

## **Response to Anonymous Referee #1 for “Radiative forcing and climate response to projected 21<sup>st</sup> 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 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<sup>st</sup> 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  $\mu\text{m}$  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 21<sup>st</sup> 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 ( $-1.0 \text{ W m}^{-2}$  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  $-1.8 \text{ W m}^{-2}$  (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  $-1.0 \text{ W m}^{-2}$ . 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  $-1.0 \text{ W m}^{-2}$ . 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 arctic (~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.

- Emissions: 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<sub>2</sub>, BC, and OC emissions in RCP6 are higher relative to the other

RCPs over roughly the same time period in (Fig. 2). SO<sub>2</sub> 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 21<sup>st</sup> 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<sub>2</sub> 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 mentioned 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<sup>st</sup> 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 21<sup>st</sup> 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 21<sup>st</sup> 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  $-0.9 \text{ W m}^{-2}$  (uncertainty range  $-1.9$  to  $-0.1 \text{ W m}^{-2}$ ).

***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 Jimenez 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<sub>2</sub> forcing is about 5 W m<sup>-2</sup> (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<sub>2</sub> 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 SO<sub>2</sub>. 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: CO<sub>2</sub> 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  $\Delta C/\Delta AOD$  (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 21<sup>st</sup> 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<sup>-2</sup> 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<sup>-2</sup> 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.

# Radiative forcing and climate response to projected 21<sup>st</sup> century aerosol decreases

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## Abstract

It is widely expected that global emissions of atmospheric aerosols and their precursors will decrease strongly throughout the remainder of the 21<sup>st</sup> century, due to emission reduction policies enacted to protect human health. For instance, global emissions of aerosols and their precursors are projected to decrease by as much as 80% by the year 2100, according to the four Representative Concentration Pathway (RCP) scenarios. The removal of aerosols will cause unintended climate consequences, including an unmasking of global warming from long-lived greenhouse gases. We use the Geophysical Fluid Dynamics Laboratory Climate Coupled Climate Model version 3 (GFDL CM3) to simulate future climate over the 21<sup>st</sup> century with and without the aerosol emission changes projected by each of the RCPs in order to isolate the radiative forcing and climate response resulting from the aerosol reductions. We find that the projected global radiative forcing and climate response due to aerosol decreases do not vary significantly across the four RCPs by 2100, although there is some mid-century variation, especially in cloud droplet effective radius, that closely follows the RCP emissions and energy consumption projections. Up to 1 W m<sup>-2</sup> of radiative forcing may be unmasked globally from 2005 to 2100 due to reductions in aerosol and precursor emissions, leading to average global temperature increases up to 1 K and global precipitation rate increases up to 0.09 mm d<sup>-1</sup>. However, when using a version of CM3 with reduced present-day aerosol

1 radiative forcing (-1.0 W m<sup>-2</sup>), the global temperature increase for RCP8.5 is about 0.5 K,  
2 with similar magnitude decreases in other climate response parameters as well. Regionally  
3 and locally, climate impacts can be much larger than the global mean, with a 2.1 K warming  
4 projected over China, Japan, and Korea due to the reduced aerosol emissions in RCP8.5, as  
5 well as nearly a 0.2 mm d<sup>-1</sup> precipitation increase, a 7 g m<sup>-2</sup> LWP decrease, and a 2 μm  
6 increase in cloud droplet effective radius. Future aerosol decreases could be responsible for  
7 30-40% of total climate warming (or 10-20% with weaker aerosol forcing) by 2100 in East  
8 Asia, even under the high greenhouse gas emissions scenario (RCP8.5). The expected  
9 unmasking of global warming caused by aerosol reductions will require more aggressive  
10 greenhouse gas mitigation policies than anticipated in order to meet desired climate targets.

## 11 1 Introduction

12 The climate effects of atmospheric aerosols represent one of the most uncertain aspects of  
13 current and future climate forcing and response estimates (IPCC Myhre et al., 2013). Whereas  
14 the greenhouse gas warming influences on climate are relatively well understood, significant  
15 questions remain regarding the magnitude, and in some cases, even the sign (cooling or  
16 warming), of aerosol-climate interactions. Aerosol radiative forcing of climate can be split  
17 into two categories: the direct effect, in which atmospheric aerosols directly scatter or absorb  
18 incoming solar radiation; and the indirect effect, in which aerosols modify cloud properties  
19 which in turn affect the radiation budget. For a fixed amount of cloud water, a more polluted  
20 air mass will have smaller and more numerous cloud droplets, leading to a larger surface area  
21 and a brighter cloud (Twomey, 1977). This is known as the cloud albedo effect. In addition,  
22 more aerosol pollution may also result in a longer cloud lifetime due to the tendency of  
23 smaller droplets to remain suspended in the atmosphere longer (Albrecht, 1989), although this  
24 cloud lifetime effect is not as well understood. Generally On a global average basis, both the  
25 direct and indirect effects tend to exert a net negative radiative forcing on present-day climate,  
26 ~~leading ultimately to a decrease in average global temperature~~ opposing the positive forcing  
27 from greenhouse gases, with the total aerosol effective radiative forcing estimated to be -0.9  
28 W m<sup>-2</sup> (uncertainty range -1.9 to -0.1 W m<sup>-2</sup>) (Myhre et al., 2013). This aerosol forcing has  
29 likely offset a significant portion of present-day CO<sub>2</sub> and other greenhouse gas-induced  
30 climate forcing and subsequent global warming. Likewise, any changes in future  
31 anthropogenic aerosols will have implications for the overall net impact on climate. Here we  
32 evaluate the changes in global and regional aerosol burden, climate forcing, and climate

1 response due to future decreases in aerosol and precursor emissions as projected by all of the  
2 Representative Concentration Pathways (RCPs). We build upon the work of Levy et al.  
3 (2013) to contrast the global and regional climate response and quantify the expected  
4 unmasking of warming due to future aerosol reductions across all of the RCPs.

5 Emissions of aerosols and their precursors have increased dramatically since the preindustrial  
6 era, due to increasing industrialization and global population. In 2012, air pollution, mostly in  
7 the form of atmospheric aerosols, was responsible for 7 million deaths (3.7 million from  
8 ambient air pollution, 3.3 million from indoor) worldwide (WHO, 2014). Due to efforts to  
9 reduce this enormous human health impact, aerosol and precursor emissions are expected to  
10 decline worldwide over the next several decades as governments enact and enforce stricter  
11 emission control policies. Emissions of sulfur dioxide (SO<sub>2</sub>, a precursor to sulfate aerosol)  
12 have already declined about 50% in North America and Western Europe and passed their peak  
13 in 2005 in China (Klimont et al., 2013). In some developing countries, emissions are still  
14 rising but are expected to begin to decline within the next few decades, as a consequence of  
15 increasing affluence and more environmental regulation (van Vuuren et al., 2011a).

16 While clearly beneficial for human health, declining aerosol emissions will result in the  
17 unintended consequence of unmasking additional climate warming, due to the reduction of the  
18 cooling effects from anthropogenic aerosols such as sulfate and organic carbon (OC). Thus,  
19 careful policy implementation is necessary in order to maximize reduction of unhealthy air  
20 pollution while also minimizing the unmasking of additional global warming. Some studies  
21 have pointed to reductions in black carbon as a possible approach to address this dilemma  
22 (Bond et al., 2013; Kopp and Mauzerall, 2010; Shindell et al., 2012). Black carbon (BC) is an  
23 aerosol species that is a strong absorber of incoming solar radiation in the troposphere and  
24 thus a climate-warming agent. It is also a major contributor to PM<sub>2.5</sub> and has an adverse effect  
25 on human health (Bond et al., 2013). Despite a clear human health benefit, questions remain  
26 whether its reduction will be an effective strategy for avoiding additional warming, due to its  
27 frequent co-emission with two strong cooling species, OC and sulfate (Chen et al., 2010;  
28 Reddington et al., 2013). Some sources of BC are not large sources of species that exert a  
29 negative radiative forcing (e.g. diesel fuel combustion), so it has been suggested that these  
30 should be preferentially targeted for control first (Kopp and Mauzerall, 2010; Shindell et al.,  
31 2012).



1 Modeling studies have suggested that aerosols also have strong impacts on precipitation,  
2 cloud cover, cloud droplet size and number, atmospheric circulation, and other climate  
3 parameters (Lohmann and Feichter, 2005; Ming and Ramaswamy, 2009, 2011; Ming et al.,  
4 2011; Ramanathan et al., 2001; Rosenfeld et al., 2008; Stevens and Feingold, 2009). An  
5 increase in aerosol emissions tends to decrease local to regional precipitation rates (warm  
6 rain), through macrophysical and microphysical processes: 1) less incoming solar radiation  
7 penetrates the troposphere and reaches the surface, resulting in less evaporation (Ramanathan  
8 et al., 2001) and 2) smaller and more abundant aerosols lead to smaller cloud droplets, which  
9 are less likely to convert to rain drops via coalescence on the local and regional scale (Radke  
10 et al., 1989; Rosenfeld, 2000). An exception to (1) is BC or other absorbing aerosols, which  
11 may have opposing effects on precipitation through heating the atmosphere (causing  
12 stabilization and reduction in precipitation) and warming the surface via redistributed thermal  
13 energy (causing an enhancement of precipitation) (Ming et al., 2010). However, the net effect  
14 of increasing aerosol concentrations tends to be suppression of precipitation. Thus, the  
15 expected reduction of aerosol concentrations should increase rainfall rates (warm rain)  
16 globally. Since aerosols serve as seeds for virtually all liquid cloud formation in the  
17 atmosphere, decreases in aerosols may also be expected to affect cloud cover, cloud liquid  
18 water path, and effective cloud droplet radius.

19 To estimate future aerosol emissions and burden, radiative forcing, and climate response, we  
20 must ~~utilize~~ rely on future projections or scenarios. The current state-of-the-art emissions  
21 scenarios for global climate modeling are the Representative Concentration Pathways (RCP)  
22 (Lamarque et al., 2011; Masui et al., 2011; Riahi et al., 2011; van Ruijven et al., 2008;  
23 Thomson et al., 2011; van Vuuren et al., 2012; Vuuren et al., 2011a, 2011b). The RCPs are  
24 different from previously developed scenarios in that they are initialized with radiative forcing  
25 beginning and endpoints (2005-2100). A consistent but non-unique pathway based on  
26 literature and integrated assessment modeling is then selected to match the prescribed  
27 endpoint. There are four scenarios, each named for a radiative forcing endpoint in 2100 of 2.6,  
28 4.5, 6.0, and 8.5 W m<sup>-2</sup> (RCP2.6, RCP4.5, RCP6, and RCP8.5). Each scenario contains  
29 emissions and/or concentrations for all major greenhouse gases and air pollutants, including  
30 emissions of three aerosol/precursor species: SO<sub>2</sub>, OC, and BC. All RCPs assume an  
31 autonomous change in future air pollution control policies in every world region, resulting in  
32 sharp decreases in regional and global emissions of SO<sub>2</sub>, OC, and BC.

1 There have been several previous studies on the effects of diminishing emissions of aerosols  
2 and their precursors on aerosol burden, radiative forcing, and climate (Arneth et al., 2009;  
3 Bellouin et al., 2011; Chalmers et al., 2012; Gillett and Von Salzen, 2013; Kloster et al., 2009;  
4 Lamarque et al., 2011; Leibensperger et al. 2012; Makkonen et al., 2011; Menon et al., 2008;  
5 Rotstayn et al., 2013; Shindell et al., 2013; Smith and Bond, 2014; Takemura, 2012; Unger et  
6 al., 2009). These studies and their results are summarized in Table 1. In order to have  
7 consistent comparisons to the present work, we focus Table 1 on recent studies that used a  
8 global climate modeling framework with the RCPs through 2100; hence, studies that may  
9 have used other scenarios are not included. ~~We compare recent studies to results from the~~  
10 ~~present work in Sect. 5.1.~~

11 Recently, Levy et al. (2013) used GFDL CM3 model simulations of RCP4.5 to analyze  
12 changes in radiative forcing, temperature, and precipitation driven by reductions of aerosol  
13 emissions. To isolate the effects of decreasing aerosols, Levy et al. (2013) compared the  
14 results of the RCP4.5 simulations with those of another set of simulations, in which all aerosol  
15 and precursor emissions were held fixed at 2005 levels throughout the remainder of the 21<sup>st</sup>  
16 century. The authors found roughly an additional 1° C warming and a 0.1 mm d<sup>-1</sup> increase in  
17 precipitation due to the decreasing aerosols in RCP4.5. Here we expand on the results from  
18 the Levy et al. (2013) study by estimating the changes in global and regional aerosol burden,  
19 climate forcing, and climate response due to projected reductions in aerosol emissions for all  
20 four RCPs using an updated version of the same GFDL CM3 model.

21 We first present the experimental design and the RCP emissions scenarios in Sect. 2. We then  
22 go beyond the previous studies described in Table 1 by performing a more exhaustive and  
23 robust analysis of the unintended climate consequences of reducing particulate air pollution  
24 levels in the future. We also test the robustness of our results using sensitivity simulations  
25 with a version of the CM3 model in which aerosol forcing is weakened significantly from its  
26 default value (Golaz et al. 2013). We present historical to present day to future results from  
27 1860 to 2100, focusing first on global changes (Sect. 3) and then on specific regions that may  
28 be most strongly impacted (Sect. 4). We also compare our results with those from previous  
29 studies and examine similarities and differences in the projected aerosol-driven changes in  
30 climate variables, climate forcing, and aerosol burden across the various RCPs. Finally, we  
31 attempt to connect changes in aerosols with changes in forcing and climate parameters (Sect.  
32 5). Conclusions are presented in Section 6.

1

## 2 **2 Models and simulations**

### 3 **2.1 GFDL Climate Model 3**

4 We use the Geophysical Fluid Dynamics Laboratory Climate Model version 3 (GFDL CM3)  
5 in this work. CM3 is a fully coupled chemistry-climate model containing atmosphere, ocean,  
6 land, and sea-ice components. We employ the C48 version of the model, which uses a finite-  
7 volume cubed-sphere horizontal grid consisting of 6 faces with roughly a 200-km by 200-km  
8 spatial resolution. [Transport of tracers uses the finite volume algorithm of Lin and Rood  
\(1996\) with updates as described by Putnam and Lin \(2007\) and Donner et al. \(2011\).](#) The  
9 vertical grid consists of 48 vertical levels extending from the surface up to about 0.01 hPa (80  
10 km). Additional details on the model configuration and performance can be found in Donner  
11 et al. (2011), Naik et al. (2013), and references therein. [Details on the ocean and sea ice model  
can be found in Griffies et al. \(2011\).](#)  
13

14 Anthropogenic emissions of aerosols and their precursors (and emissions or concentrations of  
15 all other reactive chemical species) are based on decadal estimates from Lamarque et al.  
16 (2010) for the historical period (1860-2000) and from Lamarque et al. (2011) for the RCP  
17 projections (2005-2100). Concentrations of long-lived greenhouse gases are based on  
18 Meinshausen et al. (2011). Source-specific emissions are provided for anthropogenic sources  
19 (energy use, industrial processes, and agriculture), biomass burning, shipping, and aircraft  
20 emissions. Since natural emission sources are not specified by Lamarque et al. (2010), we  
21 follow the methodology described by Naik et al. (2013), including emissions of isoprene, soil  
22 NO<sub>x</sub>, lightning NO<sub>x</sub>, marine primary organic aerosol (POA), DMS, dust, and sea salt. Changes  
23 in climate do not feed back on natural emissions except for dust and sea salt, which respond to  
24 simulated wind speeds, and lightning NO<sub>x</sub>, which responds to convective activity. The lack of  
25 temperature feedback on biogenic VOC emissions may lead to underestimates of future  
26 isoprene or other biogenic VOCs (Heald et al., 2008). Volcanic emissions are as described by  
27 Donner et al. (2011).

28 The tropospheric chemistry component in CM3 is based on Horowitz et al. (2003) with  
29 updates from Horowitz (2006) and solves the reaction rate differential equations using an  
30 implicit Euler backward method solver with Newton-Raphson iteration. There are 97 total  
31 chemical species including 16 aerosol species. Tropospheric chemical reactions, including the

1 NO<sub>x</sub>-O<sub>x</sub>-VOC system, are included in the model and fully coupled with the emissions and  
2 atmospheric radiation sections of the model. There are a total of 171 gas-phase reactions, 41  
3 photolysis reactions, and 16 heterogeneous reactions simulated in the model (Naik et al.,  
4 2012). Sulfate aerosols are formed via oxidation of SO<sub>2</sub> by the hydroxyl radical (OH), ozone  
5 (O<sub>3</sub>), and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). The oxidation of DMS to sulfate aerosols is also  
6 included. DMS emission from the oceans is parameterized based on 10-m reference height  
7 wind-speed but is independent of temperature. Carbonaceous aerosols are modeled in CM3 as  
8 primary organic aerosols (POA), secondary organic aerosols (SOA), and BC. SOA includes  
9 both natural and anthropogenic sources. Biogenic terpene oxidation is estimated to provide a  
10 directly emitted source of about 30.4 Tg C yr<sup>-1</sup> of SOA and butane oxidation by OH yields  
11 roughly another 9.6 Tg C yr<sup>-1</sup> (Dentener et al., 2006; Naik et al., 2013; Tie, 2005).  
12 Hydrophobic OC and BC aerosols are converted to hydrophilic aerosol with an e-folding time  
13 of 1.44 days. Sea salt and mineral dust aerosol are treated with a five-section size distribution  
14 ranging from 0.1 to 10 μm dry radius.

15 Aerosol optics in CM3 is based on Mie theory. Lognormal size distributions are assumed for  
16 OC, BC, and sulfate. Sulfate and hydrophilic BC are assumed to be internally-mixed and to  
17 undergo hygroscopic growth, while all other aerosols are externally mixed. New to CM3 was  
18 the addition of prognostic equations for calculation of cloud droplet number according to the  
19 Ming et al. (2006) parameterization, allowing for variable cloud droplet number and radius.

20 The cloud lifetime effect is parameterized as an increase in cloud droplet number (e.g. due to  
21 aerosol perturbations) resulting in a decrease in the autoconversion rate, thereby delaying  
22 precipitation and increasing cloud lifetime. The derivation of the parameterization is beyond  
23 the scope of this paper, but it follows the methodology of (Khairoutdinov and Kogan, (2000).

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24 An autoconversion threshold of 8.2 μm is used in CM3. In a given grid box, if the volume-  
25 mean cloud droplet radius is less than this threshold, autoconversion is suppressed (Rotstayn,  
26 2000). Sensitivity of the indirect effect to different thresholds in GFDL AM3 is explored in  
27 (Golaz et al., (2011)). Despite being internally mixed with sulfate in the radiation calculation,  
28 black carbon is assumed to be externally mixed with soluble species (sulfate, sea salt, OC) for  
29 the aerosol activation calculation. Fractions of BC and OC are emitted as hydrophobic (80%  
30 and 50%, respectively) but undergo aging to hydrophilic BC and OC with e-folding times of  
31 1.44 and 2.88 days, respectively. Sulfate (treated as pure ammonium sulfate, internally mixed  
32 with BC for optics), BC, and OC are assigned individual lognormal size distributions for both  
33 the aerosol optics and activation code (Ming et al., 2007). Size-resolved aerosol microphysics

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

5 Donner et al. (2011) showed that CM3 improved upon CM2.1 model-measurement evaluation  
6 metrics for several aerosol-relevant quantities, including aerosol optical depth, co-albedo, and  
7 clear-sky shortwave surface radiation flux. In the current model configuration, neither the  
8 radiation nor the activation code currently include nitrate aerosol. At present nitrate aerosols  
9 are estimated to have only contributed marginally to aerosol radiative forcing and climate  
10 effects. As nitrate may become a more significant contributor to aerosol radiative forcing in  
11 the future (Bauer et al., 2007; Bellouin et al., 2011; Hauglustaine et al., 2014), the chemical,  
12 radiative, and microphysical properties of nitrate aerosol are being incorporated into a new  
13 version of the GFDL atmospheric model.

## 14 **2.2 RCPs**

15 The Representative Concentration Pathways (RCPs) contain emissions projections for all  
16 long- and short-lived climate forcers, including the aerosol and aerosol precursor species SO<sub>2</sub>,  
17 OC, and BC. The RCPs were featured in the Intergovernmental Panel on Climate Change  
18 (IPCC) Fifth Assessment Report (AR5) and are the successors to the Special Report on  
19 Emissions Scenarios (SRES). The RCPs include globally gridded projections of emissions of  
20 important atmospheric constituents from 2005 to 2100 with extensions to 2300. Using  
21 literature and integrated assessment modeling (IAM), representative pathways are selected to  
22 fit the individual RCP starting and ending points for radiative forcing. Concentrations and  
23 emissions of short- and long-lived climate forcers (including aerosols) are then harmonized  
24 (made consistent with base years) and emissions are downscaled (gridded on a latitude-  
25 longitude scale). Each of the chosen final pathways are not unique; however, a consortium of  
26 experts from the IAM and IPCC communities have selected pathways with desirable qualities  
27 such as coverage of the entire literature range and significant spread in concentrations and  
28 emissions between the individual pathways. The four pathways include a strong mitigation  
29 scenario (RCP2.6), two stabilization scenarios in which radiative forcing stabilizes shortly  
30 after 2100 (RCP4.5 and RCP6), and one high emissions/low mitigation scenario (RCP8.5),  
31 which current emissions most closely track. The radiative forcing pathways in each RCP are  
32 internally consistent with concentrations of both short and long-lived climate forcers, with the

1 exception of dust and nitrate aerosol forcing (Masui et al., 2011; Riahi et al., 2007, 2011;  
2 Thomson et al., 2011; Vuuren et al., 2011a, 2011b). In addition to representing different GHG  
3 emission trajectories, the RCPs implicitly assume air pollution reduction policies in which  
4 emissions of reactive pollutants decrease as a function of increasing income, but  
5 independently of GHG mitigation levels.

6 Figure 2 shows the global total emissions (anthropogenic and biomass burning) of SO<sub>2</sub>, BC,  
7 and OC for the historical period and for each of the RCPs (see Figs. S1 and S2 of  
8 supplemental information for the split between anthropogenic and biomass burning). All four  
9 scenarios project strong decreases in globally averaged sulfur dioxide (SO<sub>2</sub>), organic carbon  
10 (OC), and black carbon (BC) emissions throughout the 21<sup>st</sup> century. SO<sub>2</sub> emissions, according  
11 to the RCPs, have already peaked globally (around 1980) and have been on the decline since,  
12 except for a slight uptick in the early 2000s. By 2100, SO<sub>2</sub> emissions will have decreased  
13 from a 2005 level of around 120 Tg yr<sup>-1</sup> to 13-26 Tg yr<sup>-1</sup> (RCP2.6 to RCP8.5), about an 80%  
14 decrease. RCP2.6 projects the largest decrease, followed by RCP4.5, RCP6, and RCP8.5. This  
15 order is consistent with each RCP's embedded climate policy, since RCP2.6 has the most  
16 stringent climate policy and RCP8.5 the least stringent, and many climate policies that curb  
17 CO<sub>2</sub> emissions can also curb SO<sub>2</sub> (e.g. replacement of coal-fired power plants with renewable  
18 sources). However, this order does not hold for all species, on either the global and regional  
19 scale. Air pollution control also should have an impact on the air pollutant emissions  
20 trajectory; however, the assumptions regarding air pollution control were too similar across  
21 each of the RCPs to wield noticeable influence (Rogelj et al., 2014). Thus, climate policies  
22 (specifically reductions in CO<sub>2</sub> emissions from reduced dependence on coal energy) tend to  
23 dominate the trend in not only emissions of aerosols and their precursors, but also in aerosol  
24 optical depth, radiative forcing, and climate response, as we will show later. Although there is  
25 some variability in global SO<sub>2</sub> emissions over the course of the time series, the RCPs all  
26 converge on a fairly narrow range of endpoints. In particular, RCP6 and RCP2.6 stand out, the  
27 former due to an increase in the rate of coal consumption around mid-century (2030-2060)  
28 and the latter due to stringent climate policy including the nearly complete phase-out of non-  
29 CCS (carbon capture and storage) coal energy by roughly 2050 (Masui et al., 2011; van  
30 Vuuren et al., 2011b). The increase in coal energy projected by RCP6 is a surprising feature  
31 that is not present in the other RCPs. As a result, SO<sub>2</sub>, BC, and OC emissions in RCP6 are  
32 higher relative to the other RCPs over roughly the same time period in (Fig. 2). SO<sub>2</sub> emissions  
33 briefly increase in absolute terms over a short period mid-century in RCP6, which drives

1 higher sulfate burdens, larger (negative) aerosol direct and indirect forcings, and noticeable  
2 changes in climate response, as we will show in the following sections. The narrow range for  
3 emissions of air pollutants and their precursors (e.g. SO<sub>2</sub>) has been attributed to similar air  
4 pollution policy assumptions in each RCP. New scenarios that strive to span the entire  
5 literature range for air pollutants in addition to greenhouse gases are undergoing development  
6 (Rogelj et al., 2014).

