

We are grateful to the reviewer for their interest and comments on the paper. These comments are very valuable and have helped improve the manuscript. Here we outline how we have addressed these comments in the revised manuscript. The newly added discussions and rephrased sentences have been highlighted in green in our replies below.

This paper assesses the influence on radiative forcing, temperature and precipitation of changes in anthropogenic aerosol emissions between 1970 and 2010 in the CESM1-CAM5 model. The paper decomposes the effects into the contribution from aerosol emissions increases due to increased energy use and the contribution from aerosol emissions decreases due to technological advances and air quality abatement initiatives over the same time period. The authors find that the two effects have partially cancelled each other over the analyzed period and do so nonlinearly and non-uniformly. The paper is well written overall. There are several useful and interesting points made. The experiment design is clean and well thought out. However, I find that the motivation and certain aspects of the analysis require additional depth, in order to make the novelty of the paper clearer. I am also concerned that certain aspects of the results seem counterintuitive, suggesting problems with the model set-up (see Other Major Comments 2 and 3 below). At present it is not entirely clear what new the authors are adding to the conversation, though I do believe that this analysis has the potential to be a useful contribution with some additional depth. I believe that major revisions to provide further depth would be needed for the paper to merit publication in ACP. These are discussed further below:

While revising the manuscript, we realised that the isolation of the effects of aerosol changes in the “best estimate” experiment was probably biased due somehow to the experimental set-up. To address this issue, we have carried out a new experiment, which is now used throughout the revised manuscript (including Figures and texts). Note that, while this has resulted in different estimates of the impacts of aerosol changes in the best estimate case, it does not have any bearing on the major findings of this study on the comparison of the two different retrospective emission scenarios.

1. The best estimate results shown in this manuscript are essentially an assessment of impact of aerosols on climate over the last 40 years, which has been conducted extensively in the existing literature. The main novelty in the paper, therefore, lies in the decomposition into the two factors described above. However, I believe the authors must do more to center and frame this decomposition.

At present, the authors do not make a strong case for why decomposing the total effect into these two components is a valuable exercise. This should be explicitly laid out in the introduction.

The authors need to be clearer that the assessment of last 20th century effects of aerosols has been widely done elsewhere and provide this more as background, acknowledging where their work is replicative of and consistent with that done elsewhere, and focus more thoroughly on unique insight gained through the de-composition.

As the reviewer points out, the main contribution of this work is the analysis of the climate changes associated with the two aerosol emission drivers, but not the historical aerosol changes

(referred to as “best estimate” in this work). Note however that one significant difference with most of the previous studies is that, even in the “best estimate” scenario, we analyse equilibrium rather than transient responses. Also, the “best estimate” experiment is thoroughly analysed in a manuscript that is under review for the *Journal of Geophysical Research: Atmosphere* (Zhao et al. Climate forcing and response to greenhouse gases, aerosols and ozone in CESM1), in which the impacts of historical aerosol changes have been explicitly discussed in the context of existing works. The ERF, as well as temperature and precipitation responses to the historical aerosol emission changes are presented here primarily to provide a reference for the two aerosol emission scenarios.

To clarify the motivation of this work, we expand P3 L22-24 into “As discussed above, energy use growth and technology advances are two of the major policy-relevant drivers of past aerosol/precursor emission changes via, for example, changes in power generation, industry and transportation. These drivers are very likely to continue to play important but competing roles in modulating future aerosol emission, as we gradually transit to a new energy structure. An analysis of the climate impact to recent changes in the two above emissions drivers is therefore critically important for future aerosol related climate projections and climate change impact reduction strategies. Here we perform time-slice model simulations using the fully-coupled Community Earth System Model (CESM1), seeking to quantify the climate forcing and impacts of aerosol changes related two the above policy-relevant emission drivers (energy use growth and technology advances) at both global and regional scales.”

