Responses to reviews of 'Effective radiative forcing from emissions of reactive gases and aerosols – a multi-model comparison' by G. Thornhill et al

We would like to thank the two referees for their helpful comments and suggestions. Our responses to the reviewers comments are below – reviewer comments in black, our responses in blue.

Changes and edits to the text are noted in blue.

Anonymous Referee #1

This paper examines the effective radiative forcing (ERF) of reactive gases (e.g., CH4, NOx, VOC, O3, N2O, HC, NH3) and aerosols (e.g., BC, OC, SO2) to the climate system using multi-model output from the AerChemMIP experiments of the CMIP6 project. The contribution of each species to the total ERF is decomposed, and the differences of the calculated ERFs by various models are discussed. The paper is overall well written and easy to follow. I have some minor comments for the authors to consider before publication:

1. It is not clear how many ensemble members are used for each model. Can you please clarify this?
We have clarified that only one ensemble member was used in these experiments in Section 2.2

2. Fixed SST and sea ice are used in the ERF simulations. Is it the climatological SST of the 1850s?
These were the fixed SSTs from the Pre-Industrial run – mentioned in L. 185.

3. The description of Eq. 5 is a bit vague. You may want to add "ERFaci" to line 240 after "The effect of the aerosol on cloud radiative forcing".
We have re-written this section and changed the nomenclature to be consistent with other work in this field, so that the descriptions of each term is clear.

4. "A_trop" is used in text while "RA Trop" is used in Fig. 1. It's better to be consistent.
We have changed the nomenclature and definitions in the text so that we use A_{total} in the text and the Fig.1 for this quantity.

5. Line 373, "Fig. 6"! "Fig. 5". And can you explain a bit more about how the total AOD is used to calculate the sum of the scaled ERFs?
We have corrected the figure number and have added additional explanation on how the AOD is used in this scaling. There is also more detailed discussion in the Supplementary material.

6.Table 6, second row, "Nox"! "NOx"
The typo has been corrected.

7. Line 465, please remove the brackets around "O'Connorl F. M. 2019"
The reference has been fixed.

8. Error bars are used in Fig. 9-10 to quantify the uncertainties due to interannual variability of model diagnostics. Is it possible to apply similar approach (error bars) to other figures where data are available?

We have added error bars to Fig. 1 and Fig. 6 to show the S.E. for the ERF individual models (for the inter-annual variability) and the S.D. for the multi-model means.

9. Line 507, “RFs”! “ERFs”
This has been corrected.

10. Line 525, “+/-“! “_”, please also add explanation about the numbers follow the sign, for instance, is it a standard deviation of multi-model output?
We have clarified where the numbers are standard error or standard deviation throughout the manuscript.

11. Lines 541-542, the overall aerosol ERFari from AR5 (-1.5 _ 0.4 Wm-2) is much larger than values reported here (-0.16 _ 0.03 Wm-2). Can you add some discussion about the differences?
In fact the comparison here was incorrect – the AR5 number quoted is the total ERF, not the IRFari (in the original manuscript this was referred to as the ERFari), so this has been corrected.

12. Lines 546-550, redundant
The redundant lines have been removed.

Anonymous Referee #2

Thornhill et al. "Effective Radiative forcing from emissions 1 of reactive gases and aerosols – a multimodel comparison"
This paper is a very important contribution to climate and atmospheric chemistry studies. It is critical for the current IPCC AR6 assessment. My apologies to the authors for the delay in my review – surprisingly, these isolation times do not make it easier to review. I am rushing to get this out and so there may be typos in this review.
AerChemMIP is a very important project that is trying to make sense of a complex, nonlinear system of chemistry and clouds. The design has some serious flaws, and we all knew that as it was being developed since there were obvious limitations in the number and complexity of experiments. First, there is the nonlinearity, which comes out clearly in these results: Exp(A) plus Exp(B) does not equal Exp(A+B). Further, and because of the nonlinear nature, the choice of reference atmosphere (1850) will produce quite different results than another (2014). Personally I would have much preferred to use the 2014 atmosphere as the reference atmosphere (at least we can compare the models to measurements) and then subtract the emission or concentration changes from 1850 to 2014. So, we live with AerChemMIP and use it. The analysis here is very good, but needs to develop more of an "assessment" view when reporting final numbers. They should reflect some (subjective) adjustment of values (for non-linearity) or bias (from 1850 atmosphere). Simply reporting the model average for Exp(A) and then Exp(B) will lead anyone who uses this paper to assume that the combined effect is the sum. That would not be good for either policy or attribution work. If the sum is the best answer (I would think that so), then the contributions of components may need to be expressed in % of total rather than in W/m2. I do not recommend a substantial rewrite, but rather a self-assessment by the author team of how to use these results.
We have added discussion of the non-linearity and where that would affect a simplistic interpretation of combined results in the relevant sections, and we have added comments on the limitations of the experimental design.

The abstract starts off being very careful and clear, but then I get lost between emissions and composition change. For example, SO2 is clearly an emission-based ERF, while CH4 is an abundance-based ERF. VOCs & NOx are obviously emissions (having direct RF) but O3 is not emitted. The HC (L40: halocarbons? as opposed to NMHC?) are negative presumably from the ozone depletion, but this is an odd way to list the CFCs. Methane is singled out at the end of the abstract as increasing ozone, but ozone is listed separately. Is the methane based only on the change in CH4 concentration? Further, I suspect that the methane includes stratospheric water vapor which is indirect as is ozone, so why the separate listings. This is just inconsistent and you really need to present the framework and rules for partitioning and assigning ERF. Basically, AerChemMIP is a complicated set of overlapping experiments and thus the abstract with simple results is very difficult to write correctly. Try to keep it clear and simple.
We have added text to the abstract to include a better description of the relationship between the ozone pre-cursors and the ERFs calculated.

L53- aerosols are chemically reactive. try again – aerosols and gases that …
We have re-worded this as chemically reactive gases to make the distinction clearer.

L56- climate feedbacks on natural emissions is a tough one for the non-CO2 species. I do not think AerChemMIP did anything on that.
The discussion of natural emissions has been removed. These are covered in Thornhill et al. 2020.

L68- "conditions" do you mean SST, I doubt you prescribed different chlorophyll or DMS?
We have clarified which conditions are meant here (SSTs and sea-ice).

L69- why aerosols here? It seems more like and ie than and eg, why not just say aerosols and gases.
This has been reworded.

L71- again the contrast of 'aerosols and chemistry' is really a poor description of AerChemMIP. Aerosols are a chemically reactive species (most of them are, they were created by gas phase chemistry). This Intro really must have a better inclusive discussion of greenhouse gases (including ozone), of indirect greenhouse gases (CO) and of aerosol (primary and secondary). This is at odds with the content of AerChemMIP.
This section has been expanded to clarify the indirect roles of precursor species as recommended by the reviewer.

L90- again, every time I read this, it sounds odd and misleading: " aerosols and reactive chemistry" are not opposites.
This has been reworded as “aerosols and reactive gases”.

L93- again, this phrasing sounds wrong: " anthropogenic and reactive species" is this two separate species or is it 'both'
This has been reworded as “aerosols and reactive gases”

L103- what does 'down to' mean? from 0.001 to 560 hPa?.
This has been reworded to clarify the levels of the atmosphere.

L107- typo 'is includes'

Typo has been corrected.

L102-L165 This information should really be in a table somewhere, not in the text. Focus on the results. In fact, most of this is already in Table 1.
The summary of different model aerosol and chemistry modules has been retained in the text, and the details moved to Supplementary Table S1.

L172-173- You should say 'emissions' with the NTCFs, also you should note that methane is an SLCF, which your statement seems to preclude. Also, I thought that SLCF was new preferred, but…
The use of NTCF was retained because the experiment was called piClim-NTCF, so this nomenclature was kept to aid in understanding which experiment was referred to. We state that the point that this nomenclature has changed and that SLCF is now preferred. In the piClim-NTCF experiment in AerChemMIP methane is deliberately excluded as part of the experimental design.

L174- There is a serious problem with the AerChemMIP as defined and we realized this at the time, but did not address it: viz. because of the large changes in atmospheric chemistry and oxidants between 1850 and 2014 (including the ozone depletion), it is not clear that the effect of today's NTCF emissions in today's atmosphere are anywhere close to those calculated here for the PI atmosphere. This needs to be addressed when compiling results and clearly adds to the uncertainty. The reason why it is dangerous is that it could totally misrepresent the magnitude of the response if we were to cut NTCF today.
The reviewer has a good point here. We have added discussion of the issue likely differences between perturbing emissions in a pre-industrial and present day atmosphere, and commented in places where this would influence the results.

L200- "ocean state" implies much more than SSTs – do you really mean that.
This has been corrected.
.
L235- I am confused here, it seems like the direct forcing would include, not ignore, the absorption and scatting by aerosols. Is this a typo.
This was a typo, and the section has been re-written in order to explain the method more clearly.

L239- this section is very confusing as written. I am sure it is simpler that it seems but some of the writing seems incorrect. In this line surely you mean the aerosol direct radiative effect, since "radiative effects due only to aerosols" would imply both ari and aci. The notation for ERFcs,af is inconsistent between eqn and text (comma or not). Eqns 4 & 5 sum to give not ERF, so where is this missing 'surface albedo' term (ERF-ERFcsaf) and does it matter? It must certainly be counted as an aerosol ERF. [OK, I see this in Table 3 later, is the cs,af just noise?]
This section has been re-written and clarified, with additional discussion and explanation of the terms and their meanings.

L254ff- This is odd, you said just above the SARF is calculated from ERF – A_trop, so of course this should give 0% difference. Are you just checking the math? Also, with SARF calculated as a residual term, and the ability to denote and sum all the A_trop being highly uncertain, SARF would not appear to be very certain. In fact the SARF term would depend on the models' ability to diagnose A_trop correctly.

This has been explained more clearly in Section 4.1.2, and the reason for the difference in the calculations clarified as due to the way the IRF is calculated in the kernel method. We have now used the IRF instead of the SARF in this section, as it is more consistent with previous work.

L260- With BC, the long-standing problem is that some models get far too much in the stratosphere and that would cause a very large SARF.
We have added more discussion of the BC results, and the reasons for model differences.

L285- in this figure and some others, please define carefully what the shorthand for the terms means.
We have clarified the terms and included the definitions in the figure captions.

L296ff- This is a very good discussion of the aerosol components!
Thank you, nice to hear what is good about the paper!

L321- This would be a much better lead off to the aerosol section and analysis, begin with the big picture before the weeds.
We have moved this to the top of the results section.

L343 – just put this table into the figure, it is just a summary of the bars anyway.
We have revised the plot, added error bars for both inter-annual variability and the multi-model mean, and moved the relevant tables to the supplementary material.

L354- Please, stop wasting space, Table 5 and Fig 4 has the same information. If you want to show AOD, then add it to the figure..
We have removed this plot and added the information to Table 4.

L370ff & Fig 5- I do not understand the purpose of this AOD scaling, it really does little to help. The figure shows the key data: ERF from parts vs ERF from all. The ERF-parts consistently over accounts for the combined ERF. This is as expected wince the cloud effects are largest in a pi atmosphere with little background aerosols. So herein lies one of the fundamental problems with the AerChemMIP that must be acknowledged and accounted for. I am not sure that scaling by AOD is any more justified than just scaling the indirect to match – An interesting question is whether the ERF-ari sum of the parts equals the whole? Keep this fudge factor simple since there is no correct way to do this.
I have moved the discussion on scaling to the Supplementary material, and removed the unnecessary figure. I have added appropriate caveats on the non-linearity and the effect of using the PI atmosphere, and the effect on these results.

L393- Again, this title jars a bit, SO2 and NH3 are reactive gases.
This has been renamed to reactive greenhouse gases.

L406- How big is the error in GHGas ERF if one ignores clouds? I would think large.
We have added a discussion of the expected size of the cloud masking effect.

Fig 6 & Table 6, can easily combine and understand the Std Dev better.
This has been done, as for the previous ERF plots. The Fig. now contains the error bars and the multi-model means, with a Table of values in the Supplemental material.

L419- Here is a case where some assessment is due as to how well these model simulations are accurate in the sense of including all the effects. As noted the N2O-O3 link is important and missing in some, and the other key link between N2O and tropospheric OH and methane (and maybe aerosols) is established but missing here.

We have added more discussion on this. For the relevant impacts on stratospheric/tropospheric ozone, and methane lifetime, only the models with appropriate chemical processes are considered.

L431-441- This is a very interesting and important discussion about the additivity of the components. I suspect that the CH4 result is similarly affected.

BTW, where is the effect of stratospheric H2o form CH4 noted or counted?

We have added plots of the water vapour changes from the CH4 experiment in the Supplementary Materials.

L480- Yes halocarbons, but also N2O, and N2O may be more important since it depletes ozone in the tropics (there are papers on this). I do not see what difference stratospheric Cl will make on the total lifetime – hopefully the Stevenson paper becomes referencable. Also, you need to be careful here since the methane feedback factors ff, apply to the PI atmosphere, and that is different from the present, particularly lower CO and NOx…. . The feedback factor used for GWPs etc, is the current one, not the PI one, so these results should NOT be used to change any previous assessments.

We have mentioned the change due to N2O as well, and clarified that the f factors are starting from a pre-industrial atmosphere.

Fig 11 is really hard to understand or see clearly, it will need a cleanup.

This figure has been revised to make it cleaner and easier to read.

L525- Here is a very important statement and I am not sure that you have put together all the reasonable uncertainties or non-linear scaling. The individual components here must be adjusted to recognize that the total ERF (with all simultaneously) is much less than the sum of the components. Thus you cannot recommend the individual results without scaling and without increasing the uncertainty.

We have revised the discussion to explain the differences between the individual components and the total, and the implications for uncertainty.

L561- Same as above. The individual values will not sum correctly and so these do NOT reflect the ERF of NOx emissions as we progress from 1850 to 2014. Thus they should not be used as part of an assessment until they are more critically evaluated and put in context.

These are all very important AerChemMIP results, and the analysis here is highly valuable, but their use in attribution and related studies should reflect the bias and uncertainties in combining nonlinear parts that were calculated separately, and in basing these all on a pi-atmosphere.

This discussion has been revised to explain the differences between the sum of NOx and VOC and the total. The ERFs are typically defined starting from a pre-industrial atmosphere, there is no unique way to reflect the ERF of NOx emissions as we progress from 1850 to 2014 (subtracting components from a present atmosphere would overestimate the ERF).

Effective radiative forcing from emissions of reactive gases and aerosols – a multi-model comparison

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Abstract

This paper quantifies effective radiative forcing (ERF) of the present-day anthropogenic emissions of NO_X, VOCs (including CO), SO_2, NH_3, black carbon and organic carbon. Effective radiative forcings from pre-industrial to present-day changes in the concentrations of methane, N_2O and ozone-depleting halocarbons are quantified. Concentration and emission changes of reactive species can cause multiple changes in the composition of radiatively active species: tropospheric ozone, stratospheric ozone, stratospheric water vapour, secondary

inorganic and organic aerosol and methane. Where possible we break down the ERFs from each emitted species into the contributions from the composition changes.

The ERFs are calculated using models that participated in the AerChemMIP experiments as part of the CMIP6 project.

The 1850 to 2014 multi-model mean ERFs (± standard deviations) are -1.03 ± 0.37 Wm^{-2} for SO_2 emissions, -0.25 ± 0.09 Wm^{-2} for organic carbon (OC), 0.15 ± 0.17 Wm^{-2} for black carbon (BC), for NH_3 it is -0.07 ± 0.01Wm^{-2} and for the aerosols combined it is -1.01 ±0.25 Wm^{-2}. The multi-model means for the reactive well-mixed greenhouse gases (including any effects on ozone and aerosol chemistry) are 0.67 ± 0.17 Wm^{-2} for methane (CH_4), 0.26 ± 0.07 Wm^{-2} for nitrous oxide (N_2O) and 0.12 ± 0.2 Wm^{-2} for ozone-depleting halocarbons (HC), Emissions of the ozone precursors nitrogen oxides (NO_x), volatile organic compounds (VOC) and both together (O_3) lead to ERFs of 0.14 ± 0.13 Wm^{-2}, 0.09 ± 0.14 Wm^{-2} and 0.20 ± 0.07 Wm^{-2} respectively. The differences in ERFs calculated for the different models reflect differences in the complexity of their aerosol and chemistry schemes, especially in the case of methane where tropospheric chemistry captures increased forcing from ozone production.

1. Introduction

The characterisation of the responses of the atmosphere, climate, and earth systems to various forcing agents is essential for understanding, and countering, the impacts of climate change. As part of this effort there have been several projects directed at using climate models from different groups around the world to produce a systematic comparison of the simulations from these models, via the Coupled Model Intercomparison Project (CMIP), which is now in its 6th iteration (Eyring et al., 2016). This CMIP work has been subdivided into different areas of interest for addressing specific questions about climate change, such as the impact of aerosols and reactive greenhouse gases, and the AerChemMIP (Collins et al., 2017) project is designed to examine the specific effects of these factors on the climate. The aerosol and aerosol precursor species considered are sulphur dioxide (SO_2), black carbon (BC), organic carbon (OC). The reactive greenhouse gases and ozone precursors are methane (CH_4), nitrogen oxide (NO_X), volatile organic compounds (VOCs – including carbon monoxide), nitrous oxide (N_2O) and ozone-depleting halocarbons (HC).

The focus of this work is to characterise the effect of the change from pre-industrial (1850) to present day (2014) in aerosols and their precursors, and chemically-reactive greenhouse gases (including species that affect ozone) on the radiation budget of the planet, referred to as radiative forcing, as an initial step to understanding the response of the atmosphere and earth system to changes in these components. In previous reports of the Intergovernmental Panel on Climate Change (IPCC) the effect of the various forcing agents on the radiation balance has been investigated in terms of the radiative forcing, (RF), which is a measure of how the radiative fluxes at the top of atmosphere (TOA) change in response to changes in, e.g., concentrations or emissions of greenhouse gases and aerosols. There have been several definitions of radiative forcing, (Forster et al., 2016;Sherwood et al., 2015), which generally considered the instantaneous radiative forcing (IRF), or a combination of the IRF including the adjustment of the stratospheric temperature to the driver, generally termed the stratospheric-temperature adjusted radiative forcing. More recently (Boucher, 2013;Chung and Soden, 2015) there has been a move towards using the effective radiative forcing (ERF) as the preferred metric, as this includes the rapid adjustments of the atmosphere to the perturbation, e.g. changes in cloud cover or type, water vapour,

tropospheric temperature, which may affect the overall radiative balance of the atmosphere. In this work, ERF is calculated using two atmospheric model simulations both with the same prescribed sea-surface temperatures (SSTs) and sea ice, but one having the perturbation we are interested in investigating, e.g. a change in emissions or concentrations of aerosols or reactive gases. The difference in the net TOA flux between these two simulations is then defined as the ERF for that perturbation.

Previous efforts to understand the radiative forcing due to aerosols and reactive gases in CMIP simulations have resulted in a wide spread of values from the different climate models, in part due to a lack of suitable model simulations for extracting the ERF from, e.g., a specific change to an aerosol species. The experiments in the AerChemMIP project have been designed to address this in part, by defining consistent model set-ups to be used to calculate the ERFs, although the individual models will still have their own aerosol and chemistry modules, with varying levels of complexity and different approaches.

There are complexities in assessing how a particular forcing agent affects the climate system due to the interactions between some of the reactive gases; for example methane and ozone are linked in complex ways, and this increases the problem of understanding the specific contribution of each to the overall ERF when one of them is perturbed. An attempt to understand some of these interactions is discussed in Section 4.2 below.

The experimental set-up and models used are described in Section 2, the methods for calculating the ERFs for the aerosol and chemistry experiments are described in Section 3, and the results are discussed in section 4. Final conclusions are drawn in Section 5.

