Responses to reviews of "Climate-driven chemistry and aerosol feedbacks in CMIP6 Earth system models" by Gillian Thornhill et al.

We would like to thank the two anonymous referees for their useful and supportive comments. Their comments are repeated below reviewer 1 in black, reviewer 2 in blue, with our responses in red.

The paper is an interesting summary of the magnitude of chemistry and aerosol feedbacks in available CMIP6 climate models. The paper is generally well-written, however in its current form the manuscript is somewhat fragmented and some important discussion about is missing. Some aspects of the methodology are described concisely, yet some important details are missing entirely, or are described only briefly. The chemical and aerosol forcing agents are considered independently which helps compartmentalise the results and some of these sections include important insights. However, other sections have not been crafted with the same care.

Thornhill et al. analyse a set of Earth System Model simulations with atmospheric chemistry and aerosol parameterisations to quantify climate feedbacks associated with aerosol and chemistry processes. The methodology allows to attribute the climate feedback to different chemical and aerosol processes and thereby provides in some cases important insights. The paper is highly relevant and fits well to the scope of ACP. The paper is generally well written, but the quality of the individual sections varies considerably.

We thank the reviewers for their positive comments. The comments regarding the fragmentation, missing discussion and individual sections are addressed in responses to specific comments below.

The paper would benefit from merging sections 4 and 5. Currently results from several CMIP6 climate models are somewhat mechanistically portrayed in section 4. Section 5 contains some context for interpreting the differences between models, but uses identical subsection headings and much of the content is more suited to the introduction of a paper on one or more of the forcing agents. The chosen format makes the manuscript unnecessarily disjointed and does not help contextualise the main results. Once sections 4 and 5 are merged, they should be revised to include discussion of the physical processes that cause differences between models. Currently, this is only achieved for one or two of the forcing agents.

I rarely recommend merging results and discussion, but I agree with reviewer #1 that in this case, where a lot of different processes are at play, it would be advisable to merge section 4 and 5 in the sense to have results and discussion for each of the different forcing agents together. The quality of the results presentation and their discussion varies substanially, and the authors should strive to be more explicit in terms of describing and explaining the important differences between models, and where possible provide an appropriate comparison to previous studies. There should then still be an overall discussion section 5/6 in the end where the overall contribution of the non-CO2 chemistry and aerosol feedbacks are discussed in the light of other climate feedbacks (physical, carbon, ...).

We have merged sections 4 and 5 as suggested by both reviewers.

The article has two main themes. Firstly, the differences in aerosol and chemistry forcing efficiency and burden sensitivity are considered. Secondly, the magnitude of feedbacks from forcing agents are contrasted. It is not clear what the authors intended the main message of the paper to be. The abstract provides very few conclusions about either of these aspects and is overly focussed on methane-specific results. If the paper is intended to focus on the second aspect, then the majority of the feedback summary tables could be moved to the SI without reducing the impact of the paper. However, I think it would be better to retain these tables and include a process-based discussion of the causes of model differences as suggested above.

We have rewritten the abstract to more closely reflect the structure and findings of the paper. The focus of the paper is on the quantification of alpha (feedback), which is the product of phi (forcing efficiency) and gamma (sensitivity to climate). Therefore, the phi and gamma terms are equally important. The focus of this paper is not a process-based discussion of model differences. Such discussions could fill whole papers themselves and are to some extent found in the model description papers. Rather the aim of this paper is to demonstrate the contribution of chemistry/aerosol feedback mechanisms to the overall climate feedback in these ESMs. This has brought out more clearly in the abstract and introduction.

The use of standard deviations to represent uncertainty in a handful of models is not appropriate. It is possible that this is not what the authors have done, but their method is currently unclear. The authors need to clarify their multi-model uncertainty calculations in the text and if they are currently using standard deviations to represent uncertainty in only 3-6 values, need to seek more appropriate ways to communicate this information. Currently multi-model uncertainties are communicated through table captions but should be described fully in the main text.

The methods section should be expanded by a description on how the authors have dealt with uncertainty in this study. What do the reported ± ranges represent for individual estimates, how are errors of the multi-model mean derived from these (error propagation of the IAV?), how is the error range of the total forcing estimate determined, how have varying estimates from emission/burden based methods been dealt with in the total feedback assessment.

We have explained more fully where we have used interannual variability and where inter-model variability.

The abstract should list the feedbacks assessed here and should be much more explicit about the major findings of this study (I would assume that this would be a summary of Figure 5). It is unclear to me why the methane effects are highlighted here, while this is not mentioned at all in the Conclusion section. In general, the authors should try to clarify the main messages from this paper in abstract, introduction and conclusions.

We realise that the overall aim of the paper was not entirely clear (see also response to reviewer 1). We have substantially revised the abstract to convey the main messages.

The introduction is somewhat simplistic in that it only lists studies that have attempted to assess non CO2 climate feedbacks. For the general audience and the orientation of the readers it would be helpful to start with a somewhat more detailed description of the major processes and feedbacks considered here and why they matter to the climate system.

We agree this is a useful addition to the introduction. We have added a few sentences of text and references to Sherwood et al. 2020 and Friedlingstein 2015.

The choice of the authors to rely on 4xCO2 experiments to diagnose climate feedback implies that some of the feedbacks considered are less climate change related, but mediated by the effect of CO2 on vegetation productivity and cover. This is an important caveat that should be explained in the Methods section for those processes that do respond to CO2 as well as climate. Also, this needs to be reflected critically in the Conclusions section/Abstract.

This was discussed to some extent in the main text. This has been brought out in the Conclusions.

All figures require subfigure labels as per ACP guidelines, to match references in the captions and main text.

These labels have been added.

Line 34: "with warmer temperatures" needs a fuller description. 4Xco2 induced warming

L35: define warmer temperatures

Accepted: "warmer" has been rephrased as "warmer surface temperatures following a quadrupling of CO_2 concentrations"

L36: positive methane feedbacks?

Accepted: This paragraph has been completely reworded.

Line 37: VOC needs to be defined.

Accepted: BVOC has been defined.

Line 40: GCMs do these things already. ESMs include the interactions between these systems, by coupling them and hence can expect a greater degree of consistency of information across model components. This needs to be clarified in the text.

Accepted: This has been clarified in the text. "Earth system models extend the complexity of physical climate models by coupling land and ocean biospheres, atmospheric chemistry and aerosols to the physical climate"

L44: consider adding Arneth et al. 2010, Nat. Geo (Doi: 10.1038/ngeo905) to this list

Accepted: This has been added.

Line 57: Here and in the conclusions, it is important to mention that some of the forcing agents considered make important climate contributions at the regions scale that are neglected when global mean temperatures are used to represent climate change.

Accepted: We have added sentences mentioning this to the introduction and conclusions.

L72: Briefly explain why you not just use one of the options. I also think that this question deserves more attention in the results section where you for some forcings can compare the magnitude of the alternative estimates more systematically to derive at a joint assessment of the individual feedback factor.

Accepted: We have added text in section 2.2 to explain the reasons for using burdens, AODs or emissions for different species.

Section 2.2: It would be helpful to know which of the feedbacks is calculated which way here. Also, given the need for standardisation here or in the discussion section, there should be a discussion about the assumption of linearity of the radiative forcing response to emissions/burden across a large range of emissions/burden.

Accepted: The discussion of feedbacks has been expanded in section 2.2 to include discussion of linearity.

Which ensemble members were selected for this study, or does the study use an ensemble mean?

Only one ensemble member was run for each of these experiments. This has been clarified in the text.

L86: It is unclear whether this is based on simulations presented in Collins et al. 2017, or based on new AerChemMIP experiments, please clarify.

We have clarified that the analysis here is based on simulations from Collins et al. 2017.

Line 102: The scale factor is not well justified. The cited document is a substantial IPCC chapter. Presumably, authors are referring to section 8.2.3.3? Including the page number would help reader. However, the derivation of the scale factor used here is unclear and some explanatory text is required.

L102: Provide an explanation for this scaling factor rather than referencing a full IPCCchapter

Accepted: More detail on this scaling factor is added to this section. The precise section (8.SM.11.3.2) is provided.

Line 105: The use of the value 9.25 also needs justification and a description of how it corresponds to values supplied in the referenced document.

L105: For completeness, give value assumed for M_atm as well as the molecular masses of CH4 and air

Accepted: The derivation of the methane lifetime has been explained and the physical constants listed with a reference to Prather et al. 2012

Section 3.1 should reference table 2 but does not.

A reference to table 2 has been added.

Section 3.2: natural emissions of what?

This has been clarified to be “of aerosols and ozone precursors”.

Line 110: “four have . . . and three have . . .” is ambiguous. “Three of these four also have. . .” is clearer. Table 2 makes this clear, but is not currently referenced.

Accepted: This has been clarified with a reference to table 2.

Line 119: Table 3 is currently referenced in a way that suggests it will compare emissions from all natural sources, whereas it actually shows differences between models for dust and BVOCs only. The text needs to be revised. This error is repeated on the first line of section 3.2.2.

L119: this sentence needs to be clarified. There are multiple climate-relevant land based emissions beyond dust and BVOCs. What do the authors want to state here? L134: same as L119

Accepted: This has been changed to make it clear that these are the emissions analysed in this study, rather than making any more general claim.

Table 3: “PAR” needs to be defined. The phrase “Not dependent on vegetation” is redundant.

Table 3: define LAI, PAR. Given an indication what LAI varies and interactive vegetation imply. The table captions says BVOC, the header VOC, which is correct?

Accepted: LAI and PAR have defined and the descriptions expanded. The header has been changed to BVOC

Given that Section 4.2.3. discusses wetland emissions, the models used should be described here briefly as well.

Accepted: Wetland models have been described as well.

Table 4: There are inconsistencies in the table. For example, sometimes “wind” is used and at other times “wind speed dependent”. Descriptions here are too brief. What is the difference between DMS emission and oceanic organic aerosol complexity for NorESM2-LM and UKESM1 for example?

Table 4: what is the difference between wind dependent and wind speed?

Accepted: This table has been reworded for consistency, and descriptions expanded.

L147: Does this sentence imply all models use the same paramterisation?

This has been clarified that the implementation of Price and Rind can vary between models.

Section 4, Line 150: Section 2.1 should be referenced in the first paragraph, so that the normalization of temperatures can be put in the context of γi as defined in that section.

L151: refer back to Section 2.1 or remove as this is partly redundant. .

Accepted: We agree reference to γ_i and section 2.1 has been added.

Line 155: For non-specialist readers an indication of the number of years required to reach equilibrium on average is needed.

L155: For the non-expert reader, explain how long the development of a new equilibrium takes and how large the difference on average would be

Accepted: Yes, we added a comment on this taking many centuries.

Table 5: No SD?

We have added standard deviations to this table.

Line 163: Figure S1 does not obviously support this claim. Global mean ERF values should be provided for each model. Also, the authors should explicitly state they are discussing "global mean" effective radiative forcing here.

L163: Figure S1 does not separate shortwave and longwave effects to make this claim.

Accepted: We have added separate maps of LW and SW in the supplement.

Line 165: The strong regional forcing over Africa should be mentioned as the primary cause of positive SW forcing. Some speculation of the process parameterisations that cause this model behavior should be given.

L165: the positive shortwave forcing OF DUST AEROSOLS? Is it possible to provide an explanation for this CNRM response?

Accepted: More description has been added on where models agree or disagree in the LW and SW forcing.

Figure 1 (and subsequent following figures): use stippling or alike to show areas of model dis-/agreement. Also revise figures to ensure the legend is readable without magnifying glasses

Accepted: We have added stippling and increased the size of the legend.

Line 182: Refer to table 6 again. Also, some speculation on the physical processes causing the increased lifetime should be given. This is a good example of the need for additional discussion and how merging, then adapting content from section 5 will improve the interpretation of results. Line 189: It is not clear what the 2nd use of "for instance" here is referring to. This sentence needs to be rewritten to improve clarity.

Accepted: Reference to table 6 has been added along with a comment on the physical processes. The 2nd "for instance" was a mistake and the sentence has been rewritten.

Table 6: The reason for missing values in this and other tables needs to be explained more clearly within the text.

Table 6 (and similar subsequent tables): Why are certain cells blank?

Accepted: Not all models provided all the diagnostics. These have been filled with N/A

Line 200: These forcing values are far larger than for dust. Are the forcing-emission-feedback relationships expected to be linear? If not, there will be discrepancies in the gamma terms across emission types, even if normalised. This assumption on linearity and its implications need to be discussed here and/or in section 2.2.

A comment on the non-linearity has been added to the introduction. For doubled emissions, errors introduced through assuming linearity are likely to be small compared to process uncertainty. Many studies use 5x or 10x emissions.

Line 201: Why 20x? Is this caused by the choice of size bins? This warrants some discussion. Why is the AOD of a similar magnitude? What model processes have been adjusted/tuned to make the AOD similar? The reasons models have similar values for very different reasons need to be better understood. This is important for understanding the causes of model diversity in climate projections.

L201: Why does this discrepancy occur, and how can the AOD be still similar? This paragraph should also have a discussion on why MIROC6 deviates in terms of the ERF response

This is indeed due to CNRM having a bin for larger particles (up to 20 microns) which add to the mass, but not to the AOD. This has been clarified in the text.

Line 205 - 209: All positive except MIROC needs to be explained/considered. What regions show a decrease in emissions that causes the global mean response to be negative? Maps for each model in the SI are needed.

Accepted: Maps for each model have been provided in the supplement. “The global mean change in emissions is positive in all models except MIROC6 and GISS-E2-1 (where the lower latitude decreases outweigh the high latitude increases). “

Line 220: It should be explained here that all models could have run the 2xdms experiment. Interactive ocean biogeochemistry is not a prerequisite, since emissions could have been scaled within the flux parameterisation as with the 2xdust experiment.

Accepted: This has been clarified.

Line 222: Fig 3 does not show the forcing values for each model as implied. Table 8 should be referenced to here.

L222: Figure 3 does not show this.

Accepted: The text will has been clarified that fig 3 shows the multi-model mean, individual maps have been added to the supplement. Table 8 has been referenced.

Line 222-224: Maps of sulphur concentrations and changes in concentrations need to be included as a figure in the SI for each model, so the reader has a clear understanding of the magnitude of regional compensation across models.

Accepted: Individual maps of emissions and ERF have been added to the supplement.

Line 225: GFDL-ESM4 values only contributes to the multi-model sensitivity to emissions/concentrations, but not to the multi-model radiative efficiency. The assumption made here is that all models have similar radiative efficiencies. This is an important assumption, given the diversity of model responses highlighted up to this point in the manuscript. Is it appropriate to assume GFDL-ESM4 has the same radiative efficiency as the two models used in the sensitivity calculation? Some justification is required if the authors want to maintain this approach. An alternative approach would be to only use the 2 models with sufficient information to calculate both the multi-model sensitivity and multi-model radiative efficiency. This subjective choice to include partial information from one model needs to be justified more clearly and the implications of extrapolating the multi-model radiative efficiency to other models needs to be considered and openly discussed.

Accepted: This is a good point, for consistency we decided to use only the 2 models with sufficient information to calculate both the multi-model sensitivity and multi-model radiative efficiency

Line 229: The magnitude of the increase should be quantified in the text.

Accepted: The increase in lifetime has been added to the text.

Table 8: Please check values, at least the alpha emission multi model mean cannot be correct.

All values have been recalculated.

Line 249: Here and elsewhere in the text, the word "significant" is used without mention of associated statistical tests. The values should be state with “significant” removed, or the methodology more accurately described.

Accepted: Significant has been replaced with more appropriate wording unless it specifically refers to a statistical test.

Line 253: Incorrect label. Figure S3 only shows the multi-model mean. Given the diversity in aerosol forcing from this source, maps of CDNC should be provided for each model. Also, interpretation of the differences between models needs to be included here.

L253: Figure S4 does not exist.

This figure came from only one model. We do not have sufficient data from all the models to make CDNC claims so this sentence and figure has been removed.

Line 253-257: Examples of regions where these behaviors are likely, with an explanation of why is needed.

We do not have sufficient data from all the models to explore these points, so these sentences have been removed.

Figure 4: Fig S3 could be a subfigure of Fig 4.

This figure has been removed.

Table 9: Uncertainty values are missing for UKESM1 and multi-model mean values are missing for Scaled Mass

This table has been revised.

