**RESPONSE TO REVIEWS**

ACPD article “Beyond direct radiative forcing: the case for characterizing the direct radiative effect of aerosols”

We thank the three reviewers for their comments and suggestions. Two of the reviews suggest accept with minor revisions (referees #2 and #3), the first referee suggests that the article should be rejected. While we respectfully disagree with referee #1, we have endeavored to clarify both the manuscript and our motivations in response to this referee’s comments. Reponses are noted below with original referee text given in italics/blue. A “track changes” version will also be submitted to the editor. Our responses are in reverse order, referees 3 through 1.

**Response to Referee #3**

*1) The current title should be abandoned in favor of something more in line with the results presented, such as “Aerosol radiative forcings and effects in GEOS-Chem”.*

We have modified the title to “Contrasting the Direct Radiative Effect and Direct Radiative Forcing of Aerosols”

*2) Statements, even if plausible, that are not directly supported by quantitative results in this paper such as “while the climate feedbacks on aerosols under rising global temperatures will likely amplify.” should be removed from the abstract.*

We have removed this phrase from the abstract.

*3) p. 32928, lines 23-25: “In this study our objective is to quantify these two metrics, and make the case that the DRE is a necessary complement to the DRF for aerosols.” The paper presents results to obtain the first objective. As already indicated, there are no concrete results for the second. The second objective should be deleted.*

We have deleted the second phrase.

*4) I don’t think it’s necessary to delete all discussion of climate feedbacks on DRE. It’s fine to cite them in the introduction as motivation for calculating DRE. It’s fine in the final discussion to speculate a little on the implications of DRE being much larger than DRF. I draw the line at anything, especially in title/abstract, that claims this paper has demonstrated that climate feedbacks on DRE are greater than DRF.*

As requested, title and abstract are modified. All text discussing implications is limited to the Discussion and Conclusions section.

*5) The literature on climate feedbacks/natural aerosols that appears in the discussion (plus other papers not cited) really should be cited up front in the introduction to avoid giving the reader the initial impression that the argument is original to this paper.*

We have moved text from the Discussion to the Introduction to clarify this.

*6) While the authors should respond to the specific "deficiencies" in radiative transfer calculations mentioned by the first reviewer, in my judgment, these are not serious enough to preclude publication. If at all possible, I do think the paper would benefit greatly from another calculation where BC is treated as internally mixed as I also believe it's more realistic despite the recent Cappa paper. At the very least, the limitation should be noted earlier (in the methods section), and the results section should discuss how much its omission is likely to affect the numbers presented (e.g. if we double BC absorption, total DRF does change notably).*

We agree that BC absorption is a very interesting topic, however it is a research topic all on its own, outside of the scope of this paper. Internal mixing is not a "standard" treatment in aerosol models (half of the AeroCom II models do not include this according to Myhre et al., 2013). We indicated our external mixing assumption in the text and that therefore "both DRE and DRF values may underestimate absorption". As suggested by the referee, we have also added a sentence on this to the methods section: "We do not include any absorption enhancement from the coating of BC in these simulations" and also in the discussion of Table 3: "If we double BC absorption, as a crude approximation of internal mixing, then the 2010 DRF would be -0.28 W/m2, a 20% change from our base estimate."

*The introduction discusses the effective radiative forcing (ERF). Since the authors are using a CTM with fixed met fields, their paper cannot really address this issue. While I don't have a problem with an introduction that strays a little from the main topic of the paper, does this discussion really need to be here?*

We completely agree with the referee that this sentence is not strictly necessary. It was added just prior to submission as the IPCC AR5 executive summary had just been released and we wanted to include the definition for completeness. We think it's worth keeping the definition for context as the ERF concept gains traction in the community following AR5.

*p. 32931, lines 22-23: "This approach greatly increases computational speed while maintaining the effective accuracy of a line-by-line calculation." Since a correlated-k method must degrade accuracy (even if by a negligible amount), this statement de serves some quantification. Alternatively, it may be deleted since the accuracy of RRTMG for line-by-line absorption calculations doesn't seem to be terribly important for aerosol effects.*

Sentence has been deleted.

