# Response to Anonymous Referee #1 for "Radiative forcing and climate response to projected 21st century aerosol decreases" by D.M. Westervelt et al.

***"This is a clear presentation of a well-designed study of the radiative and climatic impacts of future changes in global aerosol emission to the year 2100. While the model used has its limitations, the results are likely to be consistent with those from more complex and comprehensive models. Only minor modifications are need to address several comments."***

We thank referee #1 for the positive comments and provide a response below.

***"Page 9301, line 6. Since the size distributions of the accumulation mode aerosol are prescribed, some discussion of the limitations of this approach is needed. This treatment is not able to distinguish between processes that influence mass but not number (condensation) and those that effect number but not mass (coagulation). It is likely to bias the estimate of aerosol effects on clouds. Modal representations overcome this limitation."***

This is a good point. We did not mention the lack of prognostic aerosol size and aerosol microphysics. The specifics of the prescribed size distribution can be found in Donner et al. (2011), but briefly, two lognormal modes are used for carbonaceous aerosol and sulfate, whereas dust and sea salt have are broken into 5 bin each. Although a modal microphysical scheme is currently under development at GFDL, it was deemed too computationally demanding to run a coupled climate model (atmosphere, land, ocean, ice) for 100 years and have online size-resolved aerosol microphysics. However, we agree with the referee that this bears mentioning. We have added the following to the manuscript at page 9301, line 6.

"Size-resolved aerosol microphysics are not included in the model due to computational demand. This lack of both prognostic aerosol mass and number concentration may bias estimates of the effect of aerosol on clouds. Nonetheless, the aerosol activation scheme has performed well, resulting in reasonable agreement in both droplet size and droplet number concentrations (Donner et al. 2011; Ming et al., 2006, 2007)."

***Page 9301, lines 11-16. If nitrate has no optical or microphysical effect, why is so much chemistry used in the simulations? Sulfur oxidation can be treated with prescribed oxidants and diagnosed peroxide. Do the oxidant concentrations change that much in the difference scenarios? If so, then oxidants should be described more in the following RCP section. Could you also comment on the role of oxidant changes in the estimated aerosol radiative forcing?***

There is additional chemistry for organic aerosols (anthropogenic and biogenic SOA), DMS, etc., as referenced in Naik et al. (2013). Having online chemistry also allows for the feedback of meteorology on aerosols (e.g. temperature dependent reaction rates,

relative humidity changes affecting oxidant levels, etc.). Also, while older model versions had prescribed chemistry, online chemistry (MOZART) is standard for CM3 as a global chemistry-climate model. Given the state of the climate modeling community, a model without online chemistry would be rightfully criticized and perhaps not publishable due to the widespread adoption of online chemistry in climate models.

As for oxidant levels, future changes for OH, for example is a globally averaged decrease of 6.7% in RCP8.5 (2100 – 2000 difference) (Voulgarakis et al., 2013). OH increases over the 21$^{st}$ century in RCP2.6, RCP4.5, and RCP6.0, by 12.4%, 19.4%, and 8.1% respectively. However, since the major sources and sinks of OH are identical in the two sets of simulations (decreasing aerosols and fixed aerosols) this will have little effect on our aerosol forcing results.

***Page 9306, line 1. Typo.***

Thanks, fixed.

***Page 9306, line 2. Since the cloud lifetime effect is included, a description of how the cloud microphysics depends on droplet number should be added to the model description.***

Good point. We have added the following short description to the model description section, Page 9301, Line 2:

"The cloud lifetime effect is parameterized as an increase in cloud droplet number (e.g. due to aerosol perturbations) resulting in a decrease in the autoconversion rate, thereby delaying precipitation and increasing cloud lifetime. The derivation of the parameterization is beyond the scope of this paper, but it follows the methodology of Khairoutdinov and Kogan (2000). An autoconversion threshold of 8.2 μm is used in CM3. In a given grid box, if volume-mean cloud droplet radii less than this threshold, autoconversion is suppressed (Rotstayn, 2000). Sensitivity of the indirect effect to different thresholds in GFDL CM3 is explored in Golaz et al. (2011)."

***Figure 8. Aren't there any regions where the response to call forcing is opposite in sign to the global mean? If so, discuss.***

Yes, the climate response to all forcings in a certain region can be opposite in sign to the global mean. This can be seen in Fig. 11, where precipitation decreases in South America, Australia, and the Middle East, but increases strongly virtually everywhere else, leading to a global mean increase. In these grid cells, the absolute value is taken so that the ratio between aerosol-decrease and all-forcing is positive (in other words only the magnitudes of the changes are considered). We have added a sentence in Sect. 4.1.3 to clarify this:

"In instances where the all-forcing response sign differs from the aerosol-driven response, we use the absolute value (i.e. magnitude only) of the all-forcing response for the ratio calculation."

***Section 5.2. Consider showing relationships Delta C/Delta AOD instead of correlations, where C is the climate parameter. This provides a quantitative measure of the relationship.***

This is a fair point. We have added to the supplemental material the $\Delta C/\Delta AOD$ plot as requested by the reviewer as Figure S20. We have decided to leave the correlation plot in the main text, however, as we feel it conveys our point more clearly. For example, the correlation encapsulates the entire timeseries of the changes in AOD and climate variables, where as the delta method only encapsulates the beginning and end points.

***Page 9323, lines 8-22. Should also discuss the effect of precipitation changes on AOD through wet removal.***

Good point. We have changed the sentence starting on line 8 on Page 9323 to read:

"Over East Asia and parts of Europe, AOD and precipitation are somewhat strongly anticorrelated ($r < 0.5$), which is expected as wet removal by precipitation is a strong sink for aerosols."