Section 4.2 general: I think it would be easier to follow if the indirect effects of NOx and BVOC on methane were discussed jointly and possible even in one table, as they rely on the same methodology and type of experiments.

Accepted: These have put into the same table (table 12).

Line 278: Is there an hypothesis the authors could provide to explain the causes of model diversity in BVOC partitioning into ozone and aerosol forcing? This sort of discussion is essential to develop a better understanding of the importance model differences and will affect interpretation of climate feedbacks across models.

This isn't a partitioning as such as the production of ozone and SOA are through very different mechanisms. Discussion has to be added to the effect that the ozone responses are similar, but the SOA varies more.

L279: BVOC-related aerosols, or aerosols in general?

This has been clarified that this relates to the aerosols from BVOC changes.

L281: refer back to Section 2.2.

Accepted: A reference to section 2.2 has been added.

Table 10: There is no explanation of why 14% is used. This should be in the methods section, not hidden in a caption.

The reference to Etminan has been added to explain the 14% uncertainty.

L297: Methane Burden/Emissions? does not change

This will be clarified to say that the methane concentration does not change.

Line 300-302: This sentence needs to be rewritten to improve readability.

This whole discussion has been rewritten.

Line 302: It is not clear from the text as written, how BVOC burden sensitivities are used in the methane sensitivity calculation.

This has been clarified in the text – it is sensitivity of methane lifetime to BVOC emission in the 2xVOC experiment.

L304: The 0.015 Wm-2 %-1 are not described in Section 2.2. but should be

Accepted: The conversion of lifetime change to ERF change has been added to section 2.2.

Section 4.2.4: The title of this section is misleading. Several non-emission drivers are considered, not just these two.
This has been changed to be "Meteorological Drivers"

Line 265: "1" missing from UKESM1.
This has been corrected

Section 4.2.3 I find this section troublesome given the lack of explanation of the simulated methane emissions, particular because this presentation confounds the direct effects of CO2 on methane emissions (via CO2 fertilisation of wetlands) with the direct effects of temperature on methane-emissions, but exclusively attributes this to temperature. The result of which is an inflated methane-emission climate feedback compared to Ciais et al. 2013. I wonder whether there are simulations with interactive methane but no biogeochemical coupling to CO2 available from the C4MIP project that would allow to tackle this separation? As a minimum, this confounding effect needs to be explained and discussed.
Unfortunately there are no radiation-only 4xCO2 simulations. We have caveated the wetland results, particularly as we have only two models.

Table 14: What is the justification to assume at 14% uncertainty on methane radiative efficiency? Section 4.2.4 should be labelled atmospheric temperature and water vapour?
We have added a reference to section 2.2. which now describes where this uncertainty comes from. The section has ben renamed Meteorological drivers.

L356: the residual is then ASSUMED TO BE the direct effect. This statement could be backed up by a brief explanation that BVOC and NOx are the only agents affecting ozone and methane lifetimes next to climate in these models. Otherwise, it should be explained why other factors may be small and negligible.
Accepted: An explanation of this has been added to the text.

L367: Consistently use CESM-WACCM
Accepted: We have checked for naming consistency.

Section 4.3: This section needs some comment about the importance of climate forcing agents that have climatic importance at the regional scale, to prevent the results of this manuscript being interpreted incorrectly.

The focus of the paper is on the global radiative feedback per K, but we have added text "This analysis (and climate sensitivity in general) is focussed on the global mean, but it should be noted that the cooling effects of increased aerosols will be heterogenous and some regions will experience less warming than a global climate sensitivity might suggest"

Section 4.3: Figure 5 is not referenced. The text needs to be explicit that the feedbacks are the multi-model mean, and that not all feedbacks could be calculated for all processes considered. A discussion that I have been missing here is whether these terms are really additive and linear as assumed. It is possible that there is a compensation of feedbacks between models, so I wonder whether it would be possible / interesting to compare the sum of feedbacks across processes for those models that have calculated similar feedbacks

Figure 5 has been referenced. It is not obvious that there would be significant lack of additivity given these are small changes in composition, however we have added a discussion. "The totals assume that feedbacks are additive, which is the basis of the framework in section 2.1."

Line 380: The authors need to specify that these are multi-model feedbacks, here and in the table caption. Figure 5 needs to be referenced. In addition, the cancellation between models with opposite signs again needs to be mentioned within this section, as does the fact that a different number of models were used to calculate the multimodel means because of data availability.

These are all good points and have been implemented.

Figure 5: use consistent labelling of models. use consistent labelling of forcing factors (e.g. total non-CH4, wetland CH4 etc.) Use a clearer abbreviation for lightning NOx than lNOx. The figure caption should also explain, how and why feedbacks from table 16 were aggregated in the figure.

Accepted: The labelling has been changed, and the caption now describes how feedbacks are aggregated.

Line 402: Can the feedbacks be interpreted in the context of the magnitude of forcing from these forcing agents over some specified period? Uncertainty in these magnitudes should be included in the discussion with appropriate references.

The forcing responses are maintained continuously rather than being for a specific period.

Line 423: There is no use citing these values if not directly comparable. This text should be removed to avoid confusion. Further discussion of the causes of model differences is required here.

Section 5.2 is not helpful is no guidance is given as to the origin of the large range in the estimates and the plausiblity of the different model projections. The comparison to the literature numbers is insufficient in that the numbers aren't directly comparable. This section needs substanial revision.

This discussion has been removed as the numbers aren't directly comparable.

Line 433: Please clarify the difference between primary production and DMS production in the text.

Accepted: This is now referred to as biological activity

Section 5.6 response to my previous comment, but then implies that this shouldn't really be listed here as a climate feedback, but a biogeochemical carbon-methane feedback.

We describe this here as an "adjustment", but we have made it more explicit in the methods and conclusions that it is not necessarily a feedback.

Section 6: I would have liked to see a somewhat more broader discussion of the feedbacks derived here in the context of physical and other biogeochemical feedbacks, as for instance summaried in Ciais et al. 2013.

Accepted: We have compared to both Ciais 2013 and Sherwood 2020 to provide a comparison with other feedbacks.

L500: This is an important caveat that should not be left as a foot note in the conclusion section, as it is a fundamental problem of the approach. I strongly recommend to be more explicit about this in the Methods section, where relevant in Section 4 as well as specifically in the presentation of Figure 5 and Table 16.

Accepted: As with the section 5.6 comment a discussion of "adjustments" has been made more explicit in the Methods.

L503: This is a point worth discussing more. Are the feedbacks non-linear and therefore we expect them to be larger/smaller when looking at the difference between present-day and 4xCO2?

The choice of base state is likely to be important for the forcing efficiencies. We might expect aerosol forcing to be less efficient and ozone production more efficient in the present day. This is now mentioned in the text.

Line 505: This value needs context to aid interpretation. e.g. What is this as a proportion of the GHG forcing required to increase temperatures by 1 degree?

Accepted: We have compared to both Ciais 2013 and Sherwood 2020 to provide a comparison with other feedbacks.

L505 and 507: The uncertainties given are the SD of sum of the multi-model mean feedback components, but there are larger uncertainties in the derivation of these feedback that should be discussed and acknowleged.

Line 507-508: The uncertainties in these values are substantial and need to be included in this discussion and interpretation of results.

Accepted: A discussion of possible systematic uncertainties has been added to section 4.3

SI: S1, some descriptions are missing entirely and need to be included.

These descriptions have been added.

SI: All figures require subfigure labels.

We do not refer to specific subfigures individually here.

Data availability: It would be helpful if the authors would list the exact names of the experiments used, including an indication of the ensemble members selected Please carefully edits and update Table S1

The exact names are as listed in table 1. Table S2 has been added to include this information.

Climate-driven chemistry and aerosol feedbacks in CMIP6 Earth system models

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Abstract.

Feedbacks play a fundamental role in determining the magnitude of the response of the climate system to external forcing, such as from anthropogenic emissions. The latest generation of Earth system models include aerosol and chemistry components that interact with each other and with the biosphere. These interactions introduce a complex web of feedbacks which it is important to understand and quantify.

This paper addresses multiple pathways for aerosol and chemical feedbacks in Earth system models. These focus on changes in natural emissions (dust, sea salt, di-methyl sulphide, biogenic volatile organic compounds (BVOCs) and lightning) and changes in reaction rates for methane and ozone chemistry. The feedback terms are then given by the sensitivity of a pathway to climate change multiplied by the radiative effect of the change. ~~This is achieved by extending previous formalisms which include CO_2 concentrations as a state variable to a formalism which in principle includes the concentrations of all climate-active atmospheric constituents. This framework is demonstrated by applying it to the Earth system models participating in CMIP6 with a focus on the non-CO_2 reactive gases and aerosols (methane, ozone, sulphate aerosol, organic aerosol and dust).~~ We find that the overall climate feedback through chemistry and aerosols is negative in the sixth coupled model intercomparison project (CMIP6) Earth system models due to increased negative forcing from aerosols in a climate with warmer surface temperatures following a quadrupling of CO_2 concentrations. This is principally due to increased emissions of

sea salt and BVOCs which are both sensitive to climate change, and cause strong negative radiative forcings. Increased chemical loss of ozone and methane also contributes to a negative feedback. However overall methane lifetime is expected to increase in a warmer climate due to increased BVOCs. Increased emissions of methane from wetlands would also offset some of the negative feedbacks. The CMIP6 experimental design did not allow the methane lifetime or methane emission changes to affect climate so we find a robust negative contribution from interactive aerosols and chemistry to climate sensitivity in CMIP6 Earth system models. ~~Through diagnosing changes in methane emissions and lifetime we find that if Earth system models were to allow methane to vary interactively, methane positive feedbacks (principally wetland methane emissions and biogenic VOC emissions) would offset much of the aerosol feedbacks.~~

1 Introduction

Climate feedback quantifies the change in the Earth's radiation budget as the surface temperature varies. Overall this feedback must be negative for a stable climate, i.e. the net radiation budget must decrease as surface temperature increases. The dominant negative feedback comes from increased long wave emission from a warmer surface (Planck response). Warmer surface temperatures lead to changes in the physical climate system (water vapour, lapse rate, surface albedo, clouds) that further modify the radiation budget contributing additional positive and negative feedbacks (Sherwood et al., 2020). Earth system models extend the complexity of physical climate models by ~~representing~~ coupling land and ocean biospheres, atmospheric chemistry and aerosols to the physical climate. Within these models, natural processes, chemical reactions and biological transformations respond to changes in climate; and these processes in turn affect the climate. Therefore, the physical climate system and the biogeochemical cycles are coupled, leading to climate feedbacks that may act to further amplify or dampen the climate response to a climate forcing (Arneth et al., 2010; Ciais et al., 2013; Heinze et al., 2019). The importance of biogeochemical feedbacks has long been recognised for the longer timescales involved in paleoclimate studies, but the realisation of their relevance in the context of anthropogenic climate change is more recent. A multitude of biogeochemical feedbacks have been identified but the evaluation of their importance for future climate change remains very limited. A recent review of Earth system feedbacks (Heinze et al., 2019) examined the extensive range of feedbacks possible in an Earth system framework. The largest biogeochemical feedback contribution comes from the carbon cycle (Friedlingstein, 2015). Arneth et al. (2010) considered a range of terrestrial biogeochemical feedbacks interacting with the carbon cycle. O'Connor et al., (2010) reviewed potential feedbacks involving methane. Carslaw et al. (2010) reviewed climate feedbacks involving natural and anthropogenic aerosols. Climate change can impact both the source strength of natural aerosols such as sea-salt, dust, biomass burning aerosols, or their precursors (di-methyl sulphide (DMS), biogenic volatile organic compounds) and the lifetime of natural and anthropogenic aerosols through changes in transport and dry and wet deposition (Bellouin et al., 2011; Raes et al., 2010). Here we choose to focus especially on those feedbacks that are mediated through changes in the abundances of reactive

gases and aerosols, using data from CMIP6 (Coupled Model Intercomparison Project 6) (Eyring et al., 2016) Earth system models that conducted the AerChemMIP (Aerosols and Chemistry Model Intercomparison Project) simulations (Collins et al., 2017).

Note that in this paper we use change in global mean surface temperature as our measure of climate change and for simplicity assume changes in other climate variables are proportional to this. For many of the forcing agents considered here the forcing pattern varies strongly on regional scales, and would be expected to cause larger regional temperature changes than represented by the global mean.

In section 2 we describe the theoretical framework used to diagnose the feedbacks. In section 3 we describe how the different Earth ~~System~~ system models implement the biogeochemical processes. Section 4 quantifies the feedbacks as implemented in the models, and ~~. Section 5~~ compares these results with previous modelling and theoretical studies. Section ~~6~~ 5 concludes. Supplementary material contains further details of the models used, and additional figures to support the analysis in section 4~~the process analysis of responses to dust and BVOCs~~.

2 Theoretical framework to analyse biogeochemical feedbacks

2.1 Theory

In order to compare climate feedbacks we need to compare them on a common scale of the change in the top of atmosphere radiation balance following a unit warming (in W m^{-2} K^{-1}) (e.g. Gregory et al., 2009). Following Gregory et al. (2004) the radiative imbalance ΔN from an imposed forcing ΔF is given by $\Delta N = \Delta F + \alpha \Delta T$ where $\Delta T$is the global mean change in surface temperature and α is the climate feedback parameter ($= \frac{d\Delta N}{d\Delta T}$). The total derivative $\frac{d\Delta N}{d\Delta T}$ can be split into a set of partial derivatives $\frac{d\Delta N}{d\Delta T} = \sum_i \frac{\partial \Delta N}{\partial \Delta C_i}\frac{\partial \Delta C_i}{\partial \Delta T} = \sum_i \alpha_i$, where the α_i are the individual feedback terms due to a change in a climate variable C_i. For feedbacks involving changes in composition, the ΔC_i can represent changes in reactive gas or aerosol burdens or emissions. $\alpha_i = \frac{\partial \Delta N}{\partial \Delta C_i}\frac{\partial \Delta C_i}{\partial \Delta T}$ can then be expressed as $\phi_i \gamma_i$, where ϕ_i is the radiative efficiency of the species per burden (Wm^{-2} Tg^{-1}) or per emission (Wm^{-2} $(Tg\ yr^{-1})^{-1}$), and γ_i is the change in species burden or emission with climate (Tg K^{-1} or Tg yr^{-1} K^{-1}). The radiative efficiencies are based on effective radiative forcing (ERF) (Myhre et al., 2013a) to include rapid adjustments to changes in composition. Since climate change can also affect the atmospheric lifetime of a species $\frac{\partial \Delta \text{Burden}_i}{\partial \Delta T}$ does not necessarily scale with $\frac{\partial \Delta \text{Emission}_i}{\partial \Delta T}$.

2.2 Applying the theory to Earth system models

With Earth system models, the $\phi_i$and γ_i coefficients can be diagnosed from idealised simulations in which only climate or composition are changed. Here we use the set of simulations specified under the CMIP6 project (Eyring et al., 2016).

The γ_i are diagnosed from a pair of idealised climate change scenarios, a control climate *piControl* where composition is maintained at a level representative of 1850 conditions, and a warmer climate *abrupt-4xCO2* where temperatures have increased following an abrupt quadrupling of CO_2 ~~concentrations are abruptly quadrupled, but no other species are directly perturbed~~. To quantify the sensitivities to this temperature change, ~~W~~we take the 30-year time means from years 121-150 of these simulations for both the surface temperature change and the burden/emission changes. The global mean surface temperature changes are therefore not the same as the equilibrium climate sensitivities (ECSs) derived from the *abrupt-4xCO2* but are temperatures consistent with the averaging period for the burden or emissions. The γ_i are calculated from the change in emission or burden divided by the temperature change. For the dust and sea salt (these are the aerosols with single sources), rather than the burden we diagnose the AOD change (K^{-1}) where available as being the quantity most closely related to the radiative forcing (Myhre et al., 2013b). For DMS and organic aerosol emissions we use the emission change (Tg yr^{-1} K^{-1}) as changes in in aerosol lifetime will also affect AODs from other sources of sulphate and OA that we do not have ERF calculations for. For reactive gases, both emissions-based and concentration-based calculations are used. CO_2 can have climate effects beyond its global warming, for instance CO_2 directly cools the stratosphere and can affect vegetation with implications for dust and BVOCs. With the AerChemMIP setup it is not possible to distinguish these adjustments to CO_2 concentration from the impacts of surface temperature increase.

