Response to reviews of:

Solar geoengineering using solid aerosol in the stratosphere

by D. K. Weisenstein[1] and D. W. Keith

ACPD-15-11799

Response to Anonymous Referee #2

> First we would like to thank the reviewer for a careful reading of our manuscript and many constructive comments that have improved the quality and clarity of the paper. We have added a table to list the model experiments described and an appendix to detail the coagulation and condensation schemes. We added a paragraph on the limitations and inherent size broadening of a discrete sectional bin scheme. We have included a more thorough description of the AER 2-D model's dynamical fields and PSC parameterization and discuss some model comparisons with observations for both aerosols and ozone. We adjusted our RF values in Figures 7 and 13 due to a correction in the RF albedo adjustment. We added a comparison of ozone change from sulfate geoengineering to that from alumina and diamond geoengineering in Figure 12a. We also replace the unit megatons with terragrams throughout the manuscript. We have added seventeen new references and increased font size and readability of the figures. J. A. Dykema has been added as a co-author (previously recognized in a footnote and acknowledgement) due to his scientific contributions in addressing reviewer questions regarding heating rates and radiative forcing.
>
> Our answers to the reviewer's questions and comments are detailed below, with the reviewer's comments in black and our responses indented and in green.

The manuscript describes a novel approach to manipulate the stratospheric albedo to counteract global warming (geoengineering; solar radiation management, SRM) by releasing solid alumina particles into the lower stratosphere. The authors investigate the response of a two-dimensional coupled aerosol microphysics-stratospheric chemistry model to estimate the desired aerosol radiative forcing and attempt to quantify associated risks concerning the potential increase in diffuse light and impacts on the stratospheric ozone budget.

This study is a necessary step towards a better understanding of this particular research topic. The manuscript is generally well written. Although investigating a new type of aerosol in the (higher) atmosphere, the author's succeed in introducing the necessary background information without overstraining the reader. Methods are explained fairly good, the quality of figures are matching standards of a scientific journal, and the results seem plausible.

I do have, however, a few concerns which should be addressed before the manuscript is published in ACP.

General comments:

The current structure of the paper does not allow to get a quick overview about the experiments which have been conducted and are described, before one has read the entire Sect. 3. Even then it remains unclear, because on several places the author's phrases like „..we perform additional model calculations...", or „... model...for a number of parametric...scenarios...", and so on. The paper would substantially gain in quality if the experiments are briefly described en bloc before the results (even the test cases) are addressed. I suggest to introduce such a (sub-)section before the section „Model results", potentially also including an additional table presenting an overview of what has been tested and which parameter have been adjusted (in my understanding a common practice in manuscripts presenting numerical studies).

> We have added Table 1 to provide an overview of the experiments. We added a new paragraph at end of section 3.1 to reference the table and describe the 10-year model spinup and averaging period.

In this respect I also found it difficult to understand when the model was interactively coupled to a chemistry scheme and when not. Since chemistry is such an important issue for the study, and the author's relatively often emphasize that they assess potential risks associated with the method, I suggest to rephrase respective parts of the manuscript and make chemistry coupling more transparent.

> We'd initially done all the alumina calculation without full chemistry to get burdens and distributions of alumina, then repeated them with full chemistry and aerosols coupled to get the ozone response, which led to the confusion. We have modified the paper to only discuss the results with full interactive chemistry and aerosols so as to avoid confusion. Results are identical except for somewhat different OH concentrations affecting sulfate, which is evident in Figure 9. We've modified the first 2 paragraphs of Section 2 accordingly.

Concerning simulated ozone changes, I wonder why the experiments are not compared in more detail to recent studies investigating ozone changes due to proposed geoengineering scenarios using SO2 or sulfate (e.g. Tilmes et al. 2008,2009; GeoMIP)? This is a clear deficit of the manuscript. I understand that the model applied here is technically different from other models which have been used to study the geoengineered sulfate-ozone relationship. But I in my understanding of the research topic and chemistry/climate/transport/ aerosol models, several aspects could be elaborated here.