At the same time, the analysis of the decomposition should be further deepened. Although the authors report many signals, they do not provide any depth of analysis on why these signals emerge. Given how many other studies have analysed the role of aerosol increases and decreases in the second half of the 20th century and made many of the same points the authors make here, simply reporting these signals is not sufficient. Where do the two effects counteract or reinforce each other in ERF, temperature, or precipitation? Why does this occur? What are the implications for future anticipated shifts in aerosol emissions due to the two effects?

Regarding the implications for future anticipated shifts in aerosol emissions, please refer to our replies to comment #6.

Also, we have added comments on changes in low-level circulation and sea level pressure (Figure S2 in the supplement), trying to provide some speculations around the underpinning mechanisms of temperature/precipitation responses. Please refer to the new result section n the revised manuscript. However, although it is certainly interesting and relevant to shed light on

the underpinning mechanisms, we feel strongly that such analyses are speculative and with large uncertainties. Therefore, in the revised manuscript, we tried not to inflate the revised manuscript with mechanism analysis, in order also not to overwhelm the main storyline of this study which is around the climate responses to the two policy-relevant drives.

2. A main result that the authors emphasize is the result that the individual ERFs of the aerosol species do not add up linearly, giving rise to the authors' repeated statement that ERF calculations should be considered to be dependent on experiment set-up. While this is in some ways a fairly obvious statement, it would be useful to have this explored in further detail. However, the authors do not provide any explanation for this result. I believe the authors must assess the mechanism behind this more thoroughly for it to be a valuable contribution. Is the nonlinearity of the ERF when the aerosols are emitted separately versus together a result of changes in the total atmospheric burden or of changes in the spatial distribution and resulting radiative interactions? The authors allude briefly to the total burdens of each species being "identical" in the separate and combined simulations (P8, L20), but do not show it. This should be explicitly shown. The remaining explanations that are currently only suggested in Section 4.1 should be explicitly assessed.

We thank the reviewer for the valuable comments and suggestions.

Both spatial patterns and the global mean values of aerosol burden changes do not show appreciable differences between the two different experiments. This is consistent for all the three species (Figure R4.1). However, despite identical changes in aerosol burdens between the two different experiments, the diagnosed ERF do not add up, leading us to stress the nonlinearity in estimating the ERF. To make the statement clearer, we rephrase P8 L30-31 as "We note that changes in both the spatial pattern and the global mean amount (Figure S3) of the burden and AOD of the three aerosol species do not show appreciable differences to those in the experiment where all the three species change simultaneously (B10-B70)."

We also thank the reviewer for suggesting us to assess and expand the remaining discussion in Section 4.1. While this is certainly relevant, we believe such additional experiments (e.g., having sensitivity experiments where the amounts of background soluble aerosol species vary while keeping BC constant) will make the scope of this work too wide and the manuscript too heavy. Nevertheless, in combination with comment #2 from reviewer #2, we expanded the discussions into "This reflects partly the nonlinear effect associated with the mixing state of different aerosol species as well as the importance of background aerosol loadings. This is particularly important for BC whose effects depend also on the presence of sulphate and organic aerosols (Ramana et al., 2010). That is, given that aerosol species are internally-mixed in MAM3 (i.e. different chemical species are mixed within an aerosol particle), the

hygroscopicity of aerosol particles is dominated by the volume of soluble species (organic compounds and sulphate). This means that the nonlinearity in the isolated aerosol ERF may be a reflection of the aerosol scheme in CESM1. More specifically, BC particles tend to be coated with other species during ageing, thereby enhancing the absorption effects and the subsequent impacts on cloud microphysics, as well as amplifying their radiative forcing (Haywood and Ramaswamy, 1998; Kim et al., 2008; Chung et al., 2012; Wu et al., 2016). ...”

Also, we further tone down the statement in P9 L14-16 into “Overall, the above discussion illustrates the importance of background aerosol concentrations in estimating the radiative forcing of aerosols. For example, we speculate that diagnosing the ERF of BC the other way round, namely, keeping all other aerosol species at 1970 levels while changing BC to 2010 levels would likely result in a different ERF estimate.”