The ϕ_i coefficients for changes in emissions are derived from pairs of the AerChemMIP simulations defined in Collins et al. (2017), *piClim-control* where composition and climate are maintained at a level representative of 1850 conditions, and experiments *piClim-2x* (table 1) in which individual natural emission fluxes are doubled. The climate change in these simulations is restricted by using fixed sea surface temperatures and sea ice cover (Collins et al., 2017) for a 30-year mean of the *piControl* simulation. The ERFs are determined by the mean difference in top of atmosphere radiative fluxes between the *piClim-2x* and the *piClim-control* over a 30-year period. The ϕ_i are calculated from the ERF divided by either the change in AOD ~~emission~~ or change in emissions~~burden~~, depending on the units of γ_i above. The specific simulation variant numbers are listed in table S2.

The theoretical framework in section 2.1 is inherently linear whereas the Earth system may well not be. The climate changes used to diagnose γ_i are of the order 4-7 K (table 5) which are much larger than the remaining ~ 0.5-1 K goals of the Paris agreement. The doubled natural emission changes used to diagnose ϕ_i are larger than the changes found in the 4xCO_2 experiments and larger still than expected from a climate following the Paris goals.

Experiment	Flux to be doubled
piClim-control	None
*piClim-**2xdust***	Dust
*piClim-**2xss***	Sea salt
*piClim-**2xDMS***	Oceanic DMS
*piClim-**2xNOX***	Lightning NO_X
*piClim-**2xVOC***	Biogenic VOCs

Table 1: List of simulations for diagnosing ERFs of natural emitted species. The specified natural emission fluxes are doubled compared to the 1850 control.

For ϕ_{O_3}, the ozone radiative forcing (tables 10 and 11) is diagnosed from the changes in the 3D ozone distributions multiplied by a 3D kernel of ozone radiative efficiencies from Skeie et al. (2020). The uncertainty in radiative transfer modelling was estimated to be only 10% in Stevenson et al. (2013), but we increase that to 15% as a conservative estimate comparable to the 14% radiative modelling uncertainty for methane (Etminan et al., 2016). Radiative modelling uncertainties are negligible compared to the other uncertainties in section 4.~~a radiative efficiency of 0.042 W m^{-2} per Dobson Unit (DU) is used in the troposphere (Stevenson et al., 2013).~~

The ESM setups here, even with tropospheric chemistry, still constrain methane to specified concentrations at the surface. This means that any feedbacks mediated through changes in oxidising capacity have a negligible effect on methane. It is however possible to diagnose the change in methane that would be expected, if it were not constrained, from the change in its lifetime $\frac{\Delta C}{C} = \left(\frac{\Delta \tau}{\tau} + 1\right)^f - 1 \approx f\frac{\Delta \tau}{\tau}$, where C is the methane concentration, τ is the total methane lifetime (including loss to soils) and f is the feedback of methane on its own lifetime (Fiore et al., 2009). The effective radiative forcing from the change in concentration is 7.0×10^{-4} Wm^{-2} ppb^{-1}, calculated using the formula from Etminan et al.~~,~~ (2016) from a methane baseline of 802 ppb representative of 1850 (Myhre et al., 2013a), this is scaled by 1.52~~65~~ to account for the additional chemical production~~change~~ of~~in~~ ozone (0.4) and stratospheric water vapour (0.12). These values are reduced from the 0.5 and 0.15 in Myhre et al. (2013a) (section 8.SM.11.3.2) as the 25% increase in radiative efficiency from Etminan et al. (2016) does not affect the ozone or water vapour. This gives 1.~~15~~ 11 W m^{-2} per fractional change in methane lifetime~~-~~ or 0.011 W m^{-2} %$^{-1}$~~(based on 1850 baseline concentrations of methane and N_2O)~~. Changes in methane concentration due to changes in emissions ΔE are given by $\Delta C = \Delta E \tau f \left(\frac{m_{\text{air}}}{m_{\text{CH}_4}}\right) / M_{\text{atm}}$, where τ=9.~~25~~ 1 years (Prather et al., 2012), and f=1.34 (Myhre et al., 2013a). m_{air} and m_{CH_4} are the relative molecular masses of air and methane (28.97 and 16.0).

3 Model descriptions

3.1 Model implementation of aerosols, tropospheric and stratospheric chemistry

We use results from ~~6~~ 7 Earth system models that contributed simulations under the AerChemMIP *piClim-2x* experimental setup. All ~~six~~ seven models have interactive aerosol schemes, ~~four~~ five have interactive stratospheric chemistry ~~and~~ four of which~~three~~ also have interactive tropospheric chemistry (table 2). The level of sophistication of the chemistry can affect the modelled responses to the emissions of reactive gases. For instance, in models without interactive tropospheric chemistry changes in biogenic volatile organic compound emissions (BVOCs) affect only organic aerosols, whereas in models with interactive tropospheric chemistry they also affect ozone, methane lifetime, and potentially the oxidation of other aerosol precursors. For each model one ensemble member was run for each experiment.

	Tropospheric chemistry	Stratospheric chemistry	Reference
NorESM2	No	No	(Kirkevåg et al., 2018; Seland et al., 2020)
UKESM1	Interactive	Interactive	(Archibald et al., 2019; Sellar et al., 2019)
CNRM-ESM2-1	No	Interactive	(Michou et al., 2020)
MIROC6	No	No	(Tatebe et al., 2019)
GFDL-ESM4	Interactive	Interactive	(Horowitz et al., in prep)
CESM2-WACCM	Interactive	Interactive	(Gettelman et al., 2019)
GISS-E2-1	Interactive	Interactive	(Bauer et al., 2020)

Table 2 Sophistication of gas-phase chemistry used in the Earth system models (For further details see Thornhill et al. (submitted) **.**

3.2 Model implementation of natural emissions of aerosols and ozone precursors.

3.2.1 Land

The ~~principle~~ land-based natural emissions analysed here are dust, ~~and~~ BVOCs and wetland methane (table 3).

Dust emissions are parameterised as a function of surface wind speeds or wind stress, and account for the amount of bare soil, soil type, and aridity (Ackerley et al., 2012; Collins et al., 2011; Evan et al., 2014; Fiedler et al., 2016; Huneeus et al., 2011; Shao et al., 2011; Zender et al., 2004). There is a variation between the models in the sizes considered, whether binned or

modal, and the optical properties of the dust particles (Kok et al., 2018; Xie et al., 2018). Table S1 lists the parameterizations for desert-dust aerosol for the contributing models and the simulated dust-aerosol sizes.

BVOC emissions are parametrised as a function of vegetation type and cover, and also temperature and photosynthesis rates (gross primary productivity) (Guenther, 1995; Pacifico et al., 2011; Sporre et al., 2019; Unger, 2014). Some parameterisations also include dependence on CO_2 concentrations (Pacifico et al., 2012). Models differ in the speciation of the VOCs emitted but typically include isoprene and monoterpenes, with different emission parameterisations for different species. The ability of VOCs to form secondary organic aerosol are typically parameterised as a fixed yield (Mulcahy et al., 2019). For further details see table S1 and references therein.

	Dust	BVOC	Wetland methane
NorESM2	Interactive LAI, soil moisture, wind speed ~~varies~~	Dependence on PAR, temperature, LAI, vegetation type	N/A
UKESM1	Interactive vegetation (Interactive LAI, soil moisture, bare soil fraction)	Dependence on PAR, temperature, vegetation	Dependent on wetland fraction available substrate and temperature:
CNRM-ESM2-1	Prescribed annual land cover (Séférian et al, 2019)	Prescribed SOA climatology~~Prescribed~~	N/A~~Not calculated interactively~~
MIROC6	LAI from Land-surface model MATSIRO (Takata et al. 2003) ~~varies~~	Prescribed	N/A
GFDL-ESM4	Depends on simulated vegetation (LAI and SAI, used to calculate "bareness" fraction), land use, snow cover, wind speed~~Interactive vegetation~~	Externally prescribed LAI, vegetation type and PAR~~Dependence on PAR, temperature. Not dependent on vegetation.~~	N/A
CESM2-WACCM	LAI, wind friction velocity, soil moisture, vegetation/snow cover ~~varies~~	Dependence on PAR, temperature	Dependent on inundation, water table, temperature and soil decomposition.
GISS-E2-1	LAI, Vegetation, wind speed, soil moisture	Dependence on PAR, vegetation, temperature	Prescribed emissions, parameterized by temperature and precipitation

Table 3 Levels of complexity of vegetation included in the land-based emissions schemes of dust and BVOCs for the ESMs~~.~~, including dependence on photosynthetically active radiation (PAR) and leaf area index (LAI).

3.2.2 Marine

The ~~principle~~ ocean emissions analysed here are ~~of~~ sea salt, di-methyl -sulphide (DMS) and primary organic aerosols (table 4).

The air-sea exchange processes for these emissions are parameterised as a function of wind speed and sometimes temperature (Gong, 2003; Jaeglé et al., 2011).

Changes in DMS emissions can be initiated by various factors such as changes in temperature, insolation, depth of the ocean-mixed layer, sea-ice extent, wind strength, nutrient recycling, or shift in marine ecosystems (Heinze et al., 2019). The ~~surface sea-water concentrations of DMS~~DMS fluxes into the atmosphere are prescribed in some models (CNRM-ESM2-1, GFDL-ESM4, MIROC6, CESM2-WACCM) and calculated interactively from ocean biogeochemistry in others (UKESM1, NorESM2). Oceanic organic aerosol emissions are also wind-speed dependent and in addition depend on chlorophyll concentrations generated either from interactive biogeochemistry or observation-based chlorophyll concentrations in models without ocean biogeochemistry components.

	Sea salt	DMS	Oceanic organic aerosol
NorESM2-LM	Temperature and wind speed ~~dependent~~	Interactive biogeochemistry for sea water DMS concentration, wind speed and temperature for air-sea DMS flux	Climatology for chlorophyll concentration; dependent on wind speed and temperature~~Interactive biogeochemistry for Chlorophyll concentrations; wind speed; sea salt emission flux~~
UKESM1	Wind speed	Interactive biogeochemistry for sea water DMS concentration, wind speed and temperature for air-sea DMS flux	Interactive biogeochemistry, flux dependent on wind speed and temperature
CNRM-ESM2-1	Temperature, wind speed	Prescribed climatological emissions	None
MIROC6	Wind speed	Dependent on ~~wind speed and chlorophyll~~surface downward solar radiation	Climatology for chlorophyll concentration; dependent on wind speed ~~Dependent on wind speed and chlorophyll~~
GFDL-ESM4	Temperature, wind speed	Wind speed for air-sea DMS flux. Prescribed sea water concentration	Wind speed
CESM2-WACCM	Temperature, Wind speed	Wind speed and temperature for air-sea DMS flux.. Prescribed sea water concentration	Wind speed
~~CESM2-WACCM~~GISS-E2-1	Temperature, wind speed~~Wind speed~~	Wind speed and temperature for air-sea DMS flux. Prescribed sea water concentration ~~Wind speed, sea water concentration~~~~Wind speed. Prescribed sea water concentration~~	None~~Wind speed~~

Table 4 Levels of complexity of marine emissions in the ESMs

3.2.3 Lightning

The models with tropospheric chemistry (UKESM1, GFDL-ESM4, CESM2-WACCM, GISS-E2-1) all include parameterisations of the emission of nitrogen oxides (NO_X) from lightning, related to the height of the convective cloud top (Price et al., 1997; Price and Rind, 1992). The lightning frequency depends strongly on the convective cloud top height, and the ratio of cloud-to-cloud versus cloud-to-ground lightning depends on the cold cloud thickness (from 0°C to the cloud top). The precise implementation of lighting emissions and their height profile varies between the models.

4 Quantification of feedbacks

The feedbacks in this section are all derived from the difference between the *piControl* and *abrupt-4xCO2* CMIP6 experiments. The Earth system models all respond with different levels of climate change, so all climate feedbacks are normalised to the change in global mean surface temperature between *abrupt-4xCO2* and *piControl* for the 30-year period years 121-150 (table 5) to derive the γ_i (section 2.1). There is a factor of nearly two between the temperature responses of the models. Since this timeframe is not long enough for the models to have reached equilibrium (which may take many centuries) these temperatures are not the same as equilibrium climate senstivity (ECS).

	~~CNRM-ESM2~~CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	CESM2-~~-~~WACCM	GFDL-~~-~~ESM4	GISS-E2
ΔT 4xCO2 (K)	6.09 ±~~+/-~~ 0.12~~2~~	7.46 ±~~+/-~~ 0.17	4.01 ±~~+/-~~ 0.2	3.96 ±~~+/-~~ 0.19	6.~~37~~49 ±0.21	3.93 ±0.16	3.81 ±0.17

Table 5: Change in global mean surface temperature following an abrupt quadrupling of CO_2 concentrations. Difference between *abrupt-4xCO2* and *piControl* averaged over the years 121-150. Uncertainties refer to standard deviation of the interannual variability.~~, with s.d.~~.

4.1 Aerosol species

4.1.1 Desert Dust

The *2xdust* perturbation is applied by scaling the parameterisation in the emission scheme. Since changing dust emissions will affect the boundary layer meteorology the net effect is not an exact doubling of the emissions (table 6). Four of the ~~five~~ six models in AerChemMIP have a negative radiative forcing for doubled dust ~~aerosols as the negative shortwave radiative effects outweigh the positive longwave radiative effects of dust aerosols~~ (figure~~s~~ 1(a), -figures S2~~1~~-4~~(a-e);~~ table 6). The models all agree on a negative ERF over the oceans close to the source~~s~~ regions. They differ in the sign of the ERF over the deserts themselves, most (4 out of six) showing a positive longwave ERF (figure S4). The shortwave ERF is more variable (figure S3) and is also affected by any changes in low cloud amount. For~~The only exception is~~ ~~CNRM-ESM2-~~CNRM-ESM2-1 and UKESM1~~, where the global shortwave forcing is also positive, explaining the different sign of the ERF compared to the other models~~this positive ERF over the deserts outweighs the oceanic negative ERF. The ERF for GFDL-ESM4 is not significantly different from zero. UKESM1 has by far the largest dust emissions (and change from doubling) because it includes particles that are emitted and deposited in the same timestep. ~~CNRM-ESM2-~~CNRM-ESM2-1 also includes large particles (up to 2~~5~~0 μm). These models however have similar changes in dust aerosol optical depth (AOD) compared to the other models and hence the magnitude of the forcing efficiency per change in AOD (table 6) is not out of line with the others. MIROC6 has the

strongest forcing even with the lowest emissions and smallest change in AOD, thus giving it the largest forcing efficiency per AOD. ~~(figure S1 (f-j)).~~

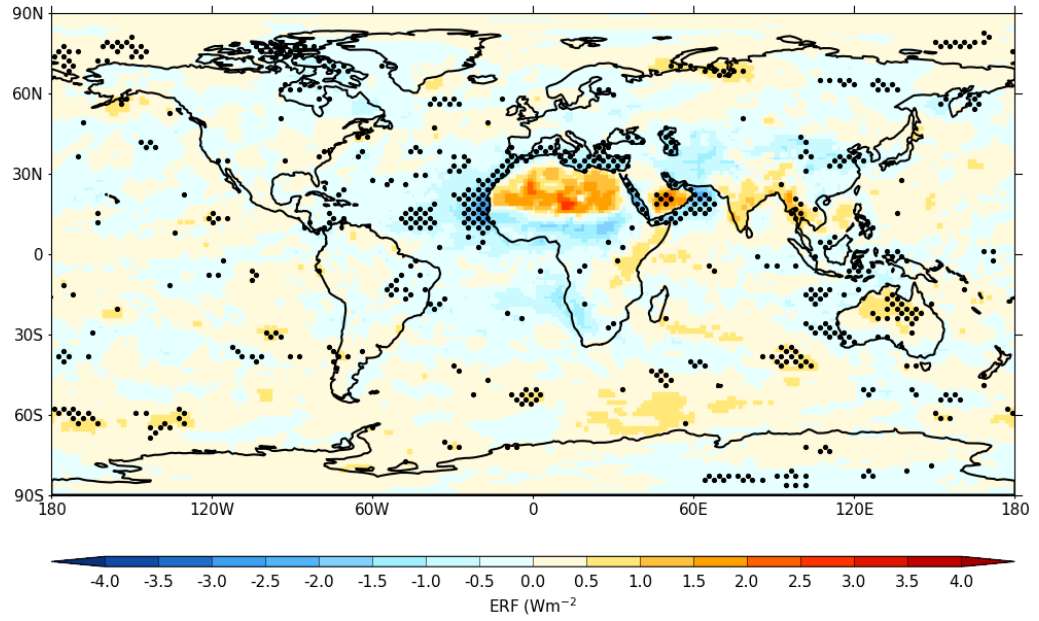
The response of dust aerosols to *abrupt-4xCO2* (figure 1(b), figure S1) is substantially different across the model ensemble. ~~Three~~ Four models (~~CNRM-ESM2~~CNRM-ESM2-1, MIROC6, ~~and~~ GFDL-ESM4 and GISS-E2) show an increase in dust emission in a *4xCO2* climate due to increased aridity and near-surface wind speeds, whereas UKESM1 has a decrease in dust emissions with more CO_2 due to increased fertilisation of the vegetation (hence less bare soil) paired with decreased near-surface winds. NorESM2 shows near zero change. The spatial pattern of the opposing response of dust emission to *4xCO2* in the two most extreme models, UKESM1 and ~~CNRM-ESM2~~CNRM-ESM2-1, is consistent with the responses in ~~10m~~near-surface-wind speed to *4xCO2* (figure ~~S2~~S5). These ~~clearly~~ reflect larger ~~(smaller)~~ increases in mean winds over regions where the mean emission amount is larger ~~(smaller)~~ for *4xCO2* compared to the pre-industrial climatology. The increase or decrease in winds is also likely to be affected by changes in vegetation in semi-arid regions, e.g., the Sahel.