7 BC emissions (middle panel in Fig. 2) increased from preindustrial times and continue to  
8 increase in RCP2.6, peaking around 2010. RCP4.5, RCP6 and RCP8.5 all peak around 2005  
9 (i.e. the beginning of the RCP projections) and decrease continuously until 2100. Present-day  
10 values of BC emissions are 8.5 Tg yr<sup>-1</sup>. By 2100, BC emissions are projected to range from  
11 less than 3.3 Tg yr<sup>-1</sup> according to RCP2.6 up to 4.3 Tg yr<sup>-1</sup> in RCP6. Here, the expected order  
12 seen in the SO<sub>2</sub> emissions reductions is not observed, but BC emissions do also converge on a  
13 relatively narrow range of endpoints.

14 Emissions of OC, sometimes co-emitted with BC, have a similar trajectory (bottom panel Fig.  
15 2). The main source of OC emissions in the RCPs is biomass burning, which makes up about  
16 60% of the total emissions of OC in present day. RCP2.6 OC emissions continue to increase  
17 in the early 21<sup>st</sup> century, peaking around 2020. Emissions of OC in RCP4.5 drop rapidly, due  
18 to the strong decrease in cropland area and increase in forested area projected by RCP4.5 (in  
19 other words, a decrease in biomass burning as a means to clear cropland), a trend that is  
20 mostly unique to RCP4.5 (van Vuuren et al. 2011a, Thomson et al., 2011). OC emissions in  
21 RCP6 remain high throughout the 21<sup>st</sup> century due to the land use assumptions embedded in  
22 RCP6, which includes shifts to larger amounts of burning to clear land for crops. CO<sub>2</sub>  
23 fertilization also plays a role in the increased emissions, as higher CO<sub>2</sub> levels can increase  
24 biomass growth and increase the amount available to be burned (Kato et al., 2011). Compared  
25 to SO<sub>2</sub> and BC, the variability and range of emissions endpoints across the different RCPs is  
26 much wider, ranging from 20 Tg yr<sup>-1</sup> in RCP4.5 in 2100 to about 32 Tg yr<sup>-1</sup> in RCP6. The  
27 final order of each RCP's global total OC emissions is different from that of both the BC and  
28 SO<sub>2</sub> final order, highlighting the difficulty in comparison between different RCPs. In  
29 particular, the lack of climate policy in RCP8.5 has little influence on OC and BC emissions  
30 in comparison to SO<sub>2</sub>.

## 1 2.3 Simulations

2 We conduct simulations using GFDL CM3 to evaluate the role of changing aerosol emissions  
3 on aerosol optical depth (AOD), aerosol radiative forcing, and climate response. Table 2  
4 summarizes the simulations performed. A series of RCP simulations (denoted RCP $x.x$  where  
5  $x.x = 2.6, 4.5$ , etc.) were run from 2006-2100 in which the RCP emissions scenarios were  
6 used for future aerosol, greenhouse gas, and other reactive species emissions or  
7 concentrations. Another series of simulations were run using the RCPs, but with the  
8 anthropogenic and biomass burning emissions of SO<sub>2</sub>, OC, and BC held constant at 2005  
9 levels. All other inputs, including greenhouse gases, were kept the same as in the full RCP  
10 scenarios. We denote these fixed emission simulations as ~~RCP $x.x$~~ FRCP $x.x$  2005AER,  
11 where the ~~F~~2005AER signifies fixed aerosol and aerosol precursor emissions at 2005 levels.  
12 Each of the RCP $x.x$  and ~~RCP $x.x$~~ FRCP $x.x$  2005AER simulations were run as a 3-member  
13 ensemble, each initialized with different initial conditions, provided by another 3-member  
14 historical ensemble (1860-2005) of GFDL CM3. The RCP4.5 and  
15 ~~RCP4.5~~FRCP4.5 2005AER simulations are not scientifically different from those presented  
16 by Levy et al. (2013); however, our simulations were run with a newer model ~~version~~  
17 ~~which~~version, which included some minor updates. We calculate the aerosol-induced climate  
18 response as the difference between the two sets (RCP $x.x$  – ~~RCP $x.x$~~ FRCP $x.x$  2005AER).  
19 Unless otherwise specified, all results presented are ensemble means. Meteorological factors,  
20 such as increasing temperatures, changes in atmospheric circulation and stability, and changes  
21 in precipitation will also induce changes in aerosol concentrations (Dawson et al., 2007; Jacob  
22 and Winner, 2009; Leibensperger et al., 2012a; Pye et al., 2009; Tai et al., 2012, 2010). The  
23 influences of greenhouse-gas driven meteorological changes are included in both our RCP $x.x$   
24 and ~~RCP $x.x$~~ FRCP $x.x$  2005AER simulations. Our methodology of running the  
25 ~~RCP $x.x$~~ FRCP $x.x$  2005AER simulations for the full 21<sup>st</sup> century allows us to simulate the  
26 climate-driven effects on aerosol abundance (AOD and concentration). Additionally, taking  
27 the difference between RCP $x.x$  and ~~RCP $x.x$~~ FRCP $x.x$  2005AER isolates the changes in  
28 aerosols (and climate response) resulting from emissions reductions alone, separate from the  
29 influence of well-mixed GHG-driven climate changes. While these  
30 ~~RCP $x.x$~~ FRCP $x.x$  2005AER simulations (or analogues) were performed in some past studies,  
31 to our knowledge this is the first study to present the results from such simulations for all four  
32 of the RCPs in a consistent framework.



1 In order to estimate aerosol effective radiative forcing, we ran additional non-coupled,  
2 atmospheric component-only simulations (AM3) in which sea surface temperatures (SST)  
3 were held constant at the 1981-2000 climatological values. Simulations were run in which the  
4 only climate forcings were aerosol emissions changes from present-day through 2100,  
5 allowing us to calculate the aerosol forcing by differencing radiative fluxes with a control  
6 simulation using 1860 emissions and the same climatological SSTs. Using the fixed SST  
7 approach allows us to estimate the radiative response to “fast” adjustments (time scale of a  
8 few years or less) to the climate system due to a forcing agent (e.g., aerosols) in addition to  
9 the full<sup>2</sup> response to the forcing agent itself. This method accounts for the indirect effects of  
10 aerosols, including the cloud lifetime effect and the cloud albedo effect. This type of fixed-  
11 SST calculation has been referred to as the “radiative flux perturbation” (Lohmann et al.,  
12 2010) or “effective radiative forcing” (Myhre et al., 2013). This series of simulations are  
13 denoted RCPx.x\_RFP for the aerosol-only simulations and RCP\_1860 for the control  
14 simulation. This pair of simulations is only used for the radiative forcing calculations, while  
15 the coupled runs described above provide AOD and climate response estimates.

16 As reported in (Levy et al., (2013), the present-day aerosol effective radiative forcing (ERF)  
17 in CM3 of  $-1.7 - -1.8 \text{ W m}^{-2}$  falls close to the upper bound of the 90% confidence interval, -  
18  $1.9 \text{ W m}^{-2}$ , set by the IPCC. To test the robustness of our results, we conduct additional  
19 experiments using an alternate model configuration with weaker present-day aerosol effective  
20 radiative forcing. With a lower “starting point”, future decreases in the magnitude of aerosol  
21 forcing will also be reduced, likely resulting in a reduced climate response. We use the  
22 “CM3w” experiment described by (Golaz et al., (2013), in which cloud parameters, most  
23 notably the autoconversion threshold, are tuned to produce the desired changes in climate  
24 forcing. In this version, the present-day aerosol effective radiative forcing is around  $-1.0 \text{ W m}^{-2}$ .  
25 To achieve this effect, we adjust the autoconversion threshold from its default value of  $8.2$   
26  $\mu\text{m}$  to  $6.0 \mu\text{m}$ . Since a lower autoconversion threshold is not consistent with observations  
27 (Rotstajn, 2000), we emphasize that this change is employed only as a means to test our  
28 results with a lower magnitude present-day aerosol ERF, and is not meant to produce a more  
29 scientifically accurate model version. The CM3w run is initialized from a historical run of  
30 CM3w, using one ensemble member each for RCP8.5 and RCP8.5 2005AER. These  
31 simulations are denoted CM3w\_RCP8.5 and CM3w\_RCP8.5\_2005AER.

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### 1 3 Global analysis

2 We first focus our analysis on area-weighted global means ranging from pre-industrial times  
3 to 2100. We analyze aerosol optical depth (sulfate, BC, and OC), aerosol climate forcing  
4 (direct and indirect), and climate response (temperature, precipitation rate, liquid water path,  
5 and effective cloud droplet radius). In subsequent sections, the analysis is extended to  
6 examine spatial distributions and mean changes over specific regions. Past, present, and future  
7 trends in global aerosol optical depth can be found in the supplemental information Sect. S2  
8 as well as Figs. S3 and S4. In short, AOD trends are well correlated with emissions trends,  
9 with correlation coefficients ranging from 0.7 to 0.9 for each species and each RCP (not  
10 shown) globally averaged, and 0.9 or greater over continental source regions. By 2100, sulfate  
11 AOD globally is projected to decrease by 50% from 2005 levels in RCP2.6, 40% in RCP4.5  
12 and RCP6, and 31% in RCP8.5 (see Supplemental Information). Figure S3 shows the  
13 RCPx.x\_FRCPx.x\_2005AER fixed emissions simulations (dashed lines) along with the  
14 RCPx.x decreasing emissions runs (solid lines) indicating that in the absence of emissions  
15 changes, future climate changes cause AOD to increase globally for each of the RCPs.  
16 However, this climate driven effect is small compared to the substantial decrease in AOD  
17 from emissions reductions (compare dashed and solid lines in Fig. S3).

#### 18 3.1 Aerosol forcing

##### 19 3.1.1 Aerosol direct and indirect forcing

20 Future cChanges (mostly decreases) in aerosol emissions and aerosol amount and optical  
21 depth lead to changes in Earth's radiative balance. Figure 3 shows the globally averaged total  
22 top-of-atmosphere (TOA) aerosol effective radiative forcing (direct and indirect) for the  
23 historical period and four RCP projections using the standard CM3 runs. From 1860 until  
24 present day, the increasing abundance of atmospheric aerosols led to a larger (more negative)  
25 aerosol forcing, peaking near present day. Preindustrial to present day aerosol forcing  
26 simulated by CM3 is about  $-1.8 \text{ W m}^{-2}$ . This large negative forcing has offset or "masked"  
27 some of the positive forcing from greenhouse gases. Although the net forcing is still positive,  
28 without the large increase in the 20<sup>th</sup> century of aerosol emissions, the net positive forcing  
29 would be much larger. As we discuss in Sect. 3.2, this masking by aerosols of the positive,  
30 greenhouse gas warming has important implications for climate. During the 21<sup>st</sup> century, the  
31 large decreases in global aerosol and aerosol precursor emissions projected by the RCPs cause

1 aerosol forcing to decrease in magnitude (become less negative). As discussed in Sect. 2.3, we  
2 also use a version of CM3 with a reduced present-day aerosol ERF, in order to test the  
3 robustness of our results to the magnitude of aerosol forcing. We present climate response  
4 results for these simulations in subsequent sections but did not run fixed-SST simulations with  
5 this adjusted version of CM3, hence we do not present forcing values associated with these  
6 runs.

7 As in many of the other trends shown thus far, there is limited spread in the various RCP  
8 projections of global aerosol effective radiative forcing (Fig. 3). For the 2096-2100 five-year  
9 average, the effective forcing (relative to 1860) is  $-0.21 \text{ W m}^{-2}$  for RCP2.6,  $-0.32 \text{ W m}^{-2}$  for  
10 RCP4.5,  $-0.46 \text{ W m}^{-2}$  for RCP6, and  $-0.53 \text{ W m}^{-2}$  for RCP8.5. RCP2.6 has the largest decrease  
11 in magnitude of aerosol forcing over the century, followed by RCP4.5, RCP6, and RCP8.5,  
12 which is the expected order according to each RCP's underlying climate policy. For example,  
13 reduction of coal energy usage, a GHG mitigation policy featured in the RCPs, also reduces  
14 the amount of  $\text{SO}_2$  emissions. As a result, total aerosol forcing trends and the end-of-century  
15 rank order for each of the RCPs can be traced back ultimately to the energy and climate  
16 mitigation policy. Furthermore, sulfate is the main contributor to the aerosol direct and  
17 indirect forcing trend for all RCPs, due to its optical properties as well as large CCN activity  
18 (hygroscopicity). Therefore, energy policies that affect sulfate will have a magnified effect on  
19 aerosol direct and indirect forcing. On the other hand, the direct climate effects of BC and OC  
20 have been reported to offset each other (OC being the negative forcing, BC positive) in  
21 previous studies with CM3 (Levy et al., 2013), and that is the case in this work as well. RCP6  
22 projects the smallest decrease in magnitude of aerosol forcing for much of the middle part of  
23 the century (2045 – 2075), despite passing RCP8.5 eventually. This is consistent with both the  
24 emissions and AOD trajectories for RCP6. RCP6 projects mid-century increases in coal for  
25 energy supply globally (Masui et al., 2011), which is visible not only in the emissions and  
26 AOD trends as described elsewhere but also the aerosol forcing trends.

### 27 3.1.2 Comparison to 2005 levels

28 The large decrease in the magnitude of aerosol forcing from present-day to 2100 represents a  
29 large *positive* forcing for 2100 relative to present. The globally averaged forcing changes from  
30 present-day to year 2100 for each RCP scenario are tabulated in Table 3. In order to have a  
31 consistent basis for comparison across the four RCPs and to account for noise in the trends,  
32 the beginning and end of century forcing values were taken as 5-year averages, 2000-2004

1 and 2096-2100. The amount of unmasked aerosol forcing follows the order expected  
2 according to each RCP's climate policy (as explained above), as is the case with SO<sub>2</sub>  
3 emissions (Fig. 2). For RCP2.6, the forcing increases by 1.37 W m<sup>-2</sup> from 2000 to 2100.  
4 RCP8.5 represents the lower part of the range at 1.05 W m<sup>-2</sup>. An additional positive forcing of  
5 at least 1 W m<sup>-2</sup> would have major climate implications. For comparison, the ~~present day~~<sup>2100</sup>  
6 ~~RCP8.5~~ CO<sub>2</sub> forcing is about ~~1.68 W m<sup>-2</sup> (range of 1.33 to 2.03 W m<sup>-2</sup>)~~<sup>5 W m<sup>-2</sup></sup> (Myhre et al.,  
7 2013). Thus, the resulting positive forcing from the decrease in aerosol emissions by 2100 is  
8 projected to be more than ~~half~~<sup>20%</sup> of the forcing of ~~present day~~<sup>CO<sub>2</sub> in 2100</sup>.

9 As a caveat, our reported aerosol effective radiative forcing values may be a ~~slight~~  
10 overestimate due to the tendency of the GFDL CM3 model to overstate the cloud lifetime  
11 effect, thus inflating the magnitude of our positive forcing due to aerosol decline (Levy et al.,  
12 2013). We test the robustness of our results with respect to the magnitude of aerosol forcing  
13 using CM3w, a version of the model with much weaker present-day ERF (see Sect. 2.3). In a  
14 multimodel evaluation of proxies for the aerosol indirect effects (both albedo and lifetime)  
15 against satellite observations, a prototype version of AM3 was found to be one of several  
16 models overestimating the strength of the relationship between cloud liquid water path (LWP)  
17 and aerosol optical depth (defined as sensitivity of LWP to AOD perturbations) (Quaas et al.,  
18 2009). Ratios for AM3 were roughly an order of magnitude larger than observations over land  
19 and ocean; however, other models performed just as poorly if not worse (all models  
20 overestimate the land ratio by at least a factor of two). Generally, a positive correlation is  
21 expected between aerosol optical depth and cloud liquid water path, since the increase in  
22 cloud droplet number concentration leads to a delay in autoconversion rate, increasing cloud  
23 lifetime and cloud liquid water path. However, some studies have found potential for a  
24 negative correlation due to a drying effect from increased entrainment of air above the clouds  
25 (Ackerman et al., 2004). Thus, the autoconversion parameterization in AM3, which is a  
26 simple implementation that does not address the effect of increased entrainment and other  
27 confounding issues, could conceivably be driving an overestimate in the cloud lifetime effect  
28 and thus the indirect effect as a whole.

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## 1 3.2 Climate response

### 2 3.2.1 Global historical and future trends

3 | The emission reductions and aerosol forcing trends discussed earlier ~~will are expected to~~ have  
4 significant effects on future climate. Figure 4 shows the 1860-2100 globally averaged time  
5 series for temperature, precipitation rate, liquid water path, and effective cloud droplet radius  
6 | for the historical, RCPx.x ensemble mean (solid lines), and ~~RCPx.x~~ FRCPx.x 2005AER  
7 ensemble mean (dashed lines). Since the RCPx.x simulations contain all climate-forcing  
8 agents (compared to the aerosol differences we will discuss later), greenhouse gases have a  
9 large influence. Temperature anomaly (relative to 1881-1920 average) is projected to range  
10 from about 2-5 K by 2100, mostly due to anthropogenic greenhouse warming. Likewise,  
11 global precipitation rates will increase, which will be shown in the following section to result  
12 partially from an increase in greenhouse gases and a decrease in aerosol emissions.  
13 Precipitation rate in the historical simulation had a significant decrease around 1950-1970;  
14 this time frame coincides with a doubling in the global anthropogenic SO<sub>2</sub> emissions (Fig. 2).  
15 Liquid water path (LWP) (Fig. 4, bottom left) steadily increases from 1860 to present day,  
16 then has four very different trajectories in the RCP future projections. The higher  
17 temperatures of RCP8.5 and RCP6 most likely lead to LWP increases, and the order of LWP  
18 values in each RCP in 2100 is consistent with the amount of temperature increase. Effective  
19 cloud droplet radius (calculated over the top two units of optical depth for liquid clouds,  
20 consistent with the Moderate Resolution Imaging Spectroradiometer (MODIS) algorithm  
21 (Donner et al., 2011; Guo et al., 2014; King et al., 2003) and weighted by cloud fraction)  
22 decreases steadily from 1950 to present day, also due to the large increase in aerosol  
23 emissions. Since many ultrafine particles are anthropogenic in nature (e.g., sulfate), an  
24 increase in their emission drives down the global average cloud droplet effective radius, since  
25 smaller sized aerosols are effective CCN that are likely to activate into cloud droplets.  
26 Likewise, with aerosol emissions declining in the 21<sup>st</sup> century, cloud droplet radius increases  
27 across all of the RCPs.

### 28 3.2.2 Climate response to aerosol forcing

29 As with aerosol optical depth, taking the difference between the time-varying simulations  
30 discussed in Sect 3.3.1 and the fixed aerosol emission simulations (RCPx.x –  
31 | ~~RCPx.x~~ FRCPx.x 2005AER), we deduce the impacts on climate driven solely by the aerosol

1 emissions reductions for each year of each future emission scenario. Fig. 5 presents the  
2 globally-averaged differences for temperature, precipitation rate, ~~cloud cover~~LWP, and cloud  
3 drop effective radius resulting from aerosol emission reductions in the RCPs (also tabulated in  
4 Table 3 for a five year average of 2096-2100 only). Additionally, the dashed lines in Fig. 4  
5 represent the fixed emission ensemble means (~~RCPx.x~~~~FRCPx.x~~ 2005AER), thus comparing  
6 these with the values from the time-varying emissions ensemble (solid lines) gives an estimate  
7 of the aerosol unmasking-driven contribution to the climate response. The projected global  
8 temperature increase due to aerosol emissions reduction alone ranges from 0.72 to 1.04 K in  
9 the standard CM3 with RCPs, compared to 2005 temperature levels. This is a significant  
10 fraction of the *total* temperature increase by 2100 (including greenhouse gas induced  
11 warming) of 1-5 K across the RCPs (compare dashed and solid lines in Fig. 4). However, in  
12 the reduced-aerosol-forcing run with RCP8.5 (CM3w RCP8.5), the globally averaged  
13 temperature increase due to aerosol decreases is only about 0.5 K, half of that in RCP8.5 with  
14 standard CM3. This result indicates that the projected climate response to aerosol decreases  
15 throughout the 21<sup>st</sup> century depends strongly on the magnitude of the present-day aerosol  
16 ERF, as a weaker ERF ( $\sim -1 \text{ W m}^{-2}$ ) can cut the global temperature response in half. This is  
17 discussed in more detail in Sect. 4.1.3 and shown in Figs. ~~87~~ and 8. The impact of ~~the RCP2.6~~  
18 aggressive phase-out of coal as an energy source in RCP2.6 can be seen from about 2020-  
19 2050 with a strong increase in aerosol driven temperature change. Likewise, the mid-century  
20 rise in coal use in RCP6 shows up as a decline in what is an otherwise consistent temperature  
21 increase throughout the century (Fig. 5). RCP4.5 and RCP8.5, on the other hand, have a  
22 steadier temperature increase that lacks the same noticeable features. There is a significant  
23 amount of overlap in climate response from decreasing aerosols among the RCPs throughout  
24 the entire century, especially considering the full range of the ensemble members of each  
25 simulation. This suggests that despite large differences in many facets of the individual RCPs  
26 (e.g., climate mitigation policies, emissions trajectories, land use), the temperature response to  
27 decreasing aerosols is relatively homogenous.

28 Although global precipitation rate (Fig. 5 upper right) is mainly controlled by the tropospheric  
29 energy balance (Ming et al., 2010), precipitation is linked to aerosols on the local scale, since  
30 aerosols serve as seeds for cloud droplet formation and the number and size of the cloud  
31 droplets influence ~~the precipitation rate~~the distribution and timing of precipitation.  
32 Additionally, aerosols can impact precipitation in other ways, including by changing  
33 atmospheric dynamics and circulation patterns, changing atmospheric heating rates, and

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1 changing the surface energy balance. We find increases of 0.08 – 0.09 mm d<sup>-1</sup> (up to 3% of  
2 2005 precipitation levels) are projected to result from the aerosol decrease in the four RCPs  
3 run with standard CM3, again representing a very narrow range of climate endpoints despite  
4 the differences among the scenarios. ~~The estimated increase of precipitation is reduced to~~  
5 ~~only about 0.04 mm d<sup>-1</sup> increase by the end of the 21<sup>st</sup> century in the CM3w RCP8.5 runs~~  
6 ~~with weaker present-day aerosol ERF. Nonetheless, s~~Since the total precipitation rate increase  
7 from all forcings ranges from 0.11 – 0.25 mm d<sup>-1</sup> (Figure 4), these aerosol driven changes are  
8 significant (discussed more in Sect. 4.1.3 and shown in Figs. 87 and 8). ~~This sensitivity of~~  
9 ~~precipitation to aerosols is consistent with the finding by Shindell et al. (2012) that~~  
10 ~~precipitation responds more strongly to aerosol forcing than an equivalent CO<sub>2</sub> forcing.~~

11 Liquid water path differences over time are shown for each of the RCPs in the lower left panel  
12 of Fig. 5. As aerosol concentrations decrease, LWP also decreases; in other words, aerosols  
13 and LWP are positively correlated. This is essentially the cloud lifetime effect acting in the  
14 opposite direction: increased aerosols cause cloud droplet concentrations to increase leading  
15 to a decrease in the autoconversion rate, which hinders precipitation formation and increases  
16 cloud lifetime and cloud liquid water path (Albrecht, 1989). The decline in aerosol emissions  
17 leads to a decrease in LWP in all of the standard CM3 runs with the RCPs, around 0.5-1.0 g  
18 m<sup>-2</sup> or 2% of 2005 levels. Accounting for the ensemble member range (shaded areas), the  
19 LWP decline in each of the RCPs is remarkably similar. As is the case with radiative forcing,  
20 temperature, and precipitation, the annual trends in the LWP values also follow the underlying  
21 RCP energy use trajectories. In particular, a rebound around 2040 in LWP in RCP6 can be  
22 seen in the bottom left of Fig. 5, analogous to the temperature decrease in RCP6 due to an  
23 increase in coal energy usage rate and ultimately aerosol and precursor emissions. LWP  
24 decreases in CM3w RCP8.5 are remarkably smaller, only about 20-40% of the decreases  
25 standard CM3 RCP runs. With a lower autoconversion threshold, the amount of liquid water  
26 held in clouds is decreased, resulting in the smaller change in CM3w RCP8.5 shown in Fig.  
27 5c.