2. Experimental Setup

2.1 Models

This analysis is based on models participating in the Coupled Model Intercomparison Project (CMIP6) (Eyring et al., 2016), which oversees climate modelling efforts from a number of centres with a view to facilitating comparisons of the model results in a systematic framework. The overall CMIP6 project has a number of sub-projects, where those with interests in specific aspects of the climate can design and request specific experiments to be undertaken by the modelling groups. To understand the effects of aerosols and reactive gases on the climate, a set of experiments was devised under the auspices of AerChemMIP (Collins et al., 2017), described in Section 2.2.

The anthropogenic emissions of the aerosols, aerosol precursors and ozone precursors (excluding methane) for use in the models are given by Hoesly et al. (2018) and van Marle et al. (2017) Models use their own natural emissions (Eyring et al., 2016). The well-mixed greenhouse gases (WMGHG), CO_2, CH_4, N_2O and halocarbons are specified as concentrations either at the surface or in the troposphere. Not all of the models include interactive aerosols, tropospheric chemistry and stratospheric chemistry, which is the ideal for the AerChemMIP experiments, but those models which do not include all these processes provide results for a subset of the experiments described in Section 2.2.

The models included in this analysis are summarised below, and in Table 1 with an overview of the model set-up, aerosol scheme and type of chemistry models used included. A more detailed description of each model and the aerosol and chemistry schemes used in each is available in the supplementary materials, Table S1.

The CNRM-ESM2-1 model (Séférian et al., 2019;Michou et al., 2020) includes an interactive tropospheric aerosol scheme, and an interactive gaseous chemistry scheme only above the level of 560 hPa. The sulfate precursors

evolve to SO_4 using a simple dependence on latitude. The cloud droplet number concentration (CDNC) depends on SO_4, organic matter and sea-salt concentrations, so the aerosol cloud-albedo effect is represented, although other aerosol-cloud interactions are not.

The UKESM1 model (Sellar et al., 2020) includes an interactive stratosphere-troposphere gas-phase chemistry scheme (Archibald et al., 2020) using the UK Chemistry and Aerosol (UKCA); (Morgenstern et al., 2009;O'Connor et al., 2014) model. The UKCA aerosol scheme, called GLOMAP-mode is two-moment simulation of tropospheric black carbon, organic carbon, SO_4 and sea salt. Dust is modelled independently using the bin scheme of Woodward (2001). A full description and evaluation of the chemistry and aerosol schemes in UKESM1 can be found in Archibald et al. (2020) and Mulcahy et al. (2020) respectively.

The MIROC6 model includes the Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) aerosol model which predicts mass mixing ratios of the main tropospheric aerosols and models aerosol-cloud interactions in which aerosols alter cloud microphysical properties and affect the radiation budget by acting as cloud condensation and ice nuclei (Takemura et al., 2005;Watanabe et al., 2010;Takemura and Suzuki, 2019;Takemura, 2018;Tatebe et al., 2019).

The MRI-ESM2 model (Yukimoto et al., 2019) has the Model of Aerosol Species in the Global Atmosphere mark-2 revision 4-climate (MASINGAR mk-2r4c) aerosol model, and a chemistry model, MRI-CCM2 (Deushi and Shibata, 2011) which models chemistry processes for ozone and other trace gases from the surface to middle atmosphere. The model includes aerosol-chemistry interactions, and aerosol-cloud interactions (Kawai et al., 2019). The ERFs of anthropogenic gases and aerosols under present-day conditions relative to preindustrial conditions estimated by MRI-ESM2 as part of the Radiative Forcing Model Intercomparison Project (RFMIP) (Pincus et al., 2016) and AerChemMIP are summarized in Oshima et al. (2020).

The BCC-ESM1 model (Wu et al., 2019;Wu et al., 2020) models major aerosol species including gas-phase chemical reactions, secondary aerosol formation, and aerosol-cloud interactions including indirect effects are represented. It does not include stratospheric chemistry, so concentrations of ozone, CH_4, and N_2O at the top two model levels are the zonally and monthly values derived from the CMIP6 data package.

The NorESM2 model contains interactive aerosols and uses the OsloAero6 aerosol module ((Seland et al., 2020), Olivié et al., in prep.) describes the formation and evolution of BC, OC, SO_4, dust, sea-salt and SOA. There is a limited gas-phase chemistry describing the oxidation of the aerosol precursors DMS, SO_2, isoprene, and monoterpenes and oxidant fields of OH, HO_2, NO_3 and ozone are prescribed climatological fields, and there is no ozone chemistry in the model.

The GFDL-ESM4 model consists of the GFDL AM4.1 atmosphere component, (Dunne et al., 2020;Horowitz et al., 2020) which includes an interactive tropospheric and stratospheric gas-phase and aerosol chemistry scheme. Nitrate aerosols are explicitly treated in this model.

The CESM2-WACCM model includes interactive chemistry and aerosols for the troposphere, stratosphere and lower thermosphere (Emmons et al., 2010); (Gettelman et al., 2019). The representation of secondary organic aerosols follows the Volatility Basis Set approached (Tilmes et al., 2019).

The IPSLCM6A-LR-INCA (referred to subsequently as IPSL-INCA) model used for this analysis has interactive aerosols but a limited gas-phase model. The aerosol scheme is based on a sectional approach with to represent the size distribution of dust, sea- salt (which has an additional super-coarse mode to model largest emission of spray-salt aerosols), BC, NH_4, NO_3, SO_4, SO_2 and OA with a combination of accumulation and coarse log-normal modes

with both soluble and insoluble treated as independent modes. DMS emissions are prescribed and not interactively calculated. BC is modelled as internally mixed with sulphate (Wang et al. (2016), where the refractive index is relies on Garnet-Maxwell method. Its emissions are derived from inventories. A new dust refractive index is implemented (Di Biagio et al., 2019). Well mixed trace gases concentrations/emissions are forced with AMIP/CMIP6 datasets (Lurton et al., 2020) ozone using Checa-Garcia et al. (2018) and solar forcing from Matthes et al. (2017).

The GISS-E2-1 model aerosol scheme (One-Moment Aerosol (OMA)) module, which includes sulfate, nitrate, ammonium, carbonaceous aerosols (BC and OC), is coupled to both the tropospheric and stratospheric chemistry scheme. For the results reported here, the physics version 3 of this model configuration was used, which includes the aerosol impacts on clouds. For details of the model, see Bauer et al. (2020).

Table 1 Components used in the Earth system models (detailed Table is in Supplementary material, Table S1)

	Aerosols	Tropospheric chemistry	Stratospheric chemistry
IPSL-CM6A-LR-INCA	Interactive	No	No
NorESM2-LM	Interactive	SOA and sulfate precursor chemistry	No
UKESM1-LL	Interactive Tropospheric. Prescribed stratospheric	Interactive	Interactive
CNRM-ESM2-1	Interactive	Chemical reactions down to 560 hPa	Interactive
MRI-ESM2	Interactive	Interactive	Interactive
MIROC6	Interactive	SOA and sulfate precursor chemistry	No
BCC-ESM1	Interactive	Interactive	No
GFDL-ESM4	Interactive	Interactive	Interactive
CESM2-WACCM	Interactive	Interactive	Interactive
GISS-E2-1	Interactive	Interactive	Interactive

2.2 Experiments

The AerChemMIP timeslice experiments (Table 2) are used to determine the present-day (2014) ERFs for the changes in emissions or concentrations of reactive gases, and aerosols or their precursors (Collins et al., 2017). The ERFs are calculated by comparing the change in net TOA radiation fluxes between two runs with the same prescribed sea surface temperatures (SSTs) and sea ice, but with near-term climate forcers (NTCFs - also referred to as short-lived climate forcers - SLCFs), reactive gas and aerosol emissions, and well-mixed greenhouse gases

(WMGHG - methane, nitrous oxide, halocarbon) concentrations perturbed. It should be noted that in AerChemMIP the NTCF experiment excludes CH_4 the experimental design The control run uses set 1850 pre-industrial values for the aerosol and aerosol precursors, CH_4 N_2O, ozone precursors and halocarbons, either as emissions or concentrations (Hoesly et al., 2018;van Marle et al., 2017;Meinshausen et al., 2017). Monthly varying prescribed SSTs and sea-ice are taken from the CMIP6 DECK coupled pre-industrial (1850) control simulation. Each experiment then perturbs the pre-industrial value by changing one (or more) of the species (emissions or concentrations) to the 2014 value, while keeping SSTs and sea-ice prescribed as in the pre-industrial control. Note adding individual species to a pre-industrial control will likely give different results to a setup where species were individually subtracted from a present-day control. The NTCFs are perturbed individually or in groups. This provides ERFs for the specific emission or concentration change, but also for all aerosol precursor or NTCFs combined (Collins et al., 2017). For models without interactive tropospheric chemistry "NTCF" and "aer" experiments are the same; in the case of NorESM2 for the NTCF experiments the model attempts to mimic the full chemistry by setting the oxidants and ozone to 2014 values. The WMGHG experiments include the effects on aerosol oxidation, tropospheric and stratospheric ozone, and stratospheric water vapour depending on the model complexity.

Thirty years of simulation are required to minimise internal variability (mainly from clouds) (Forster et al, 2016.), and one ensemble member was used for each experiment (almost all models provided only a single ensemble member).

Table 2 List of fixed SST ERF simulations. (NTCF in (Collins et al., 2017) is also referred to as 'SLCF' - short-lived climate forcers - in other publications) and for the purposes of this study excludes methane.

Experiment ID	CH_4	N_2O	Aerosol Precursors	Ozone Precursors	CFC/ HCFC	Number of models
piClim-control	1850	1850	1850	1850	1850	11
*piClim-**NTCF***	1850	1850	**2014**	**2014**	1850	8
*piClim-**aer***	1850	1850	**2014**	1850	1850	9
*piClim-**BC***	1850	1850	1850 (non BC) **2014** (BC)	1850	1850	7
*piClim-**O3***	1850	1850	1850	**2014**	1850	4
*piClim-**CH4***	**2014**	1850	1850	1850	1850	8
*piClim-**N2O***	1850	**2014**	1850	1850	1850	5
*piClim-**HC***	1850	1850	1850	1850	**2014**	6
*piClim-**NOX***	1850	1850	1850	1850 (non NO_x) **2014** (NO_x)	1850	5
*piClim-**VOC***	1850	1850	1850	1850 (non CO/VOC) **2014** (CO/VOC)	1850	5
*piClim-**SO2***	1850	1850	1850 (non SO_2) **2014** (SO_2)	1850	1850	6
*piClim-**OC***	1850	1850	1850 (non OC) **2014** (OC)	1850	1850	6

*piClim-**NH3***	1850	1850	1850 (non NH_3) **2014** (NH_3)	1850	1850	2

3. Methods

In the following analysis we use several methods to analyse the ERF and the relative contributions from different aerosols, chemistry and processes to the overall ERF for the models and experiments described above, where the appropriate model diagnostics were available.

3.1 Calculation of ERF using fixed SSTs

The ERF is calculated from the experiments described above, where the sea surface temperatures and sea-ice are fixed to climatological values. Here the ERF is defined as the difference in the net TOA flux between the perturbed experiments and the piClim-control experiment (Sherwood et al., 2015), calculated as the global mean for the 30 years of the experimental run (where the models were run longer than 30 years, only the last 30 years was used). This allows us to calculate the ERF for the individual species based on the changes to the emission or concentrations between the control and perturbed runs of the models. The assumption is that there is minimal contribution from the climate feedback when the SSTs are fixed, but the resultant ERF includes rapid adjustments to the forcing agent in the atmosphere (Forster et al., 2016).

The ERF calculated using this method includes any contributions to the ERF resulting from changes in the land surface temperature (T_s), which ideally should be removed (Shine et al., 2003;Hansen et al., 2005;Vial et al., 2013) (as the ocean temperature changes are removed by using fixed SSTs). However, there is no simple way to prescribe land surface temperatures in the models considered here analogous to the fixing the SSTs, so we make the land surface temperature correction by calculating the surface temperature adjustment from the radiative kernel (see Section 3.2) and subtracting it from the standard ERF as calculated above (see also Smith et al. (2020a);(Tang et al., 2019)). This is designated the ERF_ts to differentiate it from the standard ERF as described above.

3.2 Kernel Analysis

Where the relevant data are available, we use the radiative kernel method (Smith et al., 2018;Soden et al., 2008;Chung and Soden, 2015) to break down the ERF into the instantaneous radiative forcing (IRF) and individual rapid adjustments (designated by A) which are radiative responses to changes in atmospheric state variables that are not coupled to surface warming. In this approach, ERF is defined as:

$$ERF = IRF + A_T + A_{ts} + A_q + A_a + A_c + e \quad (1)$$

where A_T = atmospheric temperature, A_{ts} = surface temperature, A_q = water vapour, A_a = albedo, A_c = clouds, e = radiative kernel error. Individual rapid adjustments (A_x) are computed as:

$$A_x = \frac{\delta R}{\delta x} dx \quad (2)$$

where $\frac{\delta R}{\delta x}$ is the radiative kernel, a diagnostic tool typically computed with an offline version of a GCM radiative transfer model that is initialized with climatological base state data and dx is the climate response of atmospheric state variable x, diagnosed directly from each model. Cloud rapid adjustments (A_C) are estimated by diagnosing cloud radiative forcing from model flux diagnostics and correcting for cloud masking using the kernel-derived non-cloud adjustments and IRF, following common practice (e.g. (Soden et al., 2008;Smith et al., 2018)), whereby:

$$A_C = (ERF - ERF^{clr}) - (\text{IRF} - IRF^{clr}) - \sum_{x=[T,ts,q,a]} (A_x - A_x^{clr}) \quad (3)$$

For the calculation of the IRF (for aerosols this is the direct effect) here, the clear-sky IRF (IRF^{clr}) is estimated as the difference between clear-sky ERF (ERF^{clr}) and the sum of kernel-derived clear-sky rapid adjustments (A_x^{clr}). Since estimates of A_c are dependent on IRF, the same differencing method cannot be used to estimate IRF under all-sky conditions without special diagnostics (in particular the International Satellite Cloud Climatology Project diagnostics (ISCCP) diagnostics) not widely available in the AerChemMIP archive. Instead, for the calculations presented here all-sky IRF is computed by scaling IRF^{clr} by a species-specific factor to account for cloud masking (Soden, Held et al. 2008).

Kernels are available from several sources, and for this analysis we used kernels from CESM, (Pendergrass et al., 2018), GFDL (Soden et al., 2008), HadGEM3, (Smith et al., 2020b), and ECHAM6 (Block and Mauritsen, 2013) and took the mean from the four kernels for each model. Overall the individual kernels produced very similar results for each model, as reported in Smith et al. (2018).

3.3 Calculation of ERF using aerosol-free radiative fluxes

To understand the contributions of various processes to the overall ERF we can attempt to separate the ERF that is due to direct radiative forcing from that due to the effects of clouds. Greenhouse gases and aerosols can alter the thermal structure of the atmosphere and hence cloud thermodynamics (the semi-direct effect, (Ackerman et al., 2000)), and aerosols can act via microphysical effects (e.g. increasing the number of condensation nuclei and decreasing the effective radii of cloud droplets, referred to as the aerosol cloud albedo effect and the cloud lifetime effect (Twomey, 1974;Albrecht, 1989;Pincus and Baker, 1994). Following the method of Ghan (2013) the contribution of the aerosol-radiation interactions to the ERF can be distinguished from that of the aerosol-cloud interactions by using a 'double-call' method. This means that the model radiative flux diagnostics are calculated a second time but ignoring the scattering and absorption by the aerosol – referred to in the equations below with the subscript 'af'. The other effects of the aerosol on the atmosphere (i.e. cloud changes, stability changes, dynamics changes) will still be present, however. The IRFari as defined here is the direct radiative forcing from the aerosol, due to scattering and absorption of radiation. The cloud radiative forcing (ERFaci) due to the aerosol-cloud interactions is then obtained by using the difference between the aerosol-free all-sky fluxes and the aerosol-free clear-sky fluxes, which isolates the cloud effects (see eqns 4-6, where equation 6 is included for completeness). The ERFaci may include non-cloud rapid adjustments in cloudy regions of the atmosphere. The final term is the ERF as calculated from fluxes with neither clouds nor aerosols (ERFcs,af).

The ERFs are calculated in the same way as for the all-sky ERF described in Section 3.1, except that the all-sky radiative flux diagnostics are replaced by the relevant aerosol-free fluxes for both the clear-sky and all-sky cases.

IRFari = (ERF – ERFaf) (4)

ERFaci = ERFaf – ERFcs,af (5)

ERFcs,af = ERFcs,af

(6)

Separating the IRF in equation 1 into aerosols and greenhouse gas contributions, IRF= IRF_{aer}+IRF_{GHG}, we can re-write equations 4-6.

IRFari = IRF_{aer} (7)

ERFaci = $A_C + \sum_{x=[T,ts,q,a]}(A_x - A_x^{clr}) + (IRF_{GHG} - IRF_{GHG}^{clr})$ (8)

ERFcs,af = $\sum_{x=[T,ts,q,a]} A_x^{clr} + IRF_{GHG}^{clr}$ (9)

So ERFaci is equivalent to A_C in equation 3 with extra terms to account for the all-sky - clear-sky difference in the non-cloud adjustments and all-sky - clear-sky difference in any greenhouse gas IRF. With no greenhouse gas changes ERFcs,af is the total clear-sky non-cloud adjustment. Ghan (2013) attributes this mostly to the surface albedo change A_α^{clr}, however the kernel analysis shows other non-cloud adjustments are larger (table S4). For greenhouse gases ERFcs,af is the total clear-sky ERF. Assuming the non-cloud adjustments are small apart from T_{strat} (table S4), ERFcs,af is approximately $SARF_{GHG}^{clr}$. The $SARF_{GHG}^{clr}$ is expected to be an overestimate of $SARF_{GHG}$ by 10-40% due to cloud masking (Myhre and Stordal 1997). Thus for greenhouse gases the ERFaci will be a combination of the cloud adjustment and cloud-masking.

4. Results

4.1 Aerosols and precursors

4.1.1 Inter-model Variability

The ERFs are calculated as described in Section 3.1, and the summary chart of the ERFs is shown in Fig. 1 for those models with available results – it should be noted that not all models ran all the experiments. The multimodel mean is shown as a separate bar in Fig. 1, with the value given and the standard error indicated with error bars. A table of the individual values for each model and the multimodel mean are included Table S2 in the supplementary materials.

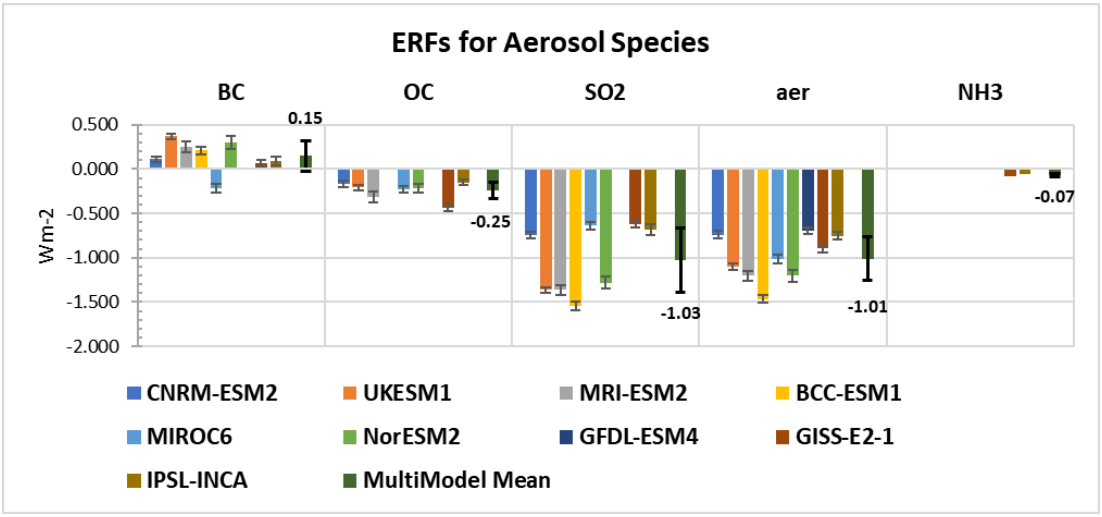

Fig. 1 Aerosol ERFs for the models with the available diagnostics for the aerosol species experiments, with interannual variability represented by error bars showing the standard error. The multimodel mean is shown with the mean value and error bars indicating the standard deviation.