As well as affecting the emissions, changing climate can also affect the removal of dust through changes in both dry and wet deposition. In all models except UKESM1 the lifetime of dust increases (table 6). The effect of an increase in lifetime can be seen by comparing the change in AOD. The modelled changes in dust AOD in the *abrupt-4xCO2* experiment are one and a half to twice as large (for those models where lifetime increases) as would be expected assuming a linear scaling with emissions across all size ranges ("scaled AOD" in table 6).

The climate feedback parameter for dust (α) is given by the product of the radiative efficiencies (ϕ) with the sensitivities to climate (γ). These vary from -0.~~016~~012 to +0.~~048~~0020 W m^{-2} K^{-1} with a multi-model mean of -0.~~003~~0026±0.~~008~~0048 W m^{-2} K^{-1}, i.e. ~~averaging to a value near~~consistent with zero. Scaling with AOD change rather than emission change gives a slightly larger magnitude with a range -0.016 to +0.0048 W m^{-2} K^{-1} and a multi-model mean of -0.0040±0.0072 W m^{-2} K^{-1} Although some models obtain similar feedback terms, this is not necessarily for the same reason. ~~For instance, CNRM-ESM2 and UKESM1 have a positive dust feedback, though for opposite reasons; an increase in positive forcing in CNRM-ESM2 and a decrease in negative forcing in UKESM1.~~ For instance GFDL-ESM4 and~~,~~ NorESM2 have small feedback terms. NorESM2-LM has a large ERF for doubled dust emissions~~,~~ but a~~the~~ small change in dust emission for *4xCO2*, whereas GFDL-ESM4 has a large change in emissions but a small ERF~~however, does not lead to a large feedback for that model~~.

Dust-aerosol feedback assessments are a relatively new area of research owing to the large uncertainties of climate models in simulating dust aerosols with changes in atmospheric composition. For instance, the spread in model estimates for dust aerosol changes in the 21st century is the largest among wildfires, biogenic SOA and DMS sulphate (Carslaw et al., 2010). Predictions for future dust emission range from an increase (Woodward et al., 2005) to a decrease (Mahowald and Luo, 2003). The modelled feedbacks in table 6 are smaller in magnitude compared to the theoretical model estimates of −0.04 to +0.02 Wm^{-2} K^{-1} by Kok et al. (2018).

(a)



(b)

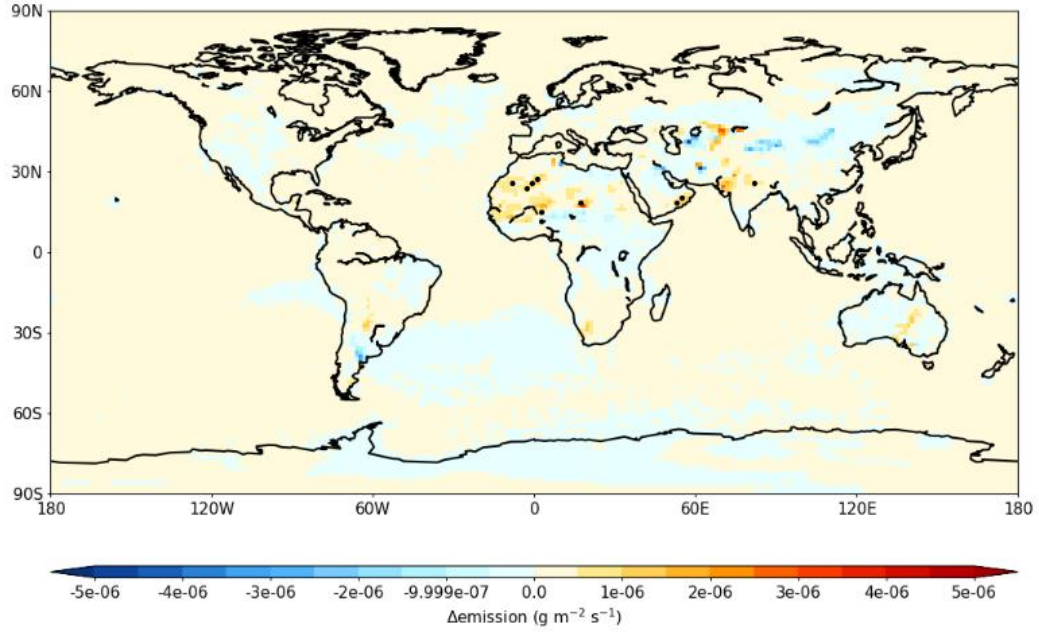


Figure 1 Multi-model mean (a) ERF from *piClim-2xdust* vs *piClim-control*, (b) Change in dust emissions for *abrupt-4xCO2* vs *piControl*. Stippling shows areas where the mean changes by more than the standard deviation across models.

	~~CNRM-ESM2~~CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2	Multi model
Emission *control* Tg yr^{-1}	2750	7875	1106	1661	1981	1765	
ΔEmission *2xdust* Tg yr^{-1}	2877	8185	1065	1397	1989	1236	
ERF *2xdust* W m^{-2}	0.09 ±0.03	0.03 ±0.03	-0.18 ±0.04	-0.14 ±0.07	-0.00 ±0.03	-0.10 ±0.04	-0.05 ±0.1
ERF/ Emission W m^{-2} $(Tg\ yr^{-1})^{-1}$	3.1 ±1.0 E-5	3.8 ±3.7 E-6	-1.7 ±0.4 E-4	-1.1 ±0.5 E-4	-0.2 ±1.5E-5	-8.2 ±3.0 E-5	
ERF/ AOD W m^{-2}	8.0 ±2.7	2.4 ±2.4	-25.6 ±5.6	-6.0 ±2.8	-0.2 ±1.6	-5.3 ±2.0	-4.4 ±10.6
ΔEmission/ ΔT Tg yr^{-1} K^{-1}	65 ±4	-109 ±15	70 ±7	-6 ±6	181 ±10	64 ±9	44 ±88
Δlifetime/ ΔT % K^{-1}	2.6 ±0.2	-0.4 ±0.4	1.9 ±0.9	1.0 ±0.5	3.7 ±0.6	1.6 ±0.8	1.7 ±1.3
scaled AOD/ ΔT K^{-1}	2.5 ±0.2 E-4	-1.7 ±0.2 E-4	4.8 ±0.4 E-4	-1.1 ±1.1E-4	17.3 ±1.0 E-4	9.8 ±1.4 E-4	5.2 ±6.6 E-4
4xCO2 ΔAOD/ΔT K^{-1}	6.0 ±0.3 E-4	-2.6 ±0.6 E-4	6.3 ±0.5 E-4	N/A	26.5 ±1.3E-4	14.6 ±1.6E-4	10.1 ±9.8 E-4
α emissions W m^{-2} K^{-1}	0.0020 ±0.0007	-0.0004 ±0.0004	-0.012 ±0.003	0.0007 ±0.0007	-0.0004 ±0.0027	-0.0052 ±0.0021	-0.0026 ±0.0048
α AOD W m^{-2} K^{-1}	0.0048 ±0.0016	-0.0006 ±0.0006	-0.016 ±0.004	N/A	-0.0006 ±0.0042	-0.0077 ±0.0030	-0.0040 ±0.0072

Table 6. Dust radiative efficiencies by emission and AOD from *2xdust* experiments. Changes in emission and AOD from *abrupt-4xCO2*. "scaled" refers to scaling the *2xdust* relations between AOD and emissions by the *4xCO2* changes in emissions. Alpha values are calculated assuming ERF is proportional to emissions or AOD. Uncertainties for each model are errors in the mean based on interannual variability. Uncertainties in the multi-model results are standard deviation across the models. "N/A" signifies that

diagnostic was not available from that model. The multi-model α terms are the average of the individual model α rather than the product of the multi-model ϕ and γ. Multi-model means are not shown for the emissions as some models include coarse particles whereas others do not.

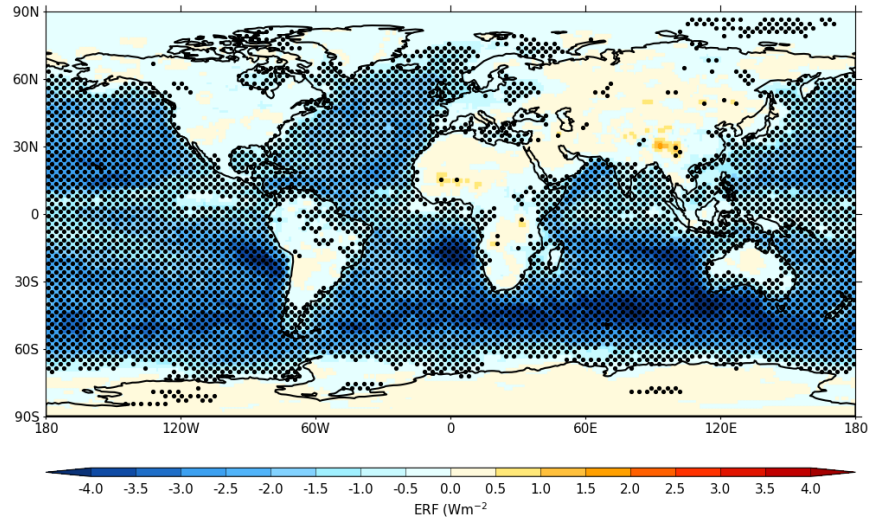
The model ranges in dust forcing and feedbacks are not surprising in light of past studies that highlight model differences in dust-emitting winds and dust-aerosol parameterizations that contribute to the model diversity in the dust-aerosol loading, optical properties, and radiative effects (Ackerley et al., 2012; Evan et al., 2014; Huneeus et al., 2011; Shao et al., 2011; Zender et al., 2004). For instance, the parameterization of the planetary boundary layer plays an important role in determining the dust loading, forcing, and regional feedbacks on winds (Alizadeh Choobari et al., 2012). Influencing factors for regional differences in the dust radiative effects are the surface albedo and aerosol size distribution (Kok et al., 2018; Xie et al., 2018), whereas feedbacks on winds depend also on meteorological factors (Heinold et al., 2008). The substantial model differences in the dust emission response to *4xCO2* paired with corresponding differences in mean 10m-wind speed in this study suggests that also the dust feedback parameter critically relies on accurately simulating atmospheric dynamics. Modelling atmospheric circulation has been identified as a grand challenge in climate research (Bony et al., 2015). Currently, we have no estimate which of the dust feedbacks shown are the most plausible, because convective dust storms are missing in such models, but this dust storm type is believed to be important for North African dust emissions (Heinold et al., 2013). Moreover, natural aerosol-climate feedbacks are thought to depend on the anthropogenic aerosol burden and might therefore be both time-dependent and underestimated in the present-day polluted atmosphere (Spracklen and Rap, 2013). Taken together, we have a low confidence in the feedback estimates for dust aerosols to increases in atmospheric concentrations of greenhouse gases.

4.1.2 Sea Salt

All models show a strong negative forcing to double sea salt emissions (figure 2(a), figure S7, table 7), although the ERF for MIROC6 is considerably~~significantly~~ smaller than the others. The emissions and mass loading for the ~~CNRM-ESM2-~~CNRM-ESM2-1 model are approximately twenty times those of the other models, largely due to including a size bin up to 20 μm. This coarse bin contains a large mass but a lower number of particles, so ~~although~~ the AOD change is similar to other models. All models show a similar forcing efficiency per AOD change. All models show an increase in sea salt emissions in the Southern Ocean in *4xCO2* (figure 2(b), S6) due to increased wind speeds, with a general tendency for decreases elsewhere due to rising temperatures (Jaeglé et al., 2011). The global mean change in emissions is positive in all models except MIROC6 and GISS-E2-1 (where the lower latitude decreases outweigh the high latitude increases). For models showing an increased sea salt lifetime in a 4xCO2 climate the modelled increase in AOD is larger than that expected from scaling the emissions change ("Scaled AOD" in table 7). Although emissions (and the mass burdens) of sea salt decrease in MIROC6 and GISS-E2-1 the AODs increase~~s~~. The mean feedback is -0.~~031~~027±0.0~~31 ~~32 W m^{-2} K^{-1} based on emissions, and -0.~~060~~049±0.05~~6~~0 W m^{-2} K^{-1}

based on the increase in AOD. The signs are consistently negative except for the emission-based feedbacks for MIROC6 and GISS-E2-1.

(a)



(b)

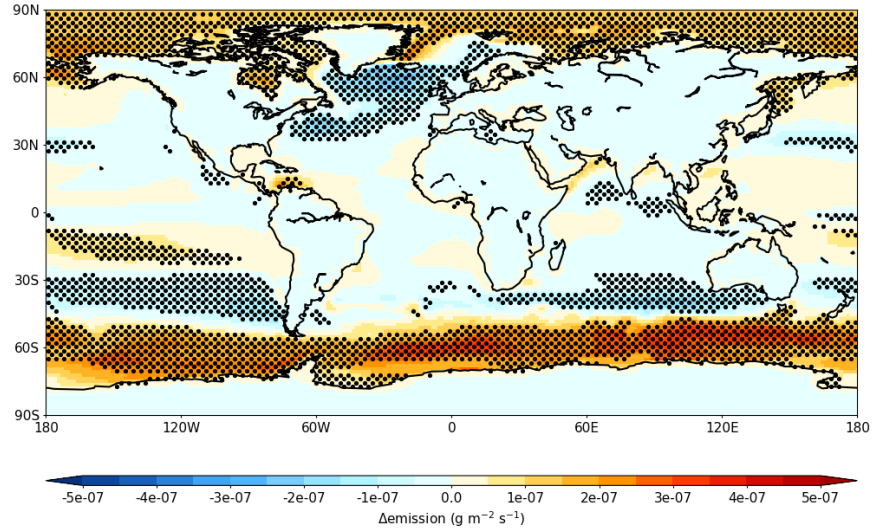


Figure 2 Multi-model mean (a) ERF from *piClim-2xss* vs *piClim-control*, (b) Change in sea-salt emissions for *abrupt-4xCO2* vs *piControl*. ~~CNRM-ESM2-~~CNRM-ESM2-1 emissions are excluded from the multi-model emissions in panel (b) as they include a coarse bin which dominates. Stippling shows areas where the mean changes by more than the standard deviation across models.