# Response to Anonymous Referee #2 for "Radiative forcing and climate response to projected 21st century aerosol decreases" by D.M. Westervelt et al.

***"In this study, the authors simulate the 21st century climate based on RCP emission scenarios and using the GFDL climate model. By fixing aerosol emissions to 2005 levels, the authors isolate the aerosol contribution to total climate response. They find that the four RCPs yield similar responses, because aerosol emissions decrease similarly in all scenarios. Aerosol decreases exert a positive radiative forcing, contribute to surface temperature rise, and increase in precipitation rates and cloud droplet radius.***

***The paper is well written and the analysis is reasonably wide-ranging, although shallow in places. Figures are well chosen and illustrate the discussion well. However, the paper suffers from two serious flaws. First, the authors take the results of their model uncritically, without giving sufficient reasons for the reader to believe the quantitative aspects of the paper. Second, the methods and results presented in the paper are not novel and the authors do not take opportunities to analyse results in a deeper, more original way. Because novelty is a criterion for publication in Atmos. Chem. Phys., I can only recommend rejection of the paper."***

We thank referee #2 for the review. We have, as the reviewer has requested, run additional simulations in which we tested a weaker aerosol forcing (-1.0 W $m^{-2}$ present-day aerosol ERF) in the GFDL climate model. This addresses both of the reviewer's criteria for rejection: 1) the large radiative forcing is now accompanied by a more reasonable simulation and 2) this adds novelty by allowing for a test of the sensitivity of aerosol-decrease-driven climate response to present-day forcing values. These results are now discussed throughout the paper (see revised manuscript), and nearly every figure in the manuscript has been updated. These new simulations should address many of the responses to the referee's points, but we provide below a point-by-point response to all of the referee's comments.

***"The authors clearly have confidence in their model and its representation of aerosol impacts on climate, even though the mechanisms of some of those impacts have not been confirmed by observations. For example, on page 9297, line 1, the authors write that "aerosols have strong impact on precipitation", but the observational evidence is mixed and the choice to represent that impact via the autoconversion rate for all cloud regimes is debated (e.g. section 7.6.4 of the IPCC report)."***

We have prepended the quoted sentence with the a short clause pointing out the lack of complete observational agreement and have removed the word "strong" so the sentence now reads:

"Modeling studies have suggested that aerosols also have impacts on precipitation, cloud cover, cloud droplet size and number, atmospheric circulation, and other climate

parameters (Lohmann and Feichter, 2005; Ming and Ramaswamy, 2009, 2011; Ming et al., 2011; Ramanathan et al., 2001; Rosenfeld et al., 2008; Stevens and Feingold, 2009)."

In regards to confidence in our model, in fairness, other mechanisms of impacts of aerosols on clouds and climate have not been robustly confirmed by observations, and many other CMIP5 models also include such mechanisms. Besides being a CMIP5 and IPCC model, GFDL CM3 has been rigorously evaluated against observations, including in two papers that are cited in the original manuscript, specifically (Donner et al., 2011; Naik et al., 2013). Additionally, Golaz et al. (2011) evaluated cloud forcing, precipitation, cloud cover, and liquid water path in CM3 against various satellite observations. The model mechanisms and their validation have been described thoroughly in these papers, and such a discussion would not be germane to nor bears repeating in the present manuscript.

***Also, the model has a strong present-day ERF of −1.8 W m−2 (page 9307, line 9) (and incidentally must therefore have a large climate sensitivity to be able to match observed warming). That strong ERF is probably due to a large sulphate AOD: the decrease in sulphate AOD given in Table 1 is larger than the median anthropogenic sulphate AOD simulated by AeroCom models (Table 4 of Myhre et al., doi:10.5194/acp-13-1853-2013, 2013) Taken together, those facts mean that the results presented here are for a strong aerosol contribution. (I do not understand why the author think their forcing is only "slightly" overestimated [page 9308, line 25].) The authors need to clearly place their findings in the context of their model's characteristics: with a weaker aerosol ERF, many of their conclusions, such as the one on Page 9317, lines 26–28, would be quite different.***

We have directly addressed the viewers concerns by running additional simulations in which present-day effective aerosol radiative forcing is about -1.0 W $m^{-2}$. These simulations are described in Sect 2.3 of the revised manuscript.

We also have removed the word "slight" from our manuscript in the quoted sentence. We have also noted in our conclusions and abstract the caveat of the large present-day aerosol forcing, for example in the line the referee mentions now reads:

"Thus, even considering the high emissions, low-mitigation RCP8.5 scenario, aerosol reductions are still a surprisingly important player in future global and regional climate change. However, an important caveat remains that our total present day aerosol effective radiative forcing is on the higher end of the IPCC range and thus may bias our results."

***Finally, the authors acknowledge that the lack of nitrate aerosol representation is a limitation of their study (Page 9301, lines 10–16 and Page 9327, lines 4–7). Indeed, other studies included nitrate aerosols and showed that it influences aerosol radiative forcing in the 21st century, because SO2 emissions decrease and NH3 emissions increase. So what is the added value of using a model that does not include nitrate aerosols?***

Indeed, (Bellouin et al., 2011) have used a CMIP5 model with RCP simulations to 2100 and included effects of ammonium nitrate, as has a newer study by (Hauglustaine et al., 2014). An older study by (Bauer et al., 2007) included projections to 2030 with nitrate. These papers are cited in our manuscript, and as the referee notes, the lack of nitrate aerosol forcing in GFDL CM3 is mentioned. Again, however, CM3 is not alone in the lack of nitrate aerosol forcing. In fact, according to (Shindell et al., 2013) and (Schmidt et al., 2014), only two models included nitrate forcing for their CMIP5 simulations. Nitrate aerosol forcing is under development in CM3 and a more robust version of nitrate thermodynamics and chemistry is now submitted to ACPD (Paulot et al., 2015), but expecting it for the present work is an unrealistic standard to hold against CM3 considering the current state of the climate modeling community.