28 With decreasing aerosols, effective cloud drop radius may increase due to the loss of smaller-  
29 sized and more numerous anthropogenic ultrafine aerosol, leaving natural aerosols (such as  
30 sea spray, which in general are fewer in number and much coarser in size) to form cloud  
31 droplets. Across the four RCPs, the globally averaged increase in cloud drop effective radius  
32 due to decreasing aerosols ranges from 0.54 μm to 0.60 μm (Fig. 5, Table 3). The increase due

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1 to all forcings ranges from about 0.60 – 0.80  $\mu\text{m}$  (Fig. 4). As expected, the increases in cloud  
2 droplet effective radius due to aerosol reductions make up a large fraction of the all-forcing  
3 increases. Cloud droplet effective radius trends follow energy usage and emissions trends in  
4 each of the RCPs very closely, with the results from the individual RCPs being quite distinct  
5 from one another through most of the century before slightly converging around 2100. Cloud  
6 droplet radius does not appear to be significantly affected by the lower autoconversion  
7 parameterization, as the differences between RCP8.5 with standard CM3 and RCP8.5 with  
8 CM3w are minimal.

## 9 **4 Spatial distribution and regional analysis**

10 Emissions of aerosols and aerosol precursors are highly heterogeneous in space. As expected,  
11 the changes of AOD, radiative forcing, and climate change in response to projected future  
12 emission changes also exhibit strong spatial structure. In this section, we first examine the  
13 spatial distributions of the changes in AOD and climate response, and then consider the  
14 average responses over two key source regions, East Asia and North America. Judging by  
15 historical emissions over the last few years since the RCPs were developed, concentrations of  
16 GHG in the atmosphere are tracking at or even above the trajectory predicted by RCP8.5 (the  
17 highest emission scenario), so we will focus mainly on that scenario (Peters et al., 2012;  
18 Sanford et al., 2014).

### 19 **4.1 Spatial distributions**

#### 20 **4.1.1 Aerosol optical depth**

21 Figure 6 shows the RCP8.5 aerosol optical depth (550 nm) differences (RCP8.5 –  
22 RCP8.5\_FRCP8.5\_2005AER) for the end of the 21<sup>st</sup> century (2096-2100 average) for sulfate,  
23 BC, OC, and total AOD. Corresponding results for the other three RCPs can be found in Figs.  
24 S6-S8 of the supplemental information. Strong reductions in AOD are simulated over most  
25 continental regions, including North America, Europe, and Asia. This coincides with the  
26 strong  $\text{SO}_2$  emissions decreases projected by RCP8.5 (and all RCPs) for much of the world.  
27 East and South Asia have the largest and most widespread decreases in AOD for all species.  
28 However, there are a few regions in which emissions changes are projected to *increase* sulfate  
29 AOD by 2100. Africa has noticeable areas of sulfate AOD increase, which can be explained  
30 by RCP8.5 projected increases in emissions at a few locales in this region (see Fig. S5 of  
31 supplementary information). Parts of Africa are expected to industrialize and grow throughout



1 the rest of the century and utilize its natural fossil fuel resources (Masui et al., 2011; Riahi et  
2 al., 2007). Similarly, RCP8.5 projects sulfate AOD increases for Indonesia, a region that is  
3 also projected to industrialize and experience an increase in biomass burning, resulting in  
4 elevated emissions of SO<sub>2</sub>, BC, and OC. There are also a few areas of increase over the  
5 tropical Pacific and Atlantic oceans for OC and SO<sub>2</sub> which are not caused by emissions  
6 increases (see Fig. S5) and instead result from meteorological changes, specifically a decrease  
7 in effectiveness of wet deposition removal of sulfate and OC. Since precipitation is increasing  
8 along with sulfate and OC aerosol optical depth, a decrease in the efficacy of wet deposition is  
9 implied. Although we do not have wet deposition fluxes archived in the model output, we  
10 note that using the same model, Fang et al. (2011) confirmed this relationship between  
11 increasing precipitation and decreasing wet deposition removal effectiveness. Using an  
12 idealized soluble tracer, the authors found that as climate warms, wet deposition of soluble  
13 pollutants decreases due to the simulated decreases in large-scale precipitation ~~over land,~~  
14 ~~suggesting that global large scale precipitation changes are not a good indicator of the~~  
15 ~~changes in wet scavenging of soluble species~~frequency. In the future, moderate and light  
16 precipitation is projected to occur less frequently, whereas heavy precipitation occurs more  
17 frequently. Since wet deposition does not depend on strength of precipitation events (i.e. same  
18 wet deposition flux for heavy or light rain), the decrease in frequency of light precipitation  
19 events leads to less wet deposition flux in the future and thus increases in AOD (Fang et al.,  
20 2011).

21 Asian BC and OC AOD decreases by 2100 are much larger than those in North America or  
22 Europe (Fig. 6). Some increases are again projected in Africa. BC and OC biomass burning  
23 emissions on average in Africa are generally expected to maintain current levels or decline  
24 slightly; however, in this particular region of Africa (mostly in Democratic Republic of the  
25 Congo and the Central African Republic), biomass burning emissions of OC and BC are  
26 projected to rise (not shown, see Fig. S5 of supplementary information). In particular,  
27 combustion and biomass burning emissions in Africa are expected to increase rapidly in the  
28 near-term (Lioussé et al., 2014). The total AOD difference in 2100 for RCP8.5 (lower right of  
29 Fig. 6) is dominated by the sulfate AOD reductions over the continents (except for in parts of  
30 Africa and other developing regions).

#### 1 4.1.2 Climate response

2 The climate responses to the aerosol reductions in RCP8.5 are shown in Fig. 7 for the end of  
3 the century and in Figs. S9-S11 of the supplemental information for the other three RCPs. As  
4 with Fig. 6, Fig. 7 shows the spatial pattern of the difference between the time-varying aerosol  
5 simulations and the fixed aerosol simulations (RCP8.5 – ~~RCP8.5~~ RCP8.5 2005AER). Areas  
6 hatched with dots in each of the figures indicate statistically significant regions at the 95%  
7 confidence level according to Student's  $t$  test. For temperature (upper left), the impact of  
8 aerosol reductions is almost entirely a warming effect, as expected, and nearly all of these  
9 increases are statistically significant. There are large areas of temperature increase in 2100 for  
10 East Asia, which is consistent with the largest decreases in AOD and aerosol emissions in this  
11 region. However, much of the strongest temperature increases driven by reduced aerosol  
12 emissions are located in or near the Arctic, suggesting that temperature change is non-local  
13 and does not necessarily occur in the areas where emissions are changing. Shindell et al.  
14 (2010) also have reported strong temperature increases in the Arctic where forcing was small  
15 in a global multimodel study.

16 Precipitation rate increases due to decreased aerosol emissions are largest over the tropical  
17 oceans, although increases are also observed over continents. In particular, some statistically  
18 significant precipitation rate increases occur over Europe, Russia, and Southeast Asia.  
19 Increases over North America are generally ~~not~~ found to be statistically significant. However,  
20 much of the areas of increase over the oceans are statistically insignificant. Simulating  
21 precipitation, especially regional details, remains challenging for most models, including  
22 CM3 (Eden et al., 2012). In our results there are two intercontinental tropical convergence  
23 zones (ITCZ) near the equator, a common feature not only identified in CM3 but in other  
24 models as well (Lin, 2007). Both of the ITCZ show strong areas of precipitation enhancement  
25 despite not coinciding with aerosol decreases. The northward shift in the ITCZ is expected  
26 when removing a negative forcing in the northern hemisphere (i.e. aerosol decreases), but this  
27 does not appear to be statistically significant (Allen and Sherwood, 2010). Precipitation in the  
28 tropics is mainly a result of deep convection, and several studies have identified the effect of  
29 aerosols on deep convective circulation and precipitation (Bell et al., 2008; Lee, 2012;  
30 Rosenfeld et al., 2008). Using a cloud-system resolving model of a large-scale deep  
31 convective system, Lee (2011) found that perturbations of aerosols in one domain can have  
32 teleconnections to other domains, acting to enhance precipitation in one domain while

1 suppressing it in another through deep convection. Since the changes in precipitation are both  
2 positive and negative (mostly positive), and regions of precipitation increase that do coincide  
3 with aerosol decrease are few, it is likely that aerosols are affecting precipitation through other  
4 means than the aerosol indirect effect alone. The aerosols are also likely exerting an influence  
5 on atmospheric dynamics and weather patterns, causing large, non-uniform increases and  
6 decreases in the tropics. In particular, we postulate that the aerosol decreases in the  
7 continental domain are ~~having teleconnections~~teleconnected to deep convection, resulting in  
8 both precipitation increases and decreases, as demonstrated by Lee (2011).

9 Liquid water path changes (Fig. 7, lower left panel) are largest over East Asia, which again is  
10 consistent with the simulated aerosol changes. Much of Europe and eastern North America  
11 also have large LWP decreases that coincide with aerosol emission decreases. However, there  
12 are large LWP increases in the Arctic region, most of which are statistically significant. These  
13 increases ~~could be~~are most likely due to a feedback from the aerosol-driven temperature  
14 increase, ~~since warmer air can hold more moisture~~as well as an increase in melting sea ice,  
15 making more water available for evaporation. Cloud droplet radius increases universally  
16 across the globe, with statistically significant changes occurring in the Northern Hemisphere,  
17 which is also where most of the aerosol reductions occur (Fig. 7, lower right). The co-location  
18 of the large increases in cloud droplet effective radius with large decreases in aerosol optical  
19 depth is a strong signal of the aerosol indirect effect, specifically the cloud albedo effect (we  
20 will explicitly show this ~~with correlations~~ in Sect. 5). Since the impacts of aerosol reductions  
21 on both liquid water path and cloud droplet effective radius are significant over the oceans in  
22 addition to the continents, we can also conclude that the aerosol reduction impacts are not  
23 necessary localized to their area of emission. Additionally, the increases of cloud droplet  
24 radius over the oceans may be amplified due to the greater susceptibility of clouds in clean  
25 environments compared to more polluted conditions.

26 Figure 8 shows the climate response in 2096-2100 for CM3w\_RCP8.5. These responses are  
27 plotted on the same scale as Fig. 7 for comparison. For temperature, the pattern is quite  
28 similar, with the largest increases over the Arctic and the continents, and statistical  
29 significance nearly everywhere. However, the magnitude of the temperature increases are  
30 noticeably smaller, owing to the reduced aerosol radiative forcing in the CM3w\_RCP8.5 runs.  
31 Precipitation changes show a vastly different spatial pattern and slightly smaller magnitude in  
32 CM3w\_RCP8.5 compared to RCP8.5 standard CM3, and overall is slightly smaller in

1 magnitude. However, much of this difference is due to the lack of a strong positive  
2 precipitation anomaly near the equator in CM3w RCP8.5, which is not statistically significant  
3 in either set of simulations. Over the continents, the precipitation change is more qualitatively  
4 similar, for example over East and South Asia, where both sets of simulations show  
5 precipitation increases. CM3w RCP8.5 produces a much larger precipitation increase over  
6 South Asia, perhaps due to the lower autoconversion threshold, although without statistical  
7 significance and only one ensemble member for CM3w, it is not possible to comment on the  
8 robustness of this result. Liquid water path decreases across Eastern North America, Europe,  
9 and East Asia, consistent with areas where aerosol decreases are largest. Many of these  
10 decreases, however, lack statistical significance, and the magnitude of the change is much  
11 smaller than in standard CM3, as discussed previously. Cloud droplet effective radius changes  
12 are qualitatively similar, with large decreases over the continents. Compared to LWP,  
13 temperature, and precipitation, cloud droplet radius is least by the lowered autoconversion  
14 threshold (see also Fig. 9).

#### 15 4.1.3 Comparison of climate response driven by aerosol decreases and by all 16 forcings

17 Although the projected absolute climate response to decreasing aerosols is similar in all the  
18 RCP simulations, the magnitude of relative effects differs. By comparing our climate response  
19 to aerosol decreases ( $RCP_{x,x} - \text{RCP}_{x,x} - \text{FRCP}_{x,x} \text{ 2005AER}$ ) to the climate response from all  
20 forcings ( $RCP_{x,x}$ ), we can roughly assess the relative importance of aerosol decreases to the  
21 total changes in climate for each RCP. The nonlinear nature of the global climate system  
22 means that these aerosol ratios are not directly additive with other ratios (say, GHG-induced  
23 climate changes) and are not the “true contributions” per se. However, comparing the  
24 magnitudes of the aerosol-induced climate changes to the total climate changes is a useful  
25 framing exercise. In instances where the all-forcing response sign differs from the aerosol-  
26 driven response, we use the absolute value (i.e. magnitude only) of the all-forcing response for  
27 the ratio calculation. For example, we We project from 2006 to the end-of-century a 0.97 K  
28 warming from aerosol emissions reductions in RCP2.6 (Fig. 5, Table 3) compared with a 1.5  
29 K warming from all climate forcings together (Fig. 4), indicating that, under this scenario,  
30 two-thirds of the warming by 2100 would result from decreases in aerosol emissions. The  
31 RCP2.6 scenario indicates that even with aggressive reductions in the emissions of  
32 greenhouse gases, the projected reduction in aerosol emissions is likely to push the climate

1 near the 2 K warming frequently cited as constituting “dangerous anthropogenic interference  
2 with the climate system” (Meinshausen et al., 2009). Since RCP2.6 projects the least warming  
3 from greenhouse gases, we find that this scenario is relatively the most susceptible (i.e. the  
4 largest percent effect) to unmasked aerosol warming as well as aerosol-driven changes in  
5 precipitation, LWP, and cloud droplet effective radius.

6 However, since GHG emissions over the past decade have been well above RCP2.6 and even  
7 slightly above RCP8.5 (Peters et al., 2012; Sanford et al., 2014), using RCP2.6 as a  
8 benchmark for the aerosol fraction of future climate change may be misleading. Figures 4 and  
9 5 show that for RCP8.5, warming from aerosol reductions is roughly 1 K globally of a total  
10 warming of nearly 5 K, or around 20%. The RCP8.5 precipitation increase of  $0.09 \text{ mm d}^{-1}$  is  
11 about 36% of the all-forcing increase of  $\sim 0.25 \text{ mm d}^{-1}$ , while the globally averaged ratios for  
12 LWP and cloud droplet effective radius are 30% and 75%, respectively. The large aerosol  
13 fraction for cloud droplet radius is expected, since cloud droplet size is highly dependent on  
14 existing aerosols, perhaps to a greater extent than the other three climate parameters presented  
15 here. Thus, even considering the high-emissions, low-mitigation RCP8.5 scenario, aerosol  
16 reductions are still a surprisingly important player in future global and regional climate  
17 change. However, an important caveat remains that our total present day aerosol effective  
18 radiative forcing is on the higher end of the IPCC range and thus may bias our results. We  
19 quantify this potential bias and discuss further below.

20 Figure 9 summarizes the 2096-2100 five-year average climate responses to decreasing  
21 aerosols (red bars, labeled RCP8.5 – RCP8.5 2005AER) for each region and puts them in the  
22 context of the all-forcing (GHG and aerosols) results (white hatched bars, labeled RCP8.5). In  
23 the standard CM3 RCP runs, Figure 8 shows the spatial distribution for 2096-2100 five-year  
24 averages of the ratio of aerosol-driven climate response to total climate response. Surface  
25 temperature increases due to aerosols are a substantial fraction of the all-forcing warming,  
26 even in RCP8.5 which features the largest warming from long-lived greenhouse gases. Much  
27 of East Asia, Australia, and the Middle East have ratios above ~~30~~20% (seen in Fig. 9,  
28 comparing red bars with white hatched bars), indicating that the large aerosol decreases in  
29 these regions will contribute significantly to projected warming. The Arctic region (defined as  
30 everything above  $66^\circ \text{ N}$ ) actually has the largest temperature increase due to decreasing  
31 aerosols, consistent with the polar amplification phenomenon. However, because climate  
32 warming from greenhouse gases is also amplified in the Arctic, the ratio of aerosol-induced

1 warming to all-forcing warming is smaller than in many other regions. Even over the oceans  
2 (not shown), where anthropogenic aerosol abundances are low, we find their global decrease  
3 accounts for more than 10–20% of the all-forcing warming in these locations. Precipitation  
4 ratios are not as smoothly distributed as temperature. Ratios are near 100% over the extra-  
5 tropical Pacific Ocean and Indian Ocean, whereas ratios over most of the Southern and Arctic  
6 Oceans are near 0. Parts of East Asia and Europe, regions for which major aerosol decreases  
7 are expected, have high ratios precipitation approaching contributions approaching 100%  
8 (red and white bars the upper right panel of Fig 9); however, this is not generalizable to all  
9 continental regions. Like precipitation, LWP ratios are close to 0 contributions are small near  
10 the poles (see Arctic region), with sporadic regions of large LWP ratios contributions in Asia,  
11 Australia, and the Middle East, indicating larger aerosol influence compared with all-forcings  
12 in these regions. Finally, ratios contributions of aerosol decreases to total for cloud droplet  
13 radius changes approach 100% for much of the globe between the latitudes 30° S and  
14 60°N. Ratios are particularly low in the Southern Ocean, where natural aerosols such as sea  
15 salt likely dominate CCN activation, and thus are less affected by decreasing anthropogenic  
16 aerosols.

17 Figure 9 also shows the comparison of aerosol decrease-driven climate response to all-forcing  
18 climate response for the CM3w RCP8.5 runs with weaker present-day aerosol ERF (yellow  
19 bars in Fig. 9, labeled CM3w RCP8.5 – CM3w RCP8.5\_2005AER). With some exceptions  
20 for precipitation in Asia, the climate responses to decreasing aerosols are always significantly  
21 less in CM3w RCP8.5 than in the standard RCP8.5 runs. This is particularly noticeable in the  
22 temperature and LWP panels of Fig. 9. The temperature response in CM3w RCP8.5 is  
23 sometimes half of the standard CM3 response, and the LWP is often an even smaller fraction.  
24 Thus, whereas RCP8.5 aerosol decreases may contribute up to 30-40% of the total warming in  
25 East Asia, this ratio is closer to 10% with the weakened aerosol forcing. While both versions  
26 of the model result in aerosol ERF within IPCC ranges, CM3w RCP8.5 simulates a present-  
27 day ERF much closer to the center of the uncertainty range, suggesting that this estimate of  
28 aerosol decrease-driven climate response may be more robust. Figure 11 summarizes the  
29 2096–2100 five year average climate responses to decreasing aerosols (blue bars, labeled  
30 AER) for each region. As previously discussed, many of the largest changes can be found in  
31 East Asia, especially for temperature (upper left), LWP (lower left), and cloud droplet radius  
32 (lower right). The Arctic region (defined as everything above 66° N) actually has the largest  
33 temperature increase due to decreasing aerosols, consistent with the polar amplification

1 phenomenon. However, because climate warming from greenhouse gases is also amplified in  
2 the Arctic, the ratio of aerosol induced warming to all forcing warming is smaller than in  
3 many other regions. Similar to Fig. 8, the AER and ALL bars allow for a comparison of the  
4 aerosol reduction driven climate response to the all forcing climate response on a regional  
5 basis. Across most of the regions, changes in cloud droplet effective radius are most  
6 susceptible to aerosol changes, followed by temperature, precipitation, and LWP.

## 8 **4.2 Regional climate response**

9 Using the regions defined in Fig. 1, we quantify changes in AOD, radiative forcing, and  
10 climate responses due to changes in aerosol concentrations on a regional scale in Tables 4 and  
11 5. We present the effective radiative forcing (or flux perturbation) as a difference between the  
12 2096-2100 value and the 2000-2004 levels (as before), which is why these values are mostly  
13 positive. The AOD and climate differences are the difference between the time-varying RCP  
14 simulations and the 2005 fixed aerosol simulations ( $RCP_{x,x} - RCP_{x,x} - FRCP_{x,x} 2005AER$ )  
15 for the end of century (2096-2100 5-year average). Values for RCP8.5 are shown in Table 4;  
16 RCP2.6 is shown in Table 5. These are chosen as the upper and lower ranges of AOD,  
17 radiative forcing, and climate response changes. Identical tables for RCP4.5 and RCP6 can be  
18 found in supplemental information (Tables S1 and S2). Bolded values in the tables represent  
19 the largest regional change for each quantity (e.g., largest  $SO_4$  AOD decrease). Table S3  
20 shows the regional changes in climate response for CM3w RCP8.5, although the AOD  
21 changes are not shown as we expect these to be minimal.

### 22 **4.2.1 East Asia**

23 Several of the largest aerosol-driven climate changes are found in the East Asian region. The  
24 region is defined to include all of China, Mongolia, Japan, and Korea (Fig. 1). East Asian  
25 emissions, AOD, radiative forcing, and climate response are analyzed below.

26 Figure S12 shows the anthropogenic  $SO_2$ , BC, and OC emissions time series from East Asia  
27 for the historical period and for each of the RCPs. For all species and all RCPs except RCP6,  
28 aerosol and aerosol precursor emissions are projected to peak in the 2010s. The trend for  
29 RCP6 for each species consists of a small increase in the 2010s, followed by a brief decrease  
30 and then a sharp increase with emissions peaking in 2050. This shifted trend results from the

1 primary energy supply projections in RCP6, in which coal energy usage in East Asia increases  
2 steadily until peaking in 2050-2060. This reliance on coal as a primary fuel source results in a  
3 mid-century peak in not only aerosol and precursor emissions, but also CO<sub>2</sub> emissions (Masui  
4 et al., 2011).

5 Figure S13 shows the East Asian region aerosol radiative forcing (calculated as a flux  
6 perturbation or effective radiative forcing) for all the RCPs and the historical period. This  
7 calculation is done by simply averaging over the region considering global aerosols as  
8 opposed to isolating the effect of East Asian aerosols alone. Radiative forcing from aerosols  
9 (direct + indirect) will continue to become more negative (larger in magnitude) until about  
10 2025, when it reaches nearly -5 W m<sup>-2</sup> over the region. For the rest of the century, the  
11 decrease in aerosol emissions (and AOD) results in a less negative radiative forcing (smaller  
12 in magnitude). Depending on the RCP, the aerosol forcing decreases in magnitude by about 2  
13 to 3 W m<sup>-2</sup> in 2100. RCP6 has the weakest recovery over most of the century. This is likely  
14 due to the increase in reliance on coal power, allowing emissions to increase and peak over  
15 the first half of the 21<sup>st</sup> century as mentioned above.

16 Climate response over East Asia is shown in Fig. 9-10 as differences between the time-  
17 varying RCP simulations and the fixed 2005 global aerosol emission simulations (RCPx.x –  
18 ~~RCPx.x - FRCPx.x 2005AER~~). Temperature increases driven by aerosol emission changes are  
19 much larger than the global average, ranging from 1.1 – 2.2. K of warming in the standard  
20 CM3 run. This is consistent with the large decrease in aerosol optical depth (-0.48 for sulfate,  
21 see Tables 4 and 5). In CM3w RCP8.5, the temperature increase is smaller than all of the  
22 standard RCPs by the end of the century. The temperature, precipitation, and especially cloud  
23 droplet effective radius all respond differently to the unique energy supply trajectory of RCP6  
24 than to the other RCPs. The warming around the middle of the century from aerosol emissions  
25 changes in RCP6 lags that in the other RCPs, and reaches only 1.1 K by 2100 (compared to  
26 2.2 K for RCP8.5). Similar trends exist for precipitation rate and cloud droplet effective  
27 radius, which lag strongly around 2050, a time period in which the rest of the RCPs steadily  
28 increase. The annual trends in the indirect forcing related quantities, LWP and cloud droplet  
29 effective radius, again bear a striking similarity to the emissions trends (Fig. S12) on the  
30 regional scale as well as the global scale, as we previously showed.



## 1 4.2.2 North America

2 The climate responses over North America to global emission changes are shown in Fig.  
3 [4011](#), and North American emissions and effective radiative forcing is shown in supplemental  
4 information Figs. S14 and S15. Unlike East Asia, anthropogenic emissions peak much earlier  
5 in North America. SO<sub>2</sub> emissions reached a maximum in roughly 1970 and have been  
6 declining steadily since then, with each of the RCPs projecting the decline to continue in a  
7 similar manner. Total aerosol forcing follows a similar qualitative pattern as in East Asia;  
8 however, aerosol forcing over North America is projected to range from about 0 to -0.5 W m<sup>-2</sup>  
9 from 2006 to 2100. Regionally averaged North American temperature increases by 1.5 to 2.0  
10 K across each of the [standard CM3 runs with the](#) RCPs, and no particular RCP stands out  
11 from another at the end of the century, especially considering the ensemble member range.  
12 [Again, temperature and precipitation response is much more muted in CM3w RCP8.5.](#)  
13 Precipitation increases by about 0.1 mm d<sup>-1</sup> over North America due to declining aerosol  
14 emissions. There is almost no change in LWP over North America, and a slight increase is  
15 even projected in RCP8.5 by the end of the century, due to the warming effect on LWP over  
16 the extreme northern regions of North America. [Despite having a major impact globally and](#)  
17 [in other regions, LWP changes are not noticeably different in CM3w RCP8.5 compared to](#)  
18 [the standard CM3 runs.](#) Cloud droplet radius increases nearly identically across the RCPs for  
19 the entire century, except for RCP2.6, which stands out as having the greatest effective radius  
20 increase for the first two-thirds of the 21<sup>st</sup> century. North American RCP2.6 SO<sub>2</sub> emissions  
21 (Fig. S14) and AOD (not shown) decrease substantially over the first half and level off for the  
22 remainder of the 21<sup>st</sup> century, which explains the trend in cloud droplet radius.