> The focus of this paper is on the microphysics of solid aerosols and on a comparative study of the ozone loss from different solid aerosols. A detailed comparison of ozone loss calculations for sulfate aerosol is far beyond this paper's scope. Nevertheless, we do cite the Tilmes references and Pitari et al. (2014) (for GeoMIP studies) and Heckendorn et al. (2009). Unfortunately, none of these studies yield direct comparisons because (1) the emission region is different (others inject at the equator only rather than 30S-30N, and at different altitudes), (2) chlorine and bromine levels are often different, (3) some contain dynamical feedbacks and some don't. However, when we run our model to match the Heckendorn results as closely as possible (narrowing the emission region and eliminating VSL bromine species), we obtain similar values of global ozone change (-4.5% vs -4.75% for 5 Tg-S/yr injections), lending confidence to our calculated ozone changes. Compared to Tilmes, our calculated ozone changes appear to be substantially larger, which may be related to dynamical responses in the WACCM model in the tropics. We have added a sentence to this effect: "Our simulations of ozone change due to SO_2 injections are similar to those of Heckendorn et al. (2009) if we compare equivalent scenarios, but larger than those of Tilmes et al. (2012)."

I also would like to see improvements towards are better discussion of aspects related to atmospheric dynamics. For instance, seasonal dependence of results, temperature dependent effects. Both are associated with the underlying meteorology (which is also unclear, see specific comments below). The author's are mentioning phenomena like QBO only briefly when the methodology is described, but never refer to them later when the results are analysed and discussed. An important part of the manuscript deals with stratospheric chemistry, which is known to be temperature dependent, but respective investigations or discussions about e.g. additional errors induced by associated impacts are not considered so far. I think it is recommended to consider such aspects in the revised manuscript.

> The purpose of this paper is to present a first look at geoengineering by solid particles, accounting for particle interactions in the stratosphere, and comparing efficacy of different particle compositions and injected monomer diameters. Aspects such as induced modifications in stratospheric temperature and dynamics await a future study with a coupled chemistry-aerosol-climate model. We discuss these limitations in Section 4.1. The dynamical fields used in our model study are now described in more detail in Section 2. We have added one sentence to Section 4.1, *Missing Feedbacks*: "However, Heckendorn et al. (2009) found that ozone loss due to heterogeneous chemistry, without the dynamical effects of changes in temperature, water vapor, and the Brewer-Dobson strength, accounted for 75% of the ozone change." which demonstrates that our study, while not perfect, is still quite useful.

With respect to diamond aerosols proposed as a forcing agent, I do not really understood the message of respective parts of the study. Does the manuscript really profits from the diamond aspects? In opinion not - instead the additionally considered diamond investigations lead to some confusion because the subject is discussed only here and there in the manuscript, only a few figures, for instance, contain data from the diamond simulations and so on. I suggest to critically examine whether it is really helpful to investigate this additional compound here.

> Diamond is an example of a class of high-index compounds (e.g., zirconia, SiC silicon carbide) that, due to more favorable upscatter to forward scatter ratios, *might* enable solar geoengineering with lower side effects than sulfate aerosol so we think it's important to keep the analysis of diamond in the paper. Table 1 helps to clarify which calculations we have performed for diamond. Diamond is omitted

> from some figures because the results are so close to alumina at 160 nm that it would clutter the figures for no reason. We do state that diamond results are not shown but similar to alumina at 160 nm for Figure 1 (sedimentation velocities) and Figure 5 (burdens), and added similar statements for Figures 3 and Figure 10.

Specific comments:

Please include a list of abbreviations (e.g. MSA, QBO, ...).

> We have spelled out abbreviations (MSA, DMS, QBO, PSC) where they are used.

p11801 lines 14-23: Is it a chemistry coupled model ? should be mentioned here.

> Yes. We have added a sentence to this paragraph: "The chemistry and aerosol schemes in the model are interactive, while dynamical fields are prescribed."

p11802 lines 2ff: The author's attest "other" models a good job in quantifying the response to idealized SRM methods - which in my opinion is too speculative since such idealized scenarios cannot be proved in reality, so that a less subjective valuation seems appropriate here.

> Given that Kravitz et al. (2014) have now repeated the results in Moreno-Cruz et al. (2011) using the entire CMIP-5 ensemble we stand by our claim of these results are both surprising and representative of the current universe of climate models used for climate science and policy. We have modified the wording of this statement to clarify that it applies to model-simulated climate changes: "Results from a large set of climate models suggest that idealized SRM can do a surprisingly good job in reducing model-simulated climate changes, both locally and globally, which, in our view, is a primary motivation for continued research on SRM (Kravitz et al., 2014; Moreno-Cruz et al., 2011)."

p11805 lines 1-4: Since also in a 2-D model the dispersion of aerosols, hence the desired aerosol effect, depend on the transport characteristics of the model, please describe in more detail the underlying methodology referring to Fleming et al (1999).