In order for inclusion of nitrate aerosol forcing to be useful, model representation of inorganic aerosol thermodynamics and chemistry must be robust and accurate. Most models, including Bellouin et al. (2011), use a simple equilibrium approach for the reaction between nitric acid and ammonia. As reported in Bellouin et al. (2011), the subsequent model-measurement agreement for nitrate aerosol mass concentration is somewhat mixed (within a factor of 2, significant scatter, bias both high and low, see Fig. 2 of Bellouin et al. (2011)). An equally legitimate question might therefore be, what is the added value of calculating nitrate forcing with a poor representation of nitrate chemistry and thermodynamics? The added value of our study is demonstrated throughout our response and in the revised manuscript, but briefly we have performed a thorough regional analysis (not seen in previous papers), presented different (new) climate response parameters, compared our of aerosol-driven forcing with total forcing (not previously done), etc.

***The main objective of the study is to expand on the findings of Levy et al. (2013) by using four, rather than only one, RCPs (page 9298, line 23). But the authors acknowledge, in a long paragraph (pages 9302-9303) and even a dedicated section (section 5.3, page 9324) that RCPs are very similar in terms of aerosol emissions, representing a "narrow range for emissions of air pollutants and their precursors" (page 9303, line 17). From this statement, it is clear that no novel insight will result from replicating Levy et al. (2013) four times.***

To contribute to the novelty of the work, we again refer to the new simulations that we have run that test our results against a more reasonable present-day aerosol effective radiative forcing of -1.0 W $m^{-2}$. That being said, there is still more to the study than simply adding additional RCPs. We feel that the referee has severely understated our work. As described above, we have presented new information (regional analysis, different variables) and presented old information in a new way (relative contribution of aerosol-decrease-driven warming to total warming). Although it may be accepted in the community, the point that RCPs are quite similar for the purposes of aerosol and air pollution has not been frequently made. While indeed the RCPs were quite similar at least on the global scale, we were careful to point out instances of difference. For example, the relative effects of the aerosol-driven climate response are quite different and

this is shown in Sect. 4.1.3. Regional differences are discussed in Sect. 4. We also note in the conclusions as well as in the main text the "mid-century variation in the climate response and radiative forcing trajectories" and discuss reasons why (i.e. elimination of coal energy use in RCP2.6, mid-century increase in coal energy in RCP6.0, etc.). To elaborate further, here are some of our more novel conclusions:

- 30-40% of warming in East Asia under RCP8.5 could be from aerosol decreases (10-20% under more reasonable aerosol forcing assumptions)
- Region-by-region analysis of climate response, including 2-3 K for East Asia, 10 K for the artcic (~half of that for weaker aerosol forcing)
- Evidence of emissions and energy use patterns in each RCP in climate variables response, including LWP and Reff.
- Spatial-temporal correlations (or anti-correlations) between changes in aerosols and changes in climate response

***Indeed, the conclusions of the study can already be read in FAQ 7.2 of the IPCC report***

The FAQ 7.2 of the IPCC report is very broad, but the referee may be referring to this line, among others:

"It is projected, however, that emissions of anthropogenic aerosols will ultimately decrease in response to air quality policies, which would suppress their cooling influence on the Earth's surface, thus leading to increased warming."

That is indeed consistent with what we found as well as what other studies have found. This is not, however, a fair representation of our conclusions. Since this is indeed a well-known finding, we have emphasized other results more, and refer the referee to the bullet points listed above and point the referee to our new simulations as well.

***One way to make an original contribution would be to analyse almost forensically the differences that arise from the slightly different trajectories taken by the RCPs***

Performing the analysis with different emissions trends from the RCPs is not possible now, but we have already presented somewhat of a "forensic" analysis of differences in RCP energy scenarios (and thus emissions) and how those differences affect AOD, forcing, and climate response. This is not a separate section, but rather mentioned in several different sections and even the abstract and conclusions. We have cited some examples from the ACPD manuscript below.

- Emisisons: Page 9303, ~line 7-18

"In particular, RCP6 and RCP2.6 stand out, the former due to an increase in the rate of coal consumption around mid-century (2030-2060) and the latter due to stringent climate policy including the nearly complete phase-out of non-CCS (carbon capture and storage) coal energy by roughly 2050 (Masui et al., 2011; van Vuuren et al., 2011b). The increase in coal energy projected by RCP6 is a surprising feature that is not present in the other RCPs. As a result, $SO_2$, BC, and OC emissions in RCP6 are higher relative to the other

RCPs over roughly the same time period in (Fig. 2). $SO_2$ emissions briefly increase in absolute terms over a short period mid-century in RCP6, which drives higher sulfate burdens, larger (negative) aerosol direct and indirect forcings, and noticeable changes in climate response, as we will show in the following sections."

- AOD: Supplemental, Section S2.2

"…there are significant deviations in the middle of the century, owing to specific features of each pathway. For example, the stringent climate policy of RCP2.6 is evident in the middle of the 21st century as RCP2.6 sulfate AOD decreases more rapidly than the others, as is the mid-century increase in coal as a primary energy supply in RCP6. Global OC AOD differences are more varied than for sulfate, due to the larger variation in land-use policy than in energy policy. However, since sulfate dominates the total AOD amount, the spread in the sum of sulfate, BC, and OC resembles sulfate more than it does OC."