	~~CNRM-ESM2~~CNRM-ESM2-1	UKESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2-1	Multi-model
ΔEmission *2xss* Tg yr^{-1}	64939	5500	3577	3771	5675	2624	
ERF *2xss* W m^{-2}	-1.04 ±0.03	-1.27 ±0.03	-0.35 ±0.04	-2.28 ±0.07	-1.84 ±0.03	-1.30 ±0.03	-1.35 ±0.61
ERF/ Emission W m^{-2} (Tg yr^{-1})$^{-1}$	-1.61 ±0.04 E-5	-2.30 ±0.05 E-4	-9.72 ±1.12 E-5	-6.0 ±0.2 E-4	-3.20 ±0.07 E-4	-5.00 ±0.13 E-4	-2.95 ±2.08 E-4
ERF/AOD W m^{-2}	-19.8 ±0.6		-25 ±3	-26 ±0.8	-38.7 ±0.8	-8.4 ±0.8	-23.5 ±9.8
ΔEmission/ΔT Tg yr^{-1} K^{-1}	2570 ±87	6.0 ±2.6	-3.93 ±2.6	72 ±4	258 ±9	-8.5 ±2.2	482 ±938
Δlifetime/ΔT % K^{-1}	0.45 ±0.13	-0.20 ±0.06	-0.68 ±0.09	-0.92 ±0.14	1.8 ±0.2	-0.61 ±0.12	-0.03 ±0.91
Scaled AOD/ΔT K^{-1}	20.8 ±0.7 E-4	N/A	-0.16 ±0.10 E-4	17 ±1 E-4	21.6 ±0.8 E-4	-5.0 ±1.3 E-4	10.8 ±11.1
4xCO2 ΔAOD/ΔT K^{-1}	24.8 ±0.8 E-4	N/A	0.62± 0.20 E-4	N/A	33.6 ±1.0 E-4	17.6 ±1.7 E-4	19.2 ±12.1
α emissions W m^{-2} K^{-1}	-0.041 ±0.002	-0.0014 ±0.0006	0.0004 ±0.0003	-0.044 ±0.003	-0.084 ±0.004	0.0042 ±0.0011	-0.027 ±0.032
α AOD W m^{-2} K^{-1}	-0.049 ±0.002	N/A	-0.0015 ±0.0005	N/A	-0.130 ±0.005	-0.015 ±0.002	-0.049 ±0.050

Table 7. Radiative efficiencies by emission and AOD from *2xss* (sea-salt). Changes in emission and AOD from *4xCO2*. "scaled" refers to scaling the *2xss* relations between AOD to emissions by the *4xCO2* changes in emissions. α values are calculated assuming ERF is proportional to emissions or AOD. Uncertainties for each model are errors in the mean based on interannual variability. Uncertainties in the multi-model results are standard deviation across the models. "N/A" signifies that diagnostic was not available from that model. The multi-model α terms are the average of the individual model α rather than the product of the multi-model ϕ and γ. Multi-model means are not shown for the emissions as these are so variable.

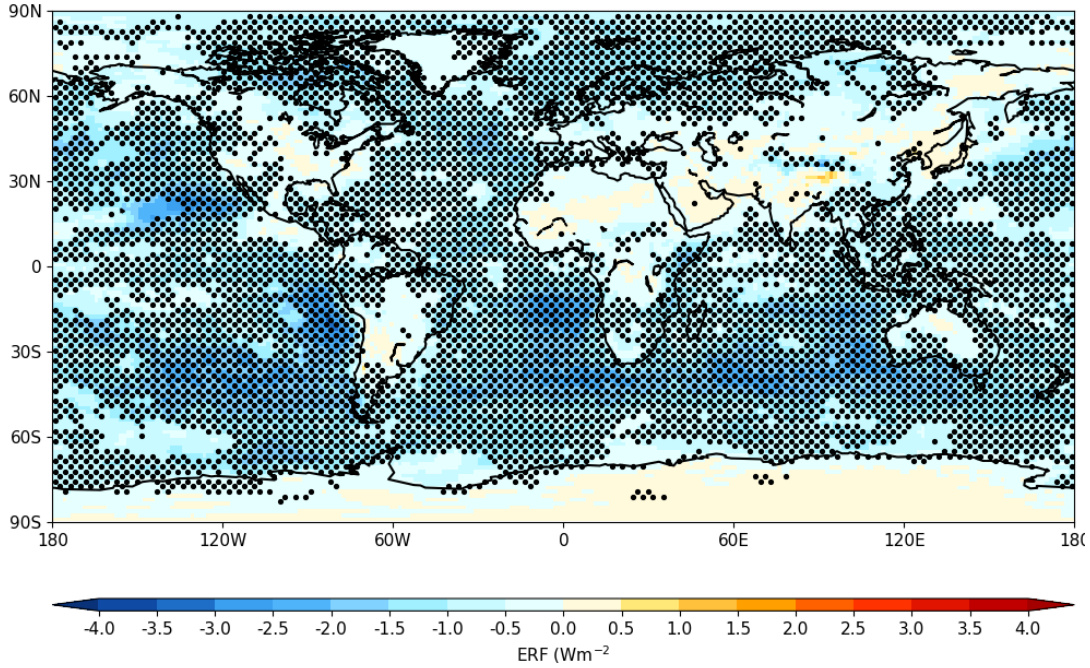
~~**Not all models provided AOD diagnostics.**~~

4.1.3 DMS

Four models ran the 2xDMS experiment. Interactive biogeochemistry or interactive DMS emissions are not a perquisite for the *2xDMS* experiment, however interactive emissions are required to calculate a feedback α hence we exclude ~~CNRM-ESM2~~

CNRM-ESM2--1 from table 8. ~~All models except CNRM-ESM2 have interactive DMS emissions that vary with climate (wind speed), and t~~Two models ~~also~~ include interactive ocean biogeochemistry (UKESM1 and NorESM2). ~~The latter two performed the *2xDMS* experiment. CNRM-ESM2 also ran the *2xDMS* experiment but uses prescribed emissions that are independent of climate.~~ The ERF for *2xDMS* is negative for all ~~three~~ three models~~models~~ that ran this experiment (figure 3(a), figure S9, table 8), though less strongly so for ~~CNRM-ESM2~~GISS-E2-1. UKESM1 and NorESM2~~Three of the models with interactive emissions~~ show a decrease in sulphur emissions in *4xCO2* where the tropical decrease more than compensates for the increase along the edge of the sea ice retreat whereas~~. GFDL-ESM4~~ GISS-E2-1 shows~~s~~ an increase in overall sulphur emissions. The multi-model mean is shown in figure 3(b) and the individual models in figure S8. ~~Since not all data is available for all models, we use the multi-model radiative efficiencies (by emission and by mass) and the multi-model sensitivities (of emissions and mass) to climate in order to calculate the multi-model feedback (table 8). The strong positive DMS increase in GFDL-ESM4 weakens the multi-model mean decrease in emission with climate.~~ The multi-model mean emission-based α is slightly positive, but ~~therefore~~ consistent with ~~near~~ zero. In spite of decreased DMS emissions in UKESM1 and NorESM2 ~~(within the uncertainty range).~~t ~~T~~here is an increased sulphur mass in all models in the *4xCO2* simulation due to an increase in the sulphate lifetime of around 2% K^{-1}~~.~~. Since this lifetime change applies to all sulphate, not just that from DMS, the radiative efficiency from 2xDMS will not necessarily apply and we do not calculate an AOD or mass-based feedback, but note that it would be negative.~~When scaled by the radiative efficiency for DMS emissions (which might not be appropriate for a lifetime increase) this leads to negative α (0.048±0.028 W m^{-2} K^{-1}).~~

(a)



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(b)

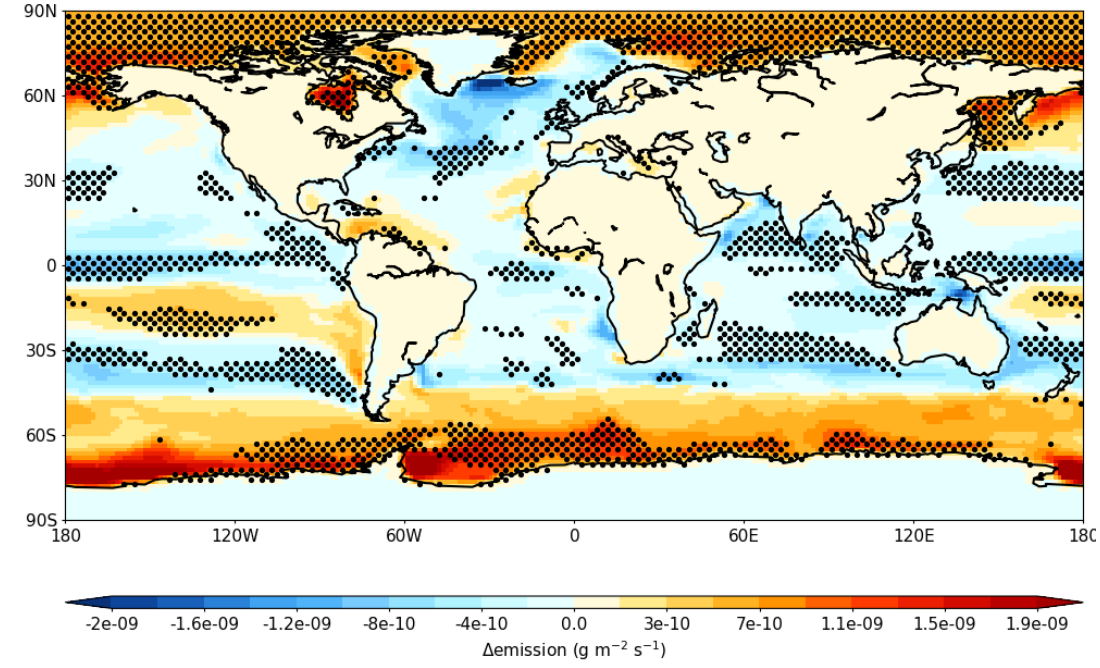


Figure 3 Multi-model mean (a) ERF from *piClim-2xDMS* vs *piClim-control*, (b) Change in DMS emissions (in g(S)) for *abrupt-4xCO2* vs *piControl* Stippling shows areas where the mean changes by more than the standard deviation across models.

	UKESM1	NorESM2	GISS-E2	Multi-model
ERF *2xDMS* W m^{-2}	-1.22 ±0.03	-1.27 ±0.07	-0.61 ±0.04	-1.02 ±0.29
ERF/ Emission W m^{-2} $(Tg(S)\ yr^{-1})^{-1}$	-0.0728 ±0.0010	-0.0674 ±0.0019	-0.0219 ±0.0012	-0.054 ±0.023
ΔEmission/ΔT Tg(S) yr^{-1} K^{-1}	-0.04 ±0.01	-0.186 ±0.02	0.02 ±0.02	-0.06 ±0.09
Δlifetime/ΔT % K^{-1}	2.48 ±0.06	2.73 ±0.11	1.13 ±0.15	2.1 ±0.7
α emissions W m^{-2} K^{-1}	0.0027 ±0.0006	0.0125 ±0.0013	-0.0006 ±0.006	0.005 ±0.006

Table 8. Radiative efficiencies by emission ~~and mass~~ from *2xDMS*. Changes in emission ~~and mass~~ from *4xCO2* experiment. ~~Emissions are for DMS or SO_2+SO_4 depending on the model. "scaled" refers to scaling the *2xDMS* relations between mass and emissions by the *4xCO2* changes in emissions.~~ α values are calculated assuming ERF is proportional to emissions ~~or mass~~. ~~Multi-model mean values of α use the multi-model mean radiative efficiencies and sensitivities to climate, rather than being an average of the individual model α values.~~ Uncertainties for each model are errors in the mean based on interannual variability. Uncertainties in the multi-model results are standard deviation across the models.~~.~~ The multi-model α terms are the average of the individual model α rather than the product of the multi-model ϕ and γ.

~~**MIROC6 and GFDL-ESM4 did not perform the *2xDMS* experiment, but DMS changes are diagnosed from their *4xCO2* experiments. DMS emissions do not vary in the CNRM-ESM2 *4xCO2* experiment.**~~

DMS is produced by marine biological activity in the ocean, and it is assumed to be the largest natural source of sulphur to the atmosphere. Up to now, there has been no comprehensive model effort to include all the important effects, and therefore the DMS emission strength change under climate change is still uncertain. The slightly positive mean~~range~~ here is in contrast to the~~here (-0.010 to -0.075 W m^{-2} K^{-1} including increases in sulphur lifetime) encompasses the~~ -0.02 W m^{-2} K^{-1} feedback from AR5 (Ciais et al., 2013), based on results from only one model (HadGEM2-ES).

~~DMS production is closely linked to primary production.~~ Modelling studies including ocean biogeochemistry have shown that under climate change, an increased stratification of the ocean at low and mid latitudes leads to a reduction in nutrients supply into the surface ocean and thus a reduction in DMS emissions, whereas at high latitudes, retreat of sea-ice can lead to increased ~~primary production~~biological activity and increase in DMS production (Kloster et al., 2007). ~~Globally, most~~Previous models which include ocean biogeochemistry have shown a slight increase in DMS production and emission to the atmosphere in a warming climate (Bopp et al., 2004; Gabric et al., 2004; Gunson et al., 2006; Vallina et al., 2007).

Some more recent studies have included the impact of ocean acidification on ocean DMS production (Schwinger et al., 2017; Six et al., 2013). Both studies used a very similar description of the ocean biogeochemistry and extended it with an observationally-based relation between ocean alkalinity and ocean DMS production. Assuming a medium sensitivity of the DMS production on pH, Six et al. (2013) found a global DMS emission decrease by 18% in 2100 under the SRES A1B scenario, and Schwinger et al. (2017) an emission reduction by 31% in 2200 under the RCP8.5 scenario. In addition recent work has provided evidence for major pathways in the oxidation of DMS in the atmosphere which are not included in any of these ESMs (Berndt et al., 2019; Wu et al., 2015).

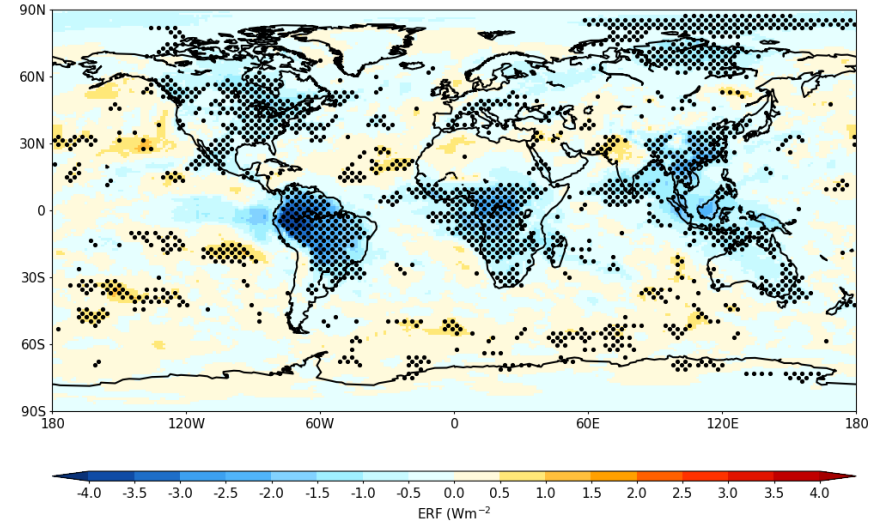
4.1.4 Organic aerosol

Biogenic VOC emissions lead to both organic aerosol and ozone production (in those models with tropospheric chemistry). It is therefore ~~difficult~~ necessary to distinguish the two in the ERFs in these models. The ozone stratospheric-temperature adjusted radiative forcing (SARF) from the ozone changes are diagnosed offline (see section 2.1). This is subtracted from the ERF to give the ERF due to aerosols only as shown in table 9 (ozone is the only non-aerosol forcing agent that varies). For NorESM2 there is no ozone change. ~~An estimate of the direct aerosol effect can be determined by additional radiation diagnostics that are run without the contribution of aerosols "aerosol free" (ERF_{af}), for clear sky conditions (ERF_{es}), and both clear sky and aerosol free (ERF_{esaf}) (Ghan, 2013). Here the aerosol direct effect is ERF- ERF_{af} and the cloud effect is ERF_{af}- ERF_{esaf} (although this may include cloud forcing due to adjustments caused by the ozone changes too).~~ The ERF before subtracting the ozone SARF~~direct aerosol and cloud radiative effects are s~~ is shown in figure 4. These estimated aerosol forcing changes are ~~significant~~ large (~~between -0.3 and~~up to -0.69 W m^{-2}). All the ERF-$SARF_{O3}$ values are negative apart from UKESM1 which has a large positive forcing from cloud changes (diagnosed from comparing all-sky and clear-sky diagnostics – not shown). ~~The most negative forcing comes from the NorESM2 model which has no changes in gas phase chemistry (table 9).~~

In terms of aerosol, there is an increase in organic aerosol (OA) mass and expected increase in AOD with very similar spatial pattern when the emission of BVOCs is doubled. ~~Changes to cloud droplet number concentration are more complex and may not be spatially co-located with the changes to BVOC emission. (figure S4~~S12~~). Whilst the additional secondary organic aerosol can grow particles to a size where they can act as cloud condensation nuclei, this process can also enhance the aging rate of particles removing them from the atmosphere more quickly. In addition, for models with interactive tropospheric chemistry, the decrease in oxidant concentrations resulting from a doubling of VOC emissions can prevent the oxidation of sulphur containing species that might otherwise have formed aerosols, leading to a reduction in CDNC.~~ The patterns of BVOC increase for the *4xCO2* experiments are much more similar between models (figure S10) in terms of pattern and sign than for the previous species (dust, sea salt, DMS), although the magnitude is considerably less for UKESM1. In the *4xCO2* experiments, these models also simulate an increase in primary organic aerosol emission from the ocean which adds to the OA mass on top of the effect of BVOC emissions. The feedback factors are negative apart from UKESM1 and are very strong in

some models NorESM2 (~~, ranging from -0.003 to~~ –0.~~276~~ 28 W m^{-2} K^{-1} ~~based on emissions and -0.025 to -0.359 W m^{-2} K^{-1} based on mass assuming all OA has the same radiative efficiency as that from vegetation~~).