> We modified the discussion of the model's transport as follows: "The model's 2-D transport is prescribed based on calculations by Fleming et al. (1999) for each

> year from 1978 to 2004, employing observed temperature, ozone, water vapor, zonal wind, and planetary waves. Different phases of the quasi-biennial oscillation (QBO) are included in the observational data employed. We average the transport fields over the years 1978-2004 into a climatology and employ that circulation each year of our ten year calculations."

p11805 line 11: In my understanding it was not the goal of the SPARC Assessment of Stratospheric Aerosol Properties (ASAP, 2006) to rate participating models or to introduce any kind of metric for them, I strongly suggest to chose a less qualitative statement instead of „best".

> We have removed the quantitative model rating attributed to SPARC (2006) and now include a few sentences describing model comparisons to observations, including mention of model deficiencies.

p11805 line 12: The understanding of mechanisms forming new aerosols in the stratosphere is far from being clear so that I suggest to rephrase the sentence, e.g. using a formulation like „is thought to form by" or „may nucleate".

> Changed wording: "Sulfate aerosol formation is thought to be initiated mainly by binary homogeneous nucleation of H_2SO_4 and H_2O vapors, primarily in the tropical tropopause region."

p11805 lines 18-20: Logarithmically spaced bins?

> Modified to read: "The model uses a sectional representation of particle sizes, with 40 logarithmically-spaced sulfate aerosol bins, representing sizes from 0.39 nm to 3.2 μm, with aerosol volume doubled between adjacent bins."

p11805 line 20: You are referring to sedimentation and wet deposition here, but only the former is described below. Please make a note that a description of sedimentation follows and add (at least) references describing how wet deposition is handled in the model.

> Added: "The sedimentation formulation is described below. Rainout/washout process are represent by a first order loss term in the troposphere with removal lifetime ranging from 5 days at the surface to 30 days at the tropopause."

p11806 lines 5-10: Please add references to the two equations.

> We moved the Filippov and Maricq references to before the equations for clarity.

p11803/4 and Sect. 3.1: The description of the use of diamond with respect to “the primary test case“ is confusing. When I interpret the figures correctly, the authors show results from a “diamond simulation“ only in the 6th Figure (and later), but describe the compound and related experiments at the beginning of the results section. Again, an experiment table would help to clarify this issue.

> We added Table 1 to list the numerical experiments performed. Diamond is omitted from some figures because the results are so similar to alumina at 160 nm (density difference is the only difference affecting aerosol distribution and mass in our simulations) that it would clutter the figures for no reason. We do state that diamond results are not shown but similar to alumina at 160 nm for Figure 1 (Sedimentation velocities) and Figure 5 (burden). We added similar statements to the discussion of Figure 3 showing the size distributions and Figure 10 showing surface area density.

Sect.3.2 and Sect. 3.3: Until the first results are described about the Figures 1 & 2, I do not find any information about the integration period of the model. When did you start the model, how long was it integrated and so on. This is a clear deficit of the manuscript and should be corrected. Please describe in detail the settings but also mention which averaging period has been used for the figures, otherwise the results are meaningless (some figures do contain this information, but some do not).

> We added to the end of section 3.1: “Each scenario is calculated with a 10 year integration period, using dynamical fields representing the 1978-2004 average repeated each year and fixed boundary conditions from approximately the year 2000, until an annually-repeating result is achieved. We analyze results from the final year of each calculation, concentrating on annual average conditions.”

p11810 referring to Fig.1: As above, for which period of integration? Shown for steady state, annual mean? Is the latitude band the same as for the upwelling?

> The original plotted sedimentation velocities were for April at 28N. These velocities are not very sensitive to latitude or season. For consistency with the tropical upwelling velocities, we now present annual averages from 20S-20N for aerosol sedimentation velocities. We added this information to the text and the figure caption.

p11811 lines 9-11: In terms of mass or mixing ratio?

> Most stratospheric sulfate exists in condensed form in terms of mass. The word “mass” has been added.

p11812 lines 6-7: Does it mean you performed a spinup over 10 years and then started to analyse the results? I assume with emissions you refer to surface and/or volcanic emissions (of S compounds).

> Yes, we do a spinup for 10 years and ignore the transient response, reporting the steady-state response, which does not change from year-to-year thereafter. Emissions referred to the stratospheric injection of particles for geoengineering. We have changed “emission” to “injection” here and in many other places in the manuscript to avoid confusion. This sentence now reads “Stratospheric particle injections are continuous in time…”

p11812 lines 21ff & Fig.2 a,b: The top panel show mixing ratios but in the paragraph the authors infer and compare the burden from the figures. In my understanding the burden is a vertically integrated quantity which is not easy to infer (visually) from a concentration altitude-latitude plot. In particular not, when the legends of the plots are different. Please adjust and/or calculate/plot the burden.