- Forcing: Page 9307, ~line 23 to Page 9308 line 9

"RCP2.6 has the largest decrease in magnitude of aerosol forcing over the century, followed by RCP4.5, RCP6, and RCP8.5, which is the expected order according to each RCP's underlying climate policy. For example, reduction of coal energy usage, a GHG mitigation policy featured in the RCPs, also reduces the amount of $SO_2$ emissions. As a result, total aerosol forcing trends and the end-of-century rank order for each of the RCPs can be traced back ultimately to the energy and climate mitigation policy. Furthermore, sulfate is the main contributor to the aerosol direct and indirect forcing trend for all RCPs, due to its optical properties as well as large CCN activity (hygroscopicity). Therefore, energy policies that affect sulfate will have a magnified effect on aerosol direct and indirect forcing.
"RCP6 projects the smallest decrease in magnitude of aerosol forcing for much of the middle part of the century (2045 – 2075), despite passing RCP8.5 eventually. This is consistent with both the emissions and AOD trajectories for RCP6. RCP6 projects mid-century increases in coal for energy supply globally (Masui et al., 2011), which is visible not only in the emissions and AOD trends as described elsewhere but also the aerosol forcing trends."

- Climate response: Page 9311 ~line 5 – 16

"The impact of the RCP2.6 aggressive phase-out of coal as an energy source can be seen from about 2020-2050 with a strong increase in aerosol driven temperature change. Likewise, the mid-century rise in coal use in RCP6 shows up as a decline in what is an otherwise consistent temperature increase throughout the century (Fig. 5). RCP4.5 and RCP8.5, on the other hand, have a steadier temperature increase that lacks the same noticeable features. "

- Climate response: Page 9312, Line 12

As is the case with radiative forcing, temperature, and precipitation, the annual trends in the LWP values also follow the underlying RCP energy use trajectories. In particular, a rebound around 2040 in LWP in RCP6 can be seen in the bottom left of Fig. 5, analogous to the temperature decrease in RCP6 due to an increase in coal energy usage rate and ultimately aerosol and precursor emissions.

- Abstract: Page 9294, line 14

"…although there is some mid-century variation, especially in cloud droplet effective radius, that closely follows the RCP emissions and energy consumption projections."

- Conclusions: Page 9326, line 9-13

"Mid-century variation in the climate response and radiative forcing trajectories closely follows the aerosol and precursor emissions trajectories (and thus the energy use trajectories), even for climate parameters such as liquid water path and cloud droplet effective radius."

***For example, it would be interesting to study why AOD trends do not correlate with emission trends in some regions (page 9306, line 17).***

Correlations with AOD and emissions are very strong (r = 0.9 or greater) over the continental regions. Where the correlations weaken is, for example, over the tropical Pacific Ocean, where we see AOD increases driven by changes in wet deposition, as explained in the manuscript on page 9314, line 8 and beyond. We have changed the text to now read:

"In short, AOD trends are well correlated with emissions trends, with globally averaged correlation coefficients ranging from 0.7 to 0.9 for each species and each RCP (not shown) globally averaged, and 0.9 or greater over continental source regions."

***Or study in details the feedback of climate change on aerosols which is apparent from Figure S3 and only briefly mentionned on page 9306, line 22.***

We have discussed the feedback of climate change on aerosols in much greater detail in the supplemental section (pasted below). Also, we are planning to do exactly what the reviewer suggests – climate effects on $PM_{2.5}$ – in a separate manuscript in a very thorough manner. We would prefer to save that analysis for the future paper.

"The reason for this AOD increase is not increasing emissions (since they are held fixed), but instead feedbacks of meteorology on aerosol burdens. In particular, temperature is projected to increase drastically as a result of anthropogenic greenhouse warming (see Fig. 6, main text), and higher temperatures may lead to larger burden by increasing the reaction rates of aerosol-forming reactions, such as sulfur dioxide oxidation. Additionally, decreases in the wet deposition efficiency, which can occur despite an increase in precipitation intensity, may lead to increases in aerosol optical depth (particularly sulfate AOD) (Fang et al. 2011). This is discussed in more detail in Sect. 4.1.1. A particularly striking example of the impact of meteorological factors on AOD can be seen in RCP6 OC AOD, in which case both the RCP6 and RCP6_F simulations have nearly identical OC AOD values for the entirety of the 21$^{st}$ century, suggesting that the trend in OC emissions is not responsible for the AOD increases."

***Another line of enquiry is to understand why RCP6.0 and RCP8.5, which have the same aerosol ERF by the end of the 21st century, end up with a 0.4 K difference in their aerosol-driven temperature anomaly (Figure 4)***

We assume the referee is referring to Fig. 5, which would be more relevant to this comment. Also, the difference between RCP6.0 and RCP8.5 at the end of the 21st century is about 0.3 K, not 0.4 K. Still, the point remains. This probably has to do with the ERF calculation, which is calculated from atmosphere-only simulations with fixed sea surface temperatures instead of CM3. Note that when considering the full ensemble range, the difference between temperature anomaly in RCP8.5 and RCP6.0 by the end of the 21st century is minimal.

***Another interesting question to answer is why LWP is little affected by aerosol changes in North America, but shows a strong trend in East Asia (Page 9321, line 1).***

LWP *is* strongly affected in Eastern North America, where some of the largest AOD decreases are. Note that as shown in Fig. 6 there are not very strong AOD decreases. The LWP changes are thus fairly consistent across both regions.

***Page 9295, line 3: Please cite IPCC chapters, rather than the whole report. Myhre et al. (2013), already cited elsewhere, is a good choice in this context.***

Done.