For the piClim-BC results, the range of values is from -0.21 Wm^{-2} to 0.37 Wm^{-2}, while the MIROC6 model has a negative ERF for BC, contrasting with the positive values from the other models - see further discussion on this in Section 4.1.2.

The experiments for the OC (organic carbon) have a range from -0.44 Wm^{-2} to -0.15 Wm^{-2}, and the variability between the models is much less than for the other experiments. The calculated ERFs for the SO2 experiment show a variation from -1.54 Wm^{-2} to -0.62 Wm^{-2}, with CNRM-ESM2-1, MIROC6, IPSL-INCA and GISS-E2-1 at the lower end of the range. These models show a smaller rapid adjustment to clouds which would account for this (see fig S1); also note that CNRM-ESM2-1 does not include aerosol effects apart from the cloud-albedo effect. The two models with results for the NH_3 (GISS-E2-1 and IPSL-INCA) experiment have ERFs of -0.08 and -0.06 Wm^{-2} respectively.

The piClim-aer experiment which uses the 2014 values of aerosol precursors and PI (pre-industrial) values for CH_4, N_2O and ozone precursors shows a range from -1.47 Wm^{-2} to -0.7 Wm^{-2} among the models, making it difficult to narrow the range of uncertainty of aerosols from global models. However, the range in the CMIP6 models is consistent with that reported in Bellouin et al. (2019), who suggest a probable range of -1.60 to -0.65 Wm^{-2} for the total aerosol ERF, and compares well with the range of -1.37 to -0.63 Wm^{-2} for the set of piClim-aer experiments considered in (Smith et al., 2020a) as part of the RFMIP project. In general, the sum of the ERFs from the individual BC, OC and SO_2 experiments does not equal the piClim-aer experiment, due to non-linearity in the aerosol-cloud interactions, particularly since the aerosol perturbation is added to the relatively pristine pre-industrial atmosphere.

The issue of the effect of perturbing the pre-industrial atmosphere with the aerosol changes is examined in more detail in the Supplementary material (see section S6) for NorESM2, where a sensitivity analysis was carried out. This analysis does not repeat the AerChemMIP experiments with the perturbation in a present-day atmosphere but examines the effect of adding the SO_2 and combined aerosol perturbation to an already polluted present day atmosphere. In this simplified sensitivity study the differences are 13% for the SO_2 experiment, and 20% for the combined aerosol experiment. However, it should be borne in mind that this is for a specific model, and the perturbed experiment still has the 1850 climate conditions.

The ERF_ts is a simplified method for corrections of land surface warming in fixed sea surface temperature simulations which in addition to land surface changes leads to changes in land surface albedo changes, tropospheric temperature, water vapor and cloud changes (Smith et al., 2020a;Tang et al., 2019).
The ERF_ts for the models where the land surface temperature adjustment is removed are also included in Supplemental Tables S2 and S3, for comparison with the standard ERF. In general, the difference between the two values is small, of the order of 5 -10%.

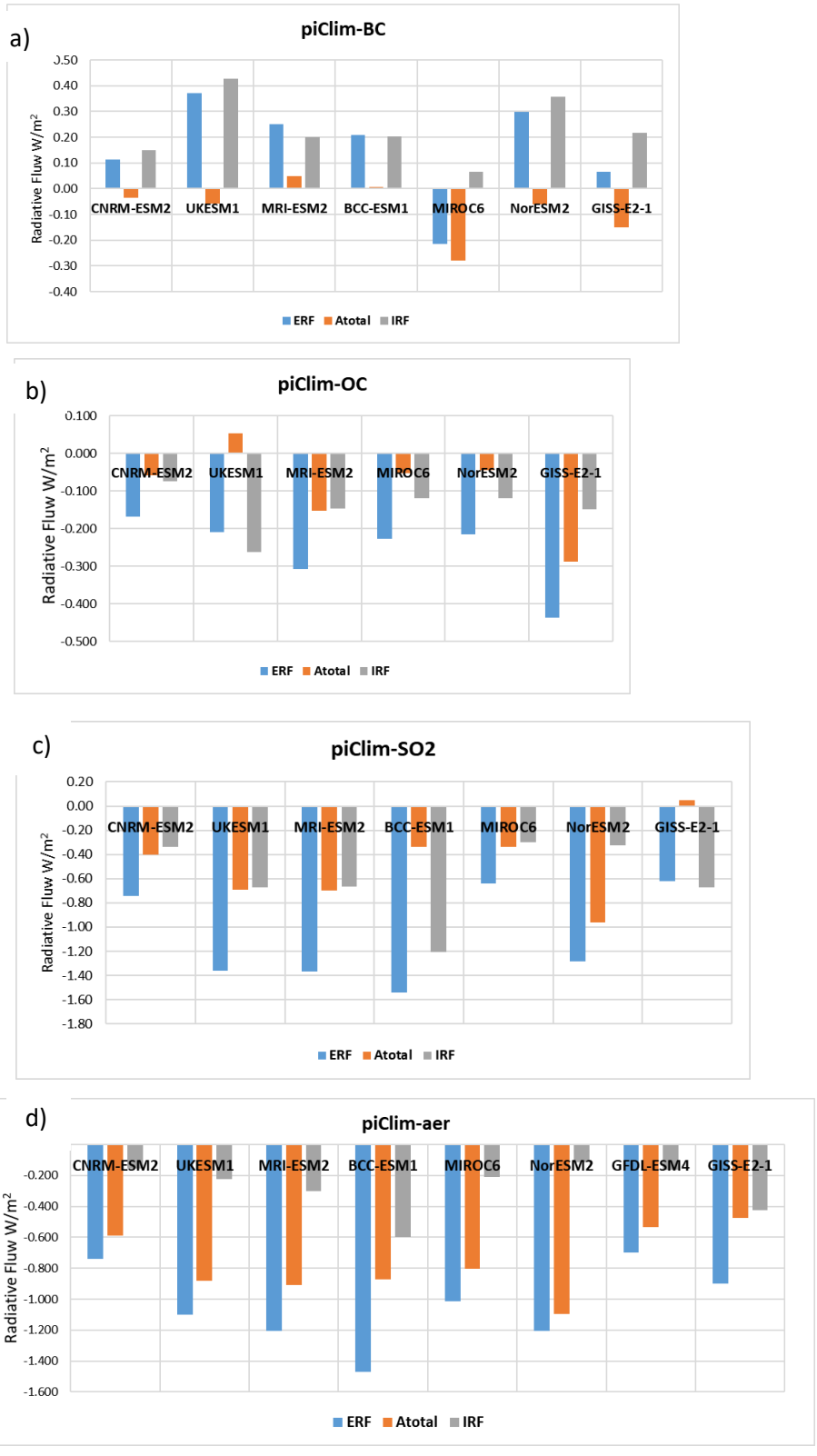

Figure 2 Breakdown of the ERFs into the atmospheric rapid adjustments (Atotal) and IRF (instantaneous radiative forcing) for the aerosols. (a) piClim-BC experiment, (b) piClim-SO2 experiment, (c) piClim-OC experiment, (d) piClim-aer experiment

629 **4.1.2 Breakdown of the ERF into atmospheric adjustments and IRF**

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The results in Fig. 2 show the ERF as calculated from the radiative fluxes in the fixed SST experiments (Section 3.1), the total of the atmospheric adjustments, A_{total}, described in Section 3.2 (where $A_{total} = A_T + A_{ts} + A_q + A_a + A_c$, c.f. eqn. 1), and the instantaneous radiative forcing (IRF).

The sum of the IRF and the atmospheric adjustments should equal the overall ERF, however as the calculation of the IRF depends upon an empirical factor for cloud masking to find the all-sky IRF from the clear-sky IRF (see Section 3.2) the sum of the IRF and the A_{total} will not necessarily equal the ERF as calculated directly from the model radiative flux diagnostics. However, in general the difference is less than 3%, suggesting that the approximation used in the calculation of the IRF is reasonable. Using the kernel method described above it is important to note that the IRF calculated here accounts for the presence of the clouds but does not include cloud changes such as the cloud albedo effect.

The models show a variability in the IRF for SO_2, (Fig. 2c) with a range of -0.3 Wm^{-2} to -1.2 Wm^{-2} with the BCC-ESM1 model being the outlier, having the largest overall ERF. The OC experiments (Fig. 2b) range from -0.08 Wm^{-2} to -0.26 Wm^{-2}, with a range for BC of 0.07 Wm^{-2} to 0.43 Wm^{-2} (Fig. 2a). In MIROC6 the treatment of BC (Takemura & Suzuki 2019; Suzuki & Takemura 2019) leads to faster wet removal of BC and hence a lower IRF. For the combined aerosols (Fig 2d) the range is from -0.1 Wm^{-2} to -0.6 Wm^{-2}.

There are significant differences between the models in the A_{total} for SO_2; these range from 0.05 Wm^{-2} to -1.0 Wm^{-2}, where the differences are dominated by the cloud adjustments which here include the cloud albedo effect as part of the adjustment (see Fig S3 for breakdowns of the atmospheric adjustments for all models). The adjustments to BC are vary in sign and magnitude, with the MRI-ESM2 and BCC-ESM1 models having a slight positive adjustment. The overall model mean has a weaker negative adjustment to that reported by (Stjern et al., 2017;Samset et al., 2016;Smith et al., 2018). The MIROC6 model has a large negative adjustment which is large

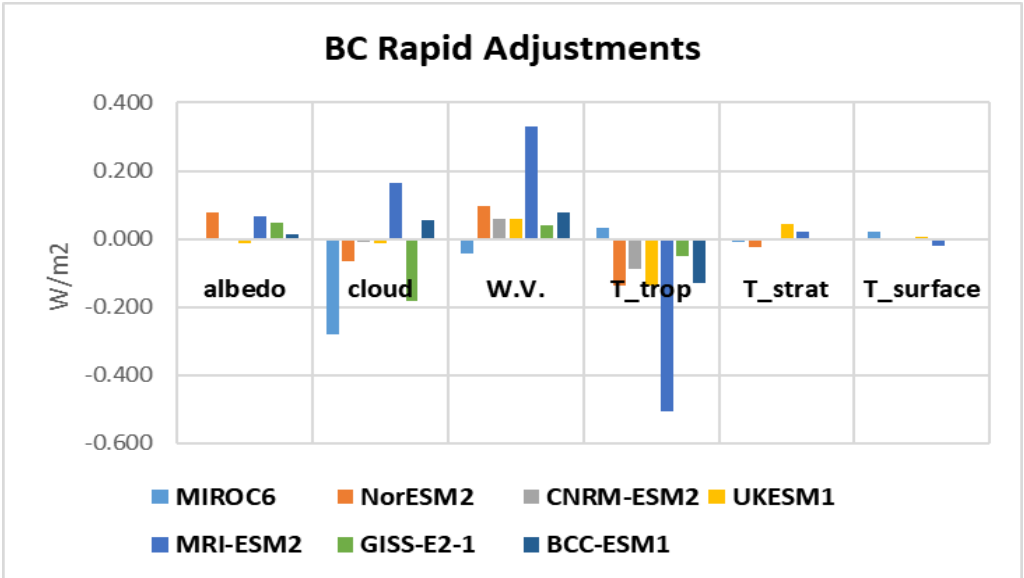

Figure 3 Breakdown of the atmospheric adjustments (albedo, cloud, water vapour, troposphere temperature, stratosphere temperature and surface temperature) for the piClim-BC experiments, showing the variability between models.

enough to lead to an overall negative ERF. We explore the contribution of the individual adjustments to BC in more detail in Fig. 3.

Examining the breakdown of the rapid adjustments for the piClim-BC experiments (Fig. 3) we see considerable variability in the relative importance of the rapid adjustments; the cloud adjustment dominates in MIROC6, consistent with the increase in low clouds reported for this model, and the treatment of BC as ice nuclei causes the large negative cloud adjustment here (Takemura and Suzuki, 2019;Suzuki and Takemura, 2019). The GISS-E2-1 model also has a strong cloud rapid adjustment, but the larger positive value of the IRF leads to an overall positive ERF for this model. With the exception of MIROC6 the negative tropospheric temperature adjustment is balanced by the water vapour (specific humidity) adjustment, although the magnitude of these adjustments for MRI-ESM2 is at least twice that for the other two models. The interaction of BC with clouds in the MRI-ESM2 model is discussed in detail in Oshima et al. (2020), in particular the impact of BC on ice nucleation in high clouds. The larger surface albedo adjustment for both NorESM2 and MRI-ESM2 is most likely due to the representation of deposition of BC on snow and ice in these models (Oshima et al., 2020).

The piClim-aer experiments (Fig. 1d) show all models have a negative A_{total}, covering a range from -0.47 to -1.1 Wm^{-2}. Overall, the cloud rapid adjustments dominate for the piClim-aer experiments, with a contribution ranging from -0.45 to -1.1 Wm^{-2} (See fig S1). Smith et al. (2020) also recently diagnosed forcing and adjustments in a similar subset of CMIP6 models for the piClim-aer experiment as part of the Radiative Forcing Model Intercomparison Project (RFMIP) efforts. While they also diagnosed IRF as a residual calculation between ERF and the sum of rapid adjustments, they estimated cloud adjustments using a modified version of the APRP method instead of radiative kernels. In their approach, the cloud albedo effect (i.e. Twomey Effect) is considered part of the IRF, whereas in the traditional kernel decomposition, it is considered a cloud adjustment. Table S5 compares the two sets of estimates, highlighting the IRF and total cloud adjustment exhibit a near equal absolute difference between the two studies and the sum of IRF and total cloud adjustment are in close agreement (Mean % difference ~ 1.0% for this subset of models). This indicates the classification of the first indirect effect is the only noticeable difference between the two approaches.

The breakdown of the rapid adjustments for all the models are included in supplementary Figure S1, showing the contributions from each type of rapid adjustment for all the experiments for which we have the relevant diagnostics.

4.1.3 Radiation and Cloud interactions

The second method of breaking down the ERF to constituents is described in Section 3.3, (the Ghan method), the results from which are shown in Table 3. The detailed ERF results for MRI-ESM2 are summarized in Oshima et al. (2020), and for UKESM1 in O'Connor et al. (2020a) . Only four of the models under consideration have so far produced the necessary diagnostics for this calculation, and the results are presented in Table 3. For the experiments on aerosols (aer, BC, SO_2, OC) the ERFcs,af (non-cloud adjustments) contribution is small, and the ERF is largely a combination of the direct radiative effect IRFari, and the cloud radiative effect, ERFaci. The IRFari is the direct effect of the aerosol due to scattering and absorption, while the ERFaci is the contribution from the aerosol-cloud interactions and is approximately equal to the rapid adjustments due to clouds (*Ac* see Section 3.2).

Table 3 Results for IRFari, ERFaci and ERFcs,af for aerosol experiments from several models

	UKESM1			CNRM-ESM2			NorESM2			MRI-ESM2		
	IRFari	cs,af	ERFaci	IRFari	cs,af	ERFaci	IRFari	cs,af	ERFaci	IRFari	cs,af	ERFaci
aer	-0.15	0.05	-1.00	-0.21	0.08	-0.61	0.03	-0.03	-1.21	-0.32	0.09	-0.98
BC	0.37	0.001	-0.005	0.13	0.01	-0.03	0.35	0.07	-0.12	0.26	0.08	-0.09
OC	-0.15	-0.01	-0.07	-0.07	0.04	-0.14	-0.07	0.02	-0.16	-0.07	-0.05	-0.21
SO2	-0.49	0.03	-0.91	-0.29	0.08	-0.53	-0.19	-0.09	-1.01	-0.48	0.05	-0.93

For the BC experiment the contribution of the aerosol-cloud interaction has a strong contribution to the overall ERF, except in the case of UKESM1 where it is much weaker; this may be due to the strong SW and LW cloud adjustments in this model cancelling out (O'Connor et al., 2020;Johnson et al., 2019). The SO_2 experiment shows a large cloud radiative effect, in fact the ERFaci is mostly double the IRFari in all the models, due to the large effect on clouds of SO_2 and sulfates through the indirect effects. For the OC experiments the ERFaci to IRFari comparison is mixed, with the ERFaci general half or less the IRFari, except in the case of UKESM1, where this ratio is reversed.

The IRFari are compared with the IRF calculated via the kernel analysis (Section 3.2) where the relevant model results are available. These are shown in fig S2(a), the agreement is generally good giving confidence in the kernel analysis. Similarly ERFaci compares well with the cloud adjustment Ac (fig S2(b)).

4.1.4 AOD Forcing Efficiencies

In order to break down the contributions of the constituent aerosol species to the overall aerosol ERF, we use the AOD (aerosol optical depth) as a forcing efficiency metric for each of the species, and use this to assess their contributions to the overall ERF. Not all models had diagnostics available for the AOD for the individual species, so the analysis uses a subset of the models.

By looking at the single species piClim-BC, piClim-OC and piClim-SO2 experiments we can find the change in the AOD for the individual species (e.g. ΔAOD for BC for the piClim-BC experiment), and use this to scale the the piClim-BC ERF by the AOD change. This assumes that the ERF in the single-species experiment is wholly due to the change in that species as indicated by the AOD, an assumption which is explored in the Supplementary material in Section S4. Table 5 shows the AOD forcing efficiency for the piClim-BC, piClim-SO2 and piClim-OC experiments for each of the five models which had the relevant optical depth diagnostics available.

Table 4 Values of ERF, ΔAOD and ERF/AOD, and IRFari/AOD change for aerosol experiments for CNRM-ESM2-, MIROC6, Nor-ESM2, GISS-E2-1 and MRI-ESM2 models.

BC Exp	BC ERF	Change in BC AOD	ERF/AOD
CNRM-ESM2	0.114	0.0015	77.64
MIROC6	-0.214	0.0006	-339.38
NorESM2	0.300	0.0019	159.75
GISS-E2-1	0.065	0.002	31.65
MRI-ESM2	0.251	0.0073	34.22
OC Exp	OC ERF	Change in OA AOD	ERF/AOD
CNRM-ESM2	-0.169	0.0030	-57.20
MIROC6	-0.227	0.0065	-35.05
NorESM2	-0.215	0.0053	-40.57
GISS-E2-1	-0.438	0.0041	-107.16
MRI-ESM2	-0.317	0.0034	-94.39
SO2 Exp	SO2 ERF	Change in SO4 AOD	ERF/AOD
CNRM-ESM2	-0.746	0.0118	-63.22
MIROC6	-0.637	0.0152	-41.91
NorESM2	-1.281	0.0099	-129.24
GISS-E2-1	-0.622	0.0308	-20.22
MRI-ESM2	-1.365	0.0279	-49.08

The MIROC6 model results in a negative scaling for BC due to the negative ERF for this experiment for this model (Takemura & Suzuki 2019; Suzuki & Takemura 2019) (see Section 4.1.1). The change in the BC AOD is similar for CNRM-ESM2-1 and Nor-ESM2, and the scale factors reflect the differences in the ERF. The scaling for the SO4 in the NorESM2 experiment is twice that of the other models, suggesting a larger impact of the SO4 AOD on the ERF in this model. These values differ somewhat from those found in Myhre et al. (2013b) where they examined the radiative forcing normalised to the AOD using models in the AeroCom Phase II experiments. They found values for sulfate ranging from -8 Wm^{-2} to -21 Wm^{-2} per unit AOD, much weaker than those in our results. However, it is important to note that in the Aerocom Phase II experiments the cloud and cloud optical properties are identical between their control and perturbed runs, so no aerosol indirect effects are included, nor

is any rapid adjustments (IRFari in equation 4). For the BC experiment their values range from 84 Wm^{-2} to 216 Wm^{-2} per unit AOD, broadly similar to the results presented here (with the exception of the negative MIROC6 result). Their results for OA (organic aerosols) which include fossil fuel and biofuel emissions have values ranging from -10 Wm^{-2} to -26 Wm^{-2} per unit AOD, weaker than our values for the piClim-OC experiments which range from -35 Wm^{-2} to -107 Wm^{-2} per unit AOD but include the cloud indirect effects here.