## 23 ~~4.2.3 Other regions~~

24 ~~Figure 11 summarizes the 2096-2100 five year average climate responses to decreasing~~  
25 ~~aerosols (blue bars, labeled AER) for each region. As previously discussed, many of the~~  
26 ~~largest changes can be found in East Asia, especially for temperature (upper left), LWP (lower~~  
27 ~~left), and cloud droplet radius (lower right). The Arctic region (defined as everything above~~  
28 ~~66° N) actually has the largest temperature increase due to decreasing aerosols, consistent~~  
29 ~~with the polar amplification phenomenon. However, because climate warming from~~  
30 ~~greenhouse gases is also amplified in the Arctic, the ratio of aerosol induced warming to all~~  
31 ~~forcing warming is smaller than in many other regions. Similar to Fig. 8, the AER and ALL~~  
32 ~~bars allow for a comparison of the aerosol reduction driven climate response to the all forcing~~

~~climate response on a regional basis. Across most of the regions, changes in cloud droplet effective radius are most susceptible to aerosol changes, followed by temperature, precipitation, and LWP.~~

## 5 Implications and discussion

### 5.1 Comparison to previous studies

Our estimated range (1.05 – 1.37 W m<sup>-2</sup>) for the effective radiative forcing change resulting from projected future aerosol and precursor emission reductions falls within the range estimated in previous literature, as shown in Table 1. In particular, Shindell et al. (2013) estimates a present-day to end-of-century aerosol forcing change of 0.68 – 1.42 W m<sup>-2</sup>. When considering our CM3w RCP8.5 simulations with lower present-day aerosol ERF, our lower estimate of 1.05 W m<sup>-2</sup> is actually much closer and in better agreement with the values in Shindell et al. (2013). Our global temperature increases from aerosol decreases across the RCPs of 0.72 – 1.04 K compares well with many previous studies (see Table 1), which have also found that temperatures will increase as much as 1 K due to aerosol decreases. Our global precipitation results also compare well with previous modeling studies. For instance, both Levy et al. (2013) and Rotstayn et al. (2013) found global increases in 2100 of 0.1 mm d<sup>-1</sup>, similar to the top of our projected range of 0.078 – 0.093 mm d<sup>-1</sup>. The effect of future aerosol decreases on liquid water path and cloud droplet effective radius were not considered by most of the previous literature, with the exception of Takemura (2012), who reports a change in LWP of around 3 g m<sup>-2</sup>, significantly higher than the 0.5-1.0 g m<sup>-2</sup> reported here. However, cloud droplet radius values compare much better. The aerosol-driven increases are very similar to those reported by Takemura (2012), who reported a difference in effective radius of about 0.6 μm at the tropopause from 2000 to 2100., compared to 0.5-0.6 μm in our study. However, Takemura (2012) did not conduct fixed aerosol simulations (~~RCPx.x\_FRCPx.x\_2005AER~~) and thus did not include the effect of climate change on future aerosol concentrations.

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### 5.2 Correlations between aerosols and climate response

It remains difficult to attribute specific changes in climate parameters to changes in aerosol emissions or optical depth, due to the complexity of global coupled chemistry-climate models

1 and the complexity of the climate system as a whole. In Fig. 12 we plot spatial correlations  
2 between RCP8.5 emission-driven changes in aerosol optical depth values and four climate  
3 parameters as an attempt to relate an aerosol quantity with corresponding climate response  
4 endpoints. Correlation plots for the three other RCPs are in the supplemental information  
5 (Figs. S16-S18). We also plot  $\Delta C/\Delta AOD$  for RCP8.5 in Figure S19, where  $\Delta C$  represents the  
6 change from the beginning to end of the 21<sup>st</sup> century for a given climate response variable,  
7 and  $\Delta AOD$  for the change in AOD. For RCP8.5 AOD-temperature correlation, there is a  
8 strong relationship ( $r < -0.75$ ) over virtually all of the continental areas and the northern  
9 Atlantic and Pacific oceans, which is expected due to aerosols having a net cooling influence  
10 on climate via the direct and indirect effects. The weak correlations over the tropical Pacific  
11 correspond to AOD increases and less extreme temperature increases over these regions,  
12 whereas the Southern Ocean positive correlations are caused by increases in temperature and  
13 very small increases (due to DMS emissions increases) in sulfate AOD (supported by the  
14 weak correlation values). AOD increases are also projected in parts of the Arctic, some of  
15 which may be related to the Arctic haze phenomenon. Coupled with strong temperature  
16 increases in the Arctic, positive AOD-temperature correlations are projected.

17 Precipitation is shown in the upper-right and is correlated both positively and negatively with  
18 changes AOD (Fig. 12). Over East Asia and parts of Europe, AOD and precipitation are  
19 somewhat strongly anti-correlated ( $r < -0.5$ ), which is expected according to the aerosol  
20 indirect effect on clouds. However, the mixed representation between positively and  
21 negatively correlated regions, as well as the overall weak correlations, suggests that it is not  
22 the aerosol indirect effect alone that is the mechanism for the projected effect of aerosols on  
23 precipitation. Instead, this is likely a signal of aerosol impacts on circulation, dynamics, and  
24 weather patterns, which can have feedback effects on precipitation. As described in Sect.  
25 4.1.2, we believe that aerosol perturbations over the continents may have teleconnections to  
26 precipitation enhancements in deep-convective cloud systems, accounting for the positive  
27 correlation over the tropics. In Fig. S20~~19~~ we also correlate AOD perturbations with deep  
28 convective precipitation and find generally stronger negative correlations ( $r \sim -0.75$ ) over  
29 much of the northern hemisphere but roughly the same positive correlations over much of the  
30 oceans, including the tropics.

31 Liquid water path (LWP) is correlated positively ( $r > 0.5$ ) with AOD over nearly the entire  
32 globe, with notable exceptions in Alaska and northern Russia. Additionally, areas with the

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1 strongest change in AOD seem to have the strongest LWP changes as well, possibly  
2 implicating the aerosol indirect effect as the mechanism behind the correlation. Similarly,  
3 cloud droplet radius is very strongly anti-correlated almost throughout the entire globe ( $r >$   
4  $0.75$ ), with the exception of the Arctic, which is also affected by the aerosol-driven increase in  
5 temperature.

### 6 **5.3 Similarity among the RCPs**

7 Our results are of course dependent on the GFDL CM3 model as well as the emissions  
8 projections provided by each of the RCPs, which have limitations. First, since recent GHG  
9 emissions are tracking at or above RCP8.5 (Peters et al., 2012; Sanford et al., 2014), the high  
10 emissions case, the RCPs may be optimistic in their greenhouse gas emissions projections.  
11 Second, the individual RCP pathways are often very similar to one another on both the global  
12 and regional scales, especially for air pollutant emissions and related quantities (see Fig. 2).  
13 This leads to a limited spread in the possible climate responses which likely does not explore  
14 the breadth of possible technology and policy driven changes in future aerosol emissions. For  
15 example, the spread in precipitation rate response driven by decreased aerosol emissions  
16 across scenarios is only  $0.015 \text{ mm d}^{-1}$ . In almost every example, the trends of AOD, radiative  
17 forcing, and climate response in each of the RCPs does not substantially differ (Table 3,  
18 Figure 5). An exception to this is with the CM3w RCP8.5 runs, which does produce a  
19 significantly different and more unique climate response to decreasing aerosols in the 21<sup>st</sup>  
20 century. Despite vastly different energy supply scenarios and climate policies, it appears to be  
21 of little consequence which RCP is used to determine the impact of the declining aerosol  
22 emissions on climate in the standard CM3 runs. Regardless of the level of projected GHG  
23 emissions in the future, decreasing aerosol concentrations will likely result in substantially  
24 warmer temperatures (see Tables 3-5) and regionally varied changes in precipitation and other  
25 climate parameters. Finally, unlike the IPCC SRES inventories, the RCPs do not begin from  
26 narrative storylines of what energy, population, and economic conditions will be like in the  
27 future making it difficult to link an RCP with a particular development pathway. However,  
28 recent work has attempted to map IPCC SRES illustrative storylines onto each of the RCPs  
29 (van Vuuren and Carter, 2013).

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## 1 6 Conclusions

2 We present a global climate modeling study of the impact of the projected future decrease of  
3 aerosol emissions using the GFDL Climate Model 3 (CM3). Future emissions follow the  
4 Representative Concentration Pathway (RCP) projections, which consist of four emissions  
5 trajectories that are consistent with global average radiative forcing values in 2100 of  
6 approximately 2.6, 4.5, 6.0, and 8.5 W m<sup>-2</sup>, respectively. In response to the continuing human  
7 health cost of air pollution, each of the RCP scenarios projects that global emissions of  
8 aerosols and their precursors will decline as the result of policy decisions to improve air  
9 quality. However, aerosols also have a net cooling effect on climate that will be reduced (i.e.,  
10 warming) as the emissions decline. Aerosols also impact clouds and precipitation, the effects  
11 of which will also change under the projected diminishing emissions. We implement the RCP  
12 emissions inventories into CM3 and run two sets of simulations for each of the pathways: one  
13 in which aerosol and aerosol precursor emissions are allowed to vary as projected by the  
14 RCPs, and another in which they are fixed at 2005 levels. The sets of fixed emissions  
15 simulations serve as controls to analyze the changes in AOD, radiative forcing, and climate  
16 solely due to the projected emission reductions of aerosols and their precursors.

17 Changes in aerosol optical depth closely follow the emissions changes spatially and  
18 temporally. Regionally, there are large AOD decreases over the continents, especially in East  
19 and South Asia. A few localities exhibit increases in aerosol optical depth, in some cases this  
20 is due to natural sources of aerosol or feedbacks on precipitation and wet deposition. In other  
21 cases, the AOD increases are indicative of local emissions increases, which are expected to  
22 occur in parts of Central Africa and Indonesia.

23 The projected decreases in aerosol emissions and optical depth lead to a decrease in the  
24 magnitude of the aerosol direct and indirect radiative forcing. CM3 predicts an aerosol direct  
25 plus indirect radiative forcing of -1.8 W m<sup>-2</sup> from pre-industrial to present-day, which is close  
26 to the upper range of -1.9 W m<sup>-2</sup> used by the IPCC (Myhre et al., 2013). To test robustness of  
27 our results to the strength of aerosol forcing, we run additional simulations in which the  
28 autoconversion threshold is lowered, resulting in a present-day aerosol ERF of about -1.0 W  
29 m<sup>-2</sup>. By 2100, the aerosol forcing is projected to diminish to between -0.21 W m<sup>-2</sup> in RCP2.6  
30 and -0.53 W m<sup>-2</sup> in RCP8.5. Our results therefore indicate that a large positive forcing, greater  
31 than up to 1 W m<sup>-2</sup> or greater, will result from the projected decrease in aerosol and aerosol  
32 precursor emissions. This forcing has a strong impact on climate, warming temperatures by as

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1 | much as 1 K globally and up to 2-3 K regionally in the standard CM3 RCP runs, with a much  
2 | more modest response in the reduced aerosol forcing runs of 0.5 K globally. Aerosol  
3 | reduction-driven surface temperature response generally accounts for a large fraction of the  
4 | total all-forcing response. Ratios over East Asia can exceed 30% in RCP8.5, and are even  
5 | higher in the other scenarios that do not feature as much greenhouse gas-induced climate  
6 | warming. However, these ratios are closer to 10-20% in most regions and globally when we  
7 | consider the reduced aerosol forcing RCP8.5 run. Global precipitation rates are projected to  
8 | increase by as much as 3% of 2005 values or 0.1 mm d<sup>-1</sup>, with greater regional impacts.  
9 | Inconsistent with Levy et al. (2013), we found significant local precipitation changes co-  
10 | located with areas of large aerosol decrease (e.g., East Asia). On the global scale, aerosol  
11 | reduction-driven changes in AOD and climate response trajectories do not vary significantly  
12 | among the four RCPs, especially towards the end of the century, despite stemming from  
13 | nominally different scenarios. Mid-century variation in the climate response and radiative  
14 | forcing trajectories closely follows the aerosol and precursor emissions trajectories (and thus  
15 | the energy use trajectories), even for climate parameters such as liquid water path and cloud  
16 | droplet effective radius.

17 | Temperature, liquid water path, and cloud droplet effective radius are strongly correlated  
18 | spatially with aerosol optical depth changes (as would be expected from their  
19 | parameterizations), confirming the localized effect that aerosols have on climate forcing.  
20 | Precipitation and AOD changes were only weakly correlated, with even the sign of the  
21 | correlation varying spatially. This suggests that local changes in aerosol quantity are not  
22 | strongly tied to local precipitation events, but that large-scale changes in atmospheric  
23 | circulation instead play a role in the effect of aerosols on precipitation. Future work is needed  
24 | to identify the mechanisms behind the precipitation response to inhomogeneous changes in  
25 | aerosol concentrations.

26 | Our results indicate the need for a tighter coupling among climate and air pollution control  
27 | policies, in order to avoid unintended warming from air pollution reductions. To  
28 | simultaneously clean the air and avoid exacerbating global warming, more aggressive  
29 | greenhouse gas mitigation will be necessary to compensate for the unintended aerosol-driven  
30 | warming. Future work on reductions of specific emissions sources (e.g., activities with large  
31 | emissions of BC) that have beneficial impacts for both human health and climate will be  
32 | beneficial. Additional work is also needed to understand the connection between changes in

1 aerosol quantities and corresponding climate responses. Future scenario development that  
2 better replicates explicit policy and technology changes driving emission reductions of air  
3 pollutants will provide scenarios that have larger variations among them, which will allow a  
4 more thorough analysis of the impacts of various mitigation efforts. Finally, the impact of  
5 nitrate aerosols on future climate needs to be explored. Many current studies have omitted  
6 nitrate, but the combination of increasing ammonia emissions and decreasing sulfate in the  
7 future will likely promote nitrate formation. If nitrate does increase in significance, society  
8 might simply replace the lost sulfate forcing and human health impact with a similar impact  
9 from nitrate aerosol.

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#### 14 **Acknowledgements**

15 D. Westervelt was supported by a fellowship from the Science, Technology and  
16 Environmental Policy (STEP) program at the Woodrow Wilson School of Public and  
17 International Affairs at Princeton University.

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## References

- [Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E. and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing., \*Nature\*, 432\(7020\), 1014–7, doi:10.1038/nature03174, 2004.](#)
- [Albrecht: Aerosols, Cloud Microphysics, and Fractional Cloudiness, \*Science\*, 245\(4923\), 1227–1230, doi:10.1126/science.245.4923.1227, 1989.](#)
- [Allen, R. J. and S. C. Sherwood, 2010: The impact of natural versus anthropogenic aerosols on atmospheric circulation in the Community Atmosphere Model, \*Climate Dyn.\*, doi:10.1007/s00382-010-0898-8.](#)
- [Arneth, A., Unger, N., Kulmala, M. and Andreae, M. O.: Atmospheric science. Clean the air, heat the planet?, \*Science\*, 326\(5953\), 672–3, doi:10.1126/science.1181568, 2009.](#)
- [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.](#)
- [Bell, T. L., Rosenfeld, D., Kim, K.-M., Yoo, J.-M., Lee, M.-I. and Hahnenberger, M.: Midweek increase in U.S. summer rain and storm heights suggests air pollution invigorates rainstorms, \*J. Geophys. Res.\*, 113\(D2\), D02209, doi:10.1029/2007JD008623, 2008.](#)



1 [Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J. and Boucher, O.: Aerosol forcing in](#)  
2 [the Climate Model Intercomparison Project \(CMIP5\) simulations by HadGEM2-ES and the](#)  
3 [role of ammonium nitrate, J. Geophys. Res., 116\(D20\), D20206, doi:10.1029/2011JD016074,](#)  
4 [2011.](#)

5 [Bollasina, M. A., Ming, Y., Ramaswamy, V., Schwarzkopf, M. D. and Naik, V.: Contribution](#)  
6 [of local and remote anthropogenic aerosols to the twentieth century weakening of the South](#)  
7 [Asian Monsoon, Geophys. Res. Lett., 41\(2\), 680–687, doi:10.1002/2013GL058183, 2014.](#)

8 [Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J.,](#)  
9 [Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim,](#)  
10 [M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N.,](#)  
11 [Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U.,](#)  
12 [Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the](#)  
13 [role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos.,](#)  
14 [118\(11\), 5380–5552, doi:10.1002/jgrd.50171, 2013.](#)

15 [Chalmers, N., Highwood, E. J., Hawkins, E., Sutton, R. and Wilcox, L. J.: Aerosol](#)  
16 [contribution to the rapid warming of near-term climate under RCP 2.6, Geophys. Res. Lett.,](#)  
17 [39\(18\), L18709, doi:10.1029/2012GL052848, 2012.](#)

18 [Chen, W.-T., Lee, Y. H., Adams, P. J., Nenes, A. and Seinfeld, J. H.: Will black carbon](#)  
19 [mitigation dampen aerosol indirect forcing?, Geophys. Res. Lett., 37\(9\), L09801,](#)  
20 [doi:10.1029/2010GL042886, 2010.](#)

21 [Dawson, J. P., Adams, P. J. and Pandis, S. N.: and Physics Sensitivity of PM 2.5 to climate in](#)  
22 [the Eastern US : a modeling case study, , 4295–4309, 2007.](#)

23 [Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S.,](#)  
24 [Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van](#)  
25 [der Werf, G. R. and Wilson, J.: Emissions of primary aerosol and precursor gases in the years](#)  
26 [2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6\(12\), 4321–4344,](#)  
27 [doi:10.5194/acp-6-4321-2006, 2006.](#)

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

37 [Eden, J. M., Widmann, M., Grawe, D. and Rast, S.: Skill, Correction, and Downscaling of](#)  
38 [GCM-Simulated Precipitation, J. Clim., 25\(11\), 3970–3984, doi:10.1175/JCLI-D-11-00254.1,](#)  
39 [2012.](#)

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

4 [Gillett, N. P. and Von Salzen, K.: The role of reduced aerosol precursor emissions in driving](#)  
5 [near-term warming, Environ. Res. Lett., 8\(3\), 034008, doi:10.1088/1748-9326/8/3/034008,](#)  
6 [2013.](#)

7 [Golaz, J.-C., Horowitz, L. W. and Levy, H.: Cloud tuning in a coupled climate model: Impact](#)  
8 [on 20th century warming, Geophys. Res. Lett., 40\(10\), 2246–2251, doi:10.1002/grl.50232,](#)  
9 [2013.](#)

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

14 [Griffies, S.M., Winton, M., Donner, L.J., Horowitz, L.W. Downes, S. M., Farneti, R.,](#)  
15 [Gnanadesikan, A., Hurlin, W.J., Lee, H., Liang, Z., Palter, J.B., Samuels, B.L, Wittenberg,](#)  
16 [A.T., Wyman, B., Yin, J., and Zadeh, N. 2011: The GFDL CM3 Coupled Climate Model:](#)  
17 [Characteristics of the Ocean and Sea Ice Simulations. \*J. Climate\*, \*\*24\*\*, 3520–](#)  
18 [3544.doi: <http://dx.doi.org/10.1175/2011JCLI3964.1>](#)

19 [Gruebler, A. and Nakicenovic, N.: Emissions scenarios: a final response, Energy Environ.,](#)  
20 [15\(1\), 11–24 \[online\] Available from:](#)  
21 [https://www.etde.org/etdeweb/details\\_open.jsp?osti\\_id=20463875](https://www.etde.org/etdeweb/details_open.jsp?osti_id=20463875) (Accessed 5 June 2014),

22 [2004.](#)

23 [Guo, H., Golaz, J.-C., Donner, L. J., Ginoux, P. and Hemler, R. S.: Multivariate Probability](#)  
24 [Density Functions with Dynamics in the GFDL Atmospheric General Circulation Model:](#)  
25 [Global Tests, J. Clim., 27\(5\), 2087–2108, doi:10.1175/JCLI-D-13-00347.1, 2014.](#)

26 [Hauglustaine, D. A., Balkanski, Y., and Schulz, M.: A global model simulation of present and](#)  
27 [future nitrate aerosols and their direct radiative forcing of climate, Atmos. Chem. Phys., 14,](#)  
28 [11031-11063, doi:10.5194/acp-14-11031-2014, 2014.](#)

29 [Heald, C. L., Henze, D. K., Horowitz, L. W., Feddema, J., Lamarque, J.-F., Guenther, A.,](#)  
30 [Hess, P. G., Vitt, F., Seinfeld, J. H., Goldstein, A. H. and Fung, I.: Predicted change in global](#)  
31 [secondary organic aerosol concentrations in response to future climate, emissions, and land](#)  
32 [use change, J. Geophys. Res. Atmos., 113\(D5\), D05211, doi:10.1029/2007JD009092, 2008.](#)

33 [Horowitz, L. W.: Past, present, and future concentrations of tropospheric ozone and aerosols:](#)  
34 [Methodology, ozone evaluation, and sensitivity to aerosol wet removal, J. Geophys. Res.,](#)  
35 [111\(D22\), D22211, doi:10.1029/2005JD006937, 2006.](#)

36 [Horowitz, L. W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie,](#)  
37 [X., Lamarque, J.-F., Schultz, M. G., Tyndall, G. S., Orlando, J. J. and Brasseur, G. P.: A](#)  
38 [global simulation of tropospheric ozone and related tracers: Description and evaluation of](#)

1 [MOZART, version 2, J. Geophys. Res. Atmos., 108\(D24\), 4784, doi:10.1029/2002JD002853,](#)  
2 [2003.](#)

3 [Jacob, D. J. and Winner, D. a.: Effect of climate change on air quality, Atmos. Environ.,](#)  
4 [43\(1\), 51–63, doi:10.1016/j.atmosenv.2008.09.051, 2009.](#)

5 [Kato, E., Kawamiya, M., Kinoshita, T. and Ito, A.: Development of spatially explicit emission](#)  
6 [scenario from land-use change and biomass burning for the input data of climate projection,](#)  
7 [Procedia Environ. Sci., 6, 146–152, doi:10.1016/j.proenv.2011.05.015, 2011.](#)

8 [Khairoutdinov, M. and Kogan, Y.: A New Cloud Physics Parameterization in a Large-Eddy](#)  
9 [Simulation Model of Marine Stratocumulus, Mon. Weather Rev., 128\(1\), 229–243,](#)  
10 [doi:10.1175/1520-0493\(2000\)128<0229:ANCPPI>2.0.CO;2, 2000.](#)

11 [King, M. D., Menzel, W. P., Kaufman, Y. J., Tanré, D., Gao, B., Platnick, S., Ackerman, S.](#)  
12 [A., Remer, L. A., Pincus, R. and Hubanks, P. A.: Cloud and Aerosol Properties , Precipitable](#)  
13 [Water , and Profiles of Temperature and Water Vapor from MODIS, , 41\(2\), 442–458, 2003.](#)

14 [Klimont, Z., Smith, S. J. and Cofala, J.: The last decade of global anthropogenic sulfur](#)  
15 [dioxide: 2000–2011 emissions, Environ. Res. Lett., 8\(1\), 014003, doi:10.1088/1748-](#)  
16 [9326/8/1/014003, 2013.](#)

17 [Kloster, S., Dentener, F., Feichter, J., Raes, F., Lohmann, U., Roeckner, E. and Fischer-Bruns,](#)  
18 [I.: A GCM study of future climate response to aerosol pollution reductions, Clim. Dyn., 34\(7-](#)  
19 [8\), 1177–1194, doi:10.1007/s00382-009-0573-0, 2009.](#)

20 [Kopp, R. E. and Mauzerall, D. L.: Assessing the climatic benefits of black carbon mitigation.,](#)  
21 [Proc. Natl. Acad. Sci. U. S. A., 107\(26\), 11703–8, doi:10.1073/pnas.0909605107, 2010.](#)

22 [Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, a., Klimont, Z., Lee, D., Liousse,](#)  
23 [C., Mieville, a., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van](#)  
24 [Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi,](#)  
25 [K. and van Vuuren, D. P.: Historical \(1850–2000\) gridded anthropogenic and biomass](#)  
26 [burning emissions of reactive gases and aerosols: methodology and application, Atmos.](#)  
27 [Chem. Phys., 10\(15\), 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.](#)

28 [Lamarque, J.-F., Kyle, G. P., Meinshausen, M., Riahi, K., Smith, S. J., Vuuren, D. P., Conley,](#)  
29 [A. J. and Vitt, F.: Global and regional evolution of short-lived radiatively-active gases and](#)  
30 [aerosols in the Representative Concentration Pathways, Clim. Change, 109\(1-2\), 191–212,](#)  
31 [doi:10.1007/s10584-011-0155-0, 2011.](#)