***Page 9295, line 15: "generally": more specifically, on a global average.***

Changed "generally" to "On a global average basis".

***Page 9295, line 17: Negative aerosol radiative forcing leads to a cooling only if it is the only radiative forcing exerted.***

Changed to "opposing the positive forcing from greenhouse gases":

On a global average basis, both the direct and indirect effects tend to exert a net negative radiative forcing on present-day climate, opposing the positive forcing from greenhouse gases, with the total aerosol effective radiative forcing estimated to be -0.9 W $m^{-2}$ (uncertainty range -1.9 to -0.1 W $m^{-2}$).

***Page 9296, line 9: Emission datasets are notoriously uncertain, especially for China, but more recent assessments seem to confirm that Chinese emissions indeed peaked around 2005, see Klimont et al., doi:10.1088/1748-9326/8/1/014003, 2013.***

We have cited the provided reference.

**Page 9297, line 12: "warming the surface": as a feedback? Because absorption of radiation will cool the surface first.**

Absorbing aerosols can cause local cooling at the surface, but at the larger scale there is net warming of the surface due to distribution and mixing of thermal energy. Clarified.

***Page 9297, line 16: This statement is only valid for liquid clouds. Homogeneous nucleation of ice crystals occurs in the atmosphere.***

The word "liquid" has been added to the sentence.

***Page 9298, line 14: Although I agree that the choice of emission dataset influences results quantitatively, studies that do not use RCP scenarios likely find the same qualitative results. So I am not sure that the authors have a good basis to exclude them from Table 1.***

The table is already quite large. Opening it up to older studies would make the table far too cumbersome. Also, as the referee mentions, since the qualitative results are similar, there is not much more value added in having these.

***Page 9301, line 2: It would be a good place to describe the representation of second indirect effects.***

Yes, we have done this. See response to similar comment from referee #1.

***Page 9301, line 4: That statement is unclear. If BC remains externally mixed, does it still become hydrophilic and act as a CCN?***

Hydrophobic black carbon can age with an e-folding time of 1.44 days. These details can be found in papers cited throughout this section. However, BC does not act as CCN in the current version of the model. Added to the manuscript:

"Fractions of BC and OC are emitted as hydrophobic (80% and 50%, respectively) but undergo aging to hydrophilic BC and OC with e-folding times of 1.44 and 2.88 days, respectively."

***Page 9301, line 12: But nitrate is a large contributor to aerosol mass in many regions (e.g. Jimenez et al., 2009). And because aerosol indirect effects are non-linear, having the right background aerosol number matters (e.g. Carslaw et al., 2013). So nitrate is important also in present-day, in spite of perhaps exerting a weak radiative forcing.***

We generally agree. We never stated that we think nitrate is unimportant currently, just that it will be more significant in the future. Also, the major result of the Jiminez paper mentioned is that organic aerosol comprises up to 90% of the total aerosol mass. According to Fig. 1 of that paper, although in certain continental polluted locations (e.g. Beijing) nitrate can comprise up to 40% of the total mass, it is often much less than that for most of the regions.

***Page 9301, line 21: Rigorously speaking, RCPs were used in CMIP5 simulations, which form the basis of parts of the IPCC assessment.***

Clarified in the text.

***Page 9305, line 6: Are those "minor updates" relevant to the results of this study? If so, we would need to know what they were.***

No, there is no scientific difference in the two sets of simulations. Otherwise they would have been discussed in the initial manuscript.

***Page 9307, line 3: "(mostly decreases)": In the future, possibly, but historical aerosol emission changes have been increases.***

Added the word "Future" to begin the sentence.

***Page 9307, line 5: More specifically, effective radiative forcing here.***

Added the word "effective".

***Page 9308, lines 21–24: That comparison is awkward. Why not compare to CO2 radiative forcing in 2100?***

The idea was to give the reader a reference point from something well known and not dependent on uncertain future projections. However, we have changed the comparison to the reviewer's suggestion. Manuscript edited to read:

"For comparison, the 2100 RCP8.5 $CO_2$ forcing is about 5 W $m^{-2}$ (Myhre et al., 2013). Thus, the resulting positive forcing from the decrease in aerosol emissions by 2100 is projected to be more than 20% of the forcing of $CO_2$ in 2100."

***Page 9309, lines 19–20: You seem to have decided that aerosol effects on climate are large before even doing the analysis. I recommend saying something like "are expected to have significant effects".***

Changed as suggested by the referee.

***Page 9311, line 19: On a global, annual average, the evaporation flux must be balanced by the precipitation flux, so aerosol impacts on cloud microphysics can only change the timing of precipitation, not global amounts. Only aerosol impacts on evaporation have that ability.***

Noted. We have clarified the statement.

***Page 9312, lines 1–2: The authors have not shown that precipitation responds more strongly to aerosol than to CO2 forcing, so is Shindell et al. (2012) really relevant***

***here?***

Fair point. We have deleted this sentence and the reference.

***Page 9313, lines 7–10: If the authors were looking at climate feedbacks on aerosols, choosing the RCP scenario that best tracks recent greenhouse emissions would be justifiable. But here, the authors would need to show that recent aerosol emissions are best represented by RCP8.5. In any case, since the authors show the other RCPs in the supplementary material, they do not really need to justify which RCP is highlighted in the main text.***

Yes, but given how similar the aerosol emissions trajectories are in the four RCPs, there is no reason not to use the most realistic scenario with respect to other trajectories.

