aerosol forcing, but little robustness has been demonstrated.
(3) Rephrased.
(4) Yes, reviewer 3 also raised this point and we have rephrased the sentence as the contrast to the natural aerosol is not necessary to justify what we do.
(5) Here we mean extensive aerosol properties, this is not stated directly.
(6) This statement was confusing, also for the other reviewer. Our point was that the aerosol forcing is not thought to lead to circulation changes which in turn act to change the forcing. To make this clearer we now state that aerosol-induced circulation changes do not feedback to cause aerosol-forcing changes.
(7) Yes we clarified this following the reviewers suggestion.
(8) This has also been clarified, as it was also confusing for the third reviewer.
(9) Yes we have circulated the code to other groups and have worked through some difficulties of interpretation, but the implementation has not been an issue. The reference to below the surface is purely a formal argument.
(10) Yes, this has also been rephrased to make it more clear.
(11) Correct, but this is stated at the end of the section.
(12) This has been rephrased to be more precise as suggested by the reviewer.
(13) We have added additional text to better reflect on the similarity and differences between the model output and the data.
(14) Reviewer 3 also had issues with our reference to Kirkby et al and we have moderated our statements accordingly.
(15) We are not sure we understand the reviewers point, but believe that our definition of IRF is sufficiently clear even if it deviates from other uses of the term.
(16) This is fair, but our statement is not exclusive and refers to the net effect.
(17) We have been far more careful than most studies, by running ten years and three to six ensemble members and characterizing variability using a 100 year ensemble.
(18) We adopt the view that the difference between a comprehensive model and a simple one is the transparency of its assumptions. We have added a qualifier to make this a bit more clear.
(19) We took care to use the word constituent because we did not mean feature. But to make this more clear we added a parenthetical.
(20) This has been rephrased.

(21) This was a typo, changed to 0.73.
(22) Sentence fragment removed.
(23) This is a question of style upon which we differ.
(24) We found another instances of repeated has, but not this one.
(25) It is not a great reference, but it is the first paper to use them and the only one to discuss them so far, we have been slow in writing these up.
(26) Rephrased
(27) This should eventually come out as a section heading.

Specific Response (Anonymous Referee #3)

1. The nature of a climatology's representation of high-frequency variability:

This basic assumption is now highlighted, even italicized, in the conclusions. However, we note that the purpose of the climatology is to fit the forcing, not the physical properties per se, the latter are a means to an end. Hence to the extent the climatology captures the forcing its physical parameters should be seen as effective parameters chosen to give the right estimate of the forcing. This is emphasized, and most relevant in the discussion of the Twomey effect, where non-linearities are expected to be strongest and is used to explain why the assumed droplet population density perturbations might appear on the small side, and it is also now emphasized again in the conclusions..

2. Assumption of constant in time aerosol properties:

We agree this merits a deeper analysis in the future. Here we had a desire to focus on first things first, namely the importance of emissions and amount, rather than type. The reviewer is right that scaling SSA could provide a way to get around the constraint of overly strong mid-century warming, and we intend to look at this more deeply, but we thought it is useful to first focus on the main argument, i.e., the effect of changing aerosol burdens. To bring the reviewer's point out more clearly, however, we have modified the discussion of this point in the manuscript to make the link to the arguments by Stevens (2015) more explicit.

3-4. The reviewer questions the transparency with respect to the derivation of the anthropogenic fraction:

To address this issue manuscript has been revised to state: "The merged fine-mode AOD (at 550 nm) along with a scaling factor (anthropogenic fraction) based on fine-mode AOD output of comprehensive aerosol-chemistry models run for present-day and pre-industrial conditions define the anthropogenic AOD (at 550 nm) map of MAC – any anthropogenic contribution to the coarse mode is neglected." This should make it clear to the reader that the fine-mode fraction is based on modelling, does not include a coarse mode contribution and is not derived from observations. These issues are however a bit muted, as are the importance of differences between MACv1 and its update, MACv2, by the fact that the purpose of the climatology is not to describe the aerosol, rather the forcing – now explicitly stated in the conclusions; and because improvements in MACv2 give anthropogenic AOD estimates much more in line with the multi-model mean from comprehensive modelling.

5. Pre 1850 aerosol contribution:

The reviewer's comments indicates that we were not clear enough in specifying that the purpose of the climatology is not to present a reference description of the aerosol, rather a reference climatology of post 1850 anthropogenic aerosol *radiative forcing*. To address this point, in the abstract we have added the statement that "[MACv2-SP] was created to provide a harmonized description of post 1850 anthropogenic-aerosol radiative forcing for climate modelling studies. MACv2-SP has been designed to be easy to implement, change and use, and thereby enable studies exploring the climatic effects of different plausible patterns of aerosol radiative forcing, including a Twomey effect." We have also re-iterated this point in the revised conclusions.

6. Equations:

The reviewer raises a number of concerns regarding details in the equations. These were very helpful and have been used as the basis for considerably revising the presentation of the equations, adopting a framework that now also includes the use of mid-visible extinction as the basis for the climatology.

Specific (more minor) Comments

Please excuse the terseness of our responses, the reviewer's comments were thoughtful and helpful and almost all led to changes.

(1) We agree: changed to 'observable response'
(2) Changed as suggested.
(3) We have rephrased to avoid having to make this distinction.
(4) Thanks ;-), changed.
(5) We have rephrased this to state: "Such an assumption is supported by the lack of evidence of strong feedback of the climate on the aerosol forcing" Using feedback makes the intent of the statement clearer, i.e., that their is little evidence that the response to the aerosol forcing is associated with a circulation or other change that in turn modifies the aerosol forcing.
(6) We are not aware of studies that make a case for co-variances amplifying radiative forcing, nor given its linearity is it clear to us how a clear-sky effect would work – through cloud masking? But to address the reviewer's comments that there could be amplifying effects, after the one example cited we now state that "these effects are accounted for ..." rather than "these moderating effects ..." to be open to the possibility of things working in a different direction.
(7) Capitalized.
(8) Revised.
(9) Rephrased.
(10) Deleted.
(11) Fig. 9 was referenced on the previous page.
(12) This has been rephrased to say that the study implies, rather than demonstrates.
(13) Rephrased, and signs corrected.
(14) True, but our statement is not exclusive.
(15) This point has become mute, as the differences vanish when we correct an analysis error (see general comments).
(16) We have modified the caption to make this more clear. Also in the manuscript we now explicitly state that unless otherwise indicated all values are valid in the mid-visible (550 nm). The comparison to other studies is performed in Table 4.
(17) Changed
(18) Changed.
(19) Yes this means no data, which we now state explicitly in the revised caption.
(20) The lines were meant to refer to the different fits, this is also now stated explicitly.
(21) Average of the ratios, now stated explicitly.
(22) Redrafted.

(23) Rephrased.
(24) Yes, but it is not as easy to do as it should be, maybe for CMIP6.