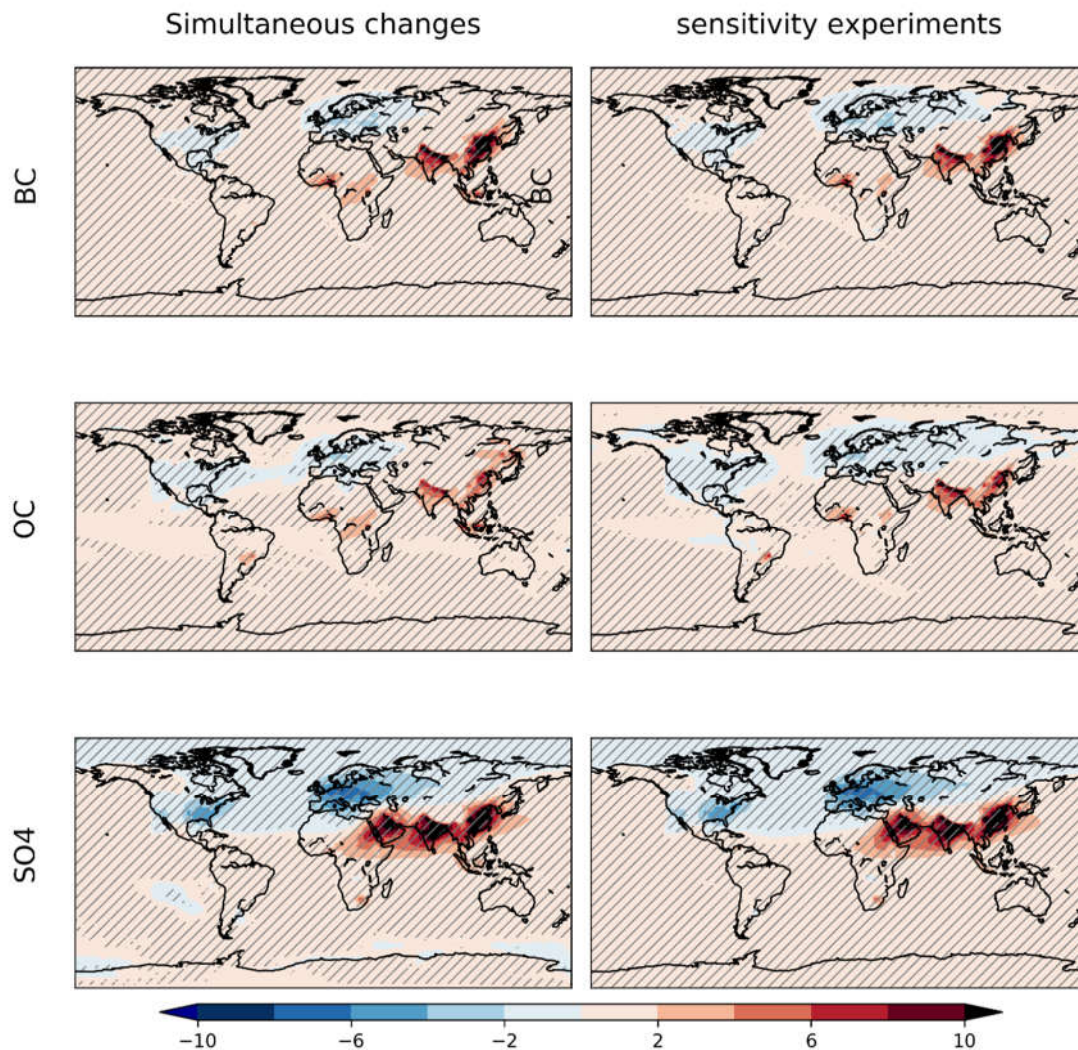


Figure R4.1 The 1970-2010 changes in the burden (mg m^{-2}) of black carbon (BC, top row), organic carbon (OC, second row) and sulphate (SO_4 , third row) in the best estimate experiment.