***Page 9314, line 2: BC is also removed by wet deposition, yet does not seem to show the same increases over tropical oceans than OC and SO2. Why not?***

BC is indeed affected in the same way, but there is so little of it over the tropical Pacific (less than 0.001 AOD) that the effect does not show up as much. Note that we do get some BC AOD increases over the tropical Atlantic, which would be attributable to wet deposition changes.

***Page 9314, line 11: The link with Fang et al. (2011) needs to be made more clearly here. Do you mean that aerosols not being deposited over land are transported in greater numbers to the oceans? Other changes could explain the observed increase: a decrease in low maritime cloud cover, or aerosols getting higher up in the atmosphere in a warmer climate, perhaps?***

Fair point, this is not 100% clear as is currently written. Some discussion about precipitation frequency is needed. Manuscript edited to read:

"Using an idealized soluble tracer, the authors found that as climate warms, wet deposition of soluble pollutants decreases due to the simulated decreases in large-scale precipitation frequency. In the future, moderate and light precipitation is projected to occur less frequently, whereas heavy precipitation occurs more frequently. Since wet deposition does not depend on strength of precipitation events (i.e. same wet deposition flux for heavy or light rain), the decrease in frequency of light precipitation events leads to less wet deposition flux in the future and thus increases in AOD (Fang et al., 2011). "

***Page 9315, line 8: CO2 is a good example of radiative forcing and temperature response not being collocated: its radiative forcing peaks in the Tropics, but the temperature response is maximal at the Poles.***

Yes. Polar amplification is already mentioned on page 9321, line 15.

***Page 9315, line 19: To be clear, having a model that simulates two ITCZs is not a good***

***thing. Correct?***

Of course. This is not unique to CM3. We mentioned and cited this: "…a common feature not only identified in CM3 but in other models as well (Lin, 2007)."

***Page 9315, line 21: In fact, the ITCZ response shown by the authors is a northward shift, which is expected when removing a negative forcing located in the north hemisphere (e.g. Allen and Sherwood, doi:10.1007/s00382-010-0898-8, 2010). However, the effect does not seem statistically significant in the GFDL model.***

Good point. Added:

The northward shift in the ITCZ is expected when removing a negative forcing in the northern hemisphere (i.e. aerosol decreases), but this does not appear to be statistically significant (Allen and Sherwood, 2010).

***Page 9316, line 20: For cloud effective radius, it is difficult to imagine a teleconnection mechanism. There are not many areas where radius changes are statistically significant, though. Is that due to a large unforced variability of effective radius in the model?***

We did not claim there was a teleconnection mechanism for cloud droplet effective radius. Perhaps it is a little difficult to see but there is a fair amount of statistical significance especially in the Northern Hemisphere. The effective radius changes are not statistically significant over polluted areas (e.g. east Asia) because these areas are already saturated with CCN at 2005 levels, so modest increases in radius due to the loss of anthropogenic nuclei is not as important.

***Page 9317, line 17: What emissions?***

Greenhouse gas emissions. Fixed.

***Page 9318, lines 9–10: Can the ratio be larger than 100%?***

Yes. Added "or greater".

***Page 9322, section 5.2: This kind of correlation analysis is rather futile because, as stated by the authors, one does not expect aerosols and their climate response to be collocated. So what could we expect to learn from correlations?***

Although precipitation is likely not co-located with aerosol forcing, other parameters may be, at least in some regions, co-located with AOD changes. For cloud droplet radius in our results, AOD changes and climate response changes are strongly anti-correlated across virtually all continental regions (see Fig. 12). (Takemura, 2012) found similar results for cloud droplet radius. Areas of strong decreases in aerosols (e.g., China), coincide with areas of strong increase in cloud droplet effective radius and liquid water

path (compare Figs. 7 and 8). However, per the advice of referee #1, we have added a figure to the supplemental presenting ΔC/ΔAOD (where C = climate parameter) to provide a more quantitative measure.

***Page 9306, line 1: Delete "?".***

Done.

***Page 9315, line 16: Should be "insignificant"?***

Correct.

***Figure 1 and Page 9318, line 21: Figure 1 is used very late in the paper. Why is it not placed after the current Figure 8?***

We put it first because it isn't really a "results" figure. We've now mentioned Fig. 1 much earlier in the paper so that it is mentioned before any other figure.

# Response to Anonymous Referee #3 for "Radiative forcing and climate response to projected 21st century aerosol decreases" by D.M. Westervelt et al.

***In this study long-term trends of radiative forcing and changes in surface air temperature, precipitation, liquid water path, and cloud droplet effective radius due to the aerosol-radiation and aerosol-cloud interactions along the RCP scenarios are estimated with a general circulation model. The authors made efforts to do the long-term ensemble simulations for analyzing comprehensive aerosol effects. However, the novelty of this study is not clear in this manuscript in comparison with past similar studies shown in Table 1.***

We have addressed the novelty issue in the response to referee #2.

***If the authors can present the novelty, it should be clearly written in Abstract, Introduction, and Conclusions.***

We have emphasized this more, and have included phrases such as "We build upon previous work…" or "we go beyond previous studies" in the sections the referee mentions.

***For example, in the latter half the Abstract, where the novelty should be stated, the authors write "we compare recent studies to results from the present work in Sect. 5.1". Readers can make a mistake to understand that the primary purpose of this study is just the comparison with past studies.***

We assume the referee is referring to the introduction here and not the abstract. Also, we find it extremely unlikely that a reader would conclude that our *primary* purpose is only to compare with past studies, given that the aims of the study are very clearly outlined on the exact same page from where this statement was made. However, to prevent any confusion we have simply deleted this sentence.