> By burden, we refer to integrated stratospheric burden which is shown in Figure 5. We have eliminated the discussion of burden in reference to Figure 2 to avoid confusion and instead discuss peak mass mixing ratio.

p11813 lines 22ff & Fig. 4: When I understand it correctly, the green curves should resemble the values of the bars of Fig. 3 b-d? Mentioning that in the paragraph would help to better understand the result.

> Yes. We added the sentence: “The 1 Tg yr^{-1} cases (green lines) match the global mass fractions shown in Figure 3.”

p11814 lines 22ff & Fig. 5b: Do you also show here sulfate from Pierce et al (2010) as in panel a? I think not - the caption is a little confusing for me. Also the beginning of the paragraph is a bit misleading, as it indicates that you are showing sulfate here (only). I suggest to introduce the burden issue with a statement about alumina in order to avoid confusion. I am also not sure why the sulfate burden increases - I thought you emit sulfur continuously (assuming it continuously reaches the stratosphere) so that the sulfur content of the stratosphere is an equilibrium state (you model quasi a background Junge layer). Then I would assume to find a constant sulfate burden in the model (assuming this is an annual mean). Or is it because I cannot distinguish thick lines from thin lines?

> No, panel (b) does not include simulations of SO_2 or H_2SO_4 geoengineering injection, as those increase sulfate burden by many times above the background level and would require a different plotting scale. The sulfate lines that increase continuously from 1 Tg/yr to 8 Tg/yr are the thin lines representing sulfate fraction on alumina particles. The thick lines contain circles and the thin lines don't (noted in caption) and there are arrows in the figure pointing to the respective y-axes to help distinguish them. The paragraph starting on line 22 of page 11814 refers to Figure 5(b) only, so discusses only sulfate burden. We clarified by stating that the scenarios presented in this figure refer to geoengineering injection of alumina and that the sulfate burdens are shown as a function of the geoengineering injection rate of alumina. Sulfate burden changes as a function of alumina injection because the sulfur coating the alumina particles sediments faster than pure sulfate particles would in most cases.

p11815 line 16/17: Please add references in order to make clear how you calculated the upscatter cross section. Same for the other parameter panel in Fig. 6 b,c.

> We now include the following description and references: "We calculated the solid particle monodisperse single scatter albedo values from Mie Theory (Bohren and Huffman 2008) using tabulated complex refractive index data for diamond (Edwards and Philipp 1985) and alumina (Thomas and Tropf 1997). The upscatter and downscatter cross sections are calculated from Wiscombe and Grams (1976), utilizing the scattering phase function from Mie Theory and the same complex refractive index data."

p11816 line 7ff & Fig. 7: Again, what is the time period you are showing? Shortwave, cloud-free forcing only?

> Time period is annual average of steady-state result. RF is for shortwave only under cloud-free conditions. We now specify "global annual average top-of-atmosphere shortwave radiative forcing … under clear sky conditions."

p11817 lines 21-24: Is there any possibility to estimate the uncertainty you mention? Wouldn't it be possible to estimate upper and lower bounds of associated errors, at least for the most „promising" scenario only?

> The error in these calculations is about 20% when compared to a

more complete RT model for the same physical assumptions. As our objective is to obtain well-founded estimates of radiative forcing for comparison with sulfate geoengineering, and relative efficiencies among solid particle scenarios, an estimate of associated errors is outside the scope of this paper. However, John Dykema of our group is preparing a detailed radiative study with RRTM to address this complex topic in a future paper.

p11818 lines 2-7: Two things are not clear:

a) I am a little confused about “per megaton“. In Fig 7a it refers to “per megaton burden“ - which is different to Fig 7b where it refers to “per megaton injection“?

Correct. The units on the y-axis of Figure 7a are watt per m^2 per Tg of atmospheric burden. We use this quantity to scale the atmospheric burden for each aerosol bin (bins shown in the x-axis of Figure 7a as “# cores in particle”) and sum over bin sizes for each scenario to obtain Figure 7b.

b) About sulfate, lines 4-7: I though that is what you show. What is the methodology of H2SO4 (and SO2) in Fig. 7b if not direct injection? And, if different from Fig. 5, please make it clear.

