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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimates of aerosol radiative forcing from the MACC re-analysis" *by* N. Bellouin et al.

Anonymous Referee #2

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This paper is an attempt to provide another forcing estimate based on assimilated aerosol data for AOD as well as anthropogenic fraction of AOD over land from a model. Over oceans, a method is used based solely on observations, but that also includes a number of unverified assumptions. My problem with the paper is that most of the justification of the assumptions in deriving these estimates is not given, and that the "uncertainty" analysis is not spelled out in detail and does not include the range of uncertainties from the literature that are know to affect such estimates. The authors need to correct the paper to address the following concerns (the majority of which and the most important of which concern the uncertainty analyses).

Abstract, line 13: "being bounded" should be "are bounded"

Page 20075, line 17: actually the susceptability of precipitation formation has been

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shown to be too high in climate models likely leading to incorrect responses to aerosol change (Wang et al., GRL, 2012)

Line 23: specify from an observational-only view

20076: define “DRF efficiencies” so that this jargon is avoided.

Line 23-24: “concludes with the TOA . . .” needs to be reworded. i.e. “concluded that the best estimate of TOA . . .”

Page 20077: line 5: wording: change to “ spread in model results”

11-13: I think the referencing here does not reflect the literature. I would suggest that the Penner et al. reference does not suggest that model estimates are necessarily correct. Only that the use of the satellite method with a model leads to a similar discrepancy, since the satellite method does not properly retrieve the pre-industrial forcing. The Quaas et al. paper does not demonstrate that model estimates of pre-industrial forcing are in error.

Page 20078: line 21-22. Diagnosis of AOD from aerosol mass needs to also specify the aerosol size distribution as well as refractive indices. In addition, one needs to know whether the separate types are internally or externally mixed.

Page 20079: line 9-10: correcting the model in proportion to the original contribution to total mass would be incorrect, if sulfate, for example, would tend to be in smaller sizes (since it is subject to nucleation, which increases aerosol number but tends to produce smaller sizes) What are the assumptions you are building into this assimilation product? This and other assumptions need to be spelled out.

Line 13-14: it corrects for left – out species, but does not correct for the fraction of nitrate that is anthropogenic (without further assumptions). This needs to be spelled out.

Line 20: “interestingly” should be “not surprisingly” since the AOD is assimilated to

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match the satellite retrieval.

Page 20080: “Unfortunately, it is not possible to completely avoid using modelled fields that are not affected by the data assimilation. For example, although the aerosol forcing estimation does not rely on aerosol speciation, it does use an associated variable, the FMF of the total AOD.” So you are not using the information about refractive index (which requires aerosol speciation) to estimate forcing?

Page 20081, line 6: Why aren’t advected natural aerosols in addition to advected anthropogenic aerosols part of what is seen over oceans?

Eq. 1, 2: The sea salt source functions are highly variable. According to Jaegle et al., ACP 2011 and Lewis and Schwartz 2004 (see Jaegle for reference), sea salt is one of the most poorly constrained aerosols. What is the uncertainty associated with this estimate? How does this uncertainty affect your conclusions? For example, in situ observations give a range of mass extinction coefficients from 3.1 to 6.6 m2/g. Thus, there is at least a factor of 2 uncertainty in this estimate, which should be factored into your analysis and conclusions.

Note: equ 3 seems odd: fine mode fraction f is difference in f *tau for non-dust and seasalt divided by difference of tau for non-dust minus seasalt. But it is FMF of tau, right?

Page 20082: Line 11, 12 and Fig.1: Very strange distribution of fine-mode natural: Where are the year-round biogenic aerosols in the tropics?

Line 19, 20: not just errors in forward modeling of AOD, but also errors in modeled/assumed component fractions.

Line 27 – 29: If there is a factor of 2 uncertainty in the seasalt optical depth (based on quotes in Jaegle et al, above), how do you obtain a 10% uncertainty on anthropogenic AOD over ocean. Also, what is justification for uncertainty estimate over land? Later (page

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Page 20083: line 1 - 7: how do you calculate the relative uncertainty? Something is being used to argue for small uncertainties here since 10% anthrop. over ocean / 21% total AOD over ocean is not 31%. It would seem Yu's method is more reasonable estimate of uncertainty. Your statemtent "The method used in this study only compares the FMF to a threshold and the impact of the FMF uncertainty is reduced." Is not clear.

Line 15 – 18: "Here, the AeroCom anthropogenic AOD of 0.029, defined with respect to pre-industrial conditions, has been multiplied by 1.25 to correct for the pre-industrial aerosol distribution, following Bellouin et al. (2008) where aerosol and precursor emissions for the year 1860 were used to represent pre-industrial aerosols." Please clarify this writing. Are you correcting the AeroCom values because their PI case was for 1850, whereas yours is 1750? Where did you get the factor of 1.25?

line 19-21 "There is therefore some agreement in the fraction of present-day total AOD that is anthropogenic be20 tween satellite-based, assimilation-based, and modelling-based estimates. However, free-running global aerosol models have lower total and anthropogenic AODs." Yes, comforting, but clearly some of this is just the choices for dust AOD and sea salt AOD you made, and other choices would not have provided a similar fraction. Also, the regional fractions may be totally different. Please provide these as well.

Page 20084: line 3, 5: "The aerosol vertical profile is a key parameter in the longwave spectrum." Actually it is a key parameter in determining the effects of BC as well. If you cannot rely on it for the longwave, why is it ok in the shortwave?