***The author should clearly write what the lacks are in the past studies and what the novelties are in this study here.***

This was done in the original manuscript:

"We then go beyond the previous studies described in Table 1 by performing a more exhaustive and robust analysis of the unintended climate consequences of reducing particulate air pollution levels in the future. We also test multiple realizations of the CM3 model in which aerosol forcing is weakened significantly from its default large estimate. We present historical to present day to future results from 1860 to 2100, focusing first on global changes (Sect. 3) and then on specific regions that may be most strongly impacted (Sect. 4). We also compare our results with those from previous studies and examine

similarities and differences in the projected aerosol-driven changes in climate variables, climate forcing, and aerosol burden across the various RCPs. Finally, we attempt to connect changes in aerosols with changes in forcing and climate parameters (Sect. 5). Conclusions are presented in Section 6."

***Also there are a few lacks of understanding on the aerosol-climate interaction.***

Of course. These are also mentioned throughout the introduction:
"significant questions remain regarding the magnitude, and in some cases, even the sign (cooling or warming), of aerosol-climate interactions."

***Therefore I suggest that the authors should make major revisions if the novelty of this study can be stated only by the revisions. Otherwise I recommend resubmission after an additional simulations and analyses of fixed emission and SST experiments (RCPx.x_F_RFP) for understanding a difference between fast and slow feedbacks on the aerosol effects, which can be a novel study.***

We have indeed run additional simulations and made major revisions as the referee suggests. However, we decided to run the simulations suggested by referee #2, as those were more germane to our current paper than what is suggested here. See response to referee #2.

***1. Page 9297, lines 5 and 15: Add "for warm rain" after "precipitation rates" and "rainfall rates", respectively.***

Done.

***2. Page 9297, line 20: "we must rely on future projections or scenarios". The RCPs are not provided under a concept whether we can trust them or not, so the authors should delete the sentence***

Changed to "We must utilize…"

***3. Section 2.1: Add description of aerosol transport processes other than emission briefly. Also add basic information on the ocean model because it is significant to evaluate the aerosol effects on whole climate change discussed in this manuscript.***

This can be found in Donner et al. (2011), but we have added the following text:
Transport of tracers follows the work of Lin and Rood (1996) with updates as described in Donner et al. (2011).

For the ocean model, we have added a citation for the MOM4 model, which is used, in our simulations:
Stephen M. Griffies, Michael Winton, Leo J. Donner, Larry W. Horowitz, Stephanie M. Downes, Riccardo Farneti, Anand Gnanadesikan, William J. Hurlin, Hyun-Chul Lee, Zhi

Liang, Jaime B. Palter, Bonita L. Samuels, Andrew T. Wittenberg, Bruce L. Wyman, Jianjun Yin, and Niki Zadeh, 2011: The GFDL CM3 Coupled Climate Model: Characteristics of the Ocean and Sea Ice Simulations. *J. Climate*, **24**, 3520–3544. doi: http://dx.doi.org/10.1175/2011JCLI3964.1

***4. Page 9310, line 4: It is interesting that the LWP is higher in RCPx.x_F than RCPx.x as shown in Fig. 4. The authors should state how aerosols contribute to increasing the LWP relative to global warming.***

This is already explained in the original manuscript:
As aerosol concentrations decrease, LWP also decreases; in other words, aerosols and LWP are positively correlated. This is essentially the cloud lifetime effect acting in the opposite direction: increased aerosols cause cloud droplet concentrations to increase leading to a decrease in the autoconversion rate, which hinders precipitation formation and increases cloud lifetime and cloud liquid water path (Albrecht, 1989). The decline in aerosol emissions leads to a decrease in LWP in all of the standard CM3 runs with the RCPs, around 0.5-1.0 g $m^{-2}$ or 2% of 2005 levels.

***5. Page 9310, line 23: Revise from "cloud cover" to "LWP"***

Done.

***6. Page 9316, lines 10-11: "These increases are most likely due to a feedback from the aerosol-driven temperature increase, since warmer air can hold more moisture." This occurs all over the globe. This trend in the Arctic region is probably from melting sea ice and consequently providing a large amount of water vapor by opened ocean. The authors should confirm the temporal trend of sea ice.***

Done. The sentence now reads:

"These increases could be due to a feedback from the aerosol-driven temperature increase, as well as an increase in melting sea ice."

***7. Page 9326, lines 13-14: "liquid water path, and cloud droplet effective radius are strongly correlated spatially with aerosol optical depth changes". It is a matter of course because the parameterization of the aerosol-cloud interaction is treated to present this relations.***

True, although AOD itself is not directly in the parameterizations. We have mentioned this caveat by inserting "as would be expected from their parameterizations" to the sentence.

**References**


Allen, R. J. and Sherwood, S. C.: The impact of natural versus anthropogenic aerosols on atmospheric circulation in the Community Atmosphere Model, Clim. Dyn., 36(9-10), 1959–1978, doi:10.1007/s00382-010-0898-8, 2010.

Bauer, S. E., Koch, D., Unger, N., Metzger, S. M., Shindell, D. T. and Streets, D. G.: Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone, Atmos. Chem. Phys., 7(19), 5043–5059, doi:10.5194/acp-7-5043-2007, 2007.

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J. and Boucher, O.: Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, J. Geophys. Res., 116(D20), D20206, doi:10.1029/2011JD016074, 2011.