Perhaps the confusion here is using the words “injection” and “emission” to both refer to geoengineering injections directly into the stratosphere. We have have replaced “emission” with “injection” to try and avoid this confusion.

p11818 lines 8-25: LW effects are estimated offline from your results with the AER model? do you show data for one year only in Fig. 8?

Correct that radiative effects are estimated offline. Our heating estimates use the annual average particle number densities in the tropical lower stratosphere from the AER model over the last year of simulation when steady-state has been reached. Figure 8 shows an annual average over the final year of simulation as well.

p11819 line 15: What context for “preliminary“?

Simply that, as this is our first paper on this topic, we have focused on the aerosol microphysics and on an investigation of the comparative ozone loss between different particle injection scenarios. A more complete study of ozone loss would require better characterization of some of the surface reaction coefficients, and this requires new laboratory studies. More detailed modeling would also require a sophisticated radiative transfer model coupled to a chemistry-climate-aerosol model and would utilize future Cl and Br abundances.

p11819 lines 16-18: I do not understand the whole sentence „To enable comparison of the ozone impact of sulfate geoengineering we use the same model to compute change in ozone abundance arising from injections of both solid particles and of sulfate aerosols.“. What do you mean with „the same model“?

We don't plot results from Pierce et al. or Heckendorn et al., but have repeated their calculations to be consistent with others in this paper. The injection method is the same as Pierce et al. (injection 30S-30N, 20-25 km) though that work did not show ozone changes. Heckendorn et al. showed ozone changes, but for injections at the equator and 20 km only, and from a 3-D coupled chemistry-climate model. Table 1 should clarify this. We have added the word "relative" in "To enable a relative comparison of the ozone impact of sulfate geoengineering…"

p11819 lines 18ff: Are there any references available for the coupled aerosol-chemistry AER model? E.g. studies showing the capability of the coupled model to simulate certain characteristics of the stratospheric chemistry system - characteristics/features which are well known and explored by a range of other studies? Please add. If such references does not exist, I suggest to include a complete table of reactions, potentially as supplementary material, in order make the respective information available for the reader.In the same paragraph, please add also references for the methodology concerning the treatment of PSCs and the underlying meteorological climatology as well (see comments made earlier).

We added four references regarding the AER model: Rinsland et al. (2003), Weisenstein et al (1998; 2004), and Anderson et al. (2006), of which only the Weisenstein et al. (1998) reference employed aerosol-chemistry coupling, while the other include details of the chemistry scheme and gas-phase comparisons with observations. We also added the sentence: "A comparison with observed ozone trends between 1979 and 2000 is presented in Anderson et al. (2006) for the AER model and several other models."

The transport and temperature climatologies are covered earlier in Section 2. We added references and more detail to our description of PSC treatment: "The model parameterizes polar stratospheric clouds (PSCs) using thermodynamic equilibrium, employing the formulas of Hanson and Mauersberger (1998) and

Marti and Mauersberger (1993) for equilibrium vapor pressures over solid HNO_3 and ice, respectively, assuming no supersaturation and prescribing the particle radii." But our results do not depend on the PSC treatment, as the polar regions represents only 13% of the global atmospheric mass, with PSCs present only seasonally. Reactions on liquid sulfate aerosols, including in the polar regions, are much more important globally.

p11820 line 4: Please add „primarily" before „Cl".

done

p11820 line 4ff: Please make clear that you refer to future conditions. In the context of the ozone Section 3.5, you refer to „present day conditions" - how does the time period(s) match with your emission scenarios of S compounds?

We changed this sentence to read: "Future concentrations of halogens are expected to decline…" We use present day concentrations of sulfur compounds as well as chlorine/bromine, N_2O, etc. for this study. Our discussion of Table 1 in Section 3.1 states that "fixed boundary conditions from approximately the year 2000" are used.

p11821 line 5-7, Fig. 9 b, c: The response of the 80 nm alumina injection is very different from that of 240 nm. To me it suggests that in the 80 nm case sulfate is more rapidly dispersed towards the poles as in the 240 nm case. Do you have any idea why the behavior is so much different, e.g. from a dynamical perspective?