(a)



(b)

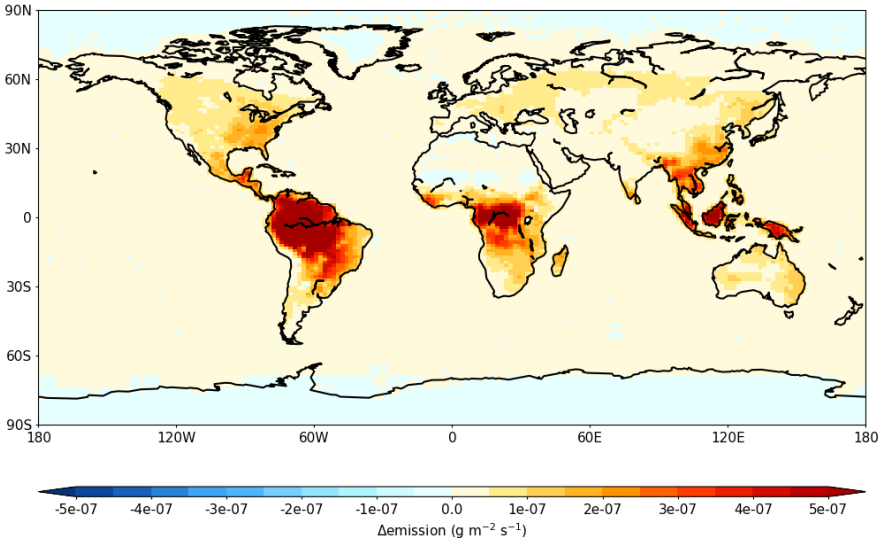


Figure 4 Multi-model mean (a) ~~Aerosol-direct-effect~~ERF from *piClim-2xVOC* vs *piClim-control*, (b) ~~cloud-radiative-effect-from *piClim-2xVOC* vs *piClim-control*, (c)~~ Change in ~~organic aerosol~~BVOC emissions for *abrupt-4xCO2* vs *piControl*. Stippling shows areas where the mean changes by more than the standard deviation across models.

	UKESM1	NorESM2	GFDL-ESM4	CESM2-WACCM	GISS-E2-1	Multi-model
ERF *(non* O3*) 2xVOC* W m^{-2}	0.03 ±0.03	-0.69 ±0.07	-0.45 ±0.03	-0.36 ±0.04	-0.24 ±0.03	-0.34 ±0.24
ERF/ Emission W m^{-2} (Tg) yr^{-1})$^{-1}$	0.4 ±0.4E-4	-11.8 ±1.2E-4	-9.7 ±0.6E-4	-5.4 ±0.6E-4	-1.3 ±0.1E-4	-5.6 ±4.7 E-4
~~ERF/mass W m^{-2}~~	~~±0.08~~	~~-0.56±0.06~~	~~-0.±0.04~~	~~-0.±0.04~~		~~-0.34±0.~~
ΔEmissionVOC/ΔT Tg yr^{-1} K^{-1}	32 ±2	234 ±7	81 ±2	156 ±2	113 ±3	123 ±69
~~Scaled mass/ΔT Tg K^{-1}~~	~~0.01~~	~~0.±0.0~~	~~0.127±0.003~~	~~0.2±0.003~~		
~~*4xCO2* Δmass/ΔTTg K^{-1}~~	~~0.135±0.004~~	~~0.644±0.018~~	~~0.022±0.001~~	~~0.5±0.01~~		~~0.±0.26~~
α emissions W m^{-2} K^{-1}	0.001 ±0.001	-0.28 ±0.03	-0.079 ±0.006	-0.084 ±0.009	-0.015 ±0.002	-0.09 ±0.10
~~α mass W m^{-2} K^{-1}~~	~~0.0±0.01~~	~~-0.359±0.03~~	~~-0.01±0.001~~	~~-0.1±0.02~~		~~-0.1±0.1~~

Table 9. Non O3 ERF (subtracting off the O3 SARF from table 10, for NorESM2 there is no O3 change). Radiative efficiencies by emission of BVOC ~~and mass~~ from *2xVOC*. Changes in emission of BVOC ~~and mass~~ from *4xCO2* experiment. ~~"scaled" refers to scaling the *2xVOC* relations between mass and emissions by the 4xCO2 changes in emissions.~~ α values are calculated assuming ERF is proportional to emissions ~~or mass~~. ~~Multi-model mean values of α use the multi-model mean radiative efficiencies and sensitivities to climate, so are different to the average of the individual model α values.~~ Uncertainties for each model are errors in the mean based on interannual variability. Uncertainties in the multi-model results are standard deviation across the models. The multi-model α terms are the average of the individual model α rather than the product of the multi-model ϕ and γ.

4.2 ~~Gas-phase~~Ozone and methane feedbacks

4.2.1 Biogenic VOCs

The ozone SARF is diagnosed offline (section 2.1) and shown in table 10, ~~To estimate the stratospheric-temperature adjusted radiative forcing (SARF) from the ozone changes, and to remove the effect of aerosols we use the clear-sky aerosol-free ERF (ERF$_{esaf}$) (table 10). However, this neglects any cloud adjustments caused by the ozone, and any cloud masking of the direct ozone SARF.~~ For ~~GFDL-ESM4 and CESM2-WACCM~~all except UKESM1 the magnitude of the ozone forcing is smaller than that for aerosols leading to a net negative ERF from BVOCs. For UKESM1 the ~~net ERF is positive due to a lower magnitude of aerosol forcing~~non-ozone forcing is positive (section 4.1.4) and the ozone adds to this. ~~The ozone contribution is also estimated assuming a radiative efficiency of 0.042 W/m^2 per Dobson Unit (Stevenson et al., 2013). This efficiency is strictly only applicable to changes in tropospheric ozone but is also applied to the stratospheric ozone since these changes occur in the lower stratosphere just above the tropopause. The estimated ozone~~

~~SARF (tropospheric + stratospheric) is within the range of the diagnosed ERF_{csaf}. CESM2-WACCM has the largest BVOC emissions and a decrease in tropospheric ozone column, although a strong increase in the stratospheric column. This is likely to be due to NOx limited chemistry near the surface and increased transport of reactive nitrogen (NO_y) away from the surface to the upper troposphere and lower stratosphere as peroxy acetyl nitrate (PAN) and other organic nitrates~~The ozone SARF per Tg VOC emission is similar between the models with CESM2-WACCM slightly lower. The overall feedback is therefore dominated by the variation in the sensitivity of BVOC emissions to climate. This ranges from 0.~~004~~005 W m^{-2} K^{-1} for UKESM1 which has the lowest ~~ozone response to BVOC emissions, and the lowest~~ BVOC increase with climate ~~(due to CO_2 inhibition)~~ to 0.~~028~~014 W m^{-2} K^{-1} for CESM2-WACCM and GISS-ES-1 which have~~s~~ the strongest BVOC response to climate.

	UKESM1		GFDL-ESM4		CESM2-WACCM		GISS-E2-1	Multi-model
~~ERF_{csaf}~~ $SARF_{O3}$ *2xVOC* W m^{-2}	0.12 ±0.02		0.07 ±0.01		0.06 ±0.01		0.23 ±0.03	0.10 ±0.08
$SARF_{O3}$ /emission W m^2(Tg yr^{-1})$^{-1}$	1.5 ±0.2 E-4		1.6 ±0.2 E-4		0.9 ±0.1 E-4		1.2 ±0.2 E-4	1.3 ±0.4 E-4
~~ERF_{csaf}/emission W m^2(Tg yr^{-1})$^{-1}$~~	~~1.2±0.4E-4~~		~~2.8±0.8E-4~~					
	~~trop~~	~~strat~~	~~trop~~	~~strat~~	~~trop~~	~~strat~~		
~~Ozone/emission DU (Tg yr^{-1})$^{-1}$~~	~~0.0015~~	~~0.0021~~	~~0.0022~~	~~0.0031~~	~~-0.0003~~	~~0.0044~~		
~~Ozone SARF /emission W m^2(Tg yr^{-1})$^{-1}$~~	~~0.63±0.09 E-4~~ ~~0.9±0.1 E-4~~		~~0.9±0.1 E-4~~ ~~1.3±0.2 E-4~~		~~-0.10±0.01 E-4~~ ~~1.8±0.3 E-4~~			
4xCO2 Tg yr^{-1} K^{-1}	32 ±2		81 ±2		156 ±2		113 ±3	95 ±45
~~α ERF_{csaf} W m^{-2} K^{-1}~~	~~0.004±0.001~~		~~0.023±0.007~~					
α $SARF_{O3}$ W m^{-2} K^{-1}	0.005 ± 0.001		0.013 ± 0.002		0.014 ± 0.002		0.014 ± 0.002	0.011 ± 0.004

Table 10. Ozone SARF and Rradiative efficiencies ~~(clear-sky aerosol-free ERF_{csaf})~~ for *2xVOC* emissions. ~~Tropospheric and stratospheric ozone column changes and their estimated radiative effects.~~ Changes in emission from *4xCO2* experiment. *α* values are calculated ~~assuming ERF_{csaf} or a~~using the ozone SARF ~~efficiency for ozone of 0.042 W m^{-2} DU^{-1}~~. Uncertainties for each model are errors in the mean based on interannual variability, and assuming a ~~14~~15% uncertainty in the ozone radiative efficiency (section 2.2). Uncertainties in the multi-model results are standard deviation across the models.

At the multi-model mean level, the cooling associated with an increase in organic aerosol (-0.04~~13~~±0.~~10~~042 W m^{-2} K^{-1} – for the 4 models with chemistry) ~~outweighs~~dominates over the warming associated with an increase in O_3 (0.011 ~~17~~±0.004~~11~~ W m^{-2} K^{-1}) leaving an overall negative feedback.~~and an increase in CH_4 lifetime (0.041±0.030 W m^{-2} K^{-1}).~~

Using multi-annual simulations of global aerosol, Scott et al. (2018) diagnosed a feedback from biogenic secondary organic aerosol of -0.06 W m^{-2} K^{-1} globally, and -0.03 W m^{-2} K^{-1} when considering only extra-tropical regions. This global feedback value was composed of a direct aerosol radiative feedback of -0.048 W m^{-2} K^{-1} and an indirect aerosol (i.e., cloud albedo) feedback of -0.013 W m^{-2} K^{-1}. Using observations from eleven sites, Paasonen et al., (2013) estimated an indirect aerosol feedback of -0.01 W m^{-2} K^{-1} due to biogenic secondary organic aerosol. The ability of models to account for changes in vegetation has a ~~significant~~large effect on the feedback. Sporre et al. (2019) found that interactive vegetation~~,~~ enhanced BVOC emissions by 63% ~~greater~~ relative to prescribed vegetation, producing more organic aerosol and an increase in (negative) aerosol forcing.

The level of compensation between increased aerosol forcing and increased ozone ~~and methane lifetime~~ is dependent on the model (here positive feedback for GFDL-ESM4, negative for UKESM1 and CESM2-WACCM). Unger (2014) found a positive feedback in NASA GISS ModelE2, whereas Scott et al. (2014) found a negative feedback in HadGEM2-ES.

4.2.2 Lightning NO_X

Lightning NO_X leads to ozone production, and changes in methane lifetime. As for BVOCs (section 4.2.1), ozone radiative kernels are used to quantify the ozone SARF. The ERF and $SARF_{O3}$ agree for all models except UKESM1 (table 11), suggesting that there is little effect on aerosols in these models. In UKESM1 NO_X is known to increase the formation of new sulphate particles (O'Connor and et al., submitted) partially offsetting the positive ozone forcing. The $SARF_{O3}$ per Tg emission varies by a factor of two (0.023 to 0.048 W m^2(Tg(N) yr^{-1})$^{-1}$) between the highest and lowest.~~To separate the ozone effect, we use ERF_{esaf} for UKESM1 as in section 4.2.1. The assumption of radiative efficiency of 0.042 W m^{-2} DU^{-1} seems to agree with the ERF for GFDL-ESM and CESM2-WACCM (table 12). For UKESM1 ERF_{esaf} is lower than expected from the ozone columns, suggesting that the clear-sky aerosol-free component misses some of the ERF due to ozone.~~

The changes in ~~L~~lightning NO_X emissions vary widely across the models, with three showing increases (~~increases in~~ UKESM1, ~~and~~ CESM2-WACCM, GISS-E2-1) but a slight decrease~~s slightly~~ in GFDL-ESM4. Although ~~although~~ they all use variations on the cloud-top height schemes ~~(Price et al., 1997; Price and Rind, 1992)~~(section 3.2.3) the differences in how this is implemented and how the modelled clouds vary with climate change all affect the emission response. T~~Hence t~~he feedback is positive for the three models with increased lightning ~~UKESM1 and CESM2-WACCM~~ (0.009 ~~and~~ to 0.01~~61~~ W m^{-2} K^{-1}), based on the ozone changes, but slightly negative for GFDL-ESM4 (-0.001 W m^{-2} K^{-1}). Including the aerosol response to lightning for UKESM1 would reduce its feedback to 0.005 W m^{-2} K^{-1} but this seems to be particular to this model.

	UKESM1	GFDL-ESM4	CESM2-WACCM	GISS-E2-1	Multi-model
ERF *2xNOX* W m^{-2}	0.12 ±0.03	0.11 ±0.04	0.15 ±0.04	0.13 ±0.03	0.13 ±0.02
ERF/emission W m^{-2}(Tg(N) yr^{-1})$^{-1}$	0.018 ±0.004	0.036 ±0.013	0.051 ±0.013	0.021 ±0.005	0.032 ±0.013
$SARF_{O3}$ *2x-NOX* W m^{-2}	0.21 ±0.02	0.10 ±0.02	0.14 ±0.02	0.14 ±0.02	0.15 ±0.04
$SARF_{O3}$ ~~Ozone SARF~~ /emission W m^{-2}(Tg(N) yr^{-1})$^{-1}$	0.031 ± 0.004	0.034 ± 0.005	0.048 ± 0.007	0.023 ± 0.003	0.034 ±0.009
4xCO2 Tg(N) yr^{-1} K^{-1}	0.27 ±0.01	-0.029 ±0.008	0.336 ±0.013	0.614 ±0.019	0.30 ±0.23
α ERF_{csaf} W m^{-2} K^{-1}	0.005 ±0.001	-0.001 ±0.0005	0.017 ±0.005	0.013 ±0.003	0.009 ±0.007
α $SARF_{O3}$ W m^{-2} K^{-1}	0.009 ± 0.001	-0.001 ± 0.0005	0.016 ± 0.002	0.014 ± 0.002	0.009 ±0.007

Table ~~12~~11. ERF and ozone SARF ~~Radiative~~ radiative efficiencies ~~(clear-sky aerosol-free ERF_{csaf} for UKESM)~~ for *2xNOX* lightning NO_X emissions. ~~Tropospheric and stratospheric ozone column changes and their estimated radiative effects.~~ Changes in emission from *4xCO2* experiment. α values are calculated assuming ERF_{csaf} or ozone~~a~~ SARF ~~efficiency for ozone of 0.042 W m^{-2} DU^{-1}~~. Uncertainties for each model are errors in the mean based on interannual variability, and assuming a ~~14~~15% uncertainty in the ozone radiative efficiency (section 2.2). Uncertainties in the multi-model results are standard deviation across the models.

