Thank you to both reviewers for their helpful comments, which we have addressed below.

Response to Reviewer 1

Major comments:

1) Normalized analysis has been shown to provide much more insight into the comparability of forcing-response relationships (Shindell and Faluvegi being a clear example). While it is clear that it is useful to provide the un-normalized numbers, the paper would greatly benefit from adding a description of the normalized results. For example, if ones takes the numbers from HadGEM/ECHAM/NorESM for BC, we see that the response in delta(T) is almost the same as the scale OC response. As it should. A table documenting the radiative forcing associated with each perturbation run should be included.

This work focusses on the climate impacts of perturbing emissions. This is a different focus from e.g. Shindell and Faluvegi who aim to assess the climate responses to regional forcing from aerosols/GHGs rather than the actual emissions.

In order to calculate climate responses for a given RF/ERF, i.e. the normalised responses, it would be necessary to run a different set of experiments (atmosphere-only rather than coupled) in order to find the RF or ERF from the emissions changes, since these cannot be derived from the coupled simulations. We therefore do not have the RF/ERF values that would be necessary to calculate normalised responses.

We can however, show the response per change in TOA SW flux. We have added this to Table 2 (change in temperature per change in TOA SW flux, and change in precipitation per change in TOA SW flux). We have also added references to these normalised values in the text throughout Section 3 in the discussion of the responses.

2) The figures only show stippling where models agree on the sign. That is a pretty low bar to pass (and I guess they still don't pass it). I would however provide estimates of the statistical significance based on the interannual variability. Similarly, zonal mean figures (5-7) are shown even for areas where models do not agree. What is the meaning of those figures in that case!?

We have changed the stippling in Figs 5-8 to denote statistical significance at the 95% confidence level (determined by a Student's t-test on all years of all models). The distribution is generally very similar to that using previous method.

The zonal mean figures show the zonal means for the individual models as well as the multi-model means. Showing the individual models is useful to see differences (and similarities) between models. The multi-model mean zonal means are useful to compare with the map figures (which show only the multi-model means), and are useful to see when the sign of the response in the different models is in agreement.

3) The control experiment is much too short for the analysis that is being performed here, where the goal is to identify the response to a forcing much smaller than 2xCO2. As one can see for ECHAM, the global surface temperature is still trending at the end of the fifty years. Knowing that, it is necessary to show and discuss the trends in the climate state for the control experiment continued over the 50 years for which the perturbation is calculated. It would not be surprising if part of the "signal" was actually present in the control experiment as well. An approach might be to take into account the model drift over the 50 years.

We agree that it is not ideal to perform these experiments against a control state that still has a slight temperature trend. However, we believe that the reviewer's concerns are due to a misunderstanding of our explanation of the experimental setup, and therefore we have amended the text to explain this more clearly. To clarify: we first ran several decades of spin-up. We then started the control simulations and the perturbed simulations both from the same point at the end of this spin-up period. Therefore the 50 year periods that we are comparing are the same for both the control and perturbed runs. Therefore any underlying temperature trend in the control will also still be present in the perturbed simulations. Since we only consider differences between the perturbed and control simulations, rather than absolute values, this will not affect the results.

There are other studies in the published literature which have used similar integration-lengths and found these to be sufficient, e.g. Pausata et al (2014), Kristjansson et al (2005). Based on this we believe that 50 years is sufficient.

Added some clarification to the first paragraph in section 2.2 (pg 3832):

The control simulations were first run for several decades using an initial ocean state based on present-day CMIP5 conditions for all models except for ECHAM-HAM, which used a preindustrial control state (see below). The control and perturbed simulations were then run from **the same point in** this spun-up state for 50 yr, in order to separate a robust signal from the interannual variability. **The climate is not necessarily expected to be stationary after the spin up, but any underlying climate trends are expected to be present in the control and perturbations. By taking the difference between the control and the perturbations we are therefore removing any underlying trends not associated with the changes in aerosol emission.** The 50-year integration length was deemed sufficient based on previous studies, e.g. Kristjansson et al (2005) performed integrations of length 40 years after 10 years of spin-up, and Pausata et al (2014) performed integrations of length 30 years after 30 years of spin-up. Furthermore, Olivie et al (2012) showed that most of the temperature response to a step CO2 perturbation in AOGCMs is achieved within around the first 10 years or so (the Cx2 case in their Fig. 1), after which the temperature remains relatively constant, with only a very gradual continued increase towards the equilibrium response temperature.

Added more to pg 3834 line 11:

These drifts are also present in the perturbation experiments, since these start from the control simulation at the beginning of the 50 yr period analysed. Therefore we do not expect any drift in the signal, i.e. in the difference between the perturbed and control simulations.

4) There is no description of how the control experiments are performed. What is the level of GHGs? Where does the ocean initial state come from?

The description of the control experimental setup and GHGs is in the paragraph starting on pg 3832

line 18. The ocean initial state is spun-up from present-day (in the case of HadGEM, NorESM and CAM4, which use present-day GHGs) and pre-industrial (in the case of ECHAM which uses preindustrial GHGs) states, based on CMIP5 initial conditions. The GHGs are fixed throughout; any differences occurring due to the different GHGs or initial ocean state will be much smaller than the differences due to different model dynamics/chemistry/aerosol/parameterisation schemes.

Amended text to describe the ocean state:

pg 3832, line 1:

The control simulations were first run for several decades using an initial ocean state based on present-day CMIP5 conditions for all models except for ECHAM-HAM, which used a preindustrial control state (see below). The control and perturbed simulations were then run from this spun-up state for 50 yr, in order to separate a robust signal from the interannual variability.