The difference in sulfate surface area density between the 80 nm and 240 nm alumina cases is due to (a) the faster sedimentation of 240 nm monomers compared to 80 nm monomers, which results in strongly decreasing concentrations of alumina away from the tropical injection region with 240 nm monomers injected (see Figure 2), and to (b) the large R_g of the complex fractal structures composed of 80 nm monomers which efficiently scavenge the smaller sulfate particles. We added the following to the paper: "With injection of 80 nm monomers, the sulfate SAD has increased by factors of 2-4 in the lower stratosphere, with maximum SAD at high latitudes where significant concentration of complex alumina fractals exist to scavenge the smaller sulfate particles. With injection of 240 nm monomers, the maximum sulfate SAD occurs in the tropics as the faster sedimentation of alumina in this case results in a smaller concentration of mostly monomers at high latitudes."

p11821 line 13-14:I do not understand the sentence „We would expect similar chemical ozone loss from similar changes in sulfate SAD whether due to geoengineering by SO_2, H_2SO_4, or alumina injection." In a general context?

> We have clarified by modifying this sentence to read: “For reactions that are dominated by liquid sulfate surface area density, we would expect similar chemical ozone loss from similar changes in sulfate SAD whether due to geoengineering by SO_2, H_2SO_4, or alumina injection.”

p11821 line 25:Please add „uncoated“ before „alumina SAD“.

> done

p11823 line 14:You are mentioning the „transport timescale“. For me it is even unclear what the timescale at all is, concerning simulation period, meteorology, analysis.

> We refer here to local balances between chemical production, destruction, and transport rates that determine ozone concentrations in different regions of the atmosphere. We have modified this to read: “The annual average ozone change as a function of latitude and altitude (Fig. 11, right hand panels) shows features linked to local balances in ozone’s formation rate, chemical destruction rate, and local transport rates.” We then explain for the tropics and midlatitude middle and lower stratosphere which ozone tendency terms are most important.

p11828 lines 9-15:Does it mean the aerosol scheme is not interactively coupled to the transport model’s radiation scheme? If so, please make it clear here and in Sect. 2 and describe briefly how heating rates are handled in the model (prescribed?) and how this relates to the transport of aerosols.

> Yes, the radiation and aerosol schemes are uncoupled. The transport and temperature fields are described in more detail now in Section 2. Here we have added: “The modeling we present utilizes temperature and transport fields uncoupled from the model’s chemistry and aerosols and is therefore missing a number of feedback processes that may be important…” We also provide a quantitative estimate of the importance of these feedbacks: “However, Heckendorn et al. (2009) found that ozone loss due to heterogeneous chemistry, without the dynamical effects of changes in temperature, water vapor, and the Brewer-Dobson strength, accounted for 75% of the ozone change.”

p11829 paragraph 2 (also affecting other sub-sections of Sect. 4):It would be helpful to insert cross references to the figures here in order to make it easier to understand what you mean, and to make it possible for the reader to find the features you discuss in the figures. In particular, I have difficulties to approve the last sentence of paragraph 2, which sounds contradictory at a first glance.

> We have added these cross-references. Thank you for the excellent suggestion.

p11829 line 28-29:Please remind the reader that the ozone changes depend on specific model assumptions, e.g. add „under the assumptions made" or so.

> Modified the sentence to read: "The annual global average ozone column is reduced by 3.6% with maximum ozone loss of 4 to 7% over polar regions for this scenario and the given modeling assumptions."

p11830 lines 8ff:Difficult to interpret. When you switch off reaction R1 or switch off the coating, to which injection scenario or alumina sizes do you refer with the calculated changes in %?

> Injection of 4 MT/yr alumina of 240 nm monomer radius. This has been clarifed in the text.

Technical comments about figures:

Please increase the readability of numbers and indices in the figures 1,2, 4, 5, 7, 8 - 13. I would like to see the line plots Fig. 1, 4, 5, 7, 10, 12 (!!), and 13 enlarged.

> We have enlarged the numbers and labels on all the plots except for Fig 3.

Figure 6: Please use a different color for the magenta curves (on poor resolving printer they may be indistinguishable from reddish colors).

> We have replaced the magenta lines with another color except where the colors are also distinguished by symbols.

Figure 9: If appropriate, please use decimal numbers for the legend in panel a. To better differentiate the contour plot in panel a from the deviations/differences shown in panels b-d, I suggest to use a different colorbar for the the latter, e.g. a gradient turning from blueish (for negative values) to reddish (for positive values) colors, with a transition around zero marked in white or very bright colors (alternatively use a contour line to mark zeros).

The colorbar labels in panel (a) are now decimal numbers. Most of the negative values in panels (b-d) were eliminated when we used a more consistent baseline (from the interactive chemistry-aerosol model) to get differences.

Figure 11: I see a similar same colorbar issue as for Fig. 9.

We have not changed the colorbars in Fig. 9.