The sum of the individual AODs from BC, SO_4, OA, dust and sea salt gives the total aerosol AOD in the piClim-aer experiment, where the various aerosols were combined. We can then use the AOD for each aerosol in the piClim-aer experiment and the forcing efficiency above to find the contribution of the individual aerosol to the overall change in ERF, providing an approximate estimate of the relative contribution of each aerosol to the overall ERF. In Fig. 4 the relative contributions to the ERF from black carbon (BC), organic aerosols (OA) and sulfate (SO_4) are shown for three of the models. The sum of the ERFs from the individual species is also compared to the ERF calculated from the piClim-aer experiment (NB the sea salt and dust contributions to the ERF are less

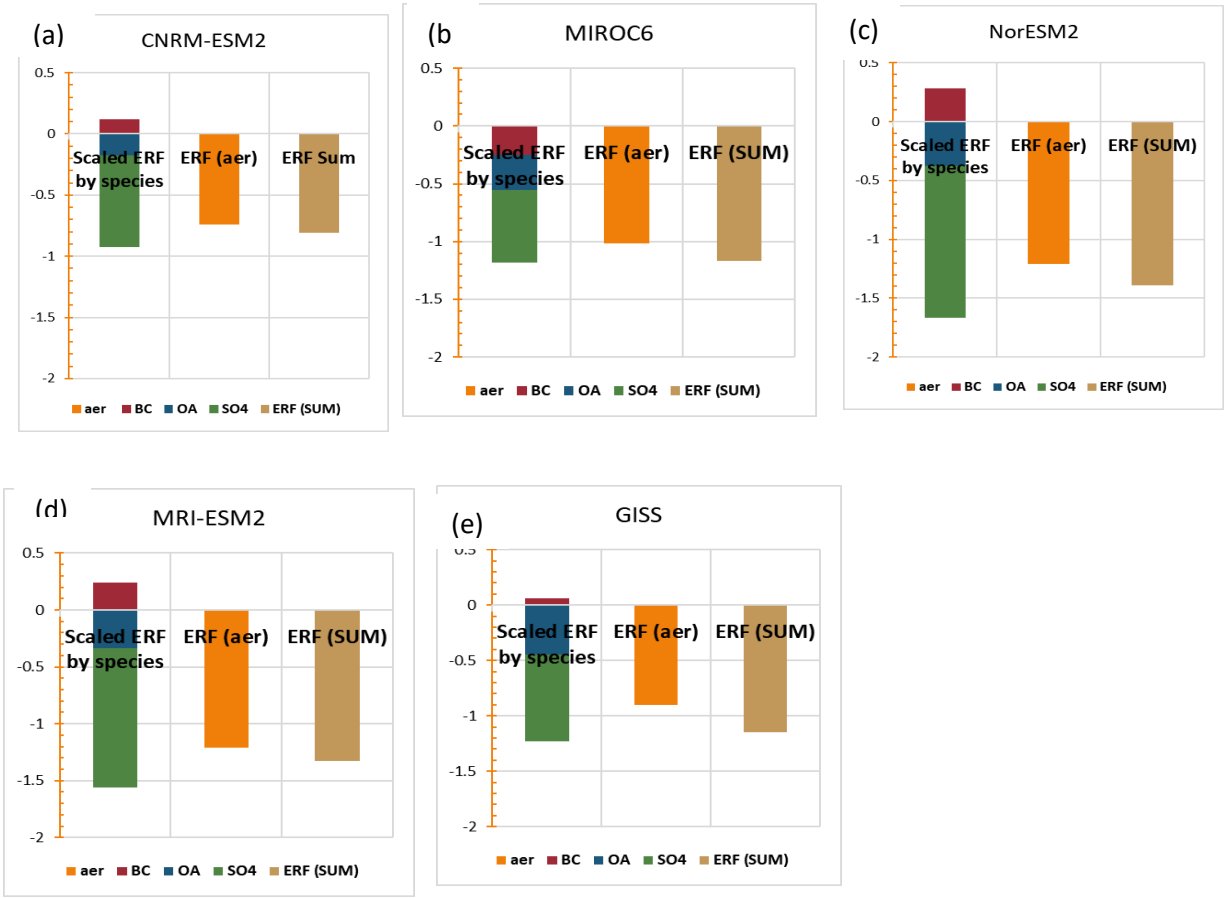

Fig. ~~5~~ 4 The contributions to the ERF for piClim-aer from the individual species, the sum of the scaled ERFs and the ERF calculated directly from the piClim-aer experiment for five of the models.

than 1%, and not shown in this figure for clarity - the ERF/AOD forcing efficiency for these is presented in (Thornhill et al., 2020). There is considerable variation in the ERF for the piClim-aer experiments between models (see Section 4.1), but from this analysis the SO_4 is the largest contributor in all cases, although in the case of the MIROC6 model its relative importance is reduced. The positive ERF contribution from the BC tends to partly

offset the negative ERF from the OA and SO_4, except in the MIROC6 model, where the BC has a negative contribution to the ERF.

The difference between the calculated ERF from the sum of the scaled ERFs is a result of the non-linearity of the aerosol-cloud interactions, a factor which is increased because the aerosols are added to the pre-industrial atmosphere. However, using the IRFari instead of the total ERF to calculate the forcing efficiency and using the same method also results in a difference between the total IRFari derived from the scaled individual experiments and the IRFari for the combined aerosol experiment, suggesting that the difference is not simply a result of the aerosol-cloud interactions.

Using the burden as a scaling factor following the same analysis as described for the AOD results in a largely similar result for the scaling factor, although interestingly the burden scaling for SO2 in the Nor-ESM2 model is similar to the other models (see Table S6 for the burden forcing efficiency).

4.2 Reactive greenhouse gases

The different Earth system models include different degrees of complexity in their chemistry, so their responses to changes in reactive gas concentrations or emissions differ. NorESM2 has no atmospheric chemistry, so there is no change to ozone (tropospheric or stratosphere) or to aerosol oxidation following changes in methane or N_2O concentrations. CNRM-ESM2-1 includes stratospheric ozone chemistry, but no non-methane hydrocarbon chemistry and so ozone is prescribed below 560 hPa. There are no effects of chemistry on aerosol oxidation. BCC-ESM1 includes tropospheric chemistry, but not stratospheric chemistry. Stratospheric concentrations are relaxed towards climatological values. UKESM1, GFDL-ESM4, CESM2-WACCM, GISS-E2 and MRI-ESM2 all include tropospheric and stratospheric ozone chemistry as well as changes to aerosol oxidation rates. The ERFs calculated for the reactive gases for several models are shown in Fig. 5, with the multi-model means given in Supplementary Table S3.

The contributions from gas-phase and aerosol changes to the ERF can be pulled apart to some extent by using the clear-sky and aerosol-free radiation diagnostics (Table 5). The direct aerosol forcing (IRFari) is diagnosed as for the aerosol experiments (section 3.3). The diagnosed changes in aerosol mass are shown in Table S8. GFDL-ESM4 and GISS-ES-1 include nitrate aerosol and show expected responses from NO_X emissions (including O3 experiment). CESM2-WACCM shows an increase in secondary organic aerosol from VOC emissions. Sulphate responses are generally inconsistent across the models. There seems little correlation between aerosol mass changes and diagnosed IRFari.

For gas-phase experiments the diagnosed cloud interactions (ERFaf-ERFcs,af) comprise the ERFaci from effects on aerosol chemistry (as in section 3.3) but also any cloud adjustments and effects of cloud masking on the gas-phase forcing (equation 8). The clear-sky aerosol-free diagnostic (ERFcs,af) is an indication of the greenhouse gas forcing however this will be an over-estimate as it neglects cloud masking effects (section 3.3).

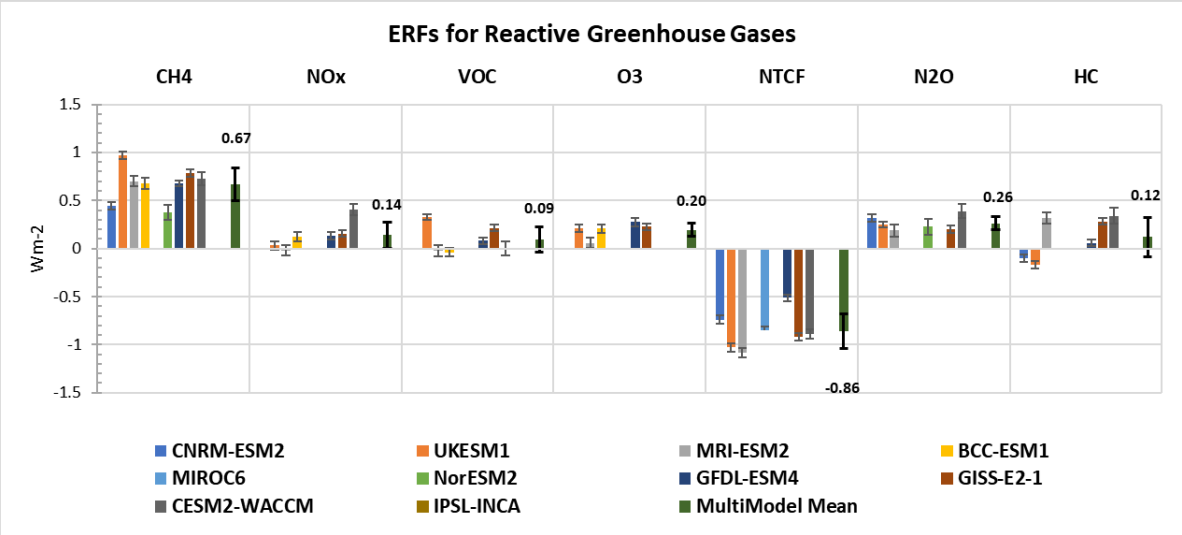

Fig. 5 Reactive gas ERFs for the models with the available diagnostics for the reactive gas experiments with interannual variability represented by error bars showing the standard error. The multimodel mean is shown with the mean value and error bars indicating the standard deviation.

4.2.1 ERF vs SARF

For methane the ERFs are largest for those models that include tropospheric ozone chemistry reflecting the increased forcing from ozone production, see section 4.2.2. The analytic calculation for CH_4-only based on Etminan et al. (2016) gives a SARF of 0.56 Wm^{-2}. The tropospheric adjustments are negative for all models except UKESM1 (Fig 6). The negative cloud adjustment comes from an increase in the LW emissions, possibly due to less high cloud. In UKESM1 (O'Connor et al., 2020b) show that methane decreases sulfate new particle formation, thus reducing cloud albedo and hence a positive cloud adjustment in that model.

For N_2O results are available for models CNRM-ESM2, NorESM2, MRI-ESM2, and GISS-E2 (the analytic N_2O-

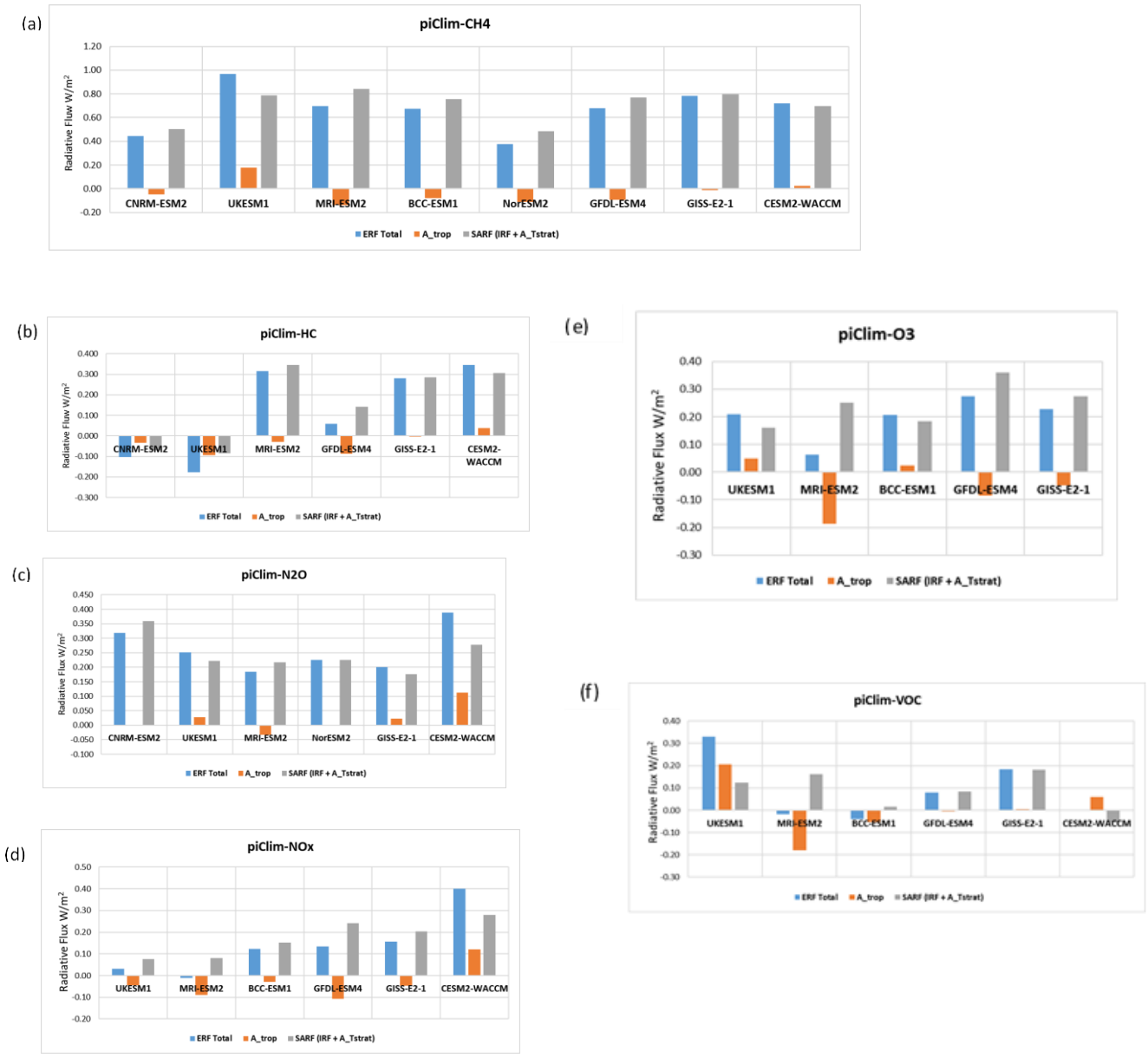

Figure 6 Breakdown of the ERF into SARF and tropospheric rapid adjustments for the chemically reactive species (a) for piClim-CH4 experiments, (b) for piClim-HC experiments, (c) for piClim-N2O experiments, (d) for piClim-NOx experiments, (e) for piClim-O3 experiments, and (f) for piClim-VOC experiments

only calculation gives a SARF of 0.17 Wm^{-2}). There appears little net rapid adjustment to N_2O apart from CESM2-WACCM. Note that due to the method of calculating the all-sky IRF (section 3.2), the IRF and the adjustment terms do not sum to give the ERF.

The models respond very differently to changes in halocarbons. The expected halocarbon-only SARF is +0.30 Wm^{-2} depending on exact speciation used in the model (WMO 2018). For CNRM-ESM2, UKESM1 and GFDL-ESM4 the ERFs are negative or only slightly positive (see also Morgenstern et al. (2020)), whereas for GISS-E2-1

and MRI-ESM2 the ERFs and SARF are both strongly positive. The differences in stratospheric ozone destruction in these models can partially explain the inter-model differences (section 4.2.2).

Table 5 Calculations of IRFari, ERFaci (cloud) and ERFcs,af for the chemically reactive species

	UKESM			GFDL-ESM4			CNRM-ESM2			NorESM2			MRI-ESM2		
	IRFari	cs,af	cloud	IRFari	cs,af	cloud	IRFari	cs,af	cloud	IRFari	cs,af	cloud	IRFari	cs,af	cloud
CH4	-0.01	0.86	0.12	-0.01	0.91	-0.22	0.00	0.56	-0.12	-0.01	0.48	-0.10	0.00	0.91	-0.21
HC	-0.02	0.02	-0.18	-0.02	0.22	-0.14	-0.01	-0.02	-0.08				-0.02	0.50	-0.17
N2O	-0.01	0.26	0.01				0.00	0.41	-0.09	-0.01	0.24	-0.00	-0.00	0.23	-0.03
O3	-0.02	0.16	0.07	-0.04	0.49	-0.18							-0.00	0.24	-0.18
NOx	-0.03	0.10	-0.05	-0.02	0.25	-0.09							-0.01	0.03	-0.04
VOC	0.00	0.13	0.20	-0.02	0.18	-0.08							0.004	0.17	-0.2

4.2.2 Ozone changes

The ozone radiative forcing is diagnosed using a kernel to scale the 3D ozone changes based on Skeie et al. (2020). This kernel includes stratospheric temperature adjustment, but not tropospheric adjustments so gives a SARF. These are shown in Fig. 7. Corresponding changes in the tropospheric and stratospheric ozone columns are shown in figure S5, Increased CH_4 concentrations give a SARF for ozone produced by methane of 0.14±0.03 W m^{-2}, anthropogenic NOx emissions and VOC (including CO) emissions give SARFs of 0.20±0.07 and 0.11±0.04 Wm^{-2} respectively. The O3 experiment comprised both NOx and VOC emission changes. The SARF in this experiment (0.31±0.05 Wm^{-2}) is close to the sum of the NOx and VOC experiments (0.30±0.05 Wm^{-2} for the same set of models) showing little non-linearity in the chemistry (Stevenson et al., 2013).

There is a larger variation across models in the stratospheric ozone depletion from halocarbons (-0.15±0.10 Wm^{-2}) with UKESM1 having noticeably larger depletion as seen in Keeble et al. (2020) giving a SARF of -0.33 Wm^{-2}. N_2O causes some stratospheric ozone depletion in these models, mainly in the tropical upper stratosphere where depletion causes a positive forcing (Skeie et al., 2020), and increases tropospheric ozone (Fig. S6) giving a small net positive SARF (0.03±0.01 Wm^{-2}).