32 [Lee, S. S.: Effect of Aerosol on Circulations and Precipitation in Deep Convective Clouds, J.](#)  
33 [Atmos. Sci., 69\(6\), 1957–1974, doi:10.1175/JAS-D-11-0111.1, 2012.](#)

34 [Leibensperger, E. M., Mickley, L. J., Jacob, D. J., Chen, W.-T., Seinfeld, J. H., Nenes, A.,](#)  
35 [Adams, P. J., Streets, D. G., Kumar, N., and Rind, D.: Climatic effects of 1950–2050 changes](#)  
36 [in US anthropogenic aerosols – Part 1: Aerosol trends and radiative forcing, Atmos. Chem.](#)  
37 [Phys., 12, 3333-3348, doi:10.5194/acp-12-3333-2012, 2012](#)

- 1 [Leibensperger, E. M., Mickley, L. J., Jacob, D. J., Chen, W.-T., Seinfeld, J. H., Nenes, a.,](#)  
2 [Adams, P. J., Streets, D. G., Kumar, N. and Rind, D.: Climatic effects of 1950–2050 changes](#)  
3 [in US anthropogenic aerosols – Part 2: Climate response, Atmos. Chem. Phys., 12\(7\), 3349–](#)  
4 [3362, doi:10.5194/acp-12-3349-2012, 2012.](#)
- 5 [Levy, H., Horowitz, L. W., Schwarzkopf, M. D., Ming, Y., Golaz, J.-C., Naik, V. and](#)  
6 [Ramaswamy, V.: The roles of aerosol direct and indirect effects in past and future climate](#)  
7 [change, J. Geophys. Res. Atmos., 118\(10\), 4521–4532, doi:10.1002/jgrd.50192, 2013.](#)
- 8 [Lin, J.-L.: The Double-ITCZ Problem in IPCC AR4 Coupled GCMs: Ocean–Atmosphere](#)  
9 [Feedback Analysis, J. Clim., 20\(18\), 4497–4525, doi:10.1175/JCLI4272.1, 2007.](#)
- 10 [Lin, S. J. and Rood, R. B.: Multidimensional flux-form semiLagrangian transport schemes,](#)  
11 [Mon. Weather Rev., 124, 2046– 2070, 1996.](#)
- 12 [Lioussé, C., Assamoi, E., Criqui, P., Granier, C. and Rosset, R.: Explosive growth in African](#)  
13 [combustion emissions from 2005 to 2030, Environ. Res. Lett., 9\(3\), 035003,](#)  
14 [doi:10.1088/1748-9326/9/3/035003, 2014.](#)
- 15 [Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys.,](#)  
16 [5\(3\), 715–737, doi:10.5194/acp-5-715-2005, 2005.](#)
- 17 [Lohmann, U., Rotstayn, L., Storelvmo, T., Jones, a., Menon, S., Quaas, J., Ekman, a. M. L.,](#)  
18 [Koch, D. and Ruedy, R.: Total aerosol effect: radiative forcing or radiative flux perturbation?,](#)  
19 [Atmos. Chem. Phys., 10\(7\), 3235–3246, doi:10.5194/acp-10-3235-2010, 2010.](#)
- 20 [Manne, A. S., Richels, R. G. and Edmonds, J. A.: Market Exchange Rates Or Purchasing](#)  
21 [Power Parity: Does The Choice Make A Difference To The Climate Debate?, Clim. Change,](#)  
22 [71\(1-2\), 1–8, doi:10.1007/s10584-005-0470-4, 2005.](#)
- 23 [Makkonen, R., Asmi, A., Kerminen, V.-M., Boy, M., Arneth, A., Guenther, A., and Kulmala,](#)  
24 [M.: BVOC-aerosol-climate interactions in the global aerosol-climate model ECHAM5.5-](#)  
25 [HAM2, Atmos. Chem. Phys., 12, 10077-10096, doi:10.5194/acp-12-10077-2012, 2012.](#)
- 26 [Masui, T., Matsumoto, K., Hijioka, Y., Kinoshita, T., Nozawa, T., Ishiwatari, S., Kato, E.,](#)  
27 [Shukla, P. R., Yamagata, Y. and Kainuma, M.: An emission pathway for stabilization at](#)  
28 [6 Wm<sup>-2</sup> radiative forcing, Clim. Change, 109\(1-2\), 59–76, doi:10.1007/s10584-011-0150-5,](#)  
29 [2011.](#)
- 30 [Meinshausen, M., Meinshausen, N., Hare, W., Raper, S. C. B., Frieler, K., Knutti, R., Frame,](#)  
31 [D. J. and Allen, M. R.: Greenhouse-gas emission targets for limiting global warming to 2](#)  
32 [degrees C., Nature, 458\(7242\), 1158–62, doi:10.1038/nature08017, 2009.](#)
- 33 [Meinshausen, M., Smith, S. J., Calvin, K., Daniel, J. S., Kainuma, M. L. T., Lamarque, J.-F.,](#)  
34 [Matsumoto, K., Montzka, S. A., Raper, S. C. B., Riahi, K., Thomson, A., Velders, G. J. M.](#)  
35 [and Vuuren, D. P. P.: The RCP greenhouse gas concentrations and their extensions from 1765](#)  
36 [to 2300, Clim. Change, 109\(1-2\), 213–241, doi:10.1007/s10584-011-0156-z, 2011.](#)

1 [Menon, S., Hansen, J., Nazarenko, L. and Luo, Y.: Climate effects of black carbon aerosols in](#)  
2 [China and India., Science, 297\(5590\), 2250–3, doi:10.1126/science.1075159, 2002.](#)

3 [Menon, S., Unger, N., Koch, D., Francis, J., Garrett, T., Sednev, I., Shindell, D. and Streets,](#)  
4 [D.: Aerosol climate effects and air quality impacts from 1980 to 2030, Environ. Res. Lett.,](#)  
5 [3\(2\), 024004, doi:10.1088/1748-9326/3/2/024004, 2008.](#)

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

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

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

13 [Ming, Y., Ramaswamy, V., Donner, L. J. and Phillips, V. T. J.: A New Parameterization of](#)  
14 [Cloud Droplet Activation Applicable to General Circulation Models, J. Atmos. Sci., 63\(4\),](#)  
15 [1348–1356, doi:10.1175/JAS3686.1, 2006.](#)

16 [Ming, Y., Ramaswamy, V., Donner, L. J., Phillips, V. T. J., Klein, S. a., Ginoux, P. a. and](#)  
17 [Horowitz, L. W.: Modeling the Interactions between Aerosols and Liquid Water Clouds with](#)  
18 [a Self-Consistent Cloud Scheme in a General Circulation Model, J. Atmos. Sci., 64\(4\), 1189–](#)  
19 [1209, doi:10.1175/JAS3874.1, 2007.](#)

20 [Ming, Y., Ramaswamy, V. and Persad, G.: Two opposing effects of absorbing aerosols on](#)  
21 [global-mean precipitation, Geophys. Res. Lett., 37\(13\), L13701, doi:10.1029/2010GL042895,](#)  
22 [2010.](#)

23 [Myhre, G., D. Shindell, F.-M. Bréon, W. Collins, J. Fuglestedt, J. Huang, D. Koch, J.-F.](#)  
24 [Lamarque, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H.](#)  
25 [Zhang, 2013: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The](#)  
26 [Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of](#)  
27 [the Intergovernmental Panel on Climate Change \[Stocker, T.F., D. Qin, G.-K. Plattner,](#)  
28 [M.Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley \(eds.\)\].](#)  
29 [Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, pp.](#)  
30 [659–740, doi:10.1017/CBO9781107415324.018.](#)

31 [Naik, V., Horowitz, L. W., Fiore, A. M., Ginoux, P., Mao, J., Aghedo, A. M. and Levy, H.:](#)  
32 [Impact of preindustrial to present-day changes in short-lived pollutant emissions on](#)  
33 [atmospheric composition and climate forcing, J. Geophys. Res. Atmos., 118, 8086–8110,](#)  
34 [doi:10.1002/jgrd.50608, 2013.](#)

35 [Peters, G. P., Andrew, R. M., Boden, T., Canadell, J. G., Ciais, P., Le Quéré, C., Marland, G.,](#)  
36 [Raupach, M. R. and Wilson, C.: The challenge to keep global warming below 2 °C, Nat.](#)  
37 [Clim. Chang., 3\(1\), 4–6, doi:10.1038/nclimate1783, 2012.](#)

- 1 [Putnam, W.M., Lin, S.-J., 2007. Finite-volume transport on various cubed-sphere grids. \*J. Comput. Phys.\* 227 \(1\), 55–78.](#)
- 2
- 3 [Pye, H. O. T., Liao, H., Wu, S., Mickley, L. J., Jacob, D. J., Henze, D. K. and Seinfeld, J. H.: Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States. \*J. Geophys. Res.\*, 114\(D1\), D01205, doi:10.1029/2008JD010701, 2009.](#)
- 4
- 5
- 6 [Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, a., Lohmann, U., Bellouin, N., Boucher, O., Sayer, a. M., Thomas, G. E., McComiskey, a., Feingold, G., Hoose, C., Kristjánsson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. a., Stier, P., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, a., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F., Iacono, M. J., Kinne, S. and Schulz, M.: Aerosol indirect effects – general circulation model intercomparison and evaluation with satellite data. \*Atmos. Chem. Phys. Discuss.\*, 9\(3\), 12731–12779, doi:10.5194/acpd-9-12731-2009, 2009.](#)
- 7
- 8
- 9
- 10
- 11
- 12
- 13
- 14 [Radke, L. F., Coakley, J. A. and King, M. D.: Direct and remote sensing observations of the effects of ships on clouds., \*Science\*, 246\(4934\), 1146–9, doi:10.1126/science.246.4934.1146, 1989.](#)
- 15
- 16
- 17 [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.](#)
- 18
- 19 [Reddington, C. L., McMeeking, G., Mann, G. W., Coe, H., Frontoso, M. G., Liu, D., Flynn, M., Spracklen, D. V. and Carslaw, K. S.: The mass and number size distributions of black carbon aerosol over Europe, \*Atmos. Chem. Phys.\*, 13\(9\), 4917–4939, doi:10.5194/acp-13-4917-2013, 2013.](#)
- 20
- 21
- 22
- 23 [Riahi, K., Grübler, A. and Nakicenovic, N.: Scenarios of long-term socio-economic and environmental development under climate stabilization, \*Technol. Forecast. Soc. Change\*, 74\(7\), 887–935, doi:10.1016/j.techfore.2006.05.026, 2007.](#)
- 24
- 25
- 26 [Riahi, K., Rao, S., Krey, V., Cho, C., Chirkov, V., Fischer, G., Kindermann, G., Nakicenovic, N. and Rafaj, P.: RCP 8.5—A scenario of comparatively high greenhouse gas emissions, \*Clim. Change\*, 109\(1-2\), 33–57, doi:10.1007/s10584-011-0149-y, 2011.](#)
- 27
- 28
- 29 [Rogelj, J., Rao, S., Mccollum, D. L., Pachauri, S., Klimont, Z., Krey, V. and Riahi, K.: Air-pollution emission ranges consistent with the representative concentration pathways., 4\(May\), doi:10.1038/NCLIMATE2178, 2014.](#)
- 30
- 31
- 32 [Rosenfeld, D.: Suppression of Rain and Snow by Urban and Industrial Air Pollution, \*Science\*, 287\(5459\), 1793–1796, doi:10.1126/science.287.5459.1793, 2000.](#)
- 33
- 34 [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.](#)
- 35
- 36
- 37 [Rotstayn, L. D.: On the “tuning” of autoconversion parameterizations in climate models, \*J. Geophys. Res.\*, 105\(D12\), 15495, doi:10.1029/2000JD900129, 2000.](#)
- 38

- 1 [Rotstayn, L. D., Collier, M. A., Chrastansky, A., Jeffrey, S. J. and Luo, J.-J.: Projected effects](#)  
2 [of declining aerosols in RCP4.5: unmasking global warming?, Atmos. Chem. Phys., 13\(21\),](#)  
3 [10883–10905, doi:10.5194/acp-13-10883-2013, 2013.](#)
- 4 [Van Ruijven, B., Urban, F., Benders, R. M. J., Moll, H. C., van der Sluijs, J. P., de Vries, B.](#)  
5 [and van Vuuren, D. P.: Modeling Energy and Development: An Evaluation of Models and](#)  
6 [Concepts, World Dev., 36\(12\), 2801–2821, doi:10.1016/j.worlddev.2008.01.011, 2008.](#)
- 7 [Sanford, T., Frumhoff, P. C., Luers, A. and Gullede, J.: The climate policy narrative for a](#)  
8 [dangerously warming world, Nat. Clim. Chang., 4\(3\), 164–166, doi:10.1038/nclimate2148,](#)  
9 [2014.](#)
- 10 [Shindell, D., Schulz, M., Ming, Y., Takemura, T., Faluvegi, G. and Ramaswamy, V.: Spatial](#)  
11 [scales of climate response to inhomogeneous radiative forcing, J. Geophys. Res., 115\(D19\),](#)  
12 [D19110, doi:10.1029/2010JD014108, 2010.](#)
- 13 [Shindell, D., Kuylenstierna, J. C. I., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z.,](#)  
14 [Anenberg, S. C., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G.,](#)  
15 [Pozzoli, L., Kupiainen, K., Höglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V.,](#)  
16 [Hicks, K., Oanh, N. T. K., Milly, G., Williams, M., Demkine, V. and Fowler, D.:](#)  
17 [Simultaneously mitigating near-term climate change and improving human health and food](#)  
18 [security., Science, 335\(6065\), 183–9, doi:10.1126/science.1210026, 2012a.](#)
- 19 [Shindell, D. T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C., Chin, M., Young, P. J.,](#)  
20 [Lee, Y. H., Rotstayn, L., Mahowald, N., Milly, G., Faluvegi, G., Balkanski, Y., Collins, W. J.,](#)  
21 [Conley, A. J., Dalsoren, S., Easter, R., Ghan, S., Horowitz, L., Liu, X., Myhre, G.,](#)  
22 [Nagashima, T., Naik, V., Rumbold, S. T., Skeie, R., Sudo, K., Szopa, S., Takemura, T.,](#)  
23 [Voulgarakis, A., Yoon, J.-H. and Lo, F.: Radiative forcing in the ACCMIP historical and](#)  
24 [future climate simulations, Atmos. Chem. Phys., 13\(6\), 2939–2974, doi:10.5194/acp-13-2939-](#)  
25 [2013, 2013.](#)
- 26 [Shindell, D. T., Voulgarakis, A., Faluvegi, G. and Milly, G.: Precipitation response to regional](#)  
27 [radiative forcing, Atmos. Chem. Phys., 12\(15\), 6969–6982, doi:10.5194/acp-12-6969-2012,](#)  
28 [2012b.](#)
- 29 [Smith, S. J., Pitcher, H. and Wigley, T. M. L.: Future Sulfur Dioxide Emissions. Clim.](#)  
30 [Change. 73, 267-318, 2005.](#)
- 31 [Smith, S. J. and Bond, T. C.: Two hundred fifty years of aerosols and climate: the end of the](#)  
32 [age of aerosols, Atmos. Chem. Phys., 14\(2\), 537–549, doi:10.5194/acp-14-537-2014, 2014.](#)
- 33 [Song, F., Zhou, T. and Qian, Y.: Responses of East Asian summer monsoon to natural and](#)  
34 [anthropogenic forcings in the 17 latest CMIP5 models, Geophys. Res. Lett., 41\(2\), 596–603,](#)  
35 [doi:10.1002/2013GL058705, 2014.](#)
- 36 [Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a](#)  
37 [buffered system., Nature, 461\(7264\), 607–13, doi:10.1038/nature08281, 2009.](#)

1 [Tai, a. P. K., Mickley, L. J., Jacob, D. J., Leibensperger, E. M., Zhang, L., Fisher, J. a. and](#)  
2 [Pye, H. O. T.: Meteorological modes of variability for fine particulate matter \(PM<sub>2.5</sub>\) air](#)  
3 [quality in the United States: implications for PM<sub>2.5</sub> sensitivity to climate change, Atmos.](#)  
4 [Chem. Phys., 12\(6\), 3131–3145, doi:10.5194/acp-12-3131-2012, 2012.](#)

5 [Tai, A. P. K., Mickley, L. J. and Jacob, D. J.: Correlations between fine particulate matter](#)  
6 [\(PM<sub>2.5</sub>\) and meteorological variables in the United States: Implications for the sensitivity of](#)  
7 [PM<sub>2.5</sub> to climate change, Atmos. Environ., 44\(32\), 3976–3984,](#)  
8 [doi:10.1016/j.atmosenv.2010.06.060, 2010.](#)

9 [Takemura, T.: Distributions and climate effects of atmospheric aerosols from the preindustrial](#)  
10 [era to 2100 along Representative Concentration Pathways \(RCPs\) simulated using the global](#)  
11 [aerosol model SPRINTARS, Atmos. Chem. Phys., 12\(23\), 11555–11572, doi:10.5194/acp-12-](#)  
12 [11555-2012, 2012.](#)

13 [Thomson, A. M., Calvin, K. V., Smith, S. J., Kyle, G. P., Volke, A., Patel, P., Delgado-Arias,](#)  
14 [S., Bond-Lamberty, B., Wise, M. a., Clarke, L. E. and Edmonds, J. a.: RCP4.5: a pathway for](#)  
15 [stabilization of radiative forcing by 2100, Clim. Change, 109\(1-2\), 77–94,](#)  
16 [doi:10.1007/s10584-011-0151-4, 2011.](#)

17 [Tie, X.: Assessment of the global impact of aerosols on tropospheric oxidants, J. Geophys.](#)  
18 [Res., 110\(D3\), D03204, doi:10.1029/2004JD005359, 2005.](#)

19 [Twomey, S.: Pollution and the Planetary Albedo, Atmos. Env., 8\(12\), 1251–1256,](#)  
20 [http://dx.doi.org/10.1016/0004-6981\(74\)90004-3,1974.](#)

21 [Unger, N., Shindell, D. T. and Wang, J. S.: Climate forcing by the on-road transportation and](#)  
22 [power generation sectors, Atmos. Environ., 43\(19\), 3077–3085,](#)  
23 [doi:10.1016/j.atmosenv.2009.03.021, 2009.](#)

24 [Van Vuuren, D. P. and Carter, T. R.: Climate and socio-economic scenarios for climate](#)  
25 [change research and assessment: reconciling the new with the old, Clim. Change, 122\(3\),](#)  
26 [415–429, doi:10.1007/s10584-013-0974-2, 2013.](#)

27 [Van Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt,](#)  
28 [G. C., Kram, T., Krey, V., Lamarque, J.-F., Masui, T., Meinshausen, M., Nakicenovic, N.,](#)  
29 [Smith, S. J. and Rose, S. K.: The representative concentration pathways: an overview, Clim.](#)  
30 [Change, 109\(1-2\), 5–31, doi:10.1007/s10584-011-0148-z, 2011a.](#)

31 [Van Vuuren, D. P., Stehfest, E., Elzen, M. G. J., Kram, T., Vliet, J., Deetman, S., Isaac, M.,](#)  
32 [Klein Goldewijk, K., Hof, A., Mendoza Beltran, A., Oostenrijk, R. and Ruijven, B.: RCP2.6:](#)  
33 [exploring the possibility to keep global mean temperature increase below 2°C, Clim. Change,](#)  
34 [109\(1-2\), 95–116, doi:10.1007/s10584-011-0152-3, 2011b.](#)

35 [Van Vuuren, D. P., Riahi, K., Moss, R., Edmonds, J., Thomson, A., Nakicenovic, N., Kram,](#)  
36 [T., Berkhout, F., Swart, R., Janetos, A., Rose, S. K. and Arnell, N.: A proposal for a new](#)  
37 [scenario framework to support research and assessment in different climate research](#)  
38 [communities, Glob. Environ. Chang., 22\(1\), 21–35, doi:10.1016/j.gloenvcha.2011.08.002,](#)  
39 [2012.](#)



- 1 [World Health Organization. Special Report on Mortality from ambient and household air](#)  
2 [pollution in 2012 – summary of results. Programme in public health, social and environmental](#)  
3 [determinants of health \(PHE\). www.who.int/phe](#)
- 4
- 5 [Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E. and Toon, O. B.: The impact of humidity](#)  
6 [above stratiform clouds on indirect aerosol climate forcing., Nature, 432\(7020\), 1014–7,](#)  
7 [doi:10.1038/nature03174, 2004.](#)
- 8 [Albrecht: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science \(80-.\),](#)  
9 [245\(4923\), 1227–1230, doi:10.1126/science.245.4923.1227, 1989.](#)
- 10 [Allen, R. J. and Sherwood, S. C.: The impact of natural versus anthropogenic aerosols on](#)  
11 [atmospheric circulation in the Community Atmosphere Model, Clim. Dyn., 36\(9–10\), 1959–](#)  
12 [1978, doi:10.1007/s00382-010-0898-8, 2010.](#)
- 13 [Anon: WHO | Ambient and household air pollution and health, \[online\] Available from:](#)  
14 [http://www.who.int/phe/health\\_topics/outdoorair/databases/en/ \(Accessed 6 May 2014\), n.d.](#)
- 15 [Arneth, A., Unger, N., Kulmala, M. and Andreae, M. O.: Atmospheric science: Clean the air,](#)  
16 [heat the planet?, Science, 326\(5953\), 672–3, doi:10.1126/science.1181568, 2009.](#)
- 17 [Bauer, S. E., Koch, D., Unger, N., Metzger, S. M., Shindell, D. T. and Streets, D. G.: Nitrate](#)  
18 [aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone,](#)  
19 [Atmos. Chem. Phys., 7\(19\), 5043–5059, doi:10.5194/acp-7-5043-2007, 2007.](#)
- 20 [Bell, T. L., Rosenfeld, D., Kim, K. M., Yoo, J. M., Lee, M. I. and Hahnenberger, M.:](#)  
21 [Midweek increase in U.S. summer rain and storm heights suggests air pollution invigorates](#)  
22 [rainstorms, J. Geophys. Res., 113\(D2\), D02209, doi:10.1029/2007JD008623, 2008.](#)
- 23 [Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J. and Boucher, O.: Aerosol forcing in](#)  
24 [the Climate Model Intercomparison Project \(CMIP5\) simulations by HadGEM2-ES and the](#)  
25 [role of ammonium nitrate, J. Geophys. Res., 116\(D20\), D20206, doi:10.1029/2011JD016074,](#)  
26 [2011.](#)
- 27 [Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J.,](#)  
28 [Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim,](#)  
29 [M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N.,](#)  
30 [Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U.,](#)  
31 [Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S.: Bounding the](#)  
32 [role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos.,](#)  
33 [118\(11\), 5380–5552, doi:10.1002/jgrd.50171, 2013.](#)
- 34 [Chalmers, N., Highwood, E. J., Hawkins, E., Sutton, R. and Wilcox, L. J.: Aerosol](#)  
35 [contribution to the rapid warming of near-term climate under RCP 2.6, Geophys. Res. Lett.,](#)  
36 [39\(18\), n/a–n/a, doi:10.1029/2012GL052848, 2012.](#)

1 [Chen, W. T., Lee, Y. H., Adams, P. J., Nenes, A. and Seinfeld, J. H.: Will black carbon](#)  
2 [mitigation dampen aerosol indirect forcing?, \*Geophys. Res. Lett.\*, 37\(9\), n/a–n/a,](#)  
3 [doi:10.1029/2010GL042886, 2010.](#)

4 [Dawson, J. P., Adams, P. J. and Pandis, S. N.: and Physics Sensitivity of PM<sub>2.5</sub> to climate](#)  
5 [in the Eastern US : a modeling case study, , 4295–4309, 2007.](#)

6 [Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S.,](#)  
7 [Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J. P., Textor, C., Schulz, M., van](#)  
8 [der Werf, G. R. and Wilson, J.: Emissions of primary aerosol and precursor gases in the years](#)  
9 [2000 and 1750 prescribed data sets for AeroCom, \*Atmos. Chem. Phys.\*, 6\(12\), 4321–4344,](#)  
10 [doi:10.5194/acp-6-4321-2006, 2006.](#)

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

20 [Eden, J. M., Widmann, M., Grawe, D. and Rast, S.: Skill, Correction, and Downscaling of](#)  
21 [GCM-Simulated Precipitation, \*J. Clim.\*, 25\(11\), 3970–3984, doi:10.1175/JCLI-D-11-00254.1,](#)  
22 [2012.](#)

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

26 [Gillett, N. P. and Von Salzen, K.: The role of reduced aerosol precursor emissions in driving](#)  
27 [near-term warming, \*Environ. Res. Lett.\*, 8\(3\), 034008, doi:10.1088/1748-9326/8/3/034008,](#)  
28 [2013.](#)

29 [Golaz, J. C., Horowitz, L. W. and Levy, H.: Cloud-tuning in a coupled climate model: Impact](#)  
30 [on 20th-century warming, \*Geophys. Res. Lett.\*, 40\(10\), 2246–2251, doi:10.1002/grl.50232,](#)  
31 [2013.](#)

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

36 [Group, W.: WORKING-GROUP I CONTRIBUTION TO THE IPCC FIFTH ASSESSMENT](#)  
37 [REPORT CLIMATE CHANGE 2013 : THE PHYSICAL SCIENCE BASIS Final Draft](#)  
38 [Underlying Scientific-Technical Assessment A report accepted by Working-Group I of the](#)  
39 [IPCC but not approved in detail ., , \(January 2014\), 2013.](#)

1 Guo, H., Golaz, J.-C., Donner, L. J., Ginoux, P. and Hemler, R. S.: Multivariate Probability  
2 Density Functions with Dynamics in the GFDL Atmospheric General Circulation Model:  
3 Global Tests, *J. Clim.*, 27(5), 2087–2108, doi:10.1175/JCLI-D-13-00347.1, 2014.

