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Interactive comment on "Global direct aerosol radiative forcing, as constrained by comprehensive observations" *by* Chul E. Chung et al.

Anonymous Referee #1

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Quantifying the direct radiative forcing of anthropogenic aerosols can be attempted either using observational or model data, or some combination of both. While models can cleanly separate anthropogenic and natural aerosols, aerosol simulations are still subject to large uncertainty. The use of observations alone is also problematic, due to issues regarding spatial coverage (AERONET) and/or accuracy (satellite observations of aerosols over land), and furthermore, assumptions are needed for separating the anthropogenic and natural aerosols. The current paper aims at providing the "most observational estimate of direct aerosol radiative forcing to date". For this end, available aerosol observations from AERONET, MISR and MODIS, along with vertical profile information from CALIPSO, are combined with GOCART simulations (and other model

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Discussion paper

results in Sect. 5). The main outcome of the paper is a best estimate of -0.10 W m^{-2} for the global mean TOA direct radiative forcing of anthropogenic aerosols, with a quoted uncertainty range of -0.28 to $+0.05$ W m^{-2}.

I find this paper an interesting contribution worth publishing in ACP. However, while the paper is concise, it is currently not well-written, and it takes the reader (or reviewer) too much effort to understand, in detail, what you actually did and why. The clarity can and must be improved before the paper can be accepted for ACP. Specific suggestions are given both in the major comments (comment 3) and minor comments.

Major comments

1. Some assumptions in this paper need more discussion. In particular, to separate the anthropogenic direct aerosol forcing from the total aerosol direct radiative effect, it is assumed that (i) fine-mode aerosols are anthropogenic, except for fine-mode sea-salt and dust aerosols, and that (ii) all coarse-mode aerosols are natural. Neither of these assumptions is strictly true. First, the fine mode also includes natural particles (e.g., biogenic SOA from natural non-combustion sources, and BC from combustion, e.g. naturally occurring forest fires). Note that Bond et al. (2013) give (in their Sect 10.4.1 and Fig. 35) a best estimate of +0.17 W m^{-2} for the preindustrial (year 1750) direct radiative effect of BC (as compared with a total natural+anthropogenic direct effect of +0.88 W m^{-2} in year 2005). Second, some of the coarse-mode aerosols are anthropogenic. In particular, note that the IPCC AR5 best estimate of the direct radiative forcing (or "aerosol-radiation interaction") of -0.35 W m^{-2} includes a contribution of -0.1 W m^{-2} from changed dust emissions due to human activities (though with a large uncertainty range, from -0.3 to +0.1 W m^{-2}). See Table 8.4 in Myhre et al. (2013).

Separating out natural BC or anthropogenic dust would most likely be very difficult in the current framework, and I'm not suggesting that this be attempted. However, these issues should be discussed. If these factors were considered, it would presumably make your best estimate of the direct radiative forcing of anthropogenic aerosols more negative, and more in line with IPCC AR5.

REFERENCES:

Bond, T. et al., 2013: Bounding the role of black carbon in the climate system: A scientific assessment. J. Geophys. Res., 118, 5380–5552.

Myhre, G., D. Shindell, et al. 2013: Anthropogenic and natural radiative forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group 1 to the Fifth Assessment Report of the Intergovermental Panel of Climate Change.

2. In section 5, the contribution of dust and sea salt to the fine-mode AOD (hereafter: $\mathrm{AOD}_{\mathrm{SD,fine}}$) is estimated as

$$\mathrm{AOD}_{\mathrm{SD,fine}} = \text{coarse-mode AOD} \times \frac{\mathrm{SD\_FMF}}{1 - \mathrm{SD\_FMF}}, \quad (1)$$

where $\mathrm{SD\_FMF}$ refers to the simulated fine-mode fraction of sea salt + dust AOD. While I can understand the reasoning behind this formula, the estimate is sensitive to errors in $\mathrm{SD\_FMF}$. Especially when $\mathrm{SD\_FMF}$ is too large (i.e., too much of simulated dust + sea salt is in the fine mode and too little in the coarse mode) $\mathrm{AOD}_{\mathrm{SD,fine}}$ can become overly large due to the factor $1 - \mathrm{SD\_FMF}$ in the denominator. In fact, nothing prevents the derived $\mathrm{AOD}_{\mathrm{SD,fine}}$ from exceeding the total fine-mode AOD, or even the total AOD in the observations! This seems to happen over Peru, where the total AOD in Fig. 1A is below 0.2, and the fine-mode AOD in Fig. 3A is around 0.10–0.15, but the fine-mode

AOD for sea salt + dust in Fig. 5A exceeds locally 0.3. This is related to a large positive anthropogenic fine-mode TOA forcing in Fig. 6A.