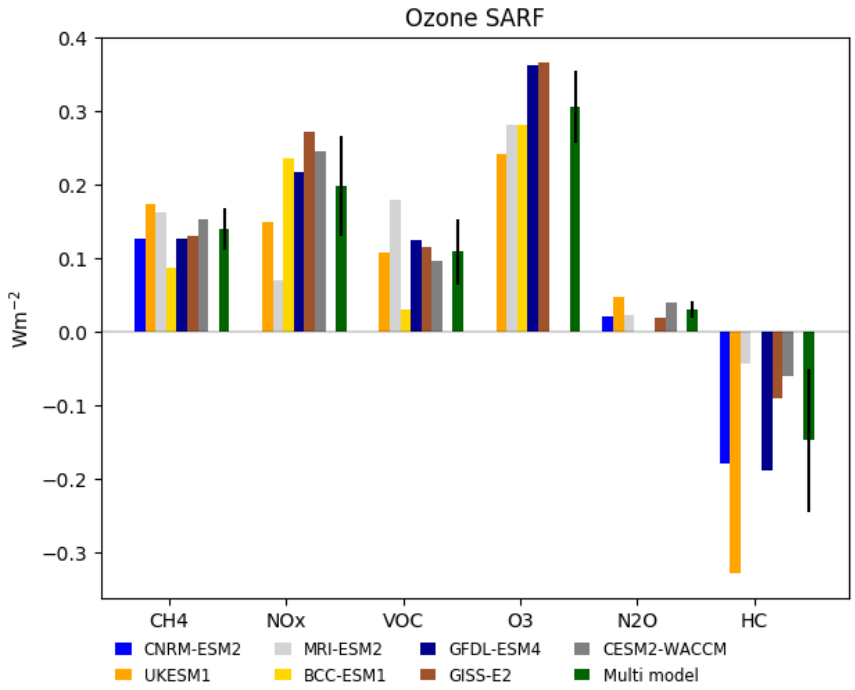

Fig. 7 Changes in ozone stratospheric-temperature adjusted radiative forcing (SARF) for each experiment, diagnosed using kernels (see text). . Uncertainties for the multi model means are standard deviations across models.

Methane oxidation also leads to water vapour production. Figure S6 shows increases in the stratosphere for the piClim-CH4 of up to 20% . The kernel analysis however finds very low radiative forcing associated with this increase (-0.002±0.003 Wm^{-2}).

4.2.3 Comparison with greenhouse gas forcings

The ERFs, ERFcs,af and SARFs diagnosed for the greenhouse gas changes (Fig. 6, Table 5) are compared with the expected greenhouse gas SARFs in Fig. 8. The expected SARFs from the well-mixed gases are given by Etminan et al. (2016) for CH_4 and N_2O, and by WMO (2018) for the halocarbons (the halocarbon changes are slightly different in each model). The expected SARFs from ozone changes are from Fig. 7.

For methane the ERFs are typically higher than the expected GHG SARF (except for CNRM-ESM2).The diagnosed ERFcs,af and SARF agree better with the expected SARF in UKESM1, BCC-ESM1 and CESM2-WACCM, but not in other models. For N_2O the modelled ERF is larger than the expected SARF for CNRM-ESM2-1 and CESM2-WACCM, this is explained by the rapid adjustments for CESM2-WACCM, but not for CNRM-ESM2. For halocarbons the stratospheric ozone depletion offsets the direct SARF and accounts for much of the spread in the model SARF, although the CNRM-ESM2-1 ERF and SARF is lower than expected. The modelled HC ERF for UKESM1 is strongly negative due to increased aerosol cloud interactions, (O'Connor et al., 2020a;Morgenstern et al., 2020) but removing cloud effects using the SARF or ERFcs,af agrees better with the expected value. The estimated ozone SARF from the NO_X, VOC and O3 experiments generally agrees with the model SARF and ERFcs,af. For CESM2-WACCM the ERF from the VOC experiment is zero, and the SARF negative even though the diagnosed ozone SARF is positive. For all experiments and models ERFcs,af is generally higher than the expected or diagnosed SARF (see section 3.3).

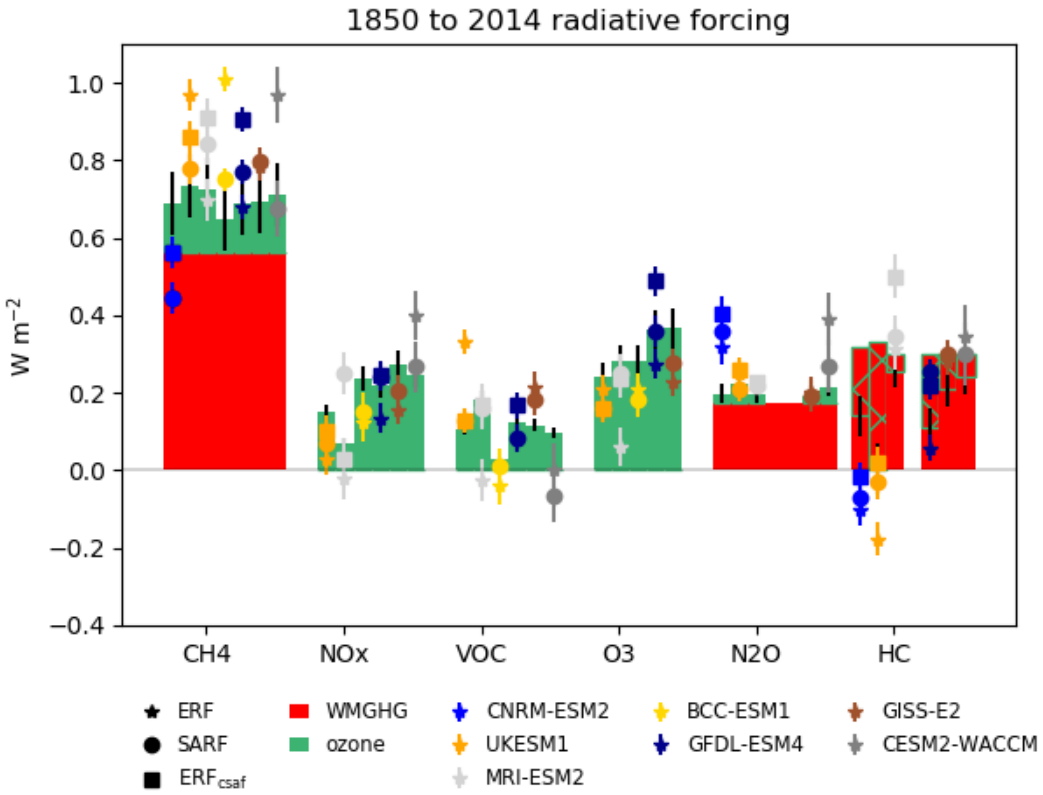

Fig. 8 Estimated SARF from the greenhouse gas changes (WMGHGs and ozone), using radiative efficiencies for the WMGHGs and kernel calculations for ozone (see text). Hatched bars show decreases in ozone SARF. Symbols show the modelled ERF, SARF and ERFcs,af (estimate of greenhouse gas clear-sky ERF). Uncertainties on the bars are due to uncertainties in radiative efficiencies. Uncertainties on the symbols are errors in the mean due to interannual variability in the model diagnostic.

4.2.4 Methane Lifetime

In the CMIP6 setup the modelled methane concentrations do not respond to changes in oxidation rates. The methane lifetime is diagnosed (which includes stratospheric loss to OH as parameterised within each model) and assuming losses to chlorine oxidation and soil uptake of 11 and 30 Tg yr^{-1} ((Saunois et al., 2020)(Myhre et al., 2013b) and this can be used to infer the methane changes that would be expected if methane were allowed to vary. Fig. 9 shows the methane lifetime response is large and negative for NO_x emissions, with a smaller positive change for VOC emissions. Halocarbon concentration increases decrease the methane lifetime, as ozone depletions leads to increased UV in the troposphere and increased methane loss to chlorine in the stratosphere (Stevenson et al., 2020). N2O also decreases the methane lifetime by depleting ozone in the tropics although the effect is less than for halocarbons. The O3 experiment has a significantly more negative effect (-27±9 %) than the sum of NOx and VOC (-16±8 %) (uncertainties are multi-model standard deviation). This suggests significant non-additivity. Note that a combined CH_4+NOx+VOC experiment is not available to test the additivity further.

The lifetime response to changing methane concentrations can be used to diagnose the methane lifetime feedback factor f ((Fiore et al., 2009). The results here give f=1.32, 1.31, 1.43, 1.30, 1.26, 1.19 (mean 1.30±0.07) for UKESM1, MRI-ESM2, BCC-ESM1, GFDL-ESM4, GISS-E2-1 and CESM-WACCM. This is in very good agreement with AR5, although their values are starting from a year 2000 baseline rather than pre-industrial.

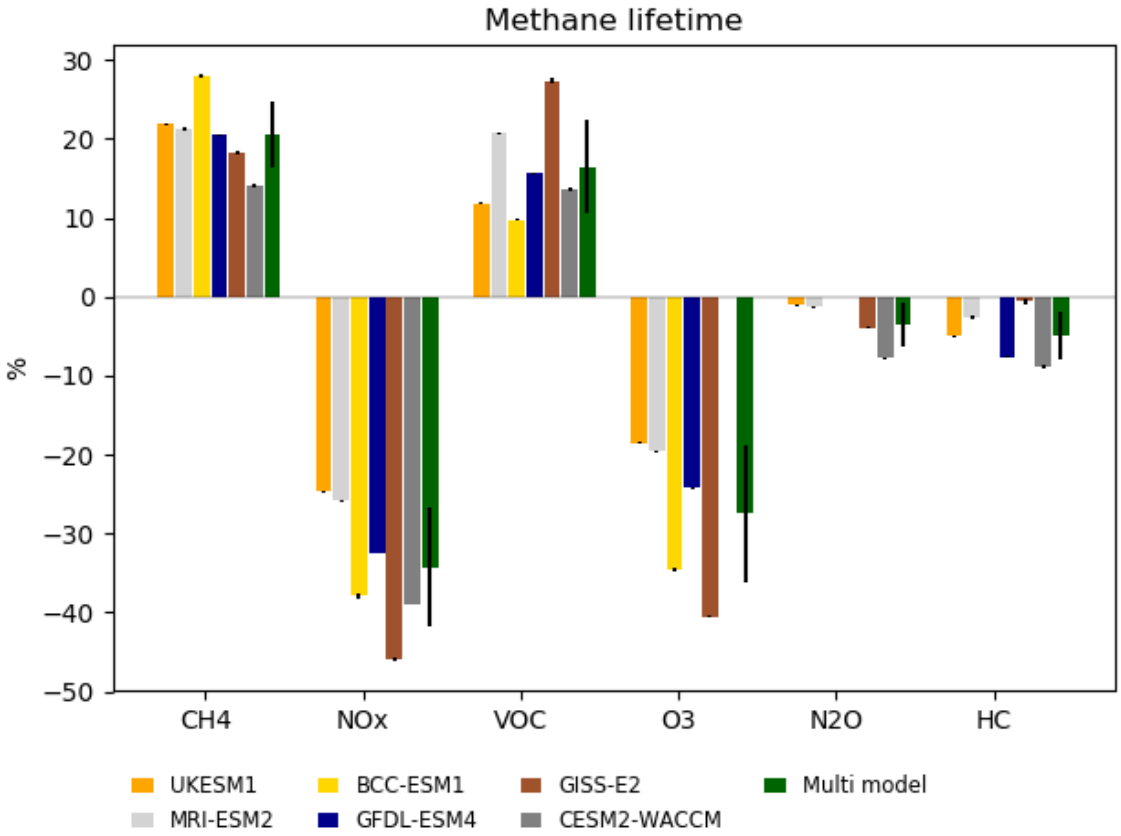

Fig. 9 Changes in methane lifetime (%), for each experiment. Uncertainties for individual models are errors on the mean from interannual variability. Uncertainties for the multi model mean are standard deviations across models.

4.2.5 Total ERFs

The methane lifetime changes can be converted to expected changes in concentration if methane were allowed to freely evolve following Fiore et al. (2009), using the *f*-factors appropriate to each model (section 3.3.4). The inferred radiative forcing is based on radiative efficiency of methane (Etminan et al., 2016). The methane changes also have implications for ozone production, so we assume an ozone SARF per ppb of CH_4 diagnosed for each model from section 4.2.

The breakdown of the information from the analyses above is shown in Fig. 10, using the SARF calculated for the gases (WMGHGs and ozone) and kernel-diagnosed cloud adjustments (which include aerosol cloud interactions). Direct contributions from the aerosols IRFari are shown for models where this is available. The contributions from methane lifetime changes have also been added to the diagnosed ERF as these aren't accounted for in the models. Differences between the diagnosed ERF (stars) and the sum of the components (crosses) then shows to what extent this decomposition into components can account for the modelled ERF. For many of the species, this breakdown is reasonable, and illustrates that cloud radiative effects can make significant contributions to the total radiative impacts of WMGHGs and ozone precursors. This analysis cannot distinguish between cloud effects due to changes in atmospheric temperature profiles or those due to increased cloud nucleation from aerosols.

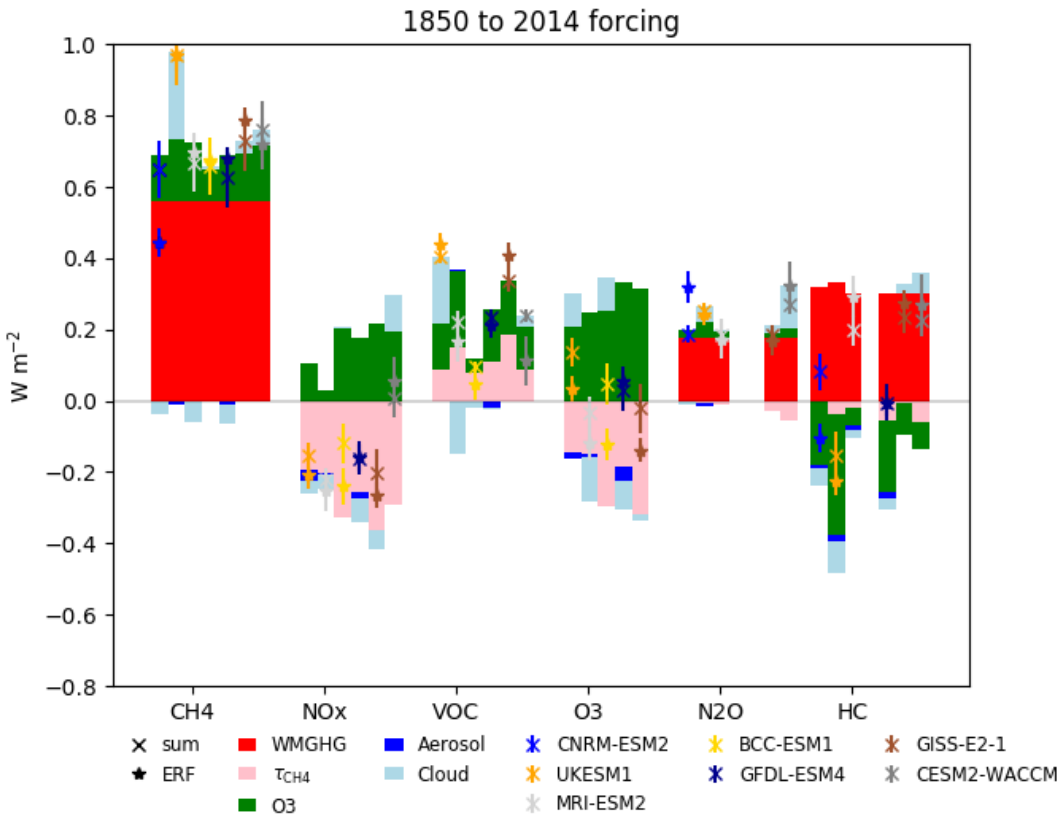

Fig. 10 SARF for WMGHGs, ozone and diagnosed changes in methane. Model diagnosed direct aerosol RF and cloud radiative effect. Crosses mark the sum of the five terms for each model. Stars mark the diagnosed ERF with the effect of methane lifetime (on methane and ozone) added. Differences between stars and crosses shows undiagnosed contributions. Uncertainties on the sum are mainly due to the uncertainties in the radiative efficiencies. Uncertainties in the ERF are errors on the mean due to interannual variability. Note for CESM2-WACCM, BCC-ESM1, GISS-E2-1 the direct aerosol effect is unavailable.

5. Discussion

For all of the species shown we see considerable variation in the calculated ERFs across the models, which is due in part to differences in the model aerosol and chemistry schemes; not all models have interactive schemes for all of the species, and whether or not chemistry is considered will impact the evolution of some of the aerosol species. We can use the differences in model complexity from the multi-model approach together with the separation of the effects of the various species in the individual AerChemMIP experiments to understand how the various components contribute to the overall ERFs we have calculated.

5.1 Aerosols

The 1850-2014 multi-model mean and standard deviation of the ERFs for SO_2, OC and BC are: -1.03 +/- 0.37 Wm^{-2} for SO_2, -0.25 +/- 0.09 Wm^{-2} for OC, and 0.15 +/- 0.17 Wm^{-2} for BC. The total ERF for the aerosols is -1.01 +/- 0.25 Wm^{-2}, within the range of -1.65 to -0.6 Wm^{-2} reported by (Bellouin et al., 2019).

The radiative kernels and double-call diagnostics are used to separate the direct and cloud effects of aerosols for those models where all the relevant diagnostics are available. These two methods broadly agree on the cloud

contribution for the BC, SO_2 and OC experiments. We generally find a weaker total adjustment to black carbon compared to other studies (Samset and Myhre, 2015;Stjern et al., 2017;Smith et al., 2018). The exceptions are MIROC6 and GISS-E2-1. These previous studies used much larger changes in black carbon (up to 10 times) which may cause non-linear effects such as self-lofting.

As the ISCCP cloud diagnostics become available for more of the CMIP6 models, it will be possible to do a direct calculation of the cloud rapid adjustments using the kernels from (Zelinka et al., 2014) and compare those with the adjustments calculated using the kernel difference method described in (Smith et al., 2018) and used here (Section 3.2; see also figure 4 and figure S2 from Smith et al. (2020a)).

The radiative efficiencies per AOD calculated here are generally larger than those from the AeroCom Phase II experiments (Myhre et al., 2013b), with the caveat that the models included here did not have fixed clouds, so that indirect effects would be included.

The values diagnosed for the IRFari (for the models we have available diagnostics for) in CMIP6 are similar to those from CMIP5 (Myhre et al., 2013a) where they reported values for sulfate of -0.4 (-0.6 to –0.2) Wm^{-2} compared to our -0.36 (-0.19 to -0.49) Wm^{-2} for the SO_2 experiment, for OC they found -0.09 (-0.16 to –0.03) Wm^{-2} compared to our value of -0.09 (-0.07 to -0.15) Wm^{-2} and for BC they had +0.4 (+0.05 to +0.80) compared to our value of 0.28 (0.13- 0.37) Wm^{-2}, so broadly the IRFari for the individual species agree with those found in the previous set of models used in CMIP5.

The overall aerosol ERF from AR5 is reported as in the range -1.5 to 0.4 Wm^{-2}, compared to ERF values reported here for the piClim-aer experiment in the range -0.7 to -1.47 Wm^{-2}.

5.2 Reactive greenhouse gases

The diagnosed ERFs from methane, N_2O, halocarbons and ozone precursors are: 0.75±0.10, 0.26±0.07, 0.12±0.21 and 0.20±0.07 W m^{-2} (excluding CNRM-ESM2-1 for methane as it cannot represent the lower tropospheric ozone changes, and excluding NorESM2 for all as it has no ozone chemistry). These compare with 0.79±0.13, 0.17±0.03, 0.18±0.15 and 0.22±0.14 W m^{-2} for 1750-2011 from AR5 (Myhre et al., 2013a) - where the effects on methane lifetime and CO_2 have been removed from the AR5 calculations, and the halocarbons are for CFCs and HCFCs only. Section 4.2.5 shows that cloud effects can make a significant contribution to the overall ERF even for WMGHGs. However, clouds cannot explain all the differences. The ERF for N_2O is larger than estimated in AR5. The ozone contribution here is estimated as 0.03±0.01 Wm^{-2} whereas it was zero in AR5, but that does not explain all the difference. The multi-model ERF for halocarbons is smaller than AR5, due to larger ozone depletion although the models have a wide spread with some showing significantly lower ERFs and some significantly higher due to varying strengths of ozone depletion in these models.