4 Hauglustaine, D. A., Balkanski, Y. and Schulz, M.: A global model simulation of present and  
5 future nitrate aerosols and their direct radiative forcing of climate, *Atmos. Chem. Phys.*,  
6 14(20), 11031–11063, doi:10.5194/acp-14-11031-2014, 2014.

7 Heald, C. L., Henze, D. K., Horowitz, L. W., Feddesma, J., Lamarque, J. F., Guenther, A.,  
8 Hess, P. G., Vitt, F., Seinfeld, J. H., Goldstein, A. H. and Fung, I.: Predicted change in global  
9 secondary organic aerosol concentrations in response to future climate, emissions, and land  
10 use change, *J. Geophys. Res. Atmos.*, 113(D5), n/a–n/a, doi:10.1029/2007JD009092, 2008.

11 Horowitz, L. W.: Past, present, and future concentrations of tropospheric ozone and aerosols:  
12 Methodology, ozone evaluation, and sensitivity to aerosol wet removal, *J. Geophys. Res.*,  
13 111(D22), D22211, doi:10.1029/2005JD006937, 2006.

14 Horowitz, L. W., Walters, S., Mauzerall, D. L., Emmons, L. K., Rasch, P. J., Granier, C., Tie,  
15 X., Lamarque, J. F., Schultz, M. G., Tyndall, G. S., Orlando, J. J. and Brasseur, G. P.: A  
16 global simulation of tropospheric ozone and related tracers: Description and evaluation of  
17 MOZART, version 2, *J. Geophys. Res. Atmos.*, 108(D24), n/a–n/a,  
18 doi:10.1029/2002JD002853, 2003.

19 Jacob, D. J. and Winner, D. a.: Effect of climate change on air quality, *Atmos. Environ.*,  
20 43(1), 51–63, doi:10.1016/j.atmosenv.2008.09.051, 2009.

21 Kato, E., Kawamiya, M., Kinoshita, T. and Ito, A.: Development of spatially explicit emission  
22 scenario from land use change and biomass burning for the input data of climate projection,  
23 *Procedia Environ. Sci.*, 6, 146–152, doi:10.1016/j.proenv.2011.05.015, 2011.

24 Khairoutdinov, M. and Kogan, Y.: A New Cloud Physics Parameterization in a Large-Eddy  
25 Simulation Model of Marine Stratocumulus, *Mon. Weather Rev.*, 128(1), 229–243,  
26 doi:10.1175/1520-0493(2000)128<0229:ANCPPI>2.0.CO;2, 2000.

27 King, M. D., Menzel, W. P., Kaufman, Y. J., Tanré, D., Gao, B., Platnick, S., Ackerman, S.  
28 A., Remer, L. A., Pincus, R. and Hubanks, P. A.: Cloud and Aerosol Properties, Precipitable  
29 Water, and Profiles of Temperature and Water Vapor from MODIS, , 41(2), 442–458, 2003.

30 Klimont, Z., Smith, S. J. and Cofala, J.: The last decade of global anthropogenic sulfur  
31 dioxide: 2000–2011 emissions, *Environ. Res. Lett.*, 8(1), 014003, doi:10.1088/1748-  
32 9326/8/1/014003, 2013.

33 Kloster, S., Dentener, F., Feichter, J., Raes, F., Lohmann, U., Roeckner, E. and Fischer Bruns,  
34 I.: A GCM study of future climate response to aerosol pollution reductions, *Clim. Dyn.*, 34(7-  
35 8), 1177–1194, doi:10.1007/s00382-009-0573-0, 2009.

36 Kopp, R. E. and Mauzerall, D. L.: Assessing the climatic benefits of black carbon mitigation.,  
37 *Proc. Natl. Acad. Sci. U. S. A.*, 107(26), 11703–8, doi:10.1073/pnas.0909605107, 2010.

- 1 Lamarque, J. F., Bond, T. C., Eyring, V., Granier, C., Heil, a., Klimont, Z., Lee, D., Liousse,  
2 C., Mieville, a., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van  
3 Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi,  
4 K. and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass  
5 burning emissions of reactive gases and aerosols: methodology and application, *Atmos.*  
6 *Chem. Phys.*, 10(15), 7017–7039, doi:10.5194/aep-10-7017-2010, 2010.
- 7 Lamarque, J. F., Kyle, G. P., Meinshausen, M., Riahi, K., Smith, S. J., Vuuren, D. P., Conley,  
8 A. J. and Vitt, F.: Global and regional evolution of short-lived radiatively-active gases and  
9 aerosols in the Representative Concentration Pathways, *Clim. Change*, 109(1-2), 191–212,  
10 doi:10.1007/s10584-011-0155-0, 2011.
- 11 Lee, S. S.: Effect of Aerosol on Circulations and Precipitation in Deep Convective Clouds, *J.*  
12 *Atmos. Sci.*, 69(6), 1957–1974, doi:10.1175/JAS-D-11-0111.1, 2012.
- 13 Leibensperger, E. M., Mickley, L. J., Jacob, D. J., Chen, W. T., Seinfeld, J. H., Nenes, a.,  
14 Adams, P. J., Streets, D. G., Kumar, N. and Rind, D.: Climatic effects of 1950–2050 changes  
15 in US anthropogenic aerosols—Part 2: Climate response, *Atmos. Chem. Phys.*, 12(7), 3349–  
16 3362, doi:10.5194/aep-12-3349-2012, 2012a.
- 17 Leibensperger, E. M., Mickley, L. J., Jacob, D. J., Chen, W. T., Seinfeld, J. H., Nenes, A.,  
18 Adams, P. J., Streets, D. G., Kumar, N. and Rind, D.: Climatic effects of 1950–2050 changes  
19 in US anthropogenic aerosols—Part 1: Aerosol trends and radiative forcing, *Atmos. Chem.*  
20 *Phys.*, 12(7), 3333–3348, doi:10.5194/aep-12-3333-2012, 2012b.
- 21 Levy, H., Horowitz, L. W., Schwarzkopf, M. D., Ming, Y., Golaz, J. C., Naik, V. and  
22 Ramaswamy, V.: The roles of aerosol direct and indirect effects in past and future climate  
23 change, *J. Geophys. Res. Atmos.*, 118(10), 4521–4532, doi:10.1002/jgrd.50192, 2013.
- 24 Lin, J. L.: The Double-ITCZ Problem in IPCC AR4-Coupled GCMs: Ocean–Atmosphere  
25 Feedback Analysis, *J. Clim.*, 20(18), 4497–4525, doi:10.1175/JCLI4272.1, 2007.
- 26 Liousse, C., Assamoi, E., Criqui, P., Granier, C. and Rosset, R.: Explosive growth in African  
27 combustion emissions from 2005 to 2030, *Environ. Res. Lett.*, 9(3), 035003,  
28 doi:10.1088/1748-9326/9/3/035003, 2014.
- 29 Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmos. Chem. Phys.*,  
30 5(3), 715–737, doi:10.5194/aep-5-715-2005, 2005.
- 31 Lohmann, U., Rotstajn, L., Storelvmo, T., Jones, a., Menon, S., Quaas, J., Ekman, a. M. L.,  
32 Koch, D. and Ruedy, R.: Total aerosol effect: radiative forcing or radiative flux perturbation?,  
33 *Atmos. Chem. Phys.*, 10(7), 3235–3246, doi:10.5194/aep-10-3235-2010, 2010.
- 34 Makkonen, R., Asmi, a., Kerminen, V. M., Boy, M., Arneth, a., Hari, P. and Kulmala, M.: Air  
35 pollution control and decreasing new-particle formation lead to strong climate warming,  
36 *Atmos. Chem. Phys. Discuss.*, 11(9), 25991–26007, doi:10.5194/aepd-11-25991-2011, 2011.
- 37 Masui, T., Matsumoto, K., Hijioka, Y., Kinoshita, T., Nozawa, T., Ishiwatari, S., Kato, E.,  
38 Shukla, P. R., Yamagata, Y. and Kainuma, M.: An emission pathway for stabilization at

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

3 Meinshausen, M., Meinshausen, N., Hare, W., Raper, S. C. B., Frieler, K., Knutti, R., Frame,  
4 D. J. and Allen, M. R.: Greenhouse gas emission targets for limiting global warming to 2  
5 degrees C., *Nature*, 458(7242), 1158–62, doi:10.1038/nature08017, 2009.

6 Meinshausen, M., Smith, S. J., Calvin, K., Daniel, J. S., Kainuma, M. L. T., Lamarque, J. F.,  
7 Matsumoto, K., Montzka, S. A., Raper, S. C. B., Riahi, K., Thomson, A., Velders, G. J. M.  
8 and Vuuren, D. P. P.: The RCP greenhouse gas concentrations and their extensions from 1765  
9 to 2300, *Clim. Change*, 109(1-2), 213–241, doi:10.1007/s10584-011-0156-z, 2011.

10 Menon, S., Unger, N., Koch, D., Francis, J., Garrett, T., Sednev, I., Shindell, D. and Streets,  
11 D.: Aerosol climate effects and air quality impacts from 1980 to 2030, *Environ. Res. Lett.*,  
12 3(2), 024004, doi:10.1088/1748-9326/3/2/024004, 2008.

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

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

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

20 Ming, Y., Ramaswamy, V., Donner, L. J. and Phillips, V. T. J.: A New Parameterization of  
21 Cloud Droplet Activation Applicable to General Circulation Models, *J. Atmos. Sci.*, 63(4),  
22 1348–1356, doi:10.1175/JAS3686.1, 2006.

23 Ming, Y., Ramaswamy, V., Donner, L. J., Phillips, V. T. J., Klein, S. a., Ginoux, P. a. and  
24 Horowitz, L. W.: Modeling the Interactions between Aerosols and Liquid Water Clouds with  
25 a Self-Consistent Cloud Scheme in a General Circulation Model, *J. Atmos. Sci.*, 64(4), 1189–  
26 1209, doi:10.1175/JAS3874.1, 2007.

27 Ming, Y., Ramaswamy, V. and Persad, G.: Two opposing effects of absorbing aerosols on  
28 global-mean precipitation, *Geophys. Res. Lett.*, 37(13), n/a–n/a, doi:10.1029/2010GL042895,  
29 2010.

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

34 Peters, G. P., Andrew, R. M., Boden, T., Canadell, J. G., Ciais, P., Le Quéré, C., Marland, G.,  
35 Raupach, M. R. and Wilson, C.: The challenge to keep global warming below 2 °C, *Nat.*  
36 *Clim. Chang.*, 3(1), 4–6, doi:10.1038/nclimate1783, 2012.

1 Pye, H. O. T., Liao, H., Wu, S., Miekley, L. J., Jacob, D. J., Henze, D. K. and Seinfeld, J. H.:  
2 Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels  
3 in the United States, *J. Geophys. Res.*, 114(D1), D01205, doi:10.1029/2008JD010701, 2009.

4 Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, a.,  
5 Lohmann, U., Bellouin, N., Boucher, O., Sayer, a. M., Thomas, G. E., McComiskey, a.,  
6 Feingold, G., Hoose, C., Kristjánsson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P.  
7 a., Stier, P., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, a.,  
8 Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J.-F.,  
9 Iacono, M. J., Kinne, S. and Schulz, M.: Aerosol indirect effects—general circulation model  
10 intercomparison and evaluation with satellite data, *Atmos. Chem. Phys. Discuss.*, 9(3),  
11 12731–12779, doi:10.5194/acpd-9-12731-2009, 2009.

12 Radke, L. F., Coakley, J. A. and King, M. D.: Direct and remote sensing observations of the  
13 effects of ships on clouds., *Science*, 246(4934), 1146–9, doi:10.1126/science.246.4934.1146,  
14 1989.

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

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

19 Reddington, C. L., McMeeking, G., Mann, G. W., Coe, H., Frontoso, M. G., Liu, D., Flynn,  
20 M., Spracklen, D. V. and Carslaw, K. S.: The mass and number size distributions of black  
21 carbon aerosol over Europe, *Atmos. Chem. Phys.*, 13(9), 4917–4939, doi:10.5194/acp-13-  
22 4917-2013, 2013.

23 Riahi, K., Grübler, A. and Nakicenovic, N.: Scenarios of long-term socio-economic and  
24 environmental development under climate stabilization, *Technol. Forecast. Soc. Change*,  
25 74(7), 887–935, doi:10.1016/j.techfore.2006.05.026, 2007.

26 Riahi, K., Rao, S., Krey, V., Cho, C., Chirkov, V., Fischer, G., Kindermann, G., Nakicenovic,  
27 N. and Rafaj, P.: RCP 8.5—A scenario of comparatively high greenhouse gas emissions,  
28 *Clim. Change*, 109(1–2), 33–57, doi:10.1007/s10584-011-0149-y, 2011.

29 Rogelj, J., Rao, S., Mccollum, D. L., Pachauri, S., Klimont, Z., Krey, V. and Riahi, K.: Air-  
30 pollution emission ranges consistent with the representative concentration pathways.,  
31 4(May), doi:10.1038/NCLIMATE2178, 2014.

32 Rosenfeld, D.: Suppression of Rain and Snow by Urban and Industrial Air Pollution, *Science*  
33 (80–), 287(5459), 1793–1796, doi:10.1126/science.287.5459.1793, 2000.

34 Rosenfeld, D., Lohmann, U., Raga, G. B., O’Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell,  
35 A. and Andreae, M. O.: Flood or drought: how do aerosols affect precipitation?, *Science*,  
36 321(5894), 1309–13, doi:10.1126/science.1160606, 2008a.

1 Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell,  
2 A. and Andreae, M. O.: Flood or drought: how do aerosols affect precipitation?, *Science*,  
3 321(5894), 1309–13, doi:10.1126/science.1160606, 2008b.

4 Rotstayn, L. D.: On the “tuning” of autoconversion parameterizations in climate models, *J.*  
5 *Geophys. Res.*, 105(D12), 15495, doi:10.1029/2000JD900129, 2000.

6 Rotstayn, L. D., Collier, M. A., Chrastansky, A., Jeffrey, S. J. and Luo, J. J.: Projected effects  
7 of declining aerosols in RCP4.5: unmasking global warming?, *Atmos. Chem. Phys.*, 13(21),  
8 10883–10905, doi:10.5194/acp-13-10883-2013, 2013.

9 Van Ruijven, B., Urban, F., Benders, R. M. J., Moll, H. C., van der Sluijs, J. P., de Vries, B.  
10 and van Vuuren, D. P.: Modeling Energy and Development: An Evaluation of Models and  
11 Concepts, *World Dev.*, 36(12), 2801–2821, doi:10.1016/j.worlddev.2008.01.011, 2008.

12 Sanford, T., Frumhoff, P. C., Luers, A. and Gullede, J.: The climate policy narrative for a  
13 dangerously warming world, *Nat. Clim. Chang.*, 4(3), 164–166, doi:10.1038/nclimate2148,  
14 2014.

15 Shindell, D., Kuylensstierna, J. C. I., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z.,  
16 Anenberg, S. C., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G.,  
17 Pozzoli, L., Kupiainen, K., Höglund Isaksson, L., Emberson, L., Streets, D., Ramanathan, V.,  
18 Hicks, K., Oanh, N. T. K., Milly, G., Williams, M., Demkine, V. and Fowler, D.:  
19 Simultaneously mitigating near term climate change and improving human health and food  
20 security., *Science*, 335(6065), 183–9, doi:10.1126/science.1210026, 2012.

21 Shindell, D., Schulz, M., Ming, Y., Takemura, T., Faluvegi, G. and Ramaswamy, V.: Spatial  
22 scales of climate response to inhomogeneous radiative forcing, *J. Geophys. Res.*, 115(D19),  
23 D19110, doi:10.1029/2010JD014108, 2010.

24 Shindell, D. T., Lamarque, J. F., Schulz, M., Flanner, M., Jiao, C., Chin, M., Young, P. J.,  
25 Lee, Y. H., Rotstayn, L., Mahowald, N., Milly, G., Faluvegi, G., Balkanski, Y., Collins, W. J.,  
26 Conley, A. J., Dalsoren, S., Easter, R., Ghan, S., Horowitz, L., Liu, X., Myhre, G.,  
27 Nagashima, T., Naik, V., Rumbold, S. T., Skeie, R., Sudo, K., Szopa, S., Takemura, T.,  
28 Voulgarakis, A., Yoon, J. H. and Lo, F.: Radiative forcing in the ACCMIP historical and  
29 future climate simulations, *Atmos. Chem. Phys.*, 13(6), 2939–2974, doi:10.5194/acp-13-2939-  
30 2013, 2013.

31 Smith, S. J. and Bond, T. C.: Two hundred fifty years of aerosols and climate: the end of the  
32 age of aerosols, *Atmos. Chem. Phys.*, 14(2), 537–549, doi:10.5194/acp-14-537-2014, 2014.

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

35 Tai, a. P. K., Mickley, L. J., Jacob, D. J., Leibensperger, E. M., Zhang, L., Fisher, J. a. and  
36 Pye, H. O. T.: Meteorological modes of variability for fine particulate matter (PM<sub>2.5</sub>) air  
37 quality in the United States: implications for PM<sub>2.5</sub> sensitivity to climate change, *Atmos.*  
38 *Chem. Phys.*, 12(6), 3131–3145, doi:10.5194/acp-12-3131-2012, 2012.

- 1 Tai, A. P. K., Mickley, L. J. and Jacob, D. J.: Correlations between fine particulate matter  
2 (PM<sub>2.5</sub>) and meteorological variables in the United States: Implications for the sensitivity of  
3 PM<sub>2.5</sub> to climate change, *Atmos. Environ.*, 44(32), 3976–3984,  
4 doi:10.1016/j.atmosenv.2010.06.060, 2010.
- 5 Takemura, T.: Distributions and climate effects of atmospheric aerosols from the preindustrial  
6 era to 2100 along Representative Concentration Pathways (RCPs) simulated using the global  
7 aerosol model SPRINTARS, *Atmos. Chem. Phys.*, 12(23), 11555–11572, doi:10.5194/acp-12-  
8 11555-2012, 2012.
- 9 Thomson, A. M., Calvin, K. V., Smith, S. J., Kyle, G. P., Volke, A., Patel, P., Delgado-Arias,  
10 S., Bond-Lamberty, B., Wise, M. a., Clarke, L. E. and Edmonds, J. a.: RCP4.5: a pathway for  
11 stabilization of radiative forcing by 2100, *Clim. Change*, 109(1-2), 77–94,  
12 doi:10.1007/s10584-011-0151-4, 2011.
- 13 Tie, X.: Assessment of the global impact of aerosols on tropospheric oxidants, *J. Geophys.*  
14 *Res.*, 110(D3), D03204, doi:10.1029/2004JD005359, 2005.
- 15 Twomey, S. A.: Pollution and Cloud Albedo, *Eos T Am Geophys Un*, 58(8), 797–797 [online]  
16 Available from: //a1977dp36300943, 1977.
- 17 Unger, N., Shindell, D. T. and Wang, J. S.: Climate forcing by the on road transportation and  
18 power generation sectors, *Atmos. Environ.*, 43(19), 3077–3085,  
19 doi:10.1016/j.atmosenv.2009.03.021, 2009.
- 20 Vuuren, D. P. and Carter, T. R.: Climate and socio-economic scenarios for climate change  
21 research and assessment: reconciling the new with the old, *Clim. Change*, 122(3), 415–429,  
22 doi:10.1007/s10584-013-0974-2, 2013.
- 23 Vuuren, D. P., Edmonds, J., Kainuma, M., Riahi, K., Thomson, A., Hibbard, K., Hurtt, G. C.,  
24 Kram, T., Krey, V., Lamarque, J. F., Masui, T., Meinshausen, M., Nakicenovic, N., Smith, S.  
25 J. and Rose, S. K.: The representative concentration pathways: an overview, *Clim. Change*,  
26 109(1-2), 5–31, doi:10.1007/s10584-011-0148-z, 2011a.
- 27 Vuuren, D. P., Stehfest, E., Elzen, M. G. J., Kram, T., Vliet, J., Deetman, S., Isaac, M., Klein  
28 Goldewijk, K., Hof, A., Mendoza Beltran, A., Oostenrijk, R. and Ruijven, B.: RCP2.6:  
29 exploring the possibility to keep global mean temperature increase below 2°C, *Clim. Change*,  
30 109(1-2), 95–116, doi:10.1007/s10584-011-0152-3, 2011b.
- 31 Van Vuuren, D. P., Riahi, K., Moss, R., Edmonds, J., Thomson, A., Nakicenovic, N., Kram,  
32 T., Berkhout, F., Swart, R., Janetos, A., Rose, S. K. and Arnell, N.: A proposal for a new  
33 scenario framework to support research and assessment in different climate research  
34 communities, *Glob. Environ. Chang.*, 22(1), 21–35, doi:10.1016/j.gloenvcha.2011.08.002,  
35 2012.
- 36 World Health Organization. Special Report on Mortality from ambient and household air  
37 pollution in 2012—summary of results. Programme in public health, social and environmental  
38 determinants of health (PHE). [www.who.int/phe](http://www.who.int/phe)



1 Table 1: Summary of previous global climate modeling studies examining future climate effects of aerosols (only those using RCP  
 2 projections)

Work	Model	RCPs	Output	Domain	Aerosol	Results
Levy et al. (2013)	GFDL CM3	4.5	Emis., AOD, T, P, RF	Global, 1860-2100	Fixed 2005 emissions	1.26 W m <sup>-2</sup> , 1° C, 0.1 mm day <sup>-1</sup>
Shindell et al. (2013)	Multimodel	All	Emis, AOD, RF	Global, 1850-2100	Fixed for RF calc.	0.68 – 1.42 W m <sup>-2</sup>
Rotstayn et al. (2013)	CSIRO-Mk3.6	4.5	Emis, burden, AOD, RF, T, P	Global, 1850-2100	Fixed 2005 emissions	1.46 W m <sup>-2</sup> , 1.1° C, 0.1 mm day <sup>-1</sup>
Gillet et al. (2013)	CanESM2	2.6, 4.5, 8.5	Emis, burden, T	Global, 1850-2100	Fixed 2000 emissions	0.4 – 0.7° C
Chalmers et al. (2012)	HadGEM2-ES	2.6, 4.5	Burden, RF, AOD, CDNC, CLD, SW	Global, 2000-2037	Compare RCP2.6 and RCP4.5	See Chalmers et al. (2012)
Takemura (2012)	SPRINTARS	All	Emis, burden, AOD, RF, R <sub>eff</sub> , LWP, IWP	Global, regional, 1850-2100	Compare to 2000 levels	1.72 – 1.96 W m <sup>-2</sup>
Lamrque et al. (2011)	CAM5	All	Emis, burden, AOD, RF, O <sub>3</sub>	Global, regional, 1850-2100	Compare to 2000 levels	0.44 - 0.57 W m <sup>-2</sup>
Bellouin et al. (2011)	HadGEM2-ES	All	Emis, , AOD, RF	Global, 2000-2100	Compare to 2000 levels	~1 W m <sup>-2</sup>
This work	GFDL CM3	All	Emis, AOD, RF, T, P, Cld, R <sub>eff</sub>	Global, regional, 1860-2100	Fixed 2005 emissions	See Table 3-5

1 Table 2: List of simulations

	RCP simulations	RCP Fixed emis. simulations	RCP Fixed SST simulations
Simulation name	RCP2.6	RCP2.6_2005AER <del>F</del>	RCP2.6_RFP
	RCP4.5	RCP4.5_2005AER <del>F</del>	RCP4.5_RFP
	RCP6	RCP6_2005AER <del>F</del>	RCP6_RFP
	RCP8.5	RCP8.5_2005AER	RCP8.5_RFP
	<u>CM3w_RCP8.5*</u>	<u>CM3w_RCP8.5_2005AER*<del>F</del></u>	RCP_1860
Aerosol setup	Time-varying	SO <sub>2</sub> , OC, BC emissions fixed at 2005 levels	Time-varying, aerosol forcing only
GHG	Time-varying	Time-varying	Time varying

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3 \*Simulation uses configuration with weaker aerosol effective radiative forcing (see Sect. 2.3)

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13 Table 3: Global average difference in aerosol optical depth (AOD), forcing (RF), temperature  
 14 (T), total precipitation rate (P), liquid water path (LWP), and cloud droplet effective radius  
 15 ( $R_{\text{eff}}$ ) due to aerosol emissions reductions at the end of the 21<sup>st</sup> century (2096-2100 average).  
 16 Values represent differences between RCP simulations and fixed 2005 simulations (base  
 17 cases) ( $\text{RCP}_{x.x} - \text{RCP}_{x.x} - \text{FRCP}_{x.x} - \text{2005AER}$ ). RF values represent the difference between  
 18 2100 and 2006 total aerosol forcing.