The left column is from the experiment where all aerosol species change simultaneously, while the right column is for those isolated between the reference 2010 run and the one where the emissions of a targeted species (e.g. BC in the top right) are fixed to its 1970 levels.

Other Major Comments:

3. Confidence intervals should be provided for all global-mean values.

Done

4. I agree with Anonymous Referee 3 that the negative global mean temperature sensitivity value in the best estimate case seems to violate basic energy conservation. How can a positive global-mean ERF result in a negative global-mean temperature change or vice versa? This suggests a major issue with the model formulation. If it is somehow robust, it needs to be explained.

As mentioned above (also see our reply to reviewer #3), we added a new model simulation to isolate the effects of aerosol changes in the “best estimates” experiment. As such, we repeated all related analysis and edited the manuscript accordingly (including Figures and texts).

The sign of global mean temperature response to aerosol changes isolated using the new experiment now consistent with that of ERF. However, there are still pronounced inconsistencies at regional scales, and the underlying mechanism has been thoroughly discussed in the revised manuscript.

5. Figure 1: Why does the large increase in SO₄ burden result in a positive ERF? This needs to be explained, particularly when compared to the larger negative ERF of the smaller (and less reflective) OC burden.

The positive global mean SO₄ ERF, as we stated in P6 Ls1-3, is due to the partial cancellation between the pronounced positive forcing from sulphate aerosol reductions over Europe and North America and the relatively confined negative forcing from sulphate aerosol increases over Asia (Figure 1f). The former is amplified over the Arctic, and results into a net global mean positive forcing.

The larger negative OC ERF explainable, since the small OC reductions over Europe and North America leads to only slight positive OC ERF (Figure 1e).

In response to this comment, we expand P6 Ls1-3 into “The global mean ERF of sulphate aerosols is **small and positive**, because of the partial cancellation between the negative forcing from sulphate aerosol increases over Asia and the pronounced positive forcing from sulphate aerosol reductions over Europe and North America **which is amplified over the Arctic** (Fig. 1f).”

6. Section 4.3 is largely a literature review on the formulation of aerosol emissions scenarios and does not explore what implications the results of this analysis have for future projections. How may their findings “help better assess and interpret such uncertainties in future climate projections”?

We thank the reviewer for these comments.

In combination with comment#1 and comments from the anonymous reviewers #2 and #3, we have expanded Section 4.3 into “Reliable projections of future climate under different but equally plausible emission pathways are of utmost importance to better constrain the range of possible societal risks and response options. Unfortunately, there are still considerable challenges due to limitations and uncertainties in our understanding of many aspects of the climate system (Knutti and Sedláček, 2013; Northrop and Chandler, 2014; Marotzke, 2019). Aerosols represent one of the largest sources of uncertainty (Boucher et al., 2013; Lee et al., 2016; Fletcher et al., 2018). Present-day anthropogenic aerosol emissions are largely influenced by sectors including power generation, industry and transport. However, in some of the future emission pathways, for example, the Tier-1 Shared Socioeconomic Pathways scenarios (SSP1; Gidden et al. (2018)), aerosol emissions are expected to decline drastically worldwide as we transit to non-fossil-fuel-based fields together with rapid implementation of air pollution control measures and new technologies. For example, mainly as a result of China’s transition to a less energy-demanding society, for the first time the global coal consumption decreased in 2015 since the 1970s (World Energy Council, 2016). However, the timing and rate of such transitions are largely uncertain. On the other hand, it is also likely that aerosol-related emissions will increase, especially over some developing regions, under scenarios where high inequality exists between and within countries. For example, in SSP3, expanding industrial sectors over Southeast Asia may continue to rely on traditional energy sources such as coal for much of the 21st century. Also, it is possible that the world may continue to rely on fossil energy sources more strongly than expected over the coming years, given the concerns about nuclear energy after the Fukushima Daiichi nuclear disaster in March 2011. As a consequence, aerosol emissions from energy use in some regions may increase and therefore offset aerosol reductions elsewhere.