*p. 32935, lines 18-24 regarding AeroCom I BC biases: the authors correctly point out that the models sometimes overpredict BC mass concentrations but neglect that they also underpredict BC over Asia. The discussion should be more objective/balanced.*

We expanded our text on the AeroCom model underestimates to include Asia: "but underestimate BC at high Northern latitudes and in Asia"

*p. 32939, lines 11-18 "Uncertainties in our estimate of DRE are likely even larger than uncertainties on DRF." (and subsequent paragraph of discussion) This is pretty speculative as the paper doesn't include any sensitivity/uncertainty calculations.*

We felt it was critical to include a discussion of potential uncertainties, but agree that we are unable to quantify these (these factors are currently too poorly understood to quantify). We have moved this paragraph (and the preceding one on uncertainties in the DRF) to the Discussion section so that the more "speculative" nature of these comments is clear to the reader.

*p. 32940, lines 10-12 "This calls into question the usefulness of separating so-called "anthropogenic" and "natural" aerosol forcing." Please be careful. There are some gray areas where the distinction is murky, but most of the time, the distinction is pretty clear and the separation is useful. Let's not throw the baby out with the bath water*

We have re-phrased: "This complicates the separation and interpretation of "anthropogenic" and "natural" aerosol forcing."

**Response to Referee #2**

*Throughout: The use of the term "pre-industrial". In the IPCC and elsewhere (e.g. AEROCOM) pre-industrial typically refers to the year 1750, though other years are occasionally used (e.g. 1800 or 1850). In 1750 (and certainly the later dates), anthropogenic emissions are far from zero (e.g. biofuel). I'm not aware of any study where "pre-industrial" is defined as shutting all anthropogenic emissions off. Thus, the "pre-industrial" simulation performed here is not "pre-industrial" in the traditional sense. Furthermore, the AEROCOM studies use 1750 as a basis, and you compare to the AEROCOM DRFs in Table 3, but this makes for an apples/oranges comparison (not quite as bad as swapping DRE for DRF, but still 2 very different DRFs!). Calling the simulation "no-anthro" would avoid confusion for the reader, which is important here since the purpose of paper is to reduce confusion between different, but related definitions of radiative differences between simulations. However, it is not obvious how best to compare directly to the AEROCOM simulations without performing simulations with 1750 emissions. Maybe the authors could determine if the differences between 1750 emissions and no-anthro are minor?*

We thank the referee for this comment. The Dentener et al., (2006) emissions used in AEROCOM estimate zero fossil fuel emissions for 1750 and very small emissions for biofuel. Thus our "zero" anthropogenic baseline is a good proxy to this. We have quantified this and added text to the manuscript with these numbers.

*P32927 L3-5. Please go into details as to the history of the usage of DRE. There are no citations here, which makes it unclear if the authors are saying this is a new term. Review 1 goes into the previous DRE vs. DRF studies in detail, so I won't say more here.*

We have added to the discussion.

*P32927 L10. In the IPCC, "pre-industrial" is defined as 1750. Good to state this explicitly, and good to make it clear that this is not what you calculate in the paper (once you introduce the simulation). The start date is certainly important for forcing (e.g. Carslaw, Nature, 2013).*

We have added this. Refer to comment above on our baseline year. We note that while IPCC defines PI as 1750, many other studies (including ACCMIP) employ 1850.

*P32928 L25, end of intro. I was missing clear statements of what the goals of this paper are. Is it to demonstrate that DRE and DRF are different? Is it to show the DRE and DRF results in the default GEOS-Chem-RT simulations.*

The last sentence of the introduction gives this information: "In this study our objective is to globally quantify and contrast these two metrics."

*P32929 L12 and throughout. When I see "GCRT" I think "GOCART", which is also a global CTM with online radiative transfer. To avoid confusion, I suggest using GEOS-Chem-RT.*

We did not think of this potential confusion! We have modified our acronym to GC-RT to preserve the short label, but remove the association with GOCART.

*P32933 L5: Since your results may depend greatly on what you assume about the BC optics (e.g. coating effects), please briefly describe here. At the end of the paper you mention ignoring coating effects, but it should be stated up front. Also, I agree with reviewer #1 that the Cappa study is currently a lone horse showing that the coating effect is negligible, and alone it probably doesn't justify ignoring coating effects.*

We have added this up-front, please see response to referee #3 for more details.