	SO <sub>4</sub> AOD	BC AOD	OC AOD	RF (W m <sup>-2</sup> )	T (K)	P (mm d <sup>-1</sup> )	LWP (g m <sup>-2</sup> )	R <sub>eff</sub> (μm)
RCP2.6	-0.034	-0.0018	-0.0055	1.37	0.97	0.080	-0.555	0.584
RCP4.5	-0.033	-0.0017	-0.0081	1.25	0.81	0.078	-0.870	0.603
RCP6.0	-0.032	-0.0011	-0.0005	1.12	0.72	0.082	-0.502	0.541
RCP8.5	-0.035	-0.0014	-0.0057	1.05	1.04	0.093	-0.748	0.585
<u>CM3w RCP8.5*</u>	=	=	=	<u>-0.5</u>	<u>0.51</u>	<u>0.048</u>	<u>-0.218</u>	<u>0.472</u>

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\*Simulation uses configuration with weaker aerosol effective radiative forcing (see Sect. 2.3)

1 Table 4: Regional average changes in aerosol optical depth (AOD), effective radiative forcing (RF), temperature (T), precipitation (P), liquid  
 2 water path (LWP), and cloud droplet effective radius ( $R_{\text{eff}}$ ) for RCP8.5 due to aerosol effects only at the end of the 21<sup>st</sup> century. Values  
 3 represent differences between RCP simulations and fixed 2005 simulations (RCP8.5–~~RCP8.5\_FRCP8.5\_2005AER~~). ~~Region definitions are in~~  
 4 ~~Fig. 1~~. Boldface values represent the largest regional change for each variable. RF values represent the difference between 2100 and 2006

	<u>SO<sub>4</sub> AOD</u>	<u>BC AOD</u>	<u>OC AOD</u>	<u>RF (W m<sup>-2</sup>)</u>	<u>T (K)</u>	<u>P (mm day<sup>-1</sup>)</u>	<u>LWP (g m<sup>-2</sup>)</u>	<u>R<sub>eff</sub> (μm)</u>
<u>NM</u>	<del>-0.13</del>	<del>-0.0029</del>	<del>-0.0039</del>	<del>1.53</del>	<del>1.81</del>	<del>0.125</del>	<del>0.832</del>	<del>0.849</del>
<u>SM</u>	<del>-0.053</del>	<del>-0.0029</del>	<del>-0.0039</del>	<del>0.853</del>	<del>0.981</del>	<del>0.0425</del>	<del>-0.552</del>	<del>0.5719</del>
<u>EA</u>	<del>-0.053</del>	<del>-0.0053</del>	<del>-0.0070</del>	<del>2.089</del>	<del>2.006</del>	<del>0.0944</del>	<del>-6.7953</del>	<del>1.8471</del>
<u>SA</u>	<del>-0.41</del>	<del>-0.027</del>	<del>-0.050</del>	<del>3.858</del>	<del>1.710</del>	<del>0.175</del>	<del>-4.379</del>	<del>1.59</del>
<u>EU</u>	<del>-0.27</del>	<del>-0.020</del>	<del>-0.022</del>	<del>2.385</del>	<del>1.451</del>	<del>0.175</del>	<del>-3.424</del>	<del>1.275</del>
<u>RU</u>	<del>-0.27</del>	<del>-0.012</del>	<del>-0.035</del>	<del>1.807</del>	<del>1.715</del>	<del>0.203</del>	<del>2.022</del>	<del>0.937</del>
<u>AU</u>	<del>-0.060</del>	<del>-0.0048</del>	<del>-0.0085</del>	<del>0.420</del>	<del>1.297</del>	<del>-0.203</del>	<del>-3.184</del>	<del>0.963</del>
<u>AF</u>	<del>-0.12</del>	<del>-0.0012</del>	<del>-0.0079</del>	<del>0.962</del>	<del>1.297</del>	<del>0.032</del>	<del>-1.318</del>	<del>0.876</del>
<u>ME</u>	<del>-0.20</del>	<del>-0.0013</del>	<del>-0.0105</del>	<del>0.420</del>	<del>2.072</del>	<del>-0.039</del>	<del>-2.241</del>	<del>1.507</del>
<u>ME</u>	<b>-0.20</b>	<b>-0.0034</b>	<b>-0.0119</b>	<b>0.42</b>	<b>2.07</b>	<b>-0.031</b>	<b>-2.24</b>	<b>1.50</b>

1 | total\_aerosol forcing.▲

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SO <sub>4</sub> AOD	BC AOD	OC AOD	RF (W m <sup>-2</sup> )	T (K)	P (mm day <sup>-1</sup> )	LWP (g m <sup>-2</sup> )	R <sub>eff</sub> (μm)
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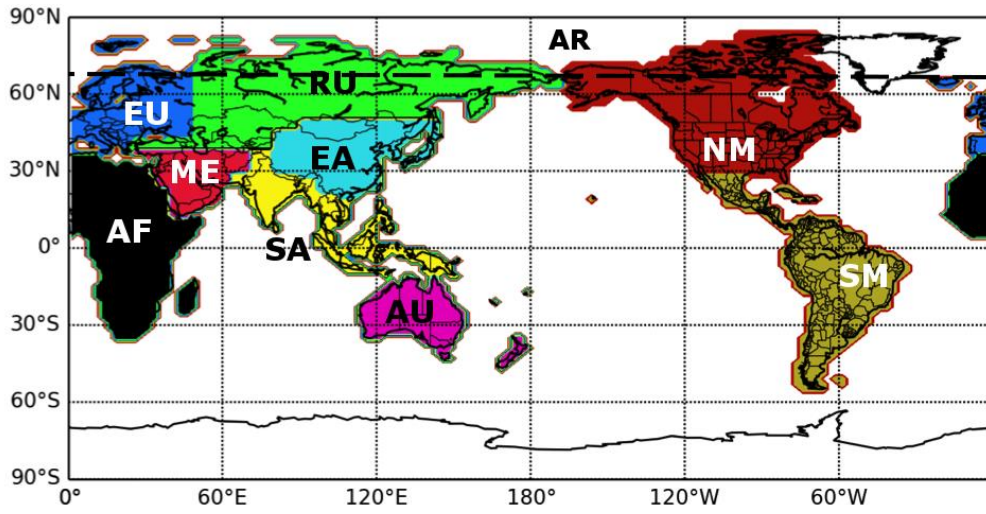
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3 | Table 5: Same as Table 4, but for RCP2.6 and ~~RCP2.6~~FRCP2.6\_2005AER

NM	-0.122	-0.0029	-0.0010	1.95	1.83	0.115	-0.057	0.756
SM	-0.068	-0.0067	-0.0275	1.16	0.95	0.0442	-1.58	0.508
EA	<b>-0.43</b>	<b>-0.033</b>	-0.061	3.06	1.71	0.33	-3.25	1.85
SA	-0.41	-0.024	-0.053	<b>4.52</b>	0.98	<b>0.36</b>	-2.93	1.35
EU	-0.269	-0.013	-0.017	3.20	1.65	0.087	-2.66	1.24
RU	-0.13	-0.0059	-0.0080	2.76	<b>2.21</b>	0.16	1.84	0.993
AU	-0.045	0.0014	0.0023	0.63	0.73	-0.107	-1.05	0.543
AF	-0.14	-0.0075	<b>-0.070</b>	0.97	1.16	0.016	-0.69	1.06
ME	-0.26	-0.0072	-0.016	0.67	1.75	0.039	-1.36	<b>2.40</b>

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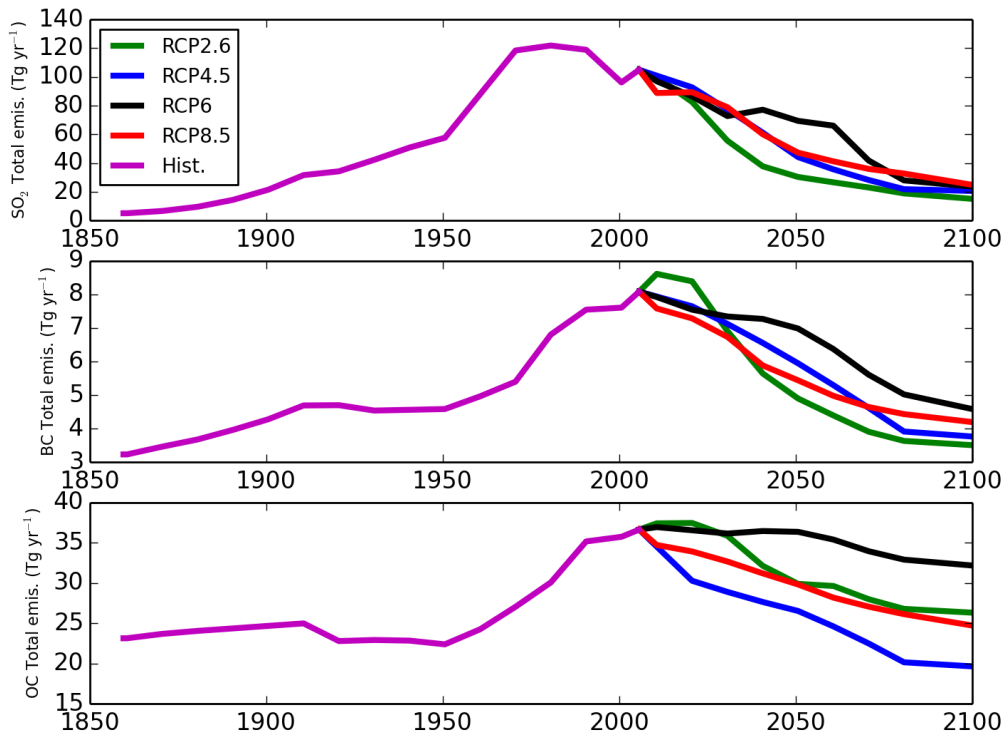
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3 Figure 1: Region definitions. North America (NM), South and Central America (SM), Europe  
4 (EU), Russia (RU), East Asia (EA), South Asia (SA), Australia (AU), Africa (AF), Middle  
5 East (ME), and Arctic (AR, everything north of the dashed line)

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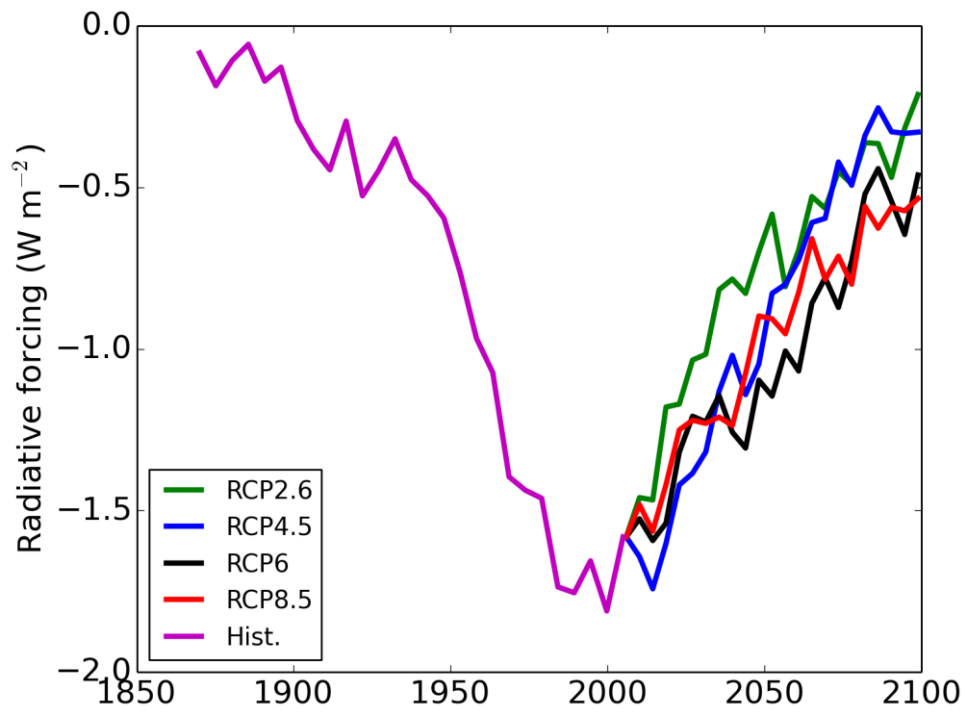
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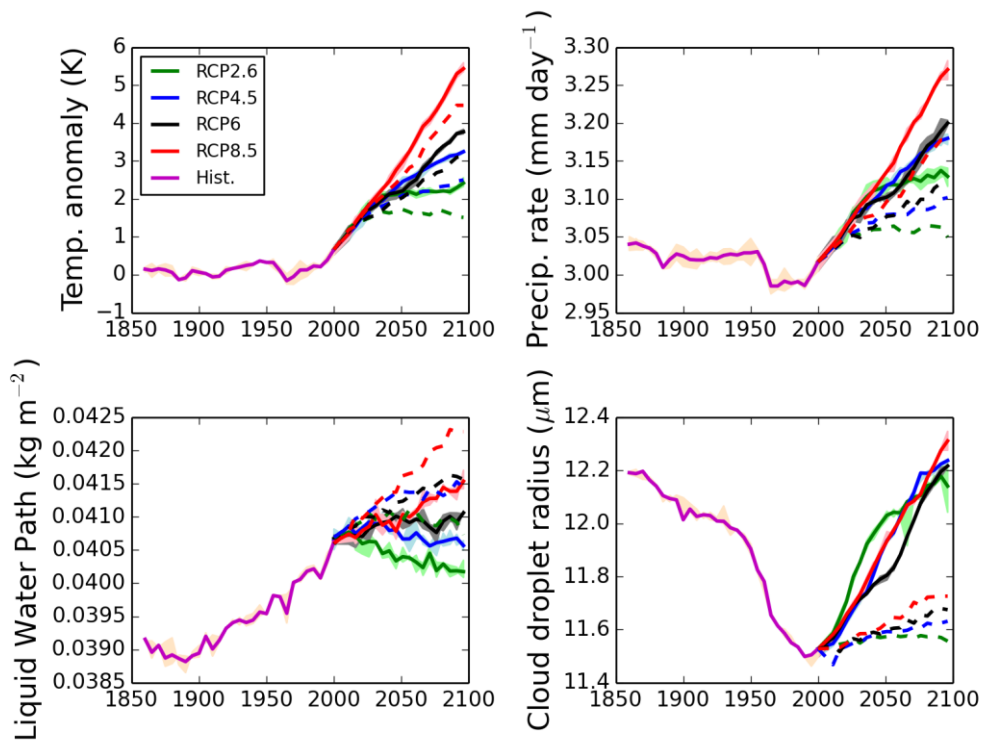


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 2 Figure 2: Global total emissions (anthropogenic + biomass burning) of sulfur dioxide (SO<sub>2</sub>),  
 3 black carbon (BC), and organic carbon (OC) from 1860-2100. Historical emissions are  
 4 colored in magenta, RCP2.6 in green, RCP4.5 in blue, RCP6 in black, and RCP8.5 in red.

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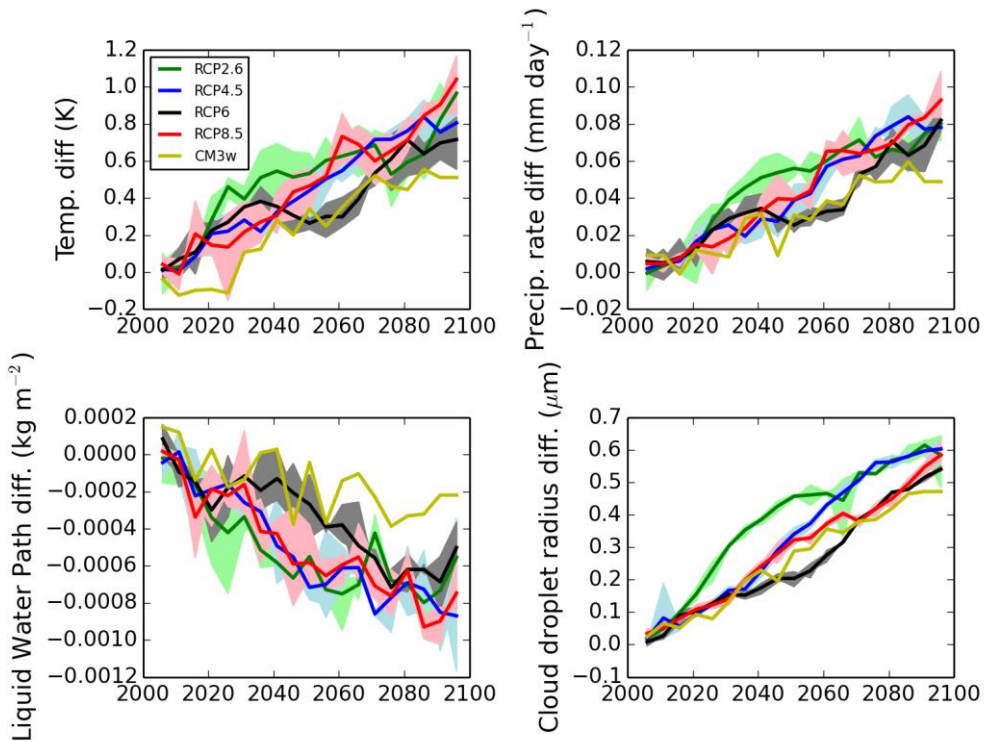


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 2 Figure 3: Globally averaged historical and future time series of top-of-atmosphere effective  
 3 radiative forcing ( $\text{W m}^{-2}$ ) from emissions of aerosols and their precursors alone.

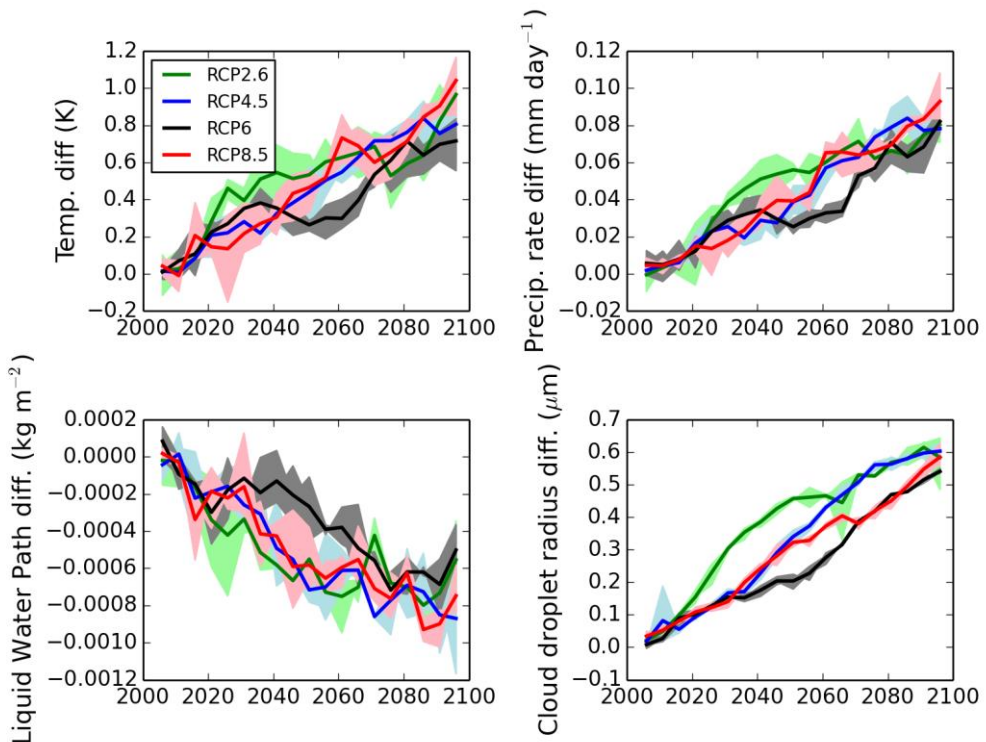


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 2 Figure 4: Globally averaged trend in climate response variables from 1860-2100. Upper left:  
 3 surface air temperature (K), upper right: total precipitation rate ( $\text{mm day}^{-1}$ ), bottom left:  
 4 liquid water path ( $\text{kg m}^{-2}$ ), bottom right: effective cloud droplet radius ( $\mu\text{m}$ ). Shaded light colors  
 5 represent the range of the three ensemble members, solid lines are the ensemble means for the  
 6 RCPx.x time-varying simulations. Dashed lines are the ensemble means for the  
 7 [RCPx.x-FRCPx.x 2005AER](#) control simulations (ensemble range not shown).

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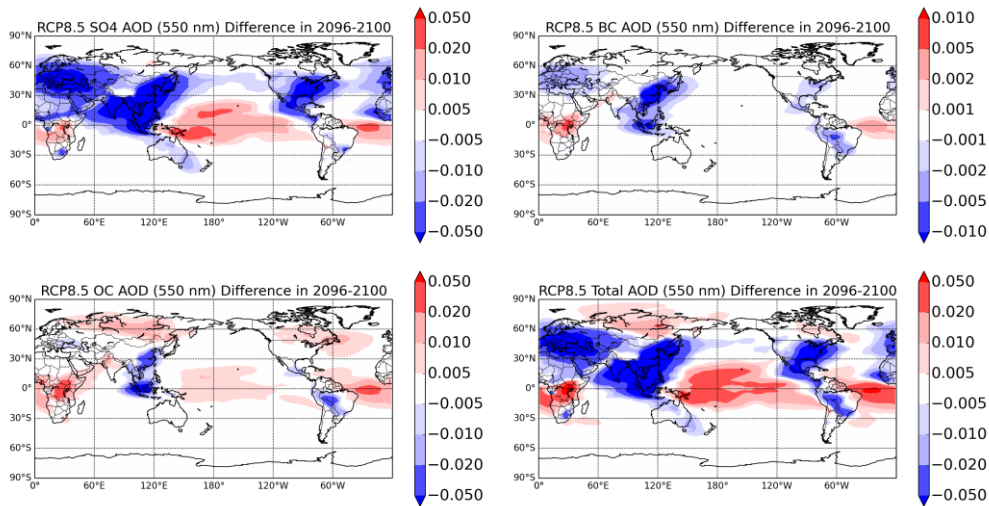


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1 Figure 5: Globally averaged trend in climate variable anomalies from 2006-2100. Values  
 2 represent differences between the projected RCP simulations and the fixed 2005 aerosol  
 3 emissions case ( $RCP_{x,x} - RCP_{x,x} - RCP_{x,x} - 2005AER$ ). Upper left: surface air temperature  
 4 (K), upper right: total precipitation rate ( $mm\ day^{-1}$ ), lower left: liquid water path (LWP) ( $kg$   
 5  $m^{-2}$ ), lower right: effective cloud droplet radius at cloud top ( $\mu m$ ). Shaded light colors  
 6 represent the range of the three ensemble members, solid lines are the ensemble means.  
 7 Yellow lines represent the CM3w\_RCP8.5 run.