At minimum, it should be ensured that the derived $\mathrm{AOD}_{\mathrm{SD,fine}}$ does not exceed the observed total fine-mode AOD. A stricter upper limit for $\mathrm{AOD}_{\mathrm{SD,fine}}$ might be derived by estimating some lower limit for the (absolute or fractional) contribution that other species than dust + sea salt make to the fine-mode AOD.

3. After reading Section 2 several times, I am still not quite sure how the aerosol optical properties were derived in this study, but I try to summarize my understanding here:

- AOD at 550 nm was derived by nudging MODIS and MISR data towards AERONET observations. Remaining data gaps were filled by a GOCART simulation.
- Single-scattering albedo and asymmetry parameter at 550 nm were derived by nudging GOGART simulations towards AERONET data.
- Fine-mode AOD at 550 nm was obtained by subtracting coarse-mode AOD at 500 nm from the AOD at 550 nm, subject to the assumption that the coarse-mode AOD is equal at 500 and 550 nm. However, it is not clear from Section 2.1 how the coarse-mode AOD was obtained. Which of AERONET, MODIS, MISR and GOCART were used (and how)?
- It is not quite clear how the fine-mode single-scattering albedo and asymmetry parameter at 550 nm were derived. But I suppose that for consistency, they were derived by subtracting the corresponding coarse-mode values (with appropriate weighting by AOD) from the total single-scattering albedo and asymmetry parameter, and that the coarse-mode single-scattering albedo and asymmetry parameter were prescribed based on GOCART as explained in Sect 2.3.

- It is stated that the spectral dependence of single-scattering albedo is treated using an coalbedo Ångström exponent CA_AE, which is derived by nudging the GOCART simulation towards AERONET data. However, nothing is said about the spectral dependence of asymmetry parameter. Was it assumed spectrally independent? More importantly, it is not clear how the spectral dependence of AOD was treated. Section 2.3 suggests that you assumed an Ångström exponent of 0 for coarse-mode (dust and sea-salt) and 1.85 for the fine-mode dust and sea-salt (but nothing said about other fine-mode species). Did you use any observations to constrain the Ångström exponent?

If this interpretation is correct, you should make an effort to rewrite Sect. 2 so that it is easy for the reader to get this information (currently it is not). If not, the need for clearer writing is even more acute.

Minor comments

1. p. 1, 8th line of abstract (and elsewhere). In IPCC reports, "forcing" refers to changes in the radiation budget, so it would be better to reserve the term "radiative forcing" to the effect of anthropogenic aerosols. Thus, when discussing the impact of all (natural + anthropogenic) or natural aerosols on the radiative budget, "aerosol radiative effect" is preferable.

2. p. 1, first paragraph of Introduction: I assume that what you consider here is the climatic effect of anthropogenic aerosols. Please state this explicitly.

3. On p. 1 and 2, there are several citations to (Myhre, 2013), while it should be (Myhre et al., 2013).

4. On p. 2: The concepts of “fine-mode” and “coarse-mode” (or “large-mode”) aerosols are central for this study, but their meaning is not explained adequately. E.g., can the modes be defined in terms of a size limit?

5. p. 3, first paragraph of Sect. 2.2: mention also the asymmetry parameter.

6. p. 3: There seems to be something wrong with the equation for $\mathrm{N\_ASY}_j$. In case that there is no difference between the AERONET and GOCART values, $\mathrm{N\_ASY}_j$ becomes 0!

7. p. 3: If the locations for AERONET stations are indexed with i and model grid boxs with j, why are there two indices (j, i) for both $\mathrm{AERONET}_{j,i}$ and $\mathrm{G\_ASY}_{j,i}$? Why is it not simply $\mathrm{AERONET}_i$ and $\mathrm{G\_ASY}_j$? The same goes for the following equation for $1 - \mathrm{N\_SSA}_j$.