The estimated ozone SARFs from the changes in levels of methane, NOx and VOC from 1850 to 2014 are 0.14±0.03, 0.20±0.07, and 0.11±0.04 W m^{-2} compared to 0.24±0.13, 0.14±0.09, and 0.11±0.05 W m^{-2} in CMIP5 (Myhre et al., 2013a). The ozone from methane contribution is smaller, here only 25% of the direct Etminan et al. (2016) methane SARF compared to 50% in AR5 (or 39% using the Etminan et al. (2016) formula). The NOx contribution is larger in this study. The CMIP5 results were based on (Stevenson et al., 2013) in which species were reduced from present day levels rather than being increased from pre-industrial levels. The NOx emission

changes are also larger for CMIP6 compared to CMIP5 (Hoesly et al. 2018). The sum of the ozone terms (CH_4+N_2O+HC+O_3) is 0.33±0.11 Wm^{-2}, agreeing well with the total 1850-2014 ozone SARF of 0.35 ±0.16 Wm^{-2} (1.s.d) from Skeie et al. (2020) which included a few additional models.

The overall effect of NTCF emissions (excluding methane and other WMGHGs) on the 1850-2014 ERF experienced by models that include tropospheric chemistry is strongly negative (-0.89±0.20 W m^{-2}) due to the dominance of the aerosol forcing over that from ozone. There is a large spread in the NTCF forcing due to the different treatment of atmospheric chemistry within these models. Models without tropospheric and/or stratospheric chemistry prescribe varying ozone levels which are not included in the NTCF experiment. Hence the overall forcing experienced by these models due to ozone and aerosols will be different from that diagnosed here.

6. Conclusion

The experimental setup and diagnostics in CMIP6 have allowed us for the first time to calculate the effective radiative forcing (ERF) for present day reactive gas and aerosol concentrations and emissions in a range of Earth system models. Quantifying the forcing in these models is an essential step to understanding their climate responses.

This analysis also allows us to quantify the radiative responses to perturbations in individual species or groups of species. These responses include physical adjustments to the imposed forcing as well as chemical adjustments and adjustments related to the emissions of natural aerosols. The total adjustment is therefore a complex combination of individual process, but the diagnosed ERF implicitly includes these and represents the overall forcing experienced by the models.

We find that the ERF from well-mixed greenhouse gases (methane, nitrous oxide and halocarbons) has significant contributions through their effects on ozone, aerosols and clouds, that vary strongly across Earth system models. This indicates that Earth system processes need to be taken into account when understanding the contribution WMGHGs have made to present climate and when projecting the climate effects of different WMGHG scenarios.

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8. Author Contributions

Manuscript preparation was done by GDT, WJC, RJK, DO and additional contributions from all co-authors. Model simulations were set up, reviewed and/or ran by DO, FMO'C, NLA, MD, LE, LH, J-FL, MMichou, MMills, JM, PN, VN, NO, MS, TT, ST, TW, GZ, JZ. Analysis was carried out by GT, WC, RK, DO.

9. Data Availability

All data from the various 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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Supplementary Material for

Effective radiative forcing from emissions of reactive gases and aerosols – a multi-model comparison

S1 Table of model characteristics

The table of models used in the paper with information on resolution and aerosol and chemistry modules.

Table S 1 Table of model properties, aerosol schemes, and chemistry (SU = sulphate; OA = organic aerosol; BC = black carbon; DU = dust; SS = sea salt; NO3 =nitrate)

Earth System Model (component models)	Resolution	Description of aerosol module	References

	IPSL-CM6A-LR (LMDz, INCA)	1.25°(lat) x 2.5°(lon) 79 vertical levels LMDzORINCA Two	LMDzORINCA Two-moment (mass and number) aerosol scheme with 5 lognormal modes. The IPSLCM6A-LR-INCA model used for this analysis has interactive aerosols but a limited gas-phase model. Aerosol scheme is based a sectional approach with to represent the size distributionof dust, Sea- salt (which has an additional super-coarse mode to model largest emission of spray-salt aerosls), BC, NH4, NO3, SO4, SO2 and OA with a combination of accumulation and coarse log-normal modes with both, soluble and insoluble, treated as independent modes. DMS emissions are prescribed and not interactively calculated. BC is modelled as internally mixed with sulphate (Wang et al., 2016), where the refractive index is relies on Garnet-Maxwell method. Its emissions are derived from inventories. A new dust refractive index is implemented (Di Biagio et al., 2019). Well mixed trace gases concentrations/emissions are forced with AMIP/CMIP6 datasets (Lurton et al., 2020) ozone by (Checa-Garcia et al., 2018) and solar forcing by (Matthes et al., 2017) Components included: SU, BC, OA, SS, DU, NO3	(Balkanski et al., 2010) (Hauglustaine et al., 2014)

UKESM11 (HadGEM3, UKCA, JULES)	1.25°(lat) x 1.88°(lon) 85 vertical levels	UKCA contains the GLOMAP-mode aerosol microphysics scheme Two-moment (mass and number) aerosol scheme with 5 lognormal modes (nucleation soluble, Aitken soluble, Aitken insoluble, accumulation soluble, coarse soluble) Components included: SU, BC, OA, SS, DU* *Dust component tracked independently in six size bins UKCA contains a stratosphere-troposphere chemistry scheme, consisting of 84 tracers, 81 species, 199 bimolecular reactions, 25 uni- or termolecular reactions, 5 heterogeneous, 3 aqueous phase reactions, and 59 photolytic reactions. Secondary aerosol formation of sulphate and secondary organic aerosol is determined by the interactive oxidants. The UKCA aerosol scheme, called GLOMAP-mode, is a two-moment scheme for the simulation of tropospheric black carbon (BC), organic carbon (OC), SO_4, and sea salt. Dust is modelled independently using the bin scheme of Woodward (2001). The UKCA chemistry and aerosol schemes are coupled such that the secondary aerosol (SO_4, OA) formation rates depend on oxidants from the stratosphere-troposphere chemistry scheme. Aerosol particles are activated into cloud droplets using the activation scheme of Abdul-Razzak and Ghan (2000) which is dependent on aerosol size distribution, aerosol composition, and meteorological conditions. Changes in CDNC affect cloud droplet effective radius ((Jones et al., 2001) and the autoconversion of cloud liquid water in to rain water (Khairoutdinov and Kogan, 2000), which both influence cloud albedo. Stratospheric aerosols (aerosol optical depth and surface area density) are prescribed in the model (Sellar et al., 2019b).	(Sellar et al.) (Williams et al., 2018) (Walters et al., 2019) (Kuhlbrodt et al., 2018) GLOMAP-mode by (Mann et al., 2010) (Mulcahy et al., 2018) (Morgenstern et al., 2009) (O'Connor et al., 2014) (Archibald et al., 2020;Sellar et al., 2020;Mulcahy et al., 2020)

CNRM-ESM2-1 (ARPEGEClimatv6.3, ISBA-CTRIP, TACTIC, REPROBUS, PISCES)	1.4°(lat) x 1.4°(lon) 91 vertical levels	Aerosols: TACTIC_v2 tropospheric aerosol bin scheme. 12 bins in total for SU, BC, OA, SS, DU, with 3 bins for SS, and 3 bins for DD. REPROBUS_v2 stratospheric chemistry scheme with 63 variables, 44 transported by the model large-scale transport scheme, and 168 chemical reactions, among which 39 photolysis and 9 heterogeneous reactions	(Séférian et al., 2016;Michou et al., 2015) (Séférian et al.) 2019 Michou et al 2019 Model description website: http://www.umrcnrm.fr/cmip6/spip.php?article10
NorESM2 CAM6-Nor, CLM5)	1.9°(lat) x 2.5°(lon) 32 vertical levels	OsloAero6 Production-tagged aerosol module with background lognormal modes (Aitken, accumulation, coarse). Process tracers can alter the shape and composition of the initially lognormal background modes to generate mixtures. OsloAero6 aerosol module which contains some slight updates since (Kirkevåg et al., 2018) describes the formation and evolution of BC, OC, SO_4, dust, sea-salt and SOA. There is a limited gas-phase chemistry describing the oxidation of the aerosol precursors DMS, SO_2, isoprene, and monoterpenes. Oxidant fields of OH, HO_2, NO_3 and O_3 are prescribed climatological fields. As there is no ozone chemistry in the model, prescribed monthly-varying ozone fields are used for the radiation. Components included: SU, BC, OA, SS, DU	(Kirkevåg et al., 2018)

MRI-ESM2	MRI-AGCM3.5: TL159; 320 x 160 lon/lat, MASINGAR mk-2r4c: TL95; 192 x 96 lon/lat, MRI-CCM2.1: T42; 128 x 64 lon/lat, with 80 vertical levels	MASINGAR mk-2r4c is an aerosol model that is a component of MRI-ESM2.0. MASINGAR mk-2r4c treats atmospheric aerosol physical and chemical processes (e.g., emission, transport, diffusion, chemical reactions, and dry and wet depositions). The size distributions of sea salt and mineral dust are divided into 10 discrete bins and those of other aerosols are represented by lognormal size distributions. Components included: SU, BC, OA, SS, DU	(Yukimoto et al., 2019) (Oshima et al., 2020)
MIROC6	1.4^{o} (lat) x 1.4^{o} (lon)	Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) predicts mass mixing ratios of the main tropospheric aerosols, and models aerosol-cloud interactions in which aerosols alter cloud microphysical properties and affect the radiation budget by acting as cloud condensation and ice nuclei. The SO_4, BC and OC aerosols are treated as externally mixed in this model. The CDNC and ice crystal number are used to calculate the aerosol indirect effect and cloud nucleation process Components included: SU, BC, OA, SS, DU	(Takemura et al., 2005;Watanabe et al., 2010;Takemura and Suzuki, 2019;Takemura, 2018;Tatebe et al., 2019)

BCC-ESM1 (BCC-AGCM3-Chem, BCC-AVIM2, MOM4-L40, SIS)	2.8125º (lat) x 2.8125º (lon) 26 vertical levels with top level at 2.914 hPa	The model prognoses mass distribution of five aerosol types including sulfate, dust, black carbon, organic carbon, and sea salt based on their emissions (and precursor emissions), chemical production for sulfate and secondary organics, dry and wet (rainout and washout) deposition, transport by advection, and dry and wet convection. It uses the BCC-AGCM3-Chem atmospheric chemistry model based on MOZART2 (Horowitz et al., 2003) It uses the BCC-AGCM3-Chem atmospheric chemistry model based on MOZART2 (Horowitz et al., 2003) which does not include stratospheric chemistry, so concentrations of O_3, CH_4, and N_2O at the top two model levels are the zonally and monthly values derived from the CMIP6 data package. Components included: SU, BC, OA, SS, DU Effects of aerosols on radiation, cloud, and precipitation are treated.	(Wu et al., 2020)

GFDL-ESM4	(C96 96x96 cells) 49 vertical levels	The model includes 56 prognostic (transported) tracers and 36 diagnostic (non-transported) chemical tracers, with 43 photolysis reactions, 190 gas-phase kinetic reactions, and 15 heterogeneous reactions. The tropospheric chemistry includes reactions for the NO_x-HO_x-O_x-CO-CH_4 system and oxidation schemes for other non-methane volatile organic compounds. The stratospheric chemistry accounts for the major ozone loss cycles (O_x, HO_x, NO_x, ClO_x, and BrO_x) and heterogeneous reactions on liquid and solid stratospheric aerosols as in Austin et al. (2013). The bulk aerosol scheme, including 18 transported aerosol tracers, is similar to that in AM4.0 (Zhao et al., 2018), with the following updates: (1) ammonium and nitrate aerosols are treated explicitly, with ISORROPIA (Fountoukis and Nenes, 2007) used to simulate the sulfate–nitrate–ammonia thermodynamic equilibrium; (2) oxidation of sulfur dioxide and dimethyl sulfide to produce sulfate aerosol is driven by the gas-phase oxidant concentrations (OH, H_2O_2, and O_3) and cloud pH simulated by the online chemistry scheme, and (3) the rate of aging of black and organic carbon aerosols from hydrophobic to hydrophilic forms varies with calculated concentrations of hydroxyl radical (OH). Aerosol species, including sulfate, BC, organic aerosols, sea-salt, dust and nitrate are treated explicitly.	(Horowitz et al., 2020)(Dunne et al., 2020)
GISS-E2-1 (p3 variant)	2° latitude by 2.5° in longitude 40 vertical layers surface to 0.1 hPa in	Aerosols and ozone are calculated prognostically using the One-Moment Aerosol (OMA). Aerosol scheme is coupled to the tropospheric chemistry scheme which includes inorganic chemistry of O_x, NO_x, HO_x, CO, and organic chemistry of CH_4 and higher hydrocarbons using the CBM4 scheme and the stratospheric chemistry scheme which includes chlorine and bromine chemistry together with polar stratospheric clouds.	(Bauer et al., 2020;Shindell et al., 2001;Shindell et al., 2003;Gery et al., 1989;Shindell et al., 2006)

CESM2-WACCM	0.9 (lat) x 1.25 (lon), 70 levels	Chemistry and aerosols for the troposphere, stratosphere, mesosphere and lower thermosphere are calculated interactively. It simulates 228 compounds, including the 4-mode Modal Aerosol Model (MAM4). This version of MAM4 is modified to allow for the simulation of stratospheric aerosols from volcanic eruptions (from their SO_2 emissions) and oxidation of OCS. The representation of secondary organic aerosols follows the Volatility Basis Set approach.	(Emmons et al., 2020;Danabasoghu, 2019;Danabasoglu, 2019;Gettelman et al., 2019;Tilmes et al., 2019) Mills et al., 2016)

(Table for European models updated from:

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S2 Tables of ERF and ERF_ts for all models analysed

By removing the adjustment due to the changes in the land surface temperature (as calculated from radiative kernels) we show the ERF_Ts for those models where the adjustment was available in the following table.

The tables below give the 1850-2014 ERF and the ERF_Ts calculated from the TOA flux differences for each model for each experiment.

Table S 2 ERFs and ERF_ts for the aerosols, including multi-model means with standard errors.

ERF	aer		BC		OC		SO2		NH3	
Wm^{-2}	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts
CNRM-ESM2	-0.74	-0.79	0.11	0.11	-0.17	-0.18	-0.75	-0.78	-	-
UKESM1	-1.10	-1.15	0.37	0.36	-0.21	-0.23	-1.36	-1.41	-	-
MRI-ESM2	-1.21	-1.24	0.25	0.27	-0.32	-0.32	-1.37	-1.42	-	-
BCC-ESM1	-1.47	-1.54	0.21	0.21	-	-	-1.54	-1.62	-	-

MIROC6	-1.01	-1.07	-0.21	-0.24	-0.23	-0.26	-0.64	-0.67		
NorESM2	-1.21	-1.21	0.30	0.30	-0.22	-0.23	-1.28	-1.29		
GFDL-ESM4	-0.70	-0.73								
GISS-E2-1	-0.90	-0.95	0.06	0.06	-0.44	-0.45	-0.62	-0.65	-0.08	
IPSL-INCA	-0.75		0.10		-0.15		-0.69		-0.06	
MultiModel Mean	-1.01	-1.09	0.15	0.16	-0.25	-0.28	-1.03	-1.12	-0.07	
S.D.	0.25	0.25	0.17	0.19	0.09	0.09	0.37	0.38	0.01	

Table S 3 ERF, ERF_ts, multimodel means and standard error for the chemically reactive gases.

ERF	CH4		HC		N2O		NTCF		O3		NOx		VOC	
Wm^{-2}	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts	ERF	ERF_ts
CNRM-ESM2	0.44	0.46	-0.10	-0.10	0.32	0.33	-0.74	-0.79						
UKESM1	0.97	1.00	-0.18	-0.19	0.25		-1.03	-1.03	0.21	0.22	0.03	0.04	0.33	0.34
MRI-ESM2	0.70	0.73	0.31	0.31	0.19		-1.08		0.06	0.15	-0.02	-0.02	-0.03	-0.03
BCC-ESM1	0.68	0.72							0.21	0.24	0.12	0.15	-0.04	-0.04
MIROC6							-0.85	-0.83						
NorESM2	0.37	0.39			0.23	0.24								
GFDL-ESM4	0.68	0.70	0.06	0.08			-0.51	-0.55	0.27	0.29	0.14	0.16	0.08	-0.08
GISS-E2-1	0.78	0.80	0.28	0.29	0.20	0.20	-0.92	-0.98	0.23	0.23	0.16	0.16	0.22	0.22
CESM2-WACCM	0.72	0.76	0.34	0.38	0.39	0.40	-0.89	-0.89			0.40	0.44	0.00	0.00
IPSL-INCA														
MultiModel Mean	0.67	0.69	0.12	0.13	0.26	0.29	-0.86	-0.85	0.20	0.23	0.14	0.15	0.09	0.07
S. D.	0.17	0.18	0.21	0.22	0.07	0.08	0.18	0.15	0.07	0.05	0.13	0.14	0.14	0.15

In Table S4 the mean ERF per emissions or concentrations is given for each experiment.

Table S 4 Table of ERF/emissions or concentrations. Emissions for NOx are scaled to Tg of NO2

ERF/emission or concentration	BC (Wm^{-2}/Tg)	SO2 (Wm^{-2}/Tg)	OC (Wm^{-2}/Tg)	NH3 (Wm^{-2}/Tg)	NOx (scaled to Wm^{-2}/Tg NO2)	CH4 (Wm^{-2}/ppb)	HC (Wm^{-2}/ppb)	N2O (Wm^{-2}/ppb)

	0.0212	-0.0094	-0.0147	-0.0013	0.0010	0.0007	0.1200	0.0048

S3 Kernel Breakdown of atmospheric adjustments for each experiment

The full breakdowns of the rapid adjustments as calculated from the kernels is shown for each of the models and experiments where the relevant data was available and shows the differences in models for how the rapid adjustments from different processes contributed to the overall rapid adjustment.