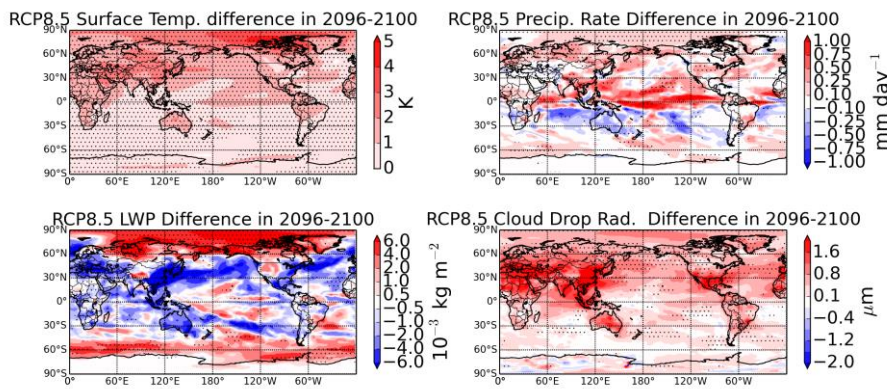
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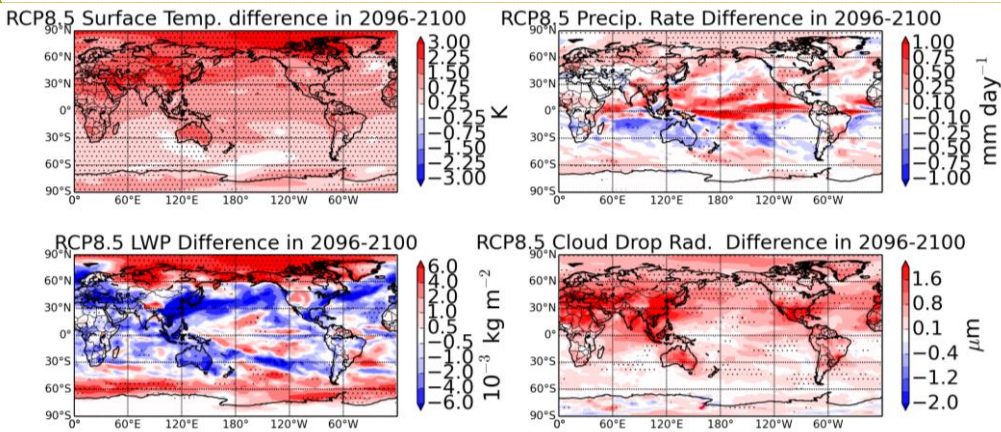
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 17 Figure 6: Anomalies in aerosol optical depth for 2096-2100 five-year average for RCP8.5.  
 18 Values represent differences between the projected RCP aerosol optical depth and the fixed

1 | 2005 aerosol emissions case (RCP8.5 – [RCP8.5\\_FRCP8.5\\_2005AER](#)). Note the different  
2 | scale for BC AOD.

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1 Figure 7: Changes in climate variables for a five-year average over 2096-2100 for RCP8.5.  
2 Values represent differences between the projected RCP simulations and the fixed 2005  
3 aerosol emissions case (RCP8.5 – ~~RCP8.5~~ FRCP8.5 2005AER). Hatched areas represent  
4 statistically significant changes at the 95% confidence level. Upper left: surface air  
5 temperature (K) , upper right: total precipitation rate (mm day<sup>-1</sup>), lower left: liquid water path  
6 (kg m<sup>-2</sup>), lower right: effective cloud droplet radius at cloud top (μm)

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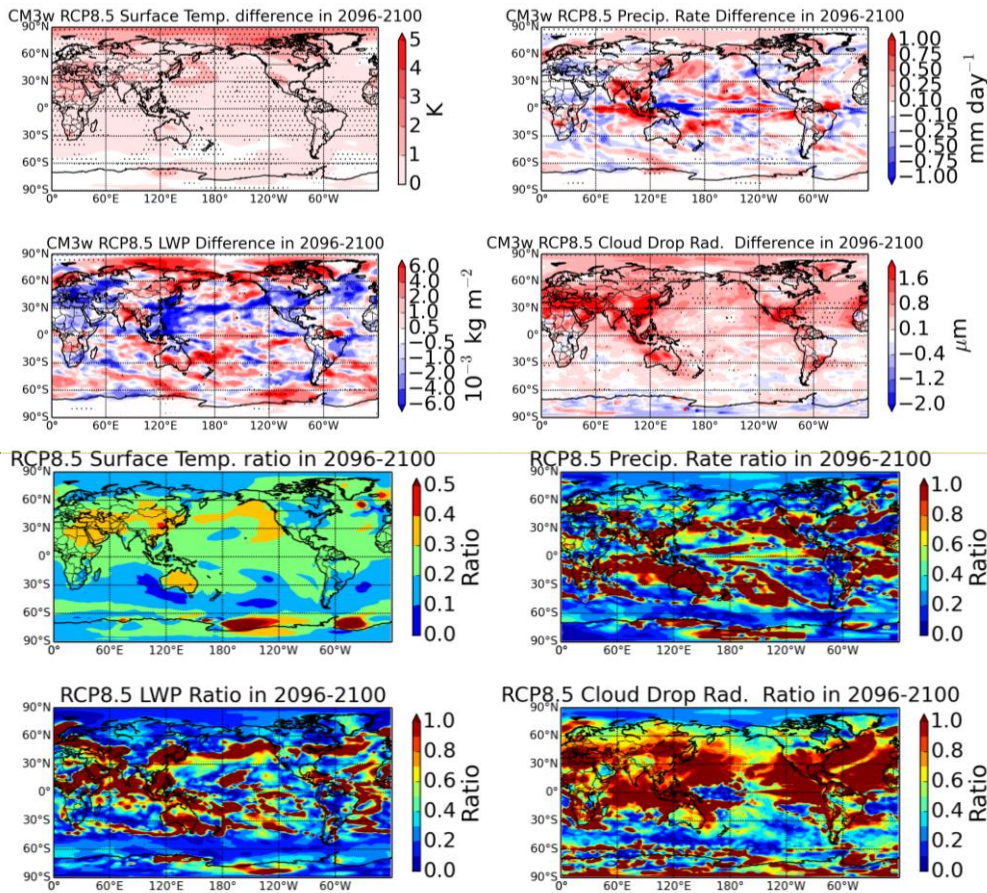
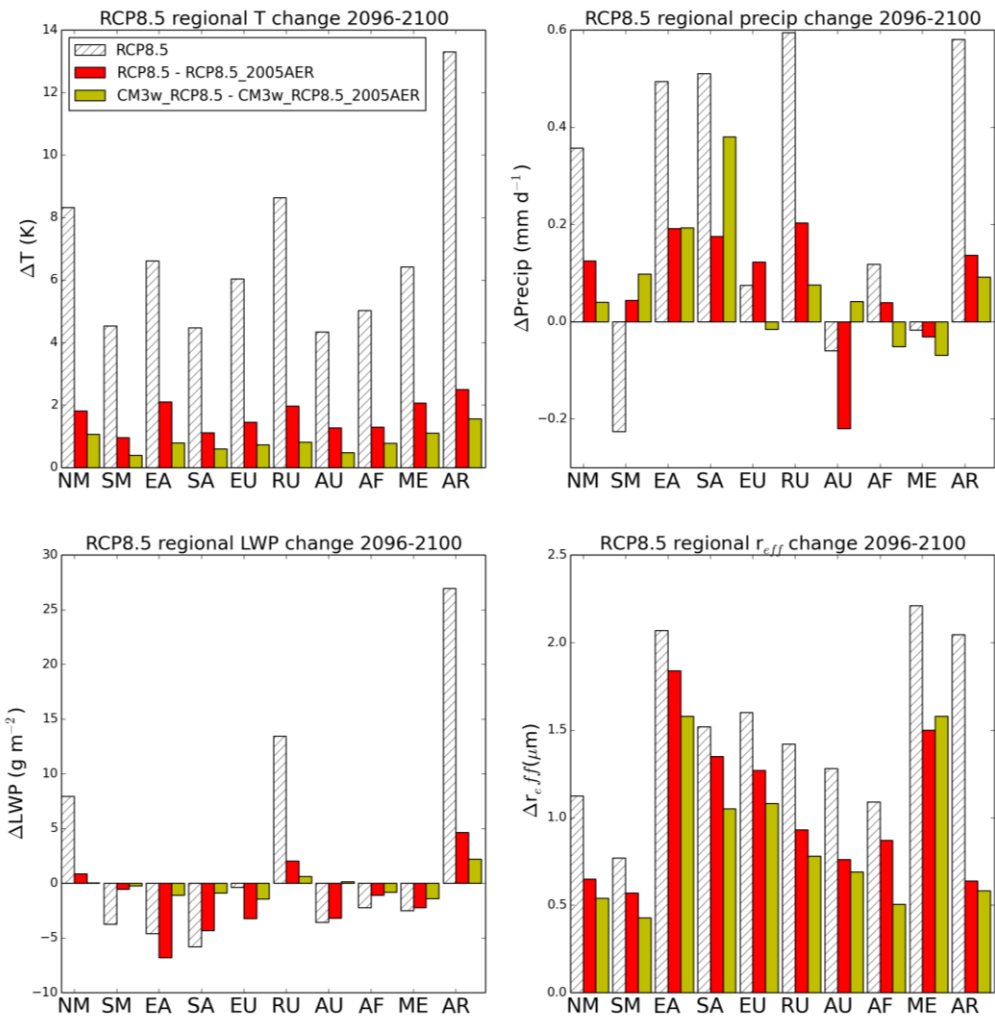


Figure 8: Same as Fig. 7, but for the case with reduced present-day effective aerosol radiative forcing (CM3w). Values represent differences between the projected RCP simulations and the fixed 2005 aerosol emissions case (CM3w RCP8.5 – CM3w RCP8.5 2005AER). Ratio of climate response to aerosol decreases and all forcing climate response for a five-year average over 2096-2100 for RCP8.5. Upper left: surface air temperature, upper right: total precipitation rate, lower left: liquid water path, lower right: effective cloud droplet radius at cloud top.

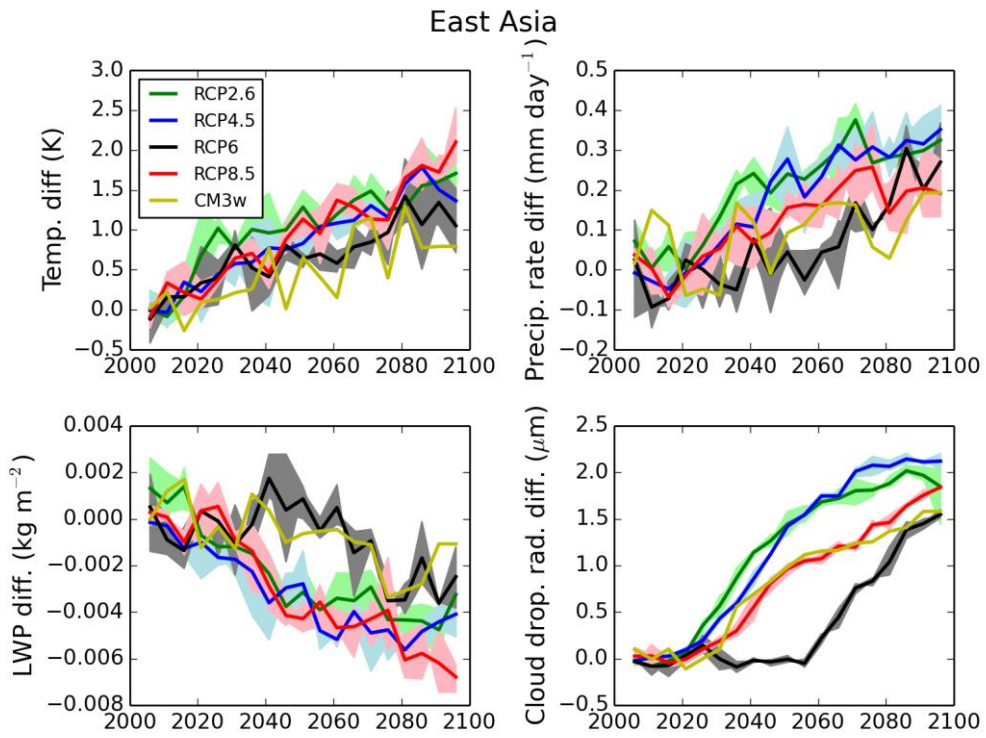




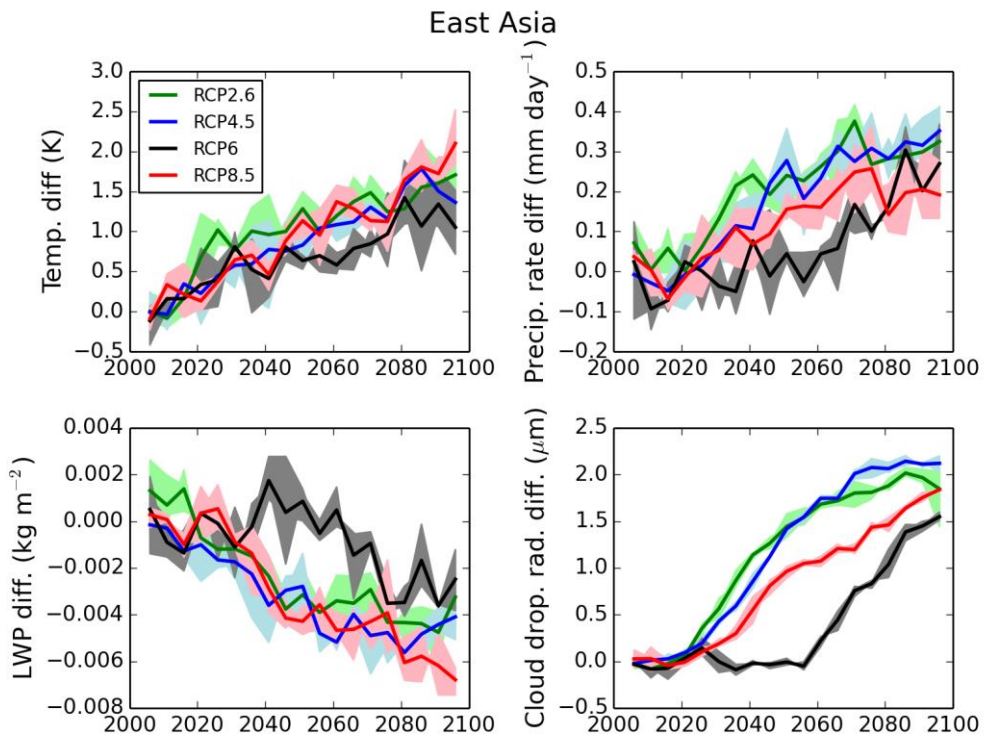
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2 Figure 9: Regional comparison of climate response to aerosol decreases (RCP8.5 –  
3 RCP8.5 2005AER, red bars), climate response to aerosol decreases under weaker aerosol  
4 forcing (CM3w RCP8.5 – CM3w RCP8.5 2005AER, yellow bars), and climate response to  
5 all-forcings (RCP8.5, white hatched bars) for a five-year average over 2096-2100 for RCP8.5.  
6 Upper left: surface air temperature (K), upper right: total precipitation rate (mm day<sup>-1</sup>), lower  
7 left: liquid water path (g m<sup>-2</sup>), lower right: effective cloud droplet radius at cloud top (μm).  
8 Regions are defined in Fig. 1.

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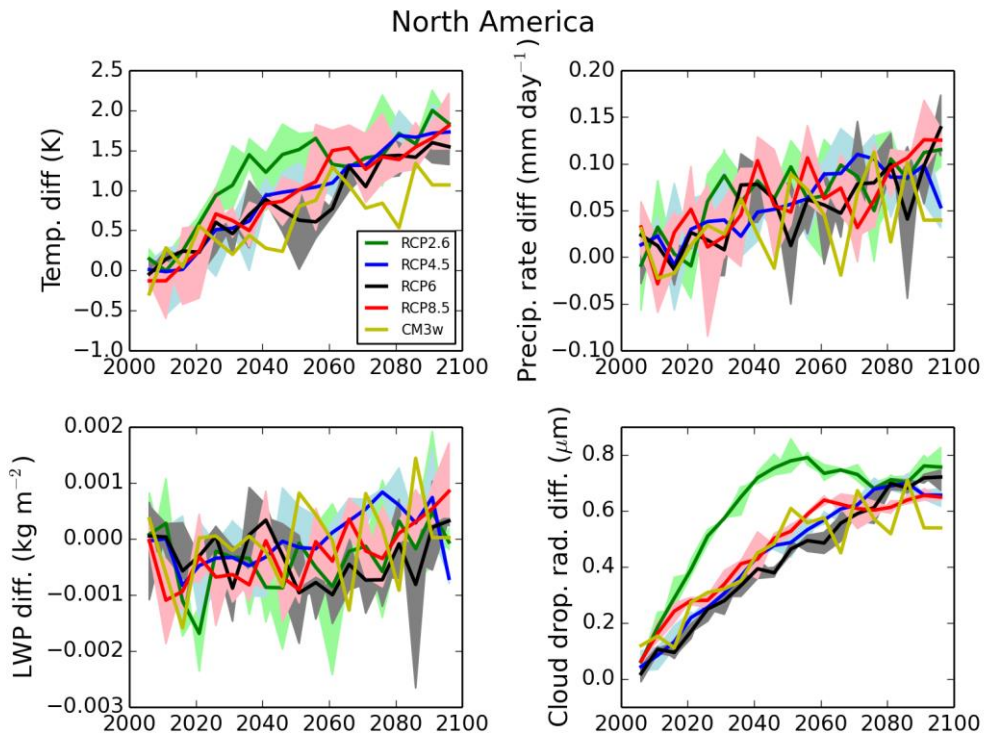


1 | Figure [910](#): Trends in climate parameter differences from 2005-2100 for the East Asia  
2 | Region. Values represent differences between the projected RCP simulations and the fixed  
3 | 2005 aerosol emissions case (base case). Shaded light colors represent the range of the three  
4 | ensemble members, solid lines are the ensemble means. Yellow lines represent the  
5 | CM3w\_RCP8.5 run.

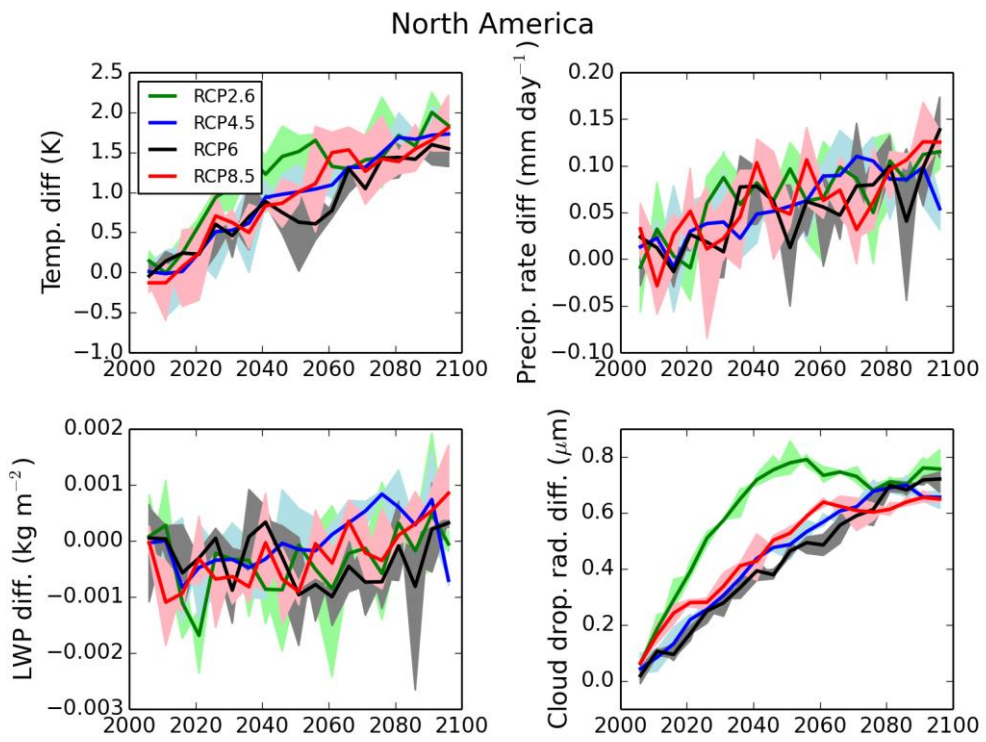
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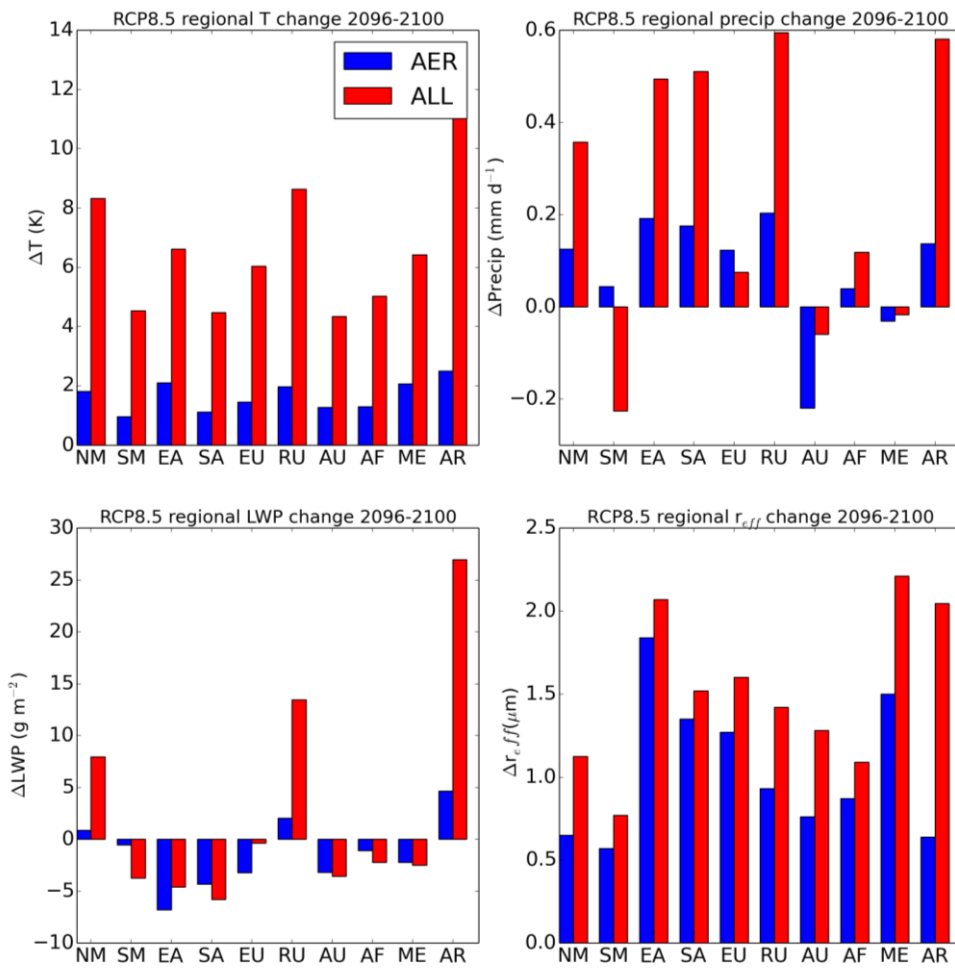


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1 | Figure 4911: Trends in climate parameter differences from 2005-2100 for the North America  
 2 | region. Values represent differences between the projected RCP simulations and the fixed  
 3 | 2005 aerosol emissions case (base case). Shaded light colors represent the range of the three  
 4 | ensemble members, solid lines are the ensemble means. Yellow lines represent the  
 5 | CM3w RCP8.5 run.

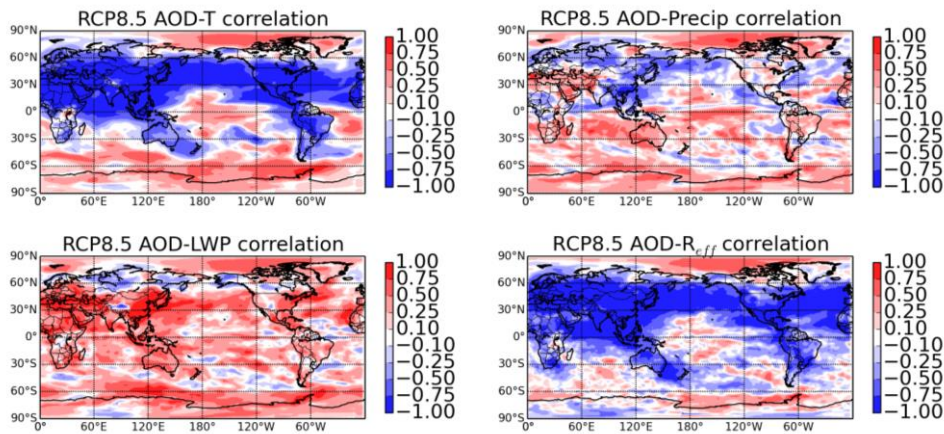
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1 Figure 11: Regional comparison of climate response to aerosol decreases (AER) and climate  
 2 response to all forcings (ALL) for a five year average over 2096-2100 for RCP8.5. Upper left:  
 3 surface air temperature (K), upper right: total precipitation rate ( $\text{mm day}^{-1}$ ), lower left: liquid  
 4 water path ( $\text{kg m}^{-2}$ ), lower right: effective cloud droplet radius at cloud top ( $\mu\text{m}$ ). Regions are  
 5 defined in Fig. 1.

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9 Figure 12: Correlations of aerosol-driven changes in temperature (T), precipitation rate  
 10 (Precip), liquid water path (LWP), and cloud droplet radius ( $R_{\text{eff}}$ ) with total anthropogenic  
 11 aerosol optical depth (sulfate, black carbon, and organic carbon AOD) for RCP8.5.

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