8. On p. 4, three different sets of aerosol optical properties are considered. While the selected parameters do not seem unreasonable, it is not clear how they were selected (no single reference is given!), nor whether the selected range represents well the real uncertainty range. For example, the single-scattering albedo of BC is taken here to be either 0.14 or 0.19. For comparison, Bond et al. (JGR 2013) cite in their Section 3.8.3 measured values between 0.10 and 0.28, and modelled values for aggregates of pure BC particles of 0.1 to 0.3. Further, a value of 0.36 based on Mie theory is applied in Appendix 3.1. The authors should either justify their values and/or test the impact of a wider parameter range.

9. p. 4. Please consider the use of a more rigorous scientific notation in your equations. Now, for example, “dust” refers to the total AOD for dust in the equations for SSA1, SSA2, and SSA3, the scattering AOD for dust in the equation for ASY, and presumably again the total AOD for dust in the equation for CA_AE. Furthermore, “BC”

refers to the GOCART simulated BC AOD in the equations for SSA1 and SSA3, but twice this value in the equation for SSA2. So apparently "total_AOD" is also a slightly different beast in the equations for SSA1 and SSA3 than in the equation for SSA2. Honestly, this becomes rather confusing. Similarly, on p. 3, it would be clearer to use AERONET_ASY in the first equation and AERONET_SSA in the second equation.

10. p. 4. Also, please consider using equation numbers to make it easier to refer to the equations. (Indeed, they would have made the previous comment shorter!).

11. p. 4. In the equation for CA_AE, "CA" refers to both "coalbedo" (on the left) and to "carbonaeous aerosol" on the right. To avoid confusion, I suggest the use of BC+OC (or, preferably, BC_AOD + OC_AOD) on the right hand side.

12. p. 4–5. Is there a reference for the equations for CA_AE in Sect. 2.2 and 2.3?

13. p. 4. While it is probably true that the uncertainty in asymmetry parameter is unimportant compared to the uncertainty in SSA, this uncertainty cannot be quantified by just doubling the BC AOD (BC makes only a minor contribution to the total AOD, so it has a small impact on the average asymmetry parameter). A more reasonable estimate might be obtained by perturbing the total aerosol asymmetry parameter by, say, ± 0.05. Obviously, even this would be a subjective choice, but since all your asymmetry parameter values are rounded to nearest 0.05, it seems reasonable to assume that the true values are not known much more precisely than that.

14. p. 4. "Global simulations" is a somewhat misleading title for Section 2.3. No global simulations are described here, just the assumptions about aerosol optical properties.

15. p. 5, 8th line of Sect. 2.4. Is "vertically summed aerosol extinction" the same as AOD, or something else?

16. p. 5–6. Section 3 should contain more information about the input data for the MACR model. Currently, it describes some details related to low clouds but does not explain how, in the first place, clouds were treated and where the cloud data came from. I realize that this information might be found in Choi and Chung (2014), but the main points should be repeated here to make this paper stand alone.

17. p. 8, first line: The criteria for selecting dust-dominated Aeronet sites are not intuitively clear. For example, why is it required that the absorption AOD exceeds 0.03? Dust is relatively weakly absorbing, with a single-scattering albedo of 0.96 at 550 nm used in this paper.

18. p. 9, last paragraph: It is concluded that the direct radiative forcing of anthropogenic aerosols is close to zero. Another important result, which would deserve some more discussion, is that the anthropogenic aerosols strongly increase atmospheric absorption (by 3.64 W m^{-2}, according to Table 1) and decrease the net solar radiation at the surface (by 3.74 W m^{-2}). This suggests that the primary climatic impact of anthropogenic aerosols is not that of cooling the planet but reducing precipitation (less solar energy available for evaporation, and less latent heating needed for balancing radiative cooling). Reduced precipitation due to absorbing aerosols has of course been discussed earlier, e.g. Ramanathan and Carmichael (2008) and Ming et al. (2010).

REFERENCES:

Ramanathan, V. and G. Carmichael, 2008: Global and regional climate changes due to black carbon. Nature Geosci., 1, 221-227.

Ming, Y., V. Ramaswamy and G. Persaad, 2010: Two opposing effects of absorb-

ing aerosols on global mean precipitation. Geophys. Res. Lett., 37, L13701, doi:10.1029/2010GL042895.

19. p. 10: Where was the review paper by Chung (2012) published?

20. p. 16: Fig. 4. The dot size scale (1, 2, 3) related to the number of AERONET climatological data is uninformative. Please be more explicit.

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