The ESMs used in CMIP6 all use a cloud-top height parameterisation of lightning. Such schemes have previously been found to increase lightning production in warmer climates whereas more sophisticated schemes based on convective updraft mass flux or ice flux show decreases in lightning with temperature~~.~~ (Clark et al., 2017; Finney et al., 2016b, 2018). The result from the Atmospheric Chemistry and Climate Model Intercomparison (ACCMIP) of 0.44 Tg(N) yr^{-1} K^{-1} (Finney et al., 2016a), lies within the range of~~Two~~ the models with increased lightning under 4xCO2 ~~(here (UKESM1 and WACCM) show increases in lightning emissions of~~ 0.27 to 0.61~~and 0.21~~ Tg(N) yr^{-1} K^{-1}~~) which is slightly lower than the results from the Atmospheric Chemistry and Climate Model Intercomparison (ACCMIP) of 0.44 Tg(N) yr^{-1} K^{-1} (Finney et al., 2016a).~~

4.2.3 Methane lifetimes

BVOC and NOx emissions also affect the methane lifetime. Methane does not change in the AerChemMIP experimental setup, but the methane changes that would be expected if methane were allowed to evolve freely can be diagnosed from the change in methane lifetime. The methane lifetime to OH (troposphere and stratosphere) is diagnosed in the models. The losses to chlorine oxidation and soil uptake are assumed to be 11 and 30 Tg yr^{-1} respectively (Saunois et al., 2020). All models show an increase in methane lifetime with BVOC emissions (0.018-0.035 % per Tg(VOC) yr^{-1} and a decrease due to lightning NO_X emissions (-2.4 - -6.8 % per Tg(N) yr^{-1} (table 12). From these the expected lifetime changes with climate can be deduced from the changes in emissions with temperature. These lifetime changes are then converted to feedbacks using the radiative efficiency (including impacts on ozone and stratospheric water vapour) for methane lifetime changes in section 2.2 (0.011 W m^{-2} $\%^{-1}$). The feedbacks range from 0.012 to 0.061 W m^{-2} K^{-1} for BVOCs and -0.042 to +0.001 W m^{-2} K^{-1} for lightning NOx where the variability is mostly due to the different sensitivities of BVOC or lightning emissions to climate in the models. For BVOC the methane lifetime feedback is larger than that due to ozone production, thus increasing the overall feedback. For lightning NOx, the methane lifetime feedback is of opposite sign to that from ozone production, with approximate compensating for UKESM1 and GFDL-ESM4 (net 0.002 and 0.000 W m^{-2} K^{-1} respectively) and an overall negative lightning feedback from CESM2-WACCM and GISS-E2-1 (-0.009 and -0.028 W m^{-2} K^{-1} respectively)~~As with BVOC emissions (above) the potential impacts of lightning on methane lifetime can be diagnosed. All models show (table 13) a decrease in methane lifetime with increased lightning NOx emission from -2.3 to -4.8 % (Tg yr^{-1})$^{-1}$. The feedbacks are negative for UKESM1 and CESM2-WACCM (0.007 and 0.012 W m^{-2} K^{-1}) and slightly positive for GFDL-ESM4 (0.001 W m^{-2} K^{-1}) and almost exactly cancel out the feedback due to the ozone column. The net (ozone + τ_{CH_4}) feedbacks for UKESM1, GFDL-ESM4 and CESM2-WACCM are -0.002, 0.000, and 0.001 W m^{-2} K^{-1}~~. For UKESM1 a feedback of -0.00~~46~~ W m^{-2} K^{-1} ~~should~~ could be added to the total lightning feedback to account for the increase in sulphate.

	UKESM1	GFDL-ESM4	CESM2-WACCM	GISS-E2-1	Multi-model
BVOC					
τ_{CH_4} /emission % (Tg(VOC) $yr^{-1})^{-1}$	0.033	0.030	0.035	0.018	0.029 ±0.007
$\tau_{CH_4}/\Delta T$ % K^{-1}	1.07 ±0.06	2.47 ±0.06	5.48 ±0.06	2.08 ±0.05	2.8 ±1.6
$\alpha\ \tau_{CH_4}$ W m^{-2} K^{-1}	0.012 ±0.002	0.028 ±0.004	0.061 ±0.009	0.023 ±0.003	0.031 ±0.018
Lightning NOx					
τ_{CH_4} /emission % (Tg(N) $yr^{-1})^{-1}$	-2.4	-3.8	-6.8	-6.1	4.8 ±1.8
$\tau_{CH_4}/\Delta T$ % K^{-1}	-0.64 ±0.02	0.11 ±0.03	-2.28 ±0.09	-3.75 ±0.12	-1.6 ±1.5
$\alpha\ \tau_{CH_4}$ W m^{-2} K^{-1}	-0.007 ±0.001	0.001 ±0.000	-0.025 ±0.004	-0.042 ±0.006	-0.018 ±0.017

Table 123. Percentage change in methane lifetime for BVOC and lightning NOx emissions. Estimated change in lifetime following changes in BVOC and NOx emission from *4xCO2* experiment. α values are calculated assuming a radiative efficiency of 0.015 W m^{-2} $\%^{-1}$. Uncertainties for each model assume a 14% uncertainty in the methane radiative efficiency (Etminan et al., 2016)**. Uncertainties in the multi-model results are standard deviation across the models.**

4.2.3 Wetland emissions

Two models diagnosed changes in wetland emissions due to 4xCO2. Although the wetland emissions do not directly affect methane concentrations in the model, changes in emission can be converted to concentration changes (section 2.2). UKESM1 and CESM2-WACCM, both of which are models with interactive wetland emissions, show strong responses to climate change, leading to a feedback of 0.16±0.03 W m^{-2} K^{-1}.

	UKESM1	CESM2-WACCM	Multi-model
4xCO2 Tg(CH4) yr^{-1} K^{-1}	40	60	
α W m^{-2} K^{-1}	0.13 ±0.02	0.19 ±0.03	0.16 ±0.03

Table ~~14~~13. Sensitivity of wetland emissions to *4xCO2* in two models. Feedback parameter assuming pre-industrial conditions. Uncertainties for each model assume a 14% uncertainty in the methane radiative efficiency (Etminan et al., 2016)**. Uncertainties in the multi-model results are standard deviation across the models.**

Wetland emissions are more strongly sensitive to CO_2 concentrations than to temperature or precipitation (Melton et al., 2013), so the values presented here are more likely to be "adjustments" to the CO_2 rather than feedbacks, and hence could be considered part of the CO_2 ERF. We find emission increases following quadrupled levels of CO_2 of 130-160%. This compares with results from the Wetland CH4 Inter-comparison of Models Project (WETCHIMP) of 20-160% following an increase in CO_2 of a factor of 2.8 (Melton et al., 2013). The CMIP6 simulation specifications do not include free-running methane concentrations therefore the effects of these increased wetland emissions will not be realised in any of the CMIP6 experiments. Outside CMIP6, ESMs are starting to include free-running methane (Ocko et al., 2018), so for these it will be important to understand the effects of changing CO_2 and meteorology on wetland emissions.

4.2.4 Meteorological drivers~~Temperature and humidity~~

As well as through changes in natural emissions, climate change can affect ozone burden and methane lifetime directly as the production and loss reactions are sensitive to temperature and water vapour(Johnson et al., 2001). Here we add the expected changes in ozone SARF and methane lifetime due to changes in BVOCs and lightning NO_X from sections 4.2.1 and 4.2.2 above and compare those to the changes diagnosed from the 4xCO2 experiments (table ~~15~~14). Since lightning NOx and BVOCs are the dominant climate-sensitive emissions of (non-methane) species affecting ozone and methane, ~~T~~the residual is then the direct effect of climate. UKESM1, GFDL-ESM4 and GISS-E2-1 all diagnosed ozone changes for the abrupt-4xCO2 experiment (figure S12). All three showed decreased tropospheric ozone and increased stratospheric ozone (apart from the tropical lower stratosphere) in the 4xCO2 climate. The ozone SARF (calculated using radiative kernels) is negative whereas the expected change from lightning NOx and BVOCs would be positive, hence the residual attributed to meteorological changes is negative.~~For CESM2-WACCM and GFDL-ESM4 most of the total increase in tropospheric ozone can be explained by the changes in natural emissions (particularly BVOC) suggesting that non-emission drivers of tropospheric ozone change (temperature, humidity, transport from the stratosphere, dry deposition) balance to have little net effect. Increases in stratospheric ozone are much larger than expected from the changes in natural emissions, suggesting that meteorological changes (principally cooling stratospheric temperatures) are the main driver. The tropospheric ozone change attributable to climate can be used to determine a feedback which is only significant for UKESM1 (-0.023 W m^{-2} K^{-1}). The stratospheric~~

~~ozone changes cannot simply be converted to an ERF, since unlike for the natural emission (where the ozone changes were close to the tropopause) the tropospheric radiative efficiency cannot be applied.~~

For ~~In~~ UKESM1, ~~and~~ GFDL-ESM4 and GISS-E2-1 the meteorological changes decrease methane lifetime leading to an overall decrease in lifetime for the 4xCO2. ~~by similar amounts (-4.5 and -4.6 % K^{-1}) and hence have similar feedbacks (-0.078 and -0.080 W m^{-2} K^{-1}). In the case of GFDL-ESM4 this leads to an overall decrease in lifetime rather than the increase expected from natural emission changes (principally BVOC).~~ In CESM2-WACCM the meteorological changes increase methane lifetime adding to the strong increase from BVOC emissions~~the overall effect of climate is to increase the methane lifetime, almost entirely due to the increased BVOC emissions with little effect of meteorological drivers~~. This is surprising since there is no known mechanism whereby temperature and humidity increases can increase the methane lifetime. This could be due to non-linearity whereby the effect of increased VOCs on methane lifetime is larger than expected from scaling the *2xVOC* experiment.

Combining the results from ozone and methane lifetime changes leads to overall feedbacks from temperature of ~~-0.101, -0.082 and +0.015 W m^{-2} K^{-1}~~ -0.15, -0.14 and -0.08 for UKESM1, GFDL-ESM4, and GISS-E2-1.~~the three models.~~

	UKESM1	GFDL-ESM4	CESM2-WACCM	GISS-E2-1	Multi-model
Ozone					
LNOx+BVOC Ozone SARF W m^{-2} K^{-1}	0.013 ±0.0005	0.012 ±0.0004	0.030 ±0.0006	0.028 ±0.0005	0.021 ±0.008
4xCO2 Ozone SARF W m^{-2} K^{-1}	-0.065 ±0.009	-0.050 ±0.007		-0.022 ±0.003	-0.046 ±0.018
α Ozone residual W m^{-2} K^{-1}	-0.079 ±0.009	-0.062 ±0.007		-0.050 ±0.003	-0.064 ±0.012
Methane lifetime					
LNOx+BVOC τ_{CH_4} % K^{-1}	0.43 ±0.07	+2.58 ±0.07	+3.20 ±0.11	-1.66 ±0.13	1.1 ±1.9
4xCO2 τ_{CH_4} % K^{-1}	-4.08 ±0.02	-2.05 ±0.06	+7.18 ±0.06	-3.33 ±0.12	-0.6 ±4.5
τ_{CH_4} residual % K^{-1}	-4.51 ±0.07	-4.63 ±0.09	+3.98 ±0.13	-1.67 ±0.02	-1.7 ±3.4
α τ_{CH_4} residual W m^{-2} K^{-1}	-0.073 ±0.~~011~~010	-0.075 ±0.011	0.064 ±0.009	-0.027 ±0.005	-0.027 ±0.056

Table ~~15~~14. Comparison of expected changes in ozone ~~column~~ SARF and methane lifetime with that diagnosed from *4xCO2*. Residual is given by the difference and is converted to a feedback using radiative efficiencies for ~~tropospheric ozone and~~ methane lifetime.

The three models showing decreased methane lifetime are in approximate agreement with ACCMIP which found a sensitivity of -3.4±1.4% K^{-1} (Naik et al., 2013; Voulgarakis et al., 2013). ACCMIP found a variation in sign of the ozone feedback amongst models −0.024±0.027 W m^{-2} for a 1850-2000 change in climate. The ACCMIP models generally did not include stratospheric chemistry so either explicitly prescribed the cross-tropopause flux of ozone or imposed a climatology of ozone above the tropopause. The four CMIP6 models here all treat the chemistry seamlessly across the troposphere and stratosphere so the impact of changes in stratosphere-troposphere exchange (STE) of ozone on the tropospheric column is likely to be different to ACCMIP.

Changes in the stratospheric ozone following a quadrupling of CO_2 are driven by cooling temperatures in the stratosphere. This is likely to be due to temperature adjustments to the stratospheric CO_2 concentrations, and so part of the ERF for CO_2 rather than a feedback ~~(Smith, submitted)~~. Feedbacks and adjustments cannot be distinguished with this experimental setup.

4.3 Overall feedback

The multi-model mean feedbacks are summarised in table 15 and figure 56. The totals assume that feedbacks are additive, which is the basis of the framework in section 2.1. The subsets of model used to generate the multi-model means are different for each process, so the total feedback is a mixture of these different subsets. The largest individual feedbacks are due to the generation of aerosols by BVOCs (-0.090 ~~113~~±0.099~~102~~ W m^{-2} K^{-1}) and the emission of methane from wetlands (0.16 ±0.03 W m^{-2} K^{-1}). The overall uncertainty is calculated by adding the inter-model uncertainty on each feedback component in quadrature. This is dominated by the uncertainty in the aerosol response to BVOC emissions. Nearly all the feedbacks are negative, most because they come from an increase in aerosol emissions with temperature and increased ozone and methane removal with temperature and humidity. For BVOC emissions, the increase in aerosols outweighs the increases in ozone and methane. For lightning NOx, the decrease in ~~ozone and~~ methane lifetime outweighs the ozone increase ~~changes cancel~~. For wetland we have attributed all the methane emission changes to temperature, whereas a significant proportion are likely to be an adjustement to CO_2 concentrations rather than a feedback (section 4.2.3). ~~A warmer and more humid climate also leads to less ozone and methane.~~

There will be additional systematic uncertainties in the overall feedback term. As described above, the use of a CO_2 perturbation to generate the climate change may lead to different feedback sensitivities compared to climate change caused by other forcing agents. There will also be an uncertainty caused by using a pre-industrial baseline atmosphere rather than present day. We are unable to quantify the likely magnitudes of these systematic uncertainties.

The ESMs that use the *abrupt-4xCO2* experiment to quantify the climate sensitivity do not allow methane to vary, so we also quantify the non-methane feedbacks that will be contributing to the diagnosed climate sensitivity in these models. This feedback is significantly negative (-0.183 ~~228~~± 0.111~~23~~ W m^{-2} K^{-1}) suggesting the climate sensitivity of ESMs might be expected to be lower than for their physical-only counterparts. This analysis (and climate sensitivity in general) is focussed on the global mean, but it should be noted that the cooling effects of increased aerosols will be heterogenous and some regions will experience less warming than a global climate sensitivity might suggest.