Table S5a Adjustments for piClim-aer experiment

piClim-aer	CNRM-ESM2	UKESM1	MRI-ESM2	BCC-ESM1	MIROC6	NorESM2	GFDL-ESM4	GISS-E2-1
albedo	-0.017	-0.049	-0.009	-0.095	-0.026	-0.015	-0.044	0.003
cloud	-0.661	-0.915	-0.842	-0.900	-0.945	-1.093	-0.452	-0.581
W.V.	-0.055	0.017	0.169	-0.008	-0.085	0.029	0.094	-0.046
T_trop	0.107	0.023	-0.243	0.092	0.183	0.038	-0.138	0.137
T_strat	-0.015	-0.006	-0.014	-0.038	0.010	-0.054	-0.027	-0.038
T_surface	0.054	0.049	0.032	0.075	0.059	0.001	0.035	0.052

Table S5b Adjustments for piClim-BC experiment

piClim-BC	CNRM-ESM2	UKESM1	MRI-ESM2	BCC-ESM1	MIROC6	NorESM2	GISS-E2-1
albedo	0.003	-0.013	0.067	0.015	-0.002	0.076	0.046
cloud	-0.010	-0.013	0.163	0.053	-0.282	-0.067	-0.184
W.V.	0.060	0.057	0.329	0.076	-0.042	0.097	0.038
T_trop	-0.088	-0.137	-0.509	-0.131	0.033	-0.137	-0.051
T_strat	-0.003	0.043	0.021	-0.004	-0.008	-0.025	-0.003
T_surface	0.001	0.005	-0.022	-0.002	0.021	-0.003	0.003

Table S5c Adjustments for piClim-OC experiment

piClim-OC	CNRM-ESM2	UKESM	MRI	MIROC6	NorESM2	GISS-E2-1
albedo	0.008	-0.015	-0.006	-0.017	0.002	0.001
cloud	-0.083	0.052	-0.129	-0.087	-0.100	-0.290
W.V.	0.000	-0.018	-0.009	-0.004	-0.008	-0.054
T_trop	0.025	0.023	-0.016	0.029	0.066	0.057
T_strat	-0.019	-0.003	-0.010	-0.008	-0.016	-0.015
T_surface	0.011	0.015	0.009	0.035	0.010	0.012

Table S5d Adjustments for piClim-SO2 experiment

piClim-SO2	CNRM-ESM2	UKESM1	MRI-ESM2	BCC-ESM1	MIROC6	NorESM2	GISS-E2-1
albedo	0.01	-0.03	-0.04	-0.10	-0.02	-0.09	-0.04
cloud	-0.49	-0.79	-0.73	-0.43	-0.40	-0.96	-0.02
W.V.	-0.04	-0.10	-0.05	-0.07	-0.06	-0.05	-0.06
T_trop	0.10	0.20	0.08	0.22	0.11	0.16	0.14
T_strat	-0.01	-0.02	-0.02	-0.03	0.00	-0.03	-0.01
T_surface	0.03	0.04	0.06	0.07	0.04	0.01	0.03

Table S5e Adjustments for piClim-CH4 experiment

piClim-CH4	CNRM-ESM2	UKESM1	MRI-ESM2	BCC-ESM1	NorESM2	GFDL-ESM4	GISS-E2-1	CESM2-WACCM
albedo	0.019	0.019	0.013	0.031	0.010	0.014	0.007	0.023
cloud	-0.038	0.242	-0.056	0.008	-0.041	-0.054	0.035	0.045
W.V.	0.071	0.109	0.070	0.055	0.018	0.117	0.021	0.068
T_trop	-0.084	-0.162	-0.138	-0.127	-0.080	-0.147	-0.056	-0.082
T_strat	0.114	0.115	0.124	0.039	0.057	0.109	0.053	0.103
T_surface	-0.017	-0.031	-0.028	-0.046	-0.018	-0.023	-0.019	-0.031

Table S5f Adjustments for piClim-HC experiment

piClim-HC	CNRM-ESM2	UKESM1	MRI-ESM2	GFDL-ESM4	GISS-E2-1	CESM2-WACCM
albedo	0.01	-0.02	0.00	0.00	-0.01	0.01
cloud	-0.05	-0.09	-0.02	-0.03	0.03	0.06
W.V.	-0.02	-0.09	0.02	-0.01	-0.01	0.01
T_trop	0.03	0.10	-0.03	-0.03	-0.01	0.00
T_strat	0.21	0.47	0.16	0.26	0.30	0.16
T_surface	0.00	0.01	0.00	-0.02	-0.01	-0.03

Table S5g Adjustments for piClim-N2O experiment

piClim-NO2	CNRM-ESM2	UKESM1	MRI-ESM2	NorESM2	GISS-E2-1	CESM2-WACCM
albedo	0.021	0.001	0.005	0.003	0.000	0.008
cloud	-0.010	0.047	0.004	0.059	0.026	0.119
W.V.	0.038	-0.002	-0.015	-0.012	-0.006	0.016
T_trop	-0.035	-0.006	-0.022	-0.037	-0.002	-0.014

T_strat	0.094	0.074	0.108	0.003	0.130	0.089
T_surface	-0.016	-0.011	-0.005	-0.014	0.005	-0.016

Table S5h Adjustments for piClim-NOx experiment

piClim-NOx	UKESM1	MRI-ESM2	BCC-ESM1	GFDL-ESM4	GISS-E2-1	CESM2-WACCM
albedo	-0.005	0.018	0.023	0.010	0.003	0.041
cloud	-0.036	-0.041	0.007	-0.065	-0.052	0.104
Spec. Hum.	-0.003	-0.040	0.031	0.029	0.005	0.062
T_trop	0.002	-0.014	-0.063	-0.057	-0.001	-0.056
T_strat	-0.024	-0.161	0.029	0.038	0.056	0.095
T_surface	-0.012	-0.012	-0.026	-0.025	-0.001	-0.030

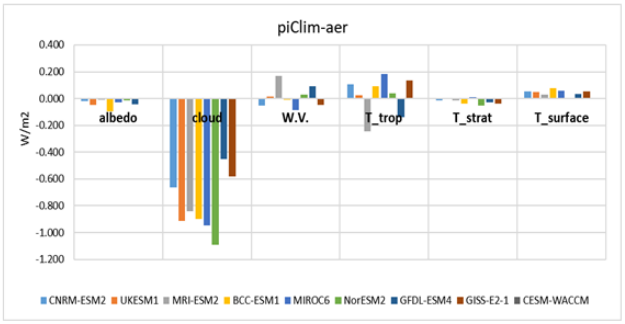
Table S5i Adjustments for piClim-O3 experiment

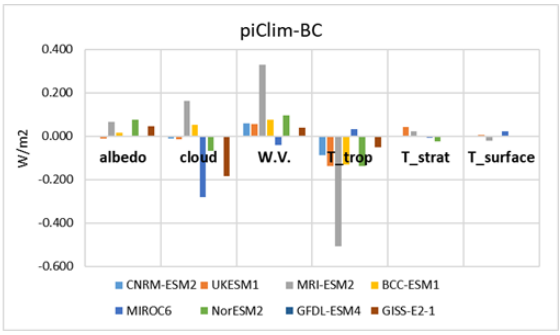
piClim-O3	UKESM1	MRI-ESM2	BCC-ESM1	GFDL-ESM4	GISS-E2-1
albedo	0.001	0.002	0.026	0.009	-0.003
cloud	0.091	-0.126	0.096	-0.079	-0.021
W.V.	0.006	0.010	0.054	0.082	-0.004
T_trop	-0.034	-0.062	-0.115	-0.083	-0.021
T_strat	0.009	-0.036	0.047	0.094	0.113
T_surface	-0.016	-0.010	-0.037	-0.015	0.002

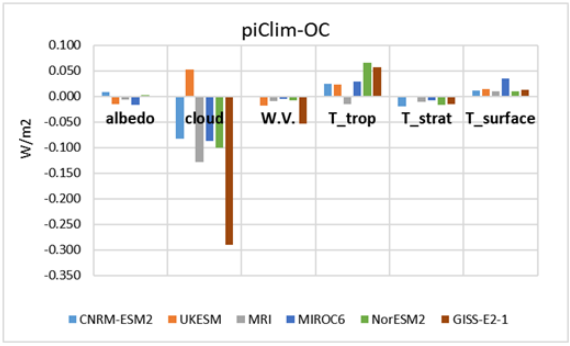
Table S5j Adjustments for piClim-VOC experiment

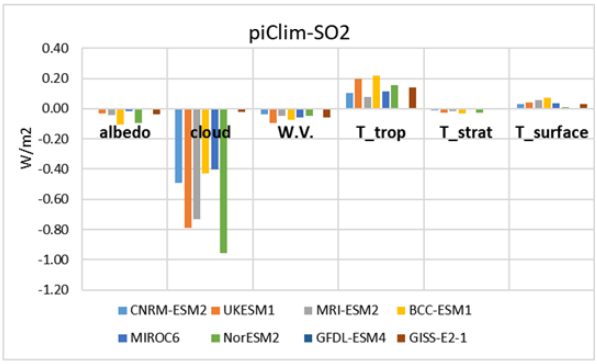
piClim-VOC	UKESM1	MRI-ESM2	BCC-ESM1	GFDL-ESM4	GISS-E2-1	CESM2-WACCM
albedo	0.008	0.006	0.000	0.002	-0.001	0.004
cloud	0.190	-0.147	-0.020	-0.004	-0.001	0.033
W.V.	0.023	0.050	0.022	0.067	0.010	-0.049
T_trop	-0.009	-0.072	-0.046	-0.059	-0.006	0.072
T_strat	0.042	0.054	-0.001	0.024	0.064	0.021
T_surface	-0.006	-0.016	-0.011	-0.011	0.002	0.000

Bar charts showing the atmospheric adjustments calculated from the kernel analysis are included below, showing adjustments for surface albedo, cloud, water vapour, tropospheric temperature, stratospheric temperature, and surface temperature.


piClim-aer
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
CNRM-ESM2 UKESM1 MRI-ESM2 BCC-ESM1 MIROC6 NorESM2 GFDL-ESM4 GISS-E2-1 CESM-WACCM


piClim-BC
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
CNRM-ESM2 UKESM1 MRI-ESM2 BCC-ESM1
MIROC6 NorESM2 GFDL-ESM4 GISS-E2-1


piClim-OC
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
CNRM-ESM2 UKESM MRI MIROC6 NorESM2 GISS-E2-1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W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
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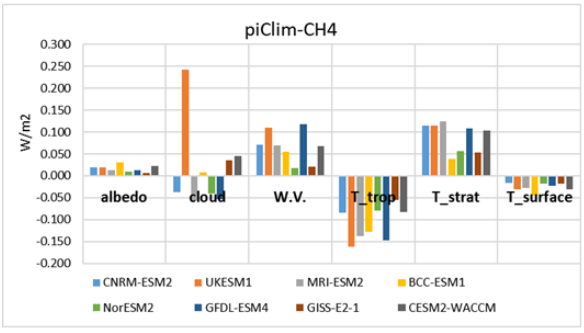
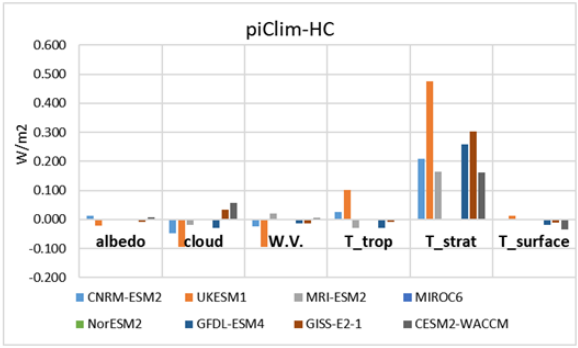
MIROC6 NorESM2 GFDL-ESM4 GISS-E2-1


piClim-CH4
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
CNRM-ESM2 UKESM1 MRI-ESM2 BCC-ESM1
NorESM2 GFDL-ESM4 GISS-E2-1 CESM2-WACCM


piClim-HC
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
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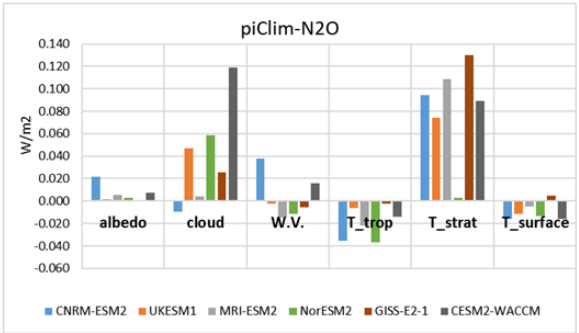
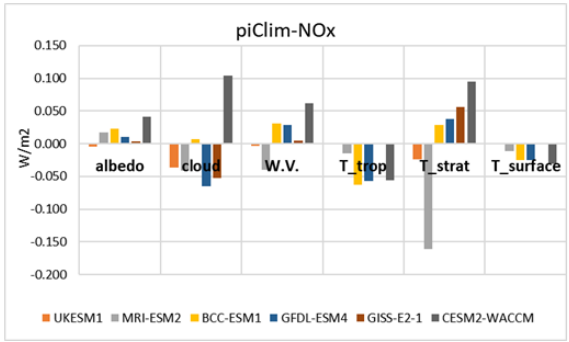
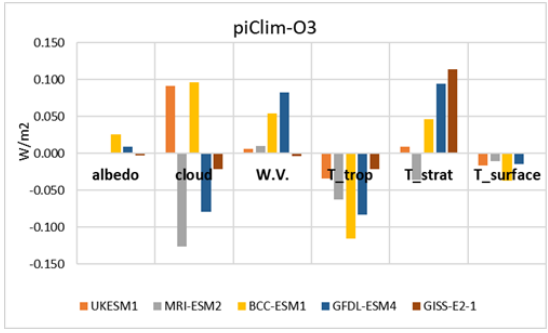
NorESM2 GFDL-ESM4 GISS-E2-1 CESM2-WACCM


piClim-N2O
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
CNRM-ESM2 UKESM1 MRI-ESM2 NorESM2 GISS-E2-1 CESM2-WACCM


piClim-NOx
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
UKESM1 MRI-ESM2 BCC-ESM1 GFDL-ESM4 GISS-E2-1 CESM2-WACCM


piClim-O3
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
UKESM1 MRI-ESM2 BCC-ESM1 GFDL-ESM4 GISS-E2-1

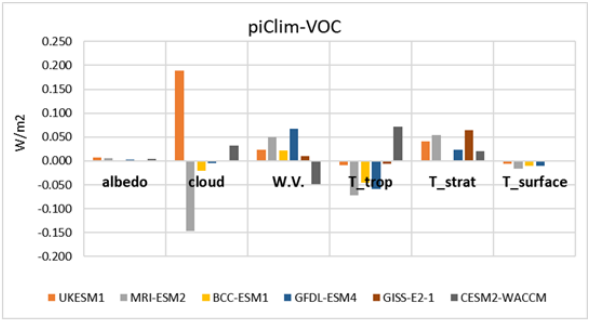
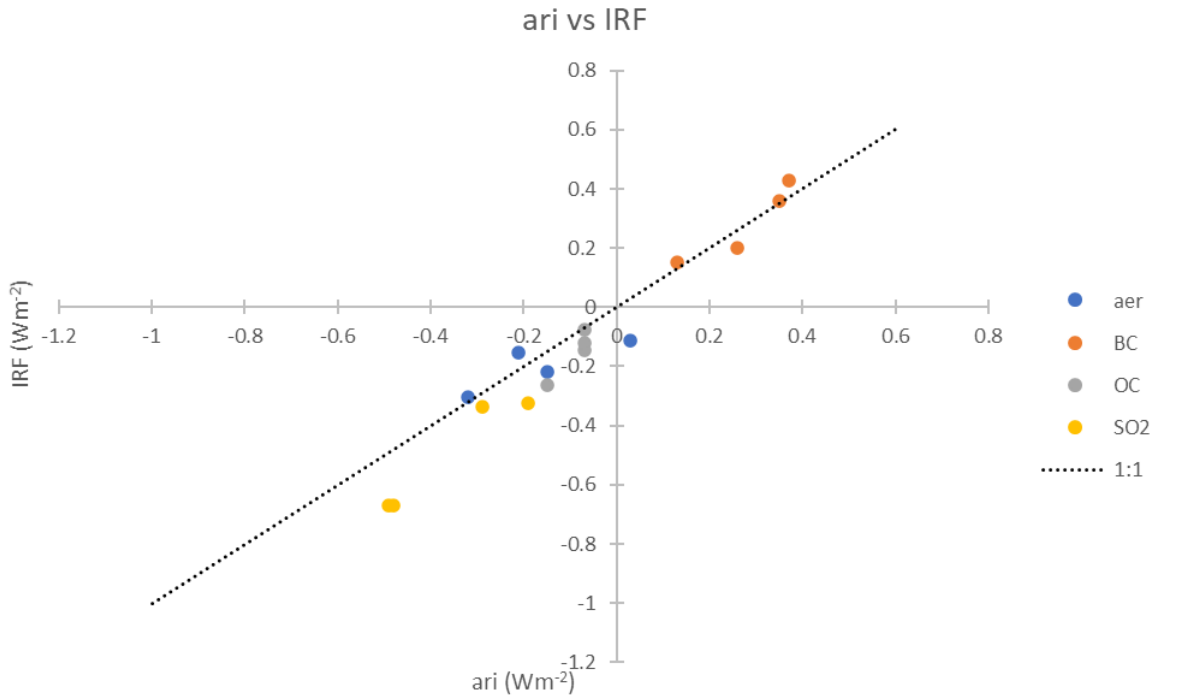

piClim-VOC
W/m2
albedo
cloud
W.V.
T_trop
T_strat
T_surface
UKESM1 MRI-ESM2 BCC-ESM1 GFDL-ESM4 GISS-E2-1 CESM2-WACCM

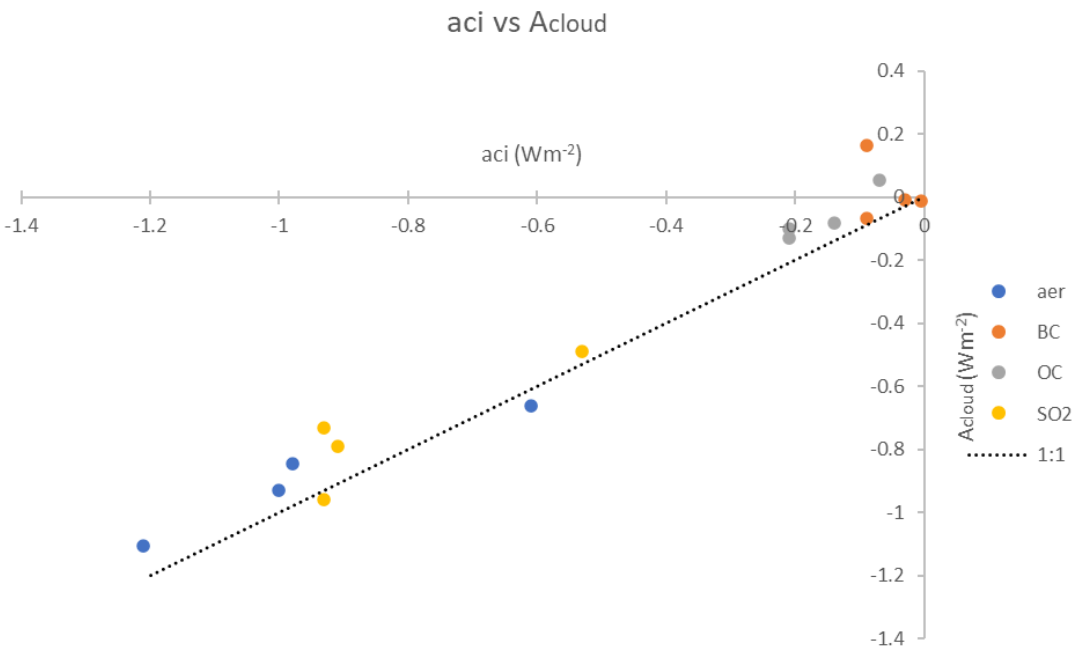
Figure S 1 Plots showing the breakdown of rapid adjustments for all experiments and models with the appropriate diagnostics

Table S 5 Comparison of IRF and cloud adjustment with Smith et al. (2020) for piClim-aer experiment.

piClim-aer (Wm^{-2})	IRF (This work)	IRF (Smith et al., 2020))	IRF Diff	Cloud adj. (This work)	Cloud adj. (Smith et al., 2020)	Cloud adj. Diff	IRF+Cld Adj (this work)	IRF+Cloud Adj. (Smith et al., 2020)	Total Diff	% Diff
CNRM-ESM2-1	-0.15	-0.75	0.60	-0.66	-0.06	-0.60	-0.82	-0.81	-0.01	0.63
GFDL-ESM4	-0.16	-0.37	0.21	-0.45	-0.26	-0.19	-0.61	-0.63	0.02	-2.77
GISS-E2-1-G (p3)	-0.42	-1.00	0.58	-0.60	-0.01	-0.59	-1.02	-1.01	-0.01	1.33
MIROC6	-0.21	-1.13	0.92	-0.95	-0.02	-0.93	-1.16	-1.15	-0.01	0.80
MRI-ESM2-0	-0.30	-0.46	0.16	-0.85	-0.68	-0.17	-1.15	-1.14	-0.01	0.79
NorESM2-LM	-0.11	-1.09	0.98	-1.11	-0.08	-1.03	-1.22	-1.17	-0.05	4.28
UKESM1-0-LL	-0.22	-0.97	0.75	-0.93	-0.18	-0.75	-1.15	-1.15	0.00	0.17
Mean	**-0.23**	**-0.82**	**0.60**	**-0.79**	**-0.18**	**-0.61**	**-1.02**	**-1.01**	**-0.01**	**1.01**

(a)





(b)

Figure S 2 Scatter plots comparing direct and indirect aerosol effects from Ghan diagnostics (x-axes) with kernel-derived breakdown (y-axes). Points are from 4 models that included Ghan diagnostics (CNRM-ESM1, UKESM1, MRI-ESM2, NorESM2) (a) Comparison of IRFari with kernel IRF. (b) Comparison of ERFaci with kernel cloud adjustment (Acloud).