*P32933 L7: Is water uptake to only sulfate considered? I'm guessing you consider water uptake to other species too, but this isn't mentioned.*

Yes, we include this. The first sentence of the paragraph indicates that aerosol refractive indices and growth factors are from GADS. We have added "hygroscopic" before growth factors so that this is explicit.

*P32934 L23-27: Can you elaborate on why aerosols produce a warming over the sahara and the middle east? Surely the single-scattering albedo of the particles is higher than 0.3-0.4 (the visible surface albedo). Is the warming because of the LW absorption effects of the aerosols?*

The referee is correct, the warming from this region is mostly from LW absorption of dust. There is a very modest contribution from dust in the SW (see Figure 2) in the highest albedo locations over the Sahara. We have edited the text so that it is clear here that we are only talking about the shortwave, not the dominant effect of longwave absorption seen further down in Section 3. We also clarified the text in the later discussion in Section 3.

*P32935 First paragraph: This seems like it belongs in the methods section.*

We have moved the paragraph as suggested.

*P32935 L13: Is it reasonable to assume that as much as 20% of the dust in the middle of major deserts (e.g. the sahara) is anthropogenic? It might be worth mentioning that there should be regional variability in the number that you're currently ignoring.*

Statement to this effect has been added.

*P32937 L3-9: Can you give more detail on how you calculated the cloud-sky TOA DRF? It it just -0.36 = 0.6*(-0.57) + 0.4*x and solve for x? I had to think for a minute to come to this.*
Almost. It is -0.36 =0.6x+0.4(-0.57). This is the standard calculation for cloud-sky TOA DRF.

*P32938 L10: Why is boreal springtime the peak DRF?*
The sulfate DRF peaks in Spring (as seen in Figure 5); this is the seasonal peak in global sulfate burden.

*P32941 L16: Could cite Carslaw, Nature, 2013 along w/ Menon*
Added.

**Response to Referee #1**

*• The authors are right that the distinction between DRE and DRF is somewhat confused in the literature. However there is whole lot of articles where the distinction is made very clearly, as it is the case in most modelling studies but also in some early observational studies (e.g. Bellouin et al., Estimate of aerosol direct effects over land and oceans from MODIS, Nature, 438, 1138-1141, 2005; Bellouin et al., Estimates of aerosol radiative forcing from the MACC re-analysis, Atmospheric Chemistry and Physics, 13, 2045-2062, 2013; etc.). The IPCC (2013, chapter 7) is also very clear on this. The title and the introduction suggest the authors are the first ones to clarify the concept when in fact they should be pointing to the right literature on the subject.*
We appreciate the referee's comments here. We have added references to the observational studies mentioned by the referee, and specifically used Bellouin et al., 2013 as an example of where this contrast between DRE and DRF has been explicitly discussed (Bellouin et al., 2005 do not make this distinction and do not discuss DRE).
Our objective in this study was to clearly distinguish the global DRE and DRF simulated for present-day and show that the DRF is likely to decline in the future, thus motivating a further need to look at DRE. We are not aware of any global modeling studies which have contrasted the DRE and DRF as we have done here. Further, given the misconceptions about this terminology in the atmospheric chemistry community, we feel that while this idea is not new (i.e. we are not inventing new terminology) it requires further reinforcement and substantiation. Following the recommendations of referee #3 we have modified the title and introduction such that the context for this work is clearer.

*• Calculations of DRE are not new, see e.g. Boucher and Tanré, Estimation of the aerosol perturbation to the Earth's radiative budget over oceans using POLDER satellite aerosol retrievals, Geophysical Research Letters, 27, 1103-1106, 2000,and many others since then. The LW contribution to the DRE was calculated by e.g. Reddy et al. (JGR, 2005) and some early papers by Jacobson among others.*
We certainly do not mean to imply that DRE has not previously been calculated. Our modified text should make this clear. We have added the reference to Boucher and Tanre in our introduction of the term DRE.