The above discussion reflects the large uncertainties (both spatially and temporally) in our understanding and estimates of future aerosol-related emission trajectories, given the possibility that very different future emission pathways may be adopted by different countries to compromise between climate/air pollution impacts and economic growth. Our findings may

help better assess and interpret such uncertainties in future climate projections associated with changes in aerosols. First of all, the large impacts of present-day aerosol emissions from the two competing drivers, as reported in this work, suggest that the major drivers (e.g., future energy structure and efficiency, air pollution control measurements, as well as technology progresses) of aerosol emission changes are likely to continue to play important roles in future climate projections. Secondly, uncertainties in future aerosol emissions pathways combine with those of other climate forcing agents (e.g., greenhouse gas emissions and land-use changes). Such uncertainties influence the impacts of aerosol forcing through changing the background climate state (see Section 4.2; e.g., (Frey et al., 2017; Nordling et al., 2019; Stolpe et al., 2019). More importantly, our results stress the importance of nonlinearities when comparing and assessing the impacts of different future aerosol emission trajectories. This adds further caveats in interpreting future climate projections related to aerosol changes in addition to uncertainties in emission pathways of both aerosols and their precursors and GHGs.”

Minor Comments:

7. The authors use a somewhat confusing formulation throughout the paper in which they refer to both the aerosol increases from increased energy use and the aerosol decreases from technological advances as “aerosol emissions” (e.g. P8, L23; P10, L26). I recommend rewording these to something like “aerosol emissions changes” throughout, to make clearer that it is in some case the absence rather than the presence of aerosol emissions that is being analysed.

We thank the reviewer for the suggestion, and have thoroughly edited the manuscript.

8. The model and experiment design section would benefit from a clearer description in the text of what the different model simulations are and what scenarios they are intended to test. This is provided at the moment entirely in the caption to Table 1, but should be at least summarized in Section 2.2.

We thank the reviewer for this suggestion. We have rephrased P5 Ls1-4 into “The baseline 2010 experiment (B10) was initialised using the year 2010 model dump from one ensemble member (No 34) of the CESM1 large ensemble (Kay et al. (2015) transient historical experiment, and was driven by the 2010 all forcing factors (Table 1). Also, we have three perturbation experiments where anthropogenic aerosols are perturbed using different emission scenarios (i.e., the 1970 best estimate, STAG_ENE and STA_TECH as described in Sect. 2.1) while all others forcing agents (e.g., GHGs, natural aerosols, land use, solar forcing) are the same as in the B10 run, in order to differentiate the impacts of the two aerosol emission drivers (refer Table 1 for more details).”

9. It is not entirely clear what the residual emissions are that are occurring in the B10 simulation but that are not captured in the SEN and STC simulations. It would help for this to be more thoroughly described and for its magnitude relative to the emissions changes to be provided, especially given the authors repeated references to it.

We agree with the reviewer that such information would be beneficial. Note, however, that the two retrospective emission scenarios were designed in a way that does not provide a linear decomposition of the total 1970-2010 emission changes. As such, it is impossible for us to quantify the residuals. To make the point clear, we added a sentence at the end of Section 2.1 to direct readers to Crippa et al. (2016) “For more details regarding the nonlinearity associated with the retrospective emission scenarios, please refer to Crippa et al. (2016).”

10. P3, L3-4: recommend replacing “will” with “may”, as certain of the Shared Socioeconomic Pathways do simulate regional increases in anthropogenic aerosol emissions.

In combination with comment #3 from reviewer #3, we tone down the statement into “Anthropogenic aerosol emissions are expected to be reduced worldwide during the 21st century (Markandya et al., 2018).”