Line 6 – 8: Are the derived optical properties consistent with the optical properties used in the satellite algorithm to obstain the optical depth? This is key to determine, since otherwise you are relying on optical properties for radiative forcing that are not consistent with your optical depths based on satellite data. I think the answer you provide is a good start: providing one method of getting forcing, but a second evaluation using the optical properties from the satellite analysis should be made to obtain a range

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of results.

Table 1 only gives one value of single scattering albedo for each aerosol type and no value for mass extinction efficiency (needed for radiative transfer calculation of forcing). Why doesn't the single scattering albedo change with relative humidity (RH)? Even if you are claiming it is fine to use the average boundary layer RH value of SSA over these large areas, I would think there should be a seasonal variation of RH over land areas at least. Also, the use of strongly absorbing anthropogenic aerosols (SSA = 0.86) over Africa for all ocean areas would seem to bias your calculation of anthropogenic aerosol effects over the oceans. Or since this table refers to properties over land areas, perhaps you use something else over oceans? What is it? Also table heading states: " Representative AERONET sites refer to the sites studied by Dubovik et al. (2002). For the other components, optical properties are prescribed globally" What exactly are you prescribing globally? I gather you do not use the regional values globally (i.e. from GSFC, Creteil, . . .) Maybe you just refer to the dust and sea salt properties?

Line 22 – 25 "Rather, it is assumed that natural aerosols are located in the first kilometre of the atmosphere, below a layer of anthropogenic aerosols. For the cloud-free DRE, the impact of this assumed vertical profile on shortwave radiative 25 fluxes is small" But, these would not be small effects if you were taking RH variations into account. What is the impact of neglecting these variations?

Page 20085: line 3 – 4: Figure 3: I still fail to understand a lack of maximum natural component of forcing in the tropics, and that is similar for all seasons.

Page 20086, line 4-6: "Uncertainties in component AOD, computed above, are translated to uncertainties in component DRE by multiplying by the component DREE." These are not explained on page 20082, so I would not say you "computed" the uncertainty. The way this is stated, it looks like you just pulled a number of 15% (for the anthropogenic component) out of the air. (see 20082 line 28)

" In addition, the uncertainty on 5 the DREE due to uncertain prescribed optical prop-

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erties has been quantified for the anthropogenic component by the Monte-Carlo approach of Bellouin et al. (2005). " What was the approach of Bellouin? Please do not make the reader go to another paper. You need to summarize this approach here. Especially, if one goes to the 2005 paper and finds numbers pulled out of the air, then we are building "science" out of thin air.

Line 17: values are not usually "stronger": use "larger"

Page 20087, line 13 -14: how different would the DRF be if you use natural aerosols over anthropogenic aerosols (i.e. one way to indicate uncertainty).

Line 25-27: Model's have shown that this area has positive forcing since 1998. This is expected theoretically for small SSA for biomass burning aerosols.

Table 3: The uncertainty analysis is crucial here. You need to provide details. I also think you need to do this regionally, since the errors must depend on the regional character of the aerosols and the choices you've made.

Page 20089: line 15, 16: dln (Nd)/dln(AOD) from the Quaas et al analysis can be very different than the change that a model would compute from dln (Nd) / dln (tau – tau(natural)), i.e. the slope of the line computed from satellite data does not necessarily capture the change in droplets based on the full optical depth and the natural optical depth because the satellite data are unable to select only the natural data (i.e. even a low optical depth cannot be considered "natural"). What is the possible error/uncertainty due to this effect?

Page 20090: line 2: the use of AOD as a proxy for aerosol number is known to be highly uncertain (better to use angstrom exponent (AE) times AOD. How different are your results if you use the latter? (The measurements by Andreae that you quote do not form a tightly fit 1:1 line.)

Line 9 – 10: actually the primary criticism of using dlnNd/dln(AOD) which applies to both the use of AOD and the use of AOD X AE is that the slopes derived from satellite

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data for present day do not represent the slopes for present day – natural (see comment above).

Line 10 – 12: Grandey and Stier mainly fault the method based on using a large area average (shown also in a previous Quaas paper). And this may indeed lead to an overestimate. But this is no reason to suggest that the estimate given here is ok, considering it is faulty in both directions! Please use the information in these papers to estimate the uncertainty in your methods.

Line 20 – 21: the use of different regions/seasons for the partial derivatives here would suggest this also for the optical properties used for the direct effect (for balance, if not a more defensible estimate).

Line 28: I am surprised the fractional cloud cover is thought to be well known from the IFS. How do these values compare to observations? We should at least be given an estimate of how "certain" they seem, since the paper makes this judgment.

Page 20091, line 1, 2: don't tell me you are just going to evaluate equ. 6 using seasonal averages and apply the range based on seasonal values as the uncertainty? What exactly do you do?

Line 10, 11: do these compilations address the issue noted above (i.e. that the primary criticism of using dlnNd/dln(AOD) which applies to both the use of AOD and the use of AOD X AE is that the slopes derived from satellite data for present day do not represent the slopes for present day – natural)? Somehow, I do not think so, since they are all based on data for the total aerosol.

Line 15: You need to provide a table with the uncertainties of the different factors that you include here and then deriving the 37%. Otherwise no one can truly know what you've done, and a scientific paper needs to have enough information for the next researchers to be able to reproduce your results.

Line 20 – 23: It is nice that you get values that are somewhat larger than IPCC, but I

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suspect a true analysis of uncertainties would be far larger.

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