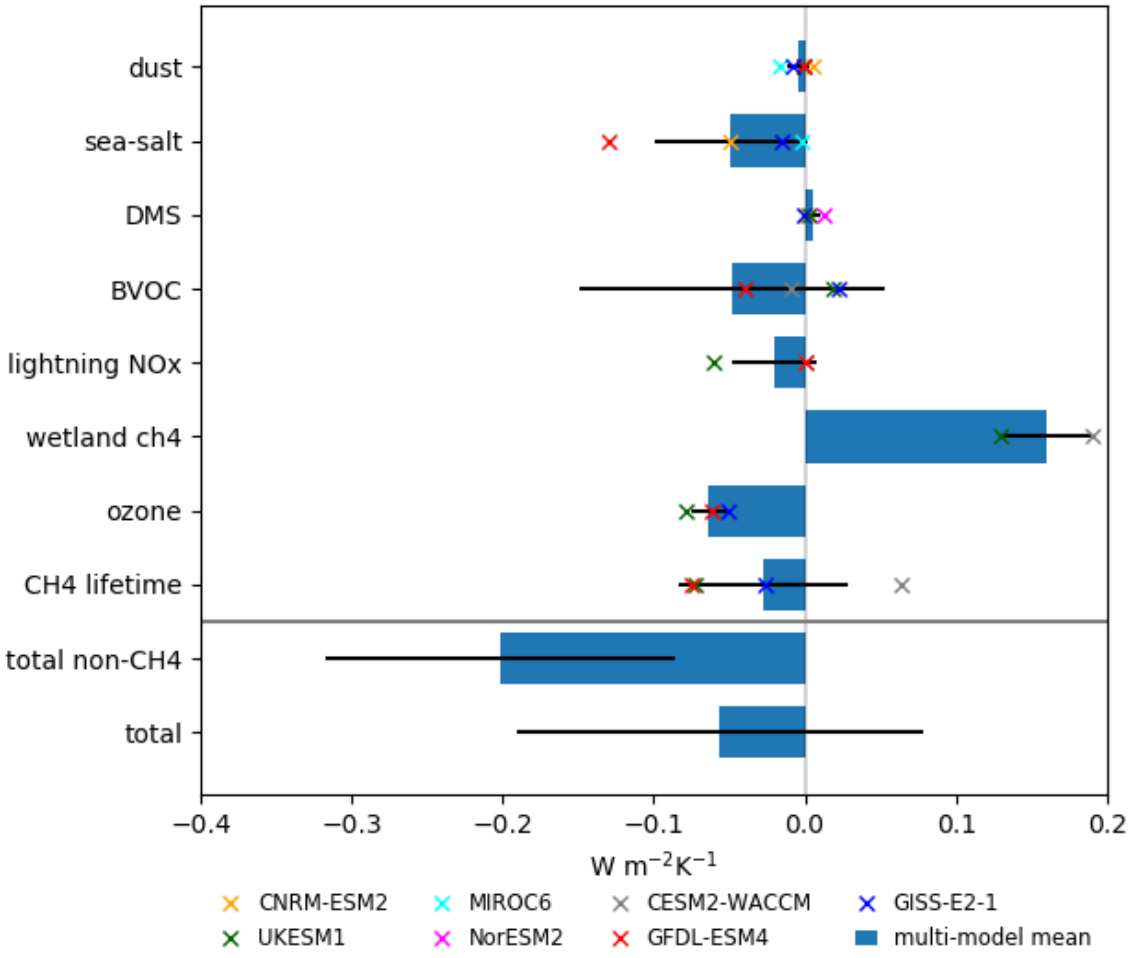


Figure 5. Feedback parameters of all the aerosol and chemical processes in table ~~16~~15. Multi-model mean and individual models. Uncertainties are inter-model standard deviations. BVOC and lightning~~NOx~~ are the sum of~~include~~ aerosol, ozone and methane lifetime effects (points are only shown for models that include all effects). Ozone and CH4-lifetime are the chemical effects (ie. excluding BVOC and lightning emissions) Non-CH4 is the sum and excludes methane lifetime effects and wetland feedback.

Process	Feedback parameter α ($Wm^{-2} K^{-1}$)
Dust (AOD)	-0.004 ± 0.007
Sea Salt (AOD)	-0.049 ± 0.050
DMS	-0.005 ± 0.006
BVOC (aAerosol mass)	-0.09 ± 0.10
BVOC (ozone)	0.011 ± 0.004
BVOC (τ_{CH_4})	0.031 ± 0.018
lNO$_x$ lightning NOx (Aerosol)	-0.0021 ± 0.0023
lightning NOxlNOx (ozone)	0.009 ± 0.007
lightning NOxlNOx (τ_{CH_4})	-0.018 ± 0.017
Wetland	0.163± 0.032
Chemistry (ozone)	-0.064 ± 0.012
Chemistry(τ_{CH_4})	-0.027 ± 0.056
Total non-methane	-0.183 2± 0.111
Total	-0.038 ± 0.131

Table 156. Feedback parameters of all the aerosol and chemical processes addressed in this study. Uncertainties are inter-model standard deviations.

5. Discussion

5.1 Dust

Dust aerosol feedback assessments are a relatively new area of research owing to the large uncertainties of climate models in simulating dust aerosols with changes in atmospheric composition. For instance, the spread in model estimates for dust aerosol changes in the 21st century is the largest among wildfires, biogenic SOA and DMS sulphate (Carslaw et al., 2010). Predictions for future dust emission range from an increase (Woodward et al., 2005) to a decrease (Mahowald and Luo, 2003). The modelled feedbacks in section 4.1.1 have a range of −0.016 to +0.048 W m^{-2} K^{-1} compared to the theoretical model estimates of −0.04 to +0.02 $Wm^{-2}K^{-1}$ by Kok et al. (2018).

The model ranges in dust forcing and feedbacks are not surprising in light of past studies that highlight model differences in dust emitting winds and dust aerosol parameterizations that contribute to the model diversity in the dust aerosol loading, optical properties, and radiative effects (Ackerley et al., 2012; Evan et al., 2014; Huneeus et al., 2011; Shao et al., 2011; Zender et al., 2004). For instance, the parameterization of the planetary boundary layer plays an important role in determining the dust

loading, forcing, and regional feedbacks on winds (Alizadeh Choobari et al., 2012). Influencing factors for regional differences in the dust radiative effects are the surface albedo and aerosol size distribution (Kok et al., 2018; Xie et al., 2018), whereas feedbacks on winds depend also on meteorological factors (Heinold et al., 2008). The substantial model differences in the dust emission response to *4xCO2* paired with corresponding differences in mean 10m wind speed in this study suggests that also the dust feedback parameter critically relies on accurately simulating atmospheric dynamics. Modelling atmospheric circulation has been identified as a grand challenge in climate research (Bony et al., 2015). Currently, we have no estimate which of the dust feedbacks shown are the most plausible, because convective dust storms are missing in such models, but this dust storm type is believed to be important for North African dust emissions (Heinold et al., 2013). Moreover, natural aerosol-climate feedbacks are thought to depend on the anthropogenic aerosol burden and might therefore be both time-dependent and underestimated in the present-day polluted atmosphere (Spracklen and Rap, 2013). Taken together, we have a low confidence in the feedback estimates for dust aerosols to increases in atmospheric concentrations of greenhouse gases.

5.2 Sea Salt

The doubled sea salt ERF in section 4.1.2 is -0.35 to -2.28 W m^{-2}, higher end than found in the literature (-0.3 to -1.1 W m^{-2} which is for direct forcing only (Yue and Liao, 2012)). The efficiency per AOD ranges from -20 to -39 W m^{-2}, again higher than the literature for direct forcing (-18 to -24 W m^{-2} (Heald et al., 2014; Yue and Liao, 2012)).

5.3 DMS

DMS is produced by marine biological activity in the ocean, and it is assumed to be the largest natural source of sulphur to the atmosphere. Up to now, there has been no comprehensive model effort to include all the important effects, and therefore the DMS emission strength change under climate change is still uncertain. The range here (-0.010 to -0.075 W m^{-2} K^{-1} including increases in sulphur lifetime) encompasses the -0.02 W m^{-2} K^{-1} from AR5 (Ciais et al., 2013), based on results from only one model (HadGEM2-ES).

DMS production is closely linked to primary production. Modelling studies including ocean biogeochemistry have shown that under climate change, an increased stratification of the ocean at low and mid latitudes leads to a reduction in nutrients supply into the surface ocean and thus a reduction in DMS emissions, whereas at high latitudes, retreat of sea-ice can lead to increased primary production and increase in DMS production (Kloster et al., 2007). Globally, most models which include ocean biogeochemistry show a slight increase in DMS production and emission to the atmosphere in a warming climate (Bopp et al., 2004; Gabric et al., 2004; Gunson et al., 2006; Vallina et al., 2007).

Some more recent studies have included the impact of ocean acidification on ocean DMS production (Schwinger et al., 2017; Six et al., 2013). Both studies used a very similar description of the ocean biogeochemistry and extended it with an observationally based relation between ocean alkalinity and ocean DMS production. Assuming a medium sensitivity of the DMS production on pH, Six et al. (2013) found a global DMS emission decrease by 18% in 2100 under the SRES A1B scenario, and Schwinger et al. (2017) an emission reduction by 31% in 2200 under the RCP8.5 scenario. In addition recent work has provided evidence for major pathways in the oxidation of DMS in the atmosphere which are not included in any of these ESMs ((Berndt et al., 2019; Wu et al., 2015).

5.4 BVOC

When emissions of BVOCs are increased we see changes to organic aerosol concentration and (in some models) the atmospheric concentrations or lifetime of O_3 and CH_4, with competing effects on climate. At the multi-model mean level, the cooling associated with an increase in organic aerosol (-0.113 ± 0.102 W m^{-2} K^{-1}) outweighs the warming associated with an increase in O_3 (0.017 ± 0.011 W m^{-2} K^{-1}) and an increase in CH_4 lifetime (0.041 ± 0.030 W m^{-2} K^{-1}).

Using multi-annual simulations of global aerosol, Scott et al. (2018) diagnosed a feedback from biogenic secondary organic aerosol of -0.06 W m^{-2} K^{-1} globally, and -0.03 W m^{-2} K^{-1} when considering only extra-tropical regions. This global feedback value was composed of a direct aerosol radiative feedback of -0.048 W m^{-2} K^{-1} and an indirect aerosol (i.e., cloud albedo) feedback of -0.013 W m^{-2} K^{-1}. Using observations from eleven sites, Paasonen et al., (2013) estimated an indirect aerosol feedback of -0.01 W m^{-2} K^{-1} due to biogenic secondary organic aerosol. The ability of models to account for changes in vegetation has a significant effect on the feedback. Sporre et al (2019) found interactive vegetation, enhanced BVOC emissions by 63% greater relative to prescribed vegetation, producing more organic aerosol and an increase in (negative) aerosol forcing. The level of compensation between increased aerosol forcing and increased ozone and methane lifetime is dependent on the model (here positive feedback for GFDL, negative for UKESM1 and WACCM). Unger (2014) found a positive feedback in NASA GISS ModelE2, whereas Scott et al. (2014) found a negative feedback in HadGEM2-ES.

5.5 Lightning

The ESMs used in CMIP6 all use a cloud-top height parameterisation of lightning. Such schemes have previously been found to increase lightning production in warmer climates whereas more sophisticated schemes based on convective updraft mass flux or ice flux show decreases in lightning with temperature. (Clark et al., 2017; Finney et al., 2016b, 2018). Two models here (UKESM1 and WACCM) show increases in lightning emissions of 0.27 and 0.21 Tg(N) yr^{-1} K^{-1} which is slightly lower than the results from the Atmospheric Chemistry and Climate Model Intercomparison (ACCMIP) of 0.44 Tg(N) yr^{-1} K^{-1} (Finney et al., 2016a).

5.6 Wetland methane

Wetland emissions are more strongly sensitive to CO_2 concentrations than to temperature or precipitation (Melton et al., 2013), so the values presented here are more likely to be "adjustments" to the CO_2 rather than feedbacks, and hence could be considered part of the CO_2 ERF. We find emission increases following quadrupled levels of CO_2 of 130-160%. This compares with results from the Wetland CH4 Inter-comparison of Models Project (WETCHIMP) of 20-160% following an increase in CO_2 of a factor of 2.8 (Melton et al., 2013). The CMIP6 simulation specifications do not include free-running methane concentrations therefore the effects of these increased wetland emissions will not be realised in any of the CMIP6 experiments. Outside CMIP6, ESMs are starting to include free-running methane (Ocko et al., 2018), so for these it will be important to understand the effects of changing CO_2 and meteorology on wetland emissions.

5.7 Temperature and humidity discussion

We find a decrease in methane lifetime of -4.5 to -4.6 % K^{-1} in UKESM1 and GFDL-ESM4, but an increase of 0.9 % K^{-1} in WACCM. The first two models compare well with ACCMIP which found a sensitivity of 3.4±1.4% K^{-1} (Naik et al., 2013; Voulgarakis et al., 2013). The impact of climate (including natural emission changes) on tropospheric ozone varies from negative in UKESM1 (-0.33 DU K^{-1}) to positive in GFDL-ESM4 and WACCM (0.18 and 0.16 DU K^{-1}). ACCMIP also found a variation in sign amongst models −0.024±0.027 W m^{-2} for a 1850-2000 change in climate (equivalent to -0.57±0.64 DU using the same radiative efficiency as table 15). The ACCMIP models generally did not include stratospheric chemistry so either explicitly prescribed the cross-tropopause flux of ozone or imposed a climatology of ozone above the tropopause. The three CMIP6 models here all treat the chemistry seamlessly across the troposphere and stratosphere so the impact of changes in stratosphere-troposphere exchange (STE) of ozone on the tropospheric column is likely to be different to ACCMIP.

Changes in the stratospheric ozone column following a quadrupling of CO_2 are driven by cooling temperatures in the stratosphere. This is likely to be due to temperature adjustments to the stratospheric CO_2 concentrations, and so part of the ERF for CO_2 rather than a feedback (Smith, submitted). Feedbacks and adjustments cannot be distinguished with this experimental setup.

6 Conclusions

Earth system models include more processes than physical-only climate models. These models will inherently include additional climate feedbacks, and so have a different overall climate feedback (and climate sensitivity) to their physical counterparts. In this study we consider seven~~six~~ earth system models (~~CNRM-ESM2~~CNRM-ESM2-1, UKESM1, MIROC6, NorESM2, GFDL-ESM4, ~~and~~ CESM2-WACCM and GISS-E2-1). Six~~Five~~ of these (~~CNRM-ESM2~~CNRM-ESM2-1, UKESM1, MIROC6, NorESM2~~, and~~ GFDL-ESM4 and GISS-E2-1) participated in the aerosol-related feedback experiments,

and four~~three~~ (UKESM1, GFDL-ESM4, ~~and~~ CESM2-WACCM and GISS-E2-1) in the ozone and methane~~chemistry~~-related feedback experiments.

We focus in this study on the responses to an abrupt forcing of quadrupled CO_2 concentrations as that is the usual method to diagnose climate feedbacks. By convention the feedbacks are quantified as a response to temperature (in W m^{-2} K^{-1}), but they may not necessarily be applicable to drivers of climate change other than CO_2 as some of the "feedbacks" may be instead adjustments to CO_2 concentrations. It should also be noted that *abrupt-4xCO2* feedbacks are based on atmospheric conditions representative of 1850s and thus may not be applicable to future responses starting from present day conditions. For many of the forcing agents considered here the forcing pattern varies strongly on regional scales, and would be expected to cause larger regional temperature changes than represented by the global mean. Thus aerosol-mediated feedbacks may alter the pattern of climate response as well as the magnitude.

Here we find that the dominant feedbacks are negative i.e. that they act to dampen the response to an imposed forcing. The total feedback, excluding inferred changes in methane, is -0.183~~228~~±0.1112~~3~~ Wm^{-2} K^{-1}. The increase in organic aerosols from increase emission of volatile organic compounds (VOCs) from vegetation makes the largest contribution to both the magnitude of the feedback and its uncertainty (-0.09~~113~~±0.10~~102~~ Wm^{-2} K^{-1}) with increases in sea salt and DMS emissions~~sulphate aeroso l~~also contributing. ~~The increase in sulphate comes both from an increase in DMS emissions and a decrease in sulphate removal.~~

Contributions from increases in ozone production from biogenic VOCs and lightning NOx are ~~partially~~ offset by decreased tropospheric ozone lifetime in a warmer climate leading to an overall negative feedback through ozone. ~~Stratospheric ozone does substantially increase.~~ Diagnoses of changes in wetland emissions of methane indicate that if ESMs did allow methane to vary interactively the combined aerosol and chemical feedbacks would be substantially less negative and consistent with zero.

The aerosol and chemistry feedbacks listed here contribute up to the order of -0.2 Wm^{-2} K^{-1} . This is smaller in magnitude than the carbon cycle response to climate (of order 0.5 Wm^{-2} K^{-1} (Ciais et al., 2013)) or the physical climate feedbacks (of order 1-2 Wm^{-2} K^{-1} (Sherwood et al., 2020)).

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Author Contributions

Manuscript preparation was by WC, GT, DO, RC-G, CES, SF and additional contributions from all co-authors. Model simulations were provided by SB, GF, AG, , J-FL, MM, JM, PN, TT. Analysis was carried out by GT, WC, DO, SF, RC-G, JW. Ozone radiative forcing was generated by RBS

Data Availability

All data from the Earth system models used in this paper are available on the Earth System Grid Federation Website, and can be downloaded from there. https://esgf-index1.ceda.ac.uk/search/cmip6-ceda/

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