11. P5, L5: “where the climate system equilibrates to imposed perturbations but the deep ocean” ←this phrase is unclear

The sentence has been rewritten into “where the surface climate system equilibrates to imposed perturbations; NB the deep ocean may take longer to equilibrate”

12. P7, L30: The drying over Europe as well as Asia seems counterintuitive, since all the other analysis suggested opposite trends (in burden, ERF, and temperature) over Europe and Asia. This bears further explanation.

We apologise for misleading the reviewer to a wrong interpretation. Note our results agree with previous studies in producing a wetting over Europe and a drying over Asia in the best-estimate emission scenario (Figure 6a).

L30 here is for the “energy use” experiment where aerosols increase globally. To make the statements even clearer, we rephrase P7 Ls28-30 into “The globe, especially land areas, gets drier in response to aerosol emission changes from energy use growth (Fig. 6b). The precipitation change in Asia (-0.11 ± 0.30 mm day⁻¹) is close to that associated with the best estimate of 1970-2010 aerosol emission changes (-0.13 ± 0.28 mm day⁻¹).”

13. P8, L7-9: I’m fairly certain the estimate from this paper and from PDRMIP have almost entirely overlapping error bounds, making this difference hardly worth noting. I would rather

read this as suggesting that the precipitation sensitivity is very well aligned with that found by PDRMIP. This is one reason why the authors need to provide confidence intervals on their global-mean values.

First, note our new model experiment to isolate the effects of aerosol changes in the “best estimates” scenario.

P8 Ls7-8 has been rewritten into “Generally, precipitation changes with temperature at a rate of 0.09-0.15 mm day⁻¹ K⁻¹. This is slightly larger than the estimate (~28.6 mm yr⁻¹ K⁻¹, i.e., ~0.08 mm day⁻¹ K⁻¹) for the slow climate response component derived from the Precipitation Driver Response Model Intercomparison Project (PDRMIP; Samset et al. (2016)).”

14. P8, L10-12: “This may suggest that regional..” This statement seems deeply obvious and well-established in the community. Recommend cutting.

We tend to agree with the reviewer that this is “well-established”, but we still feel necessary to stress the point here with our results.

We tone down P8Ls10-12 into “This supports previous studies demonstrating that regional precipitation responses are not simply linked to temperature through energy budget constraints, but also depend on other factors such as adjustments in the atmospheric circulation and remote teleconnections (Bollasina et al., 2014; Wilcox et al., 2018; Lewinschal et al., 2019)”

15. Figure 1: It is not clear that the presence of hatching means that it is statistically significant rather than no statistically significant until one reads the text. Recommend rewording L5-6 to be clearer. It is also quick difficult to see the statistical significance hatching over the ERF values – recommend a different colour (e.g. light grey) for the hatch lines.

To make the hatches more readable, we change the black “//” hatches into grey “//” in Figure 1,2,4 and 6.

Ls5-6 are rewritten into “The statistical significance at 5% level is calculated using the two-tailed student t-test and is denoted by the grey hatches.”

16. Figure 5: It is incredibly difficult to visually parse the symbols. The Global and Global Ocean symbols are essentially indistinguishable in the crowded areas of the graph. Very few of these are actually discussed in the manuscript, and I recommend removing some of the symbols (perhaps the latitude band symbols) to make this figure legible.

We appreciate that Figure 5 is not easy to read, but feel necessary to keep the latitude band values to support our statement in P7 L14.

We have updated Figure 5 with the new experiment included, making it much more readable than before.

Typographic comments:

17. There are several instances of in-line citations still being enclosed by parentheses that should be corrected (P4: L27, L28, L31)

Corrected.

18. P7, L13: “domain ”→“domains”

Corrected.

19. P9, L12-13: “is difficult to be quantified ”→“are difficult to quantify”

Corrected.

20. The paper contains a couple stub sentences without verbs: p4, L14-15 and p8, L27-28

Corrected.

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