S4 AOD scaling

In Fig (S2) we compare the ERF originally calculated from the radiative fluxes for the (piClim-xx –piClim-control) experiments – referred to as Calc ERF to the ERF contributions obtained from using the AOD scaling,

Figure S 3 Comparison of the ERF calculated from radiative fluxes with that from the ERF from AOD-scaled values.

e.g. the BC AOD in the piClim-BC experiment. In general, the change in the single species is responsible for most of the change in the ERF in these experiments, however in the MIROC6 piClim-OC experiment there is a significant contribution from the organic carbon, indicating this is not as clean a method for obtaining the scaling in this case as for the other models and experiments. In the case of NorESM2 for the SO_2 experiment we also have some contribution from the OA, which may be attributable to the way the nucleation scheme works in NorESM2. Their nucleation scheme looks at the combination of H2SO4 and low-volatile organic vapours (precursors of SOA), so changing the SO2 emissions might therefore indirectly change the pathway for the SOA precursors, leading to a shift in how much nucleates and how much condensates. This might lead to a difference in lifetime of SOA (which is part of OM), leading to differences in the OM burden or AOD. (Dirk Olive, pers. Communication).

In Table S6 the ERF per Tg burden is shown for the piClim-BC, piClim-SO2 and piClim-OC experiments.

Table S 6 Table of ERF/burden for individual aerosol experiments

ERF/burden (Wm^{-2} Tg^{-1})	CNRM-ESM2	MIROC6	NorESM2	UKESM1	GISS-E2-1	MRI-ESM2	BCC-ESM1	IPSL-INCA
piClim-BC	1.43	-2.49	2.38	4.07	0.92	1.74	1.63	0.90
piClim-OC	-0.68	-0.67	-0.55	-0.45	-1.42	-1.02		-0.35
piClim-SO2	-1.12	-0.93	-1.17	-1.34	-1.01	-1.47	-1.33	-0.64

S5 Detailed plot of the atmospheric adjustments for the piClim-CH4 model results

The rapid adjustments for the CH4 experiment are broken down to show the model differences and the contributions of the individual rapid adjustments to the overall rapid adjustment contribution to the ERF.

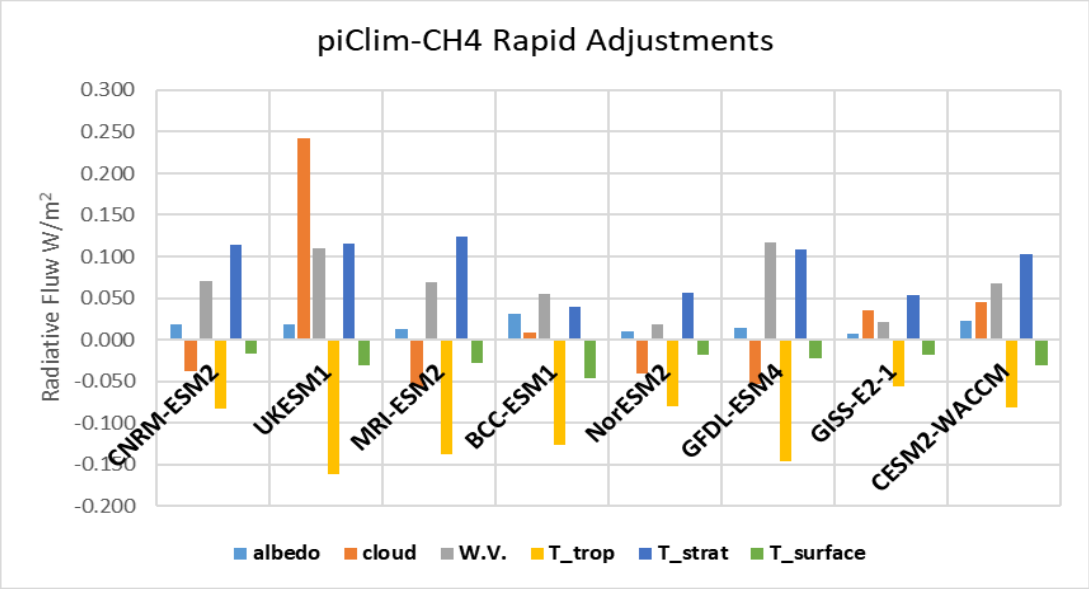


Figure S 4 Plots showing the rapid adjustments for the piClim-CH4 experiments

S5 Plots of the Ghan Calculations

We also plotted the breakdown of the ERF into the ERFari, ERFcloud and the ERFcs,af (clear sky, no aerosol) for models with the appropriate diagnostics, shown in Fig. S4 below.

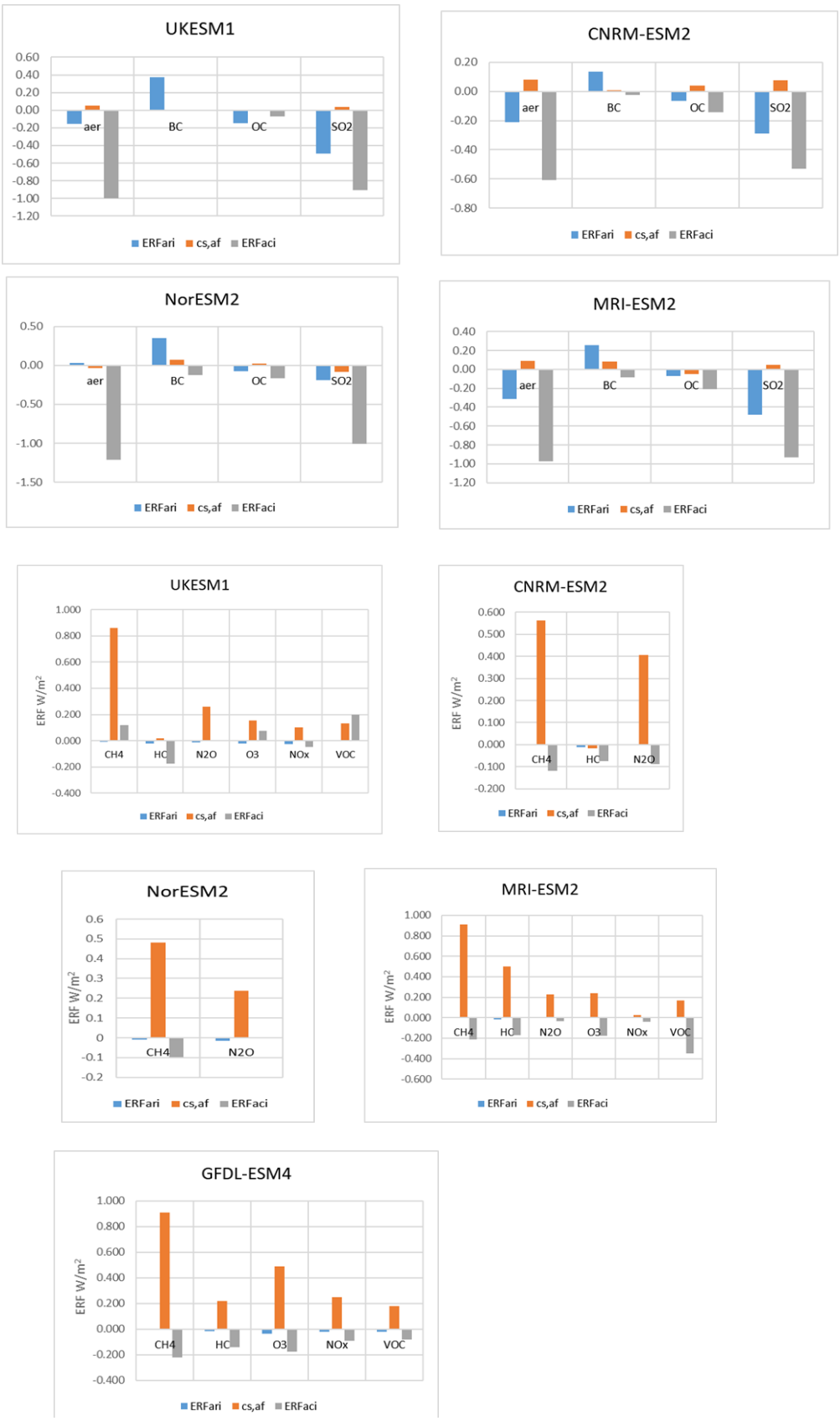


Figure S 5 Breakdown of the ERFs using the double-call method, showing IRFari, ERFcs,af and ERFaci

S6 Experiments using NorESM2 to examine adding 2014 aerosols to an atmosphere with 2014 oxidants.

The following sensitivity experiments were done with the NorESM aerosol scheme. To study the effect of SO_2 emissions, we have done a few extra simulations in addition to piClim-control and piClim-SO2 : two additional experiments which we called piClim-oxid and piClim-oxidSO2.

These experiments are :

(1) piClim-control : SO_2 emissions are 1850, oxidants are 1850

(2) piClim-SO_2 : SO_2 emissions are 2014, oxidants are 1850

(3) piClim-oxid : SO_2 emissions are 1850, oxidants are 2014

(4) piClim-oxid+SO_2 : SO_2 emissions are 2014, oxidants are 2014

The standard results in this paper compare (2) with (1) : this gives ERF = -1.303 W/m^2 (N.B the calculations here were done over 25 years, not 30 years as in the rest of the paper). It reflects the impact of adding SO_2 emissions in a clean pre-industrial atmosphere (both (1) and (2) have the oxidants on 1850 levels, as if NOx, CO, VOC, ... emissions are all 1850).

However, if we compare (4) with (3) : this gives -1.479 W/m^2. It is the impact of adding SO_2 emissions, already in a polluted atmosphere where NOx, CO, VOC, ... emissions are at 2014 levels, and therefore high oxidant values.

It shows that we have differences of the order of 13% : ERF = -1.303 W/m^2 compared to -1.479 W/m2.

Similar experiments for all aerosols together result in the following :

(1) piClim-control : aerosol emissions are 1850, oxidants are 1850

(2) piClim-aer : aerosol emissions are 2014, oxidants are 1850

(3) piClim-oxid : aerosol emissions are 1850, oxidants are 2014

(4) piClim-oxid+aer : aerosol emissions are 2014, oxidants are 2014

Comparing here (2) with (1) gives ERF = -1.214 W/m^2 and comparing (4) with (3) gives ERF = -1.458 W/m^2. This gives a difference of around 20%.

This result is only obtained in a simplified setup (prescribed oxidants), but it might give an indication of how the "chemical climate" affects the result.

The climate conditions (different temperature and deposition rates in 1850 and 2014) are of course not covered by the above experiment. It remains in an 1850 climate.

Finally, the impact of large emission reductions (like 100% for SO_2) can show a different sensitivity than smaller mitigation-type reduction sizes due to non-linearity.

(D. Olivie, pers. Comm).

S7 Breakdown of Ozone changes

Table S 7 Column ozone, and ozone changes resulting from changes concentrations (CH4, N2O, HC) or emissions (NOX, VOC, O3, NTCF) of reactive gases. The multi model mean does not include the results for CNRM-ESM2 for tropospheric ozone.

Experiment	CNRM-ESM2		UKSM1		MRI-ESM2		BCC-ESM1		GFDL-ESM4		GISS-E2		CESM2-WACCM		Multi-model	
	trop	strat	trop	strat	trop	Strat	trop	strat	trop	strat	trop	strat	trop	strat	trop	strat
Control DU		303.0 ±0.2	25.71 ±0.06	313.2 ±0.6	19.88 ±0.04	294.8 ±0.4	23.20 ±0.03		20.15 ±0.02	267.0 ±0.2	20.45 ±0.04	258.5 ±0.1	20.33 ±0.04	260.3 ±0.2	22.3 ±2.60	283 ±20
CH4 DU		+6.1±0.3	3.02 ±0.08	+2.0 ±0.6	+2.48 ±0.04	+2.9 ±0.5	+2.42 ±0.03		2.50 ±0.04	+2.1 ±0.2	2.17 ±0.05	+5.3 ±0.2	+3.15 ±0.04	+2.9±0.2	+2.6 ±0.3	+4 ±2
NOx DU			5.20 ±0.08	+4.6 ±0.6	+4.09 ±0.05	+10.5 ±0.5	7.23 ±0.03		6.61 ±0.03	+1.1 ±0.2	9.19 ±0.05	+1.0 ±0.2	+6.97 ±0.04	+0.1 ±0.2	+6.5 ±1.6	+3 ±3
VOC DU			1.47 ±0.08	+1.6 ±0.6	+1.99 ±0.05	+2.0 ±0.5	0.79 ±0.03		1.94 ±0.03	+2.5 ±0.2	1.90 ±0.05	-1.9 ±0.3	+1.57 ±0.05	+2.0 ±0.2	+1.6 ±0.4	+1 ±1
O3 DU			6.86 ±0.08	+5.1 ±0.6	+7.51 ±0.04	7.2 ±0.5	8.52 ±0.03		9.46 ±0.03	+2.8 ±0.2	11.38 ±0.06	-0.6 ±0.3			+8.7 ±1.6	+4 ±3
N2O DU		-6.7 ±0.3	0.16 ± 0.08	-3.1 ±0.6	--0.05 ±0.04	-4.7 ±0.5					0.23 ±0.05	-7.6 ±0.2	+0.41 ±0.04	-4.5 ±0.2	+0.2 ±0.2	-5 ±1
HC DU		-23.4 ±0.8	-2.12± 0.08	-38.2 ±0.6	-0.41 ±0.05	-13.4 ±0.5			-1.51 ±0.0	-23.3 ±0.2	-2.54 ±0.05	-24.2 ±0.2	-0.61 ±0.06	-22.7 ±0.4	-1.4 ±0.8	-23 ±8

Table S 8 Percentage change in total aerosol mass (sulphate, nitrate and secondary organic) from the reactive gas experiments.

Experiment	UKESM	MRI-ESM2	BCC-ESM2	GFDL-ESM4			GISS-E2			CESM2-WACCM	
	SO4	SO4	SO4	SO4	NO3	SOA	SO4	NO3	SOA	SO4	SOA
CH4	-2	+2	+1	+6	0	0	-1	-11	+1	-1	-1
NOx	-2	0	+9	-8	+200	+1	-19	+120	+19	+2	-10
VOC	-3	+1	0	+4	-5	+8	-2	0	+1	-2	+34
O3	-4	+1	+10	-3	+190	+7	-21	+130	+21		
N2O	+1	+1					+1	+7	+1	+1	+1
HC	+2	+1		+1	0	+1	+1	-11	+1	-1	+1

S8 Methane Lifetime

Table S 9 Methane lifetime (years), and change due to each experiment (%). Multi-model mean and standard deviation. Lifetimes assume a soil loss of 120 years. Stratospheric loss is included in the model calculations.

Experiment	UKESM1	CESM2-WACCM	GFDL-ESM4	BCC	GISS-E2	MRI-ESM2	Multi-model
Control years	8.0	8.7	9.6	6.3	13.4	10.1	10.0 ±1.9
CH4 %	+22	+22	+21	+26	+18	+22	22±3
NOx %	-25	-35	-33		-46	-26	-33±8
VOC %	+11		+15		+27	+21	+19±6
O3 %	-19		-24		-40	-20	-16±9
HC %	-4.9		-7.5		-0.6	-2.4	-3.7 ±2.4
N2O	-1.2	-2.8			-3.9	-1.3	-2.0 ±1.1

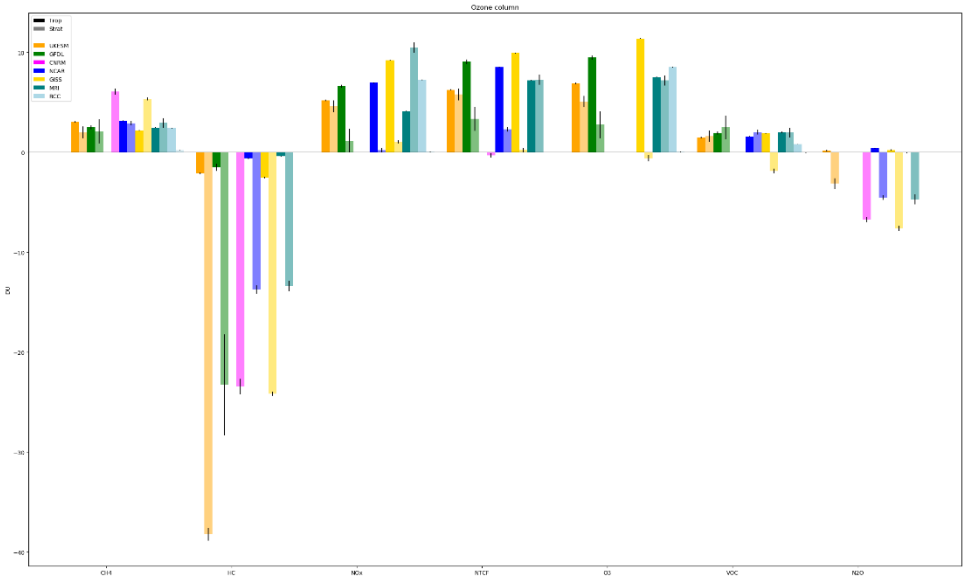


Figure S 6 Ozone column values for the troposphere and stratosphere for the reactive greenhouse gas experiments

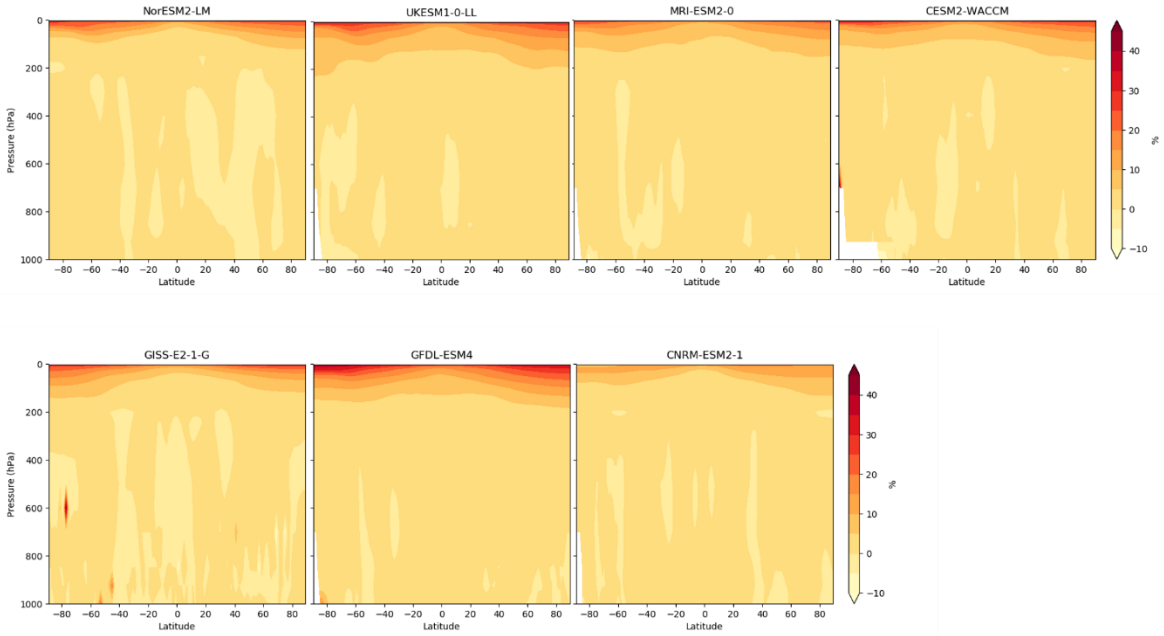


Figure S 7 Percentage changes in water vapour from the piClim-CH4 experiments.

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