*• There is some literature on how to differentiate feedbacks from forcing in relation to biogeochemical cycles (e.g. Gregory et al., Journal of Climate, 2009; Raes et al., JGR, 2010). This literature is ignored here*

*although the authors frame their paper around the importance of aerosol feedbacks. A similar framework to that of Gregory et al. was used by Carslaw et al (ACP, 2010, cited in the present manuscript) and feedback parameters in unit $Wm^{-2}K^{-1}$were provided. This metric, also used in Chapter 7 of the IPCC (2013), seems more appropriate than DRE alone when it comes to quantify aerosol feedbacks.*

We agree that these are useful metrics and have added a discussion of this to the text. We argue only in our paper that quantifying the DRE is a first step towards a more complete picture of aerosol impacts on radiation, and should be compared between model studies. As the DRE does not require quantification of the climate response it is a metric that can be estimated for a broader suite of models (including CTMs).

*The authors perform their radiative calculations with a rather accurate radiative transfer code. This is an interesting feature of their study. However it is well known that DRE and DRF are very sensitive to the model's surface albedo, relative humidity and cloud properties. In this respect the authors are not doing any better than other publications, and they miss here an opportunity to set a higher standard in aerosol DRE and DRF calculations.*

*• Little information is provided on the cloud distribution and SW/LW properties beyond the assumed size of the cloud droplets and ice crystals. It is clear that a large fraction of the spread in aerosol DRF is due to the input cloud climatology (e.g., Stier et al., Host model uncertainties in aerosol forcing estimates: Results from the AeroCom Prescribed Intercomparison Study, Atmospheric Chemistry and Physics, 13, 3245-3270, 2013).*

The focus of this study is not the cloud distribution. We note that we are not using a climatology here, but rather the distribution from the GEOS-5 assimilated meteorology for 2010. We have added a reference to Bosilovich et al., 2011 which discusses the MERRA product water budget (including clouds). MERRA is based on the same GEOS-5 assimilation.

*• The authors do not consider diurnal variations in the surface albedo. It is well known that the ocean surface albedo has large diurnal variations (because of diurnal variations in the solar zenith angle) that are positively correlated with the aerosol upscatter fraction (which also varies with solar zenith angle). Neglecting this covariance results in systematic biases in aerosol DRE estimates, while assuming a Lambertian surface introduces further uncertainties (see Bellouin, et al., Estimating the aerosol direct radiative perturbation: impact of the ocean surface representation and aerosol non-sphericity, Quaterly Journal of the Royal Meteorological Society, 130, 2217-2232, 2004).*

We have added a statement to the text on this.

*• The authors consider aerosol to form an external mixture, when it is well known and for a long time that internally-mixed BC increases absorption substantially (IPCC, chapter 7, 2013, and many references therein). The reference to Cappa et al. (2013) to justify the uncertainty on the absorption enhancement is misleading as there is plenty of evidence in favour of an enhancement effect.*

See response to referee #3 – we have extended our description of this in the paper.

*It is not clear how the conclusion that "SW-only aerosol DRE or DRF estimate would overestimate the cooling effect by 5–10%" comes from (page 32937, line 15). The 10% upper bound is about correct for the DRE but is an overestimate for the DRF because most of the LW effect comes from non-anthropogenic aerosols such as dust and volcanic stratospheric aerosols.*
We have removed DRF from this sentence.

*page 32927, line 20: The sentence has no meaning. Should DRF be ERF here? This would make more sense, although still confusing I think.*
We have edited the sentence for clarity.

*page 32927, line 23: I do not see how something published in 2007 can confuse something written in 2013. IPCC (2013) is very clear about the distinction between DRE and DRF (they are called REari and RFari by the way).*
We have re-phrased to further clarify. We agree that these definitions are much clearer in AR5. However the executive summary of the AR5 was published only weeks prior to the submission of this article; the full report only became citable while in review. We feel that the historical perspective on these IPCC definitions is useful to explain (to some degree) why DRF has been used incorrectly in the community.

*page 32934, lines 25-27: the sentence "aerosols are typically more scattering than surface albedo" does not mean anything. The authors are comparing apples and oranges here.*
We have deleted this sentence.

*page 32946: the reference to the IPCC SPM 2013 is inadequate as RF/ERF and RFari/REari are not discussed in much details there. The authors should refer specifically to Myhre et al, chapter 8, 2013 and Boucher et al., Chapter 7, 2013.*
The AR5 full report was not citable when the article was submitted. We do not discuss the RFari/REari definitions of AR5 here, thus we feel that further detailed citations are not required.