Minor comments:

1) Author list: It is CICERO, not CISERO

Changed

 Page 3825, line 5: There are much better and recent references to the impact of ozone on health and agriculture than HTAP. For example Tai et al., Nature Climate Change, 2014.

Added two references: Tai et al 2014 and Amann et al 2013

3) Page 3825, line 19-21: it might be useful to use the AR5 nomenclature (ACI-ARI) Changed sentence to read:

"The **aerosol-radiation interactions and aerosol-cloud interactions** bring futher inhomogeneities,..."

4) Page 3826, line 23: there is a wealth of recent papers highlighting those connections. Please provide a better list of references.

Added references: Boucher et al 2013, Osborne and Lambert, 2014 More references are included in the rest of the paragraph about precipitation effects.

5) Page 3831: it is not really clear what the added value of including CAM4 is. This version does not have indirect effects. Why bother? Why only BC?

We agree that our reasoning was not clearly explained. The purpose of including the CAM4 simulations and the extra NorESM simulation was to explore further the BC results, to understand them further. This work was part of a project of which BC was one of the key focusses, and hence the CAM4 experiments were available to add to this study. The fact that CAM4 does not include the indirect effects is not a problem since the BC indirect effects are small relative to the direct/semi-direct effects

Added sentence to text on pg 3829 line 5:

The extra BC simulations were included in order to explore the BC results further, as this work was part of a larger project of which BC was a key focus.

Page 3833, line 5: ozone would be affected though, because of the methane impact.

Oxidant fields for the sulphate aerosol production calculation were a 2003-2010 average from the MACC reanalysis (Inness et al. 2013)

Added the above sentence to the text on pg 3038, end of line 11.

7) Page 3833: this whole section need to include a documentation of the aerosol budget. Also, it needs to include a discussion of the differences in precipitation between the models. Finally, the aerosols should be compared to existing papers such as Samset et al., Observational evidence for overestimation of modeled black carbon radiative forcing. Atmos. Chem. Phys., 14, 12465-12477, doi:10.5194/acp-14-12465-2014, 2014.

We have added the lifetimes of BC,OC and SO4 to Table 1 (in addition to the burdens which were already shown). We have added more discussion to the paragraph starting on pg 3833 line 13, including discussion of existing papers:

"Despite all the models having the same emissions input, there is a large discrepancy between models in the vertical distribution of aerosols in the atmosphere, and in the total aerosol burden, which is typical for current global aerosol models (Textor et al 2007). HadGEM and ECHAM-HAM have relatively low total burdens of BC, and short atmospheric lifetimes, compared with NorESM and CESM-CAM4 (Table 1). Figure 2 shows vertical sections of the annual average, zonal mean BC mass mixing ratio in the control simulation for each of the models considered. HadGEM and ECHAM-HAM (Fig. 2a and b) have low concentrations of BC at high altitude, which means there is less BC above clouds. In contrast, NorESM and CESM-CAM4 show high BC concentrations extending to above 200 hPa throughout most of the northern hemisphere and southern hemisphere tropics (Fig. 2c and d). This has implications for the impact that removing anthropogenic BC emissions may have. BC at high altitude can have very strong direct effects if it is located above high-albedo cloud surfaces. In the models with higher concentrations of BC at high levels in the control simulations, more of this high-level BC can be removed in the BC perturbation experiment, leading to a larger change in BC direct forcing. The larger amount of high-level BC in NorESM and CESM-CAM4 (which uses aerosol input from OsloCTM2) is consistent with the AeroCom models discussed by Schwarz et al. 2013 and Samset et al. 2014 who found that these models have too much BC at high altitudes when compared with observations over the Pacific in the HIPPO campaign (Wofsy, 2011), and overestimate the BC lifetime. In HadGEM, the lower concentrations of BC at high altitudes and shorter BC lifetimes are likely due to recent modifications to the convective scavenging scheme, which were implemented in order to improve the correspondence with these observations. However, the BC lifetime of 3.4 days is rather short, so in this case the model is underestimating the high-level BC concentration. The true BC distribution is therefore somewhere in between that of HadGEM and **NorESM/CESM-CAM4.** The OC burden in NorESM is considerably higher than in the other three models, and its lifetime is correspondingly longer. The range of OC burdens between models is expected due to differences in OA burdens and OA/OC ratios between models (Tsigaridis et al. 2014). NorESM and CESM-CAM4 have relatively low burdens of SO_4, and short lifetimes, compared with HadGEM and ECHAM-HAM. There are also differences in the vertical distribution of OC and SO 4 between models (not shown) but as these are scattering, rather than absorbing, aerosols the impact of the vertical distribution of the aerosol will have less of an impact on the results. A more detailed evaluation of the models used here against observations is given in Stohl et al. 2015 and references therein."

We have added a supplementary figure showing observed (GPCP) precipitation for 2000-2010, the annual average precipitation in the control simulation for the multi-model mean and each individual model. We have added a paragraph to the end of Section 2 describing these:

"There are some differences between models in the precipitation patterns, particularly in the tropics (Fig. S1). All models suffer from the "double ITCZ" problem (i.e. there is an overly strong band of precipitation to south of the equator) which is a known problem in CMIP5 AOGCMs (Li and Xie, 2014). This is most pronounced in ECHAM-HAM (Fig. S1d). ECHAM-HAM and HadGEM also have region of very low precipitation around the Equator in the Pacific (Figs. S1c and d). There is some variation in the north-eastward extent of the North Atlantic storm track: in NorESM it extends too far north-east, while in CAM4 it does not extend far enough (Figs. S1e and f); in HadGEM and ECHAM-HAM it matches the observations well. All models have too much precipitation over the Himalayas and the Andes, which is probably due to inaccuracies in