Donner, L. J., Wyman, B. L., Hemler, R. S., Horowitz, L. W., Ming, Y., Zhao, M., Golaz, J.-C., Ginoux, P., Lin, S.-J., Schwarzkopf, M. D., Austin, J., Alaka, G., Cooke, W. F., Delworth, T. L., Freidenreich, S. M., Gordon, C. T., Griffies, S. M., Held, I. M., Hurlin, W. J., Klein, S. a., Knutson, T. R., Langenhorst, A. R., Lee, H.-C., Lin, Y., Magi, B. I., Malyshev, S. L., Milly, P. C. D., Naik, V., Nath, M. J., Pincus, R., Ploshay, J. J., Ramaswamy, V., Seman, C. J., Shevliakova, E., Sirutis, J. J., Stern, W. F., Stouffer, R. J., Wilson, R. J., Winton, M., Wittenberg, A. T. and Zeng, F.: The Dynamical Core, Physical Parameterizations, and Basic Simulation Characteristics of the Atmospheric Component AM3 of the GFDL Global Coupled Model CM3, J. Clim., 24(13), 3484–3519, doi:10.1175/2011JCLI3955.1, 2011.

Fang, Y., Fiore, A. M., Horowitz, L. W., Gnanadesikan, A., Held, I., Chen, G., Vecchi, G. and Levy, H.: The impacts of changing transport and precipitation on pollutant distributions in a future climate, , 116, 1–14, doi:10.1029/2011JD015642, 2011.

Golaz, J.-C., Salzmann, M., Donner, L. J., Horowitz, L. W., Ming, Y. and Zhao, M.: Sensitivity of the Aerosol Indirect Effect to Subgrid Variability in the Cloud Parameterization of the GFDL Atmosphere General Circulation Model AM3, J. Clim., 24(13), 3145–3160, doi:10.1175/2010JCLI3945.1, 2011.

Hauglustaine, D. A., Balkanski, Y. and Schulz, M.: A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate, Atmos. Chem. Phys., 14(20), 11031–11063, doi:10.5194/acp-14-11031-2014, 2014.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5(3), 715–737, doi:10.5194/acp-5-715-2005, 2005.

Masui, T., Matsumoto, K., Hijioka, Y., Kinoshita, T., Nozawa, T., Ishiwatari, S., Kato, E., Shukla, P. R., Yamagata, Y. and Kainuma, M.: An emission pathway for stabilization

at 6 Wm−2 radiative forcing, Clim. Change, 109(1-2), 59–76, doi:10.1007/s10584-011-0150-5, 2011.

Ming, Y. and Ramaswamy, V.: Nonlinear Climate and Hydrological Responses to Aerosol Effects, J. Clim., 22(6), 1329–1339, doi:10.1175/2008JCLI2362.1, 2009.

Ming, Y. and Ramaswamy, V.: A Model Investigation of Aerosol-Induced Changes in Tropical Circulation, J. Clim., 24(19), 5125–5133, doi:10.1175/2011JCLI4108.1, 2011.

Ming, Y., Ramaswamy, V. and Chen, G.: A Model Investigation of Aerosol-Induced Changes in Boreal Winter Extratropical Circulation, J. Clim., 24(23), 6077–6091, doi:10.1175/2011JCLI4111.1, 2011.

Naik, V., Horowitz, L. W., Fiore, A. M., Ginoux, P., Mao, J., Aghedo, A. M. and Levy, H.: Impact of preindustrial to present-day changes in short-lived pollutant emissions on atmospheric composition and climate forcing, J. Geophys. Res. Atmos., 118, n/a–n/a, doi:10.1002/jgrd.50608, 2013.

Paulot, F., Ginoux, P., Cooke, W. F., Donner, L. J., Fan, S., Lin, M., Mao, J., Naik, V. and Horowitz, L. W.: Sensitivity of nitrate aerosols to ammonia emissions and to nitrate chemistry: implications for present and future nitrate optical depth, Atmos. Chem. Phys. Discuss., 15(18), 25739–25788, doi:10.5194/acpd-15-25739-2015, 2015.

Ramanathan, V., Crutzen, P. J., Kiehl, J. T. and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle., Science, 294(5549), 2119–24, doi:10.1126/science.1064034, 2001.

Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A. and Andreae, M. O.: Flood or drought: how do aerosols affect precipitation?, Science, 321(5894), 1309–13, doi:10.1126/science.1160606, 2008.

Schmidt, G. A., Shindell, D. T. and Tsigaridis, K.: Reconciling warming trends, Nat. Publ. Gr., 7(3), 158–160, doi:10.1038/ngeo2105, 2014.

Shindell, D. T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C., Chin, M., Young, P. J., Lee, Y. H., Rotstayn, L., Mahowald, N., Milly, G., Faluvegi, G., Balkanski, Y., Collins, W. J., Conley, A. J., Dalsoren, S., Easter, R., Ghan, S., Horowitz, L., Liu, X., Myhre, G., Nagashima, T., Naik, V., Rumbold, S. T., Skeie, R., Sudo, K., Szopa, S., Takemura, T., Voulgarakis, A., Yoon, J.-H. and Lo, F.: Radiative forcing in the ACCMIP historical and future climate simulations, Atmos. Chem. Phys., 13(6), 2939–2974, doi:10.5194/acp-13-2939-2013, 2013.

Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system., Nature, 461(7264), 607–13, doi:10.1038/nature08281, 2009.

Takemura, T.: Distributions and climate effects of atmospheric aerosols from the preindustrial era to 2100 along Representative Concentration Pathways (RCPs) simulated

using the global aerosol model SPRINTARS, Atmos. Chem. Phys., 12(23), 11555–11572, doi:10.5194/acp-12-11555-2012, 2012.

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P. J., Prather, M. J., Wild, O., Field, R. D., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Folberth, G. A., Horowitz, L. W., Josse, B., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S. and Zeng, G.: Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, Atmos. Chem. Phys., 13(5), 2563–2587, doi:10.5194/acp-13-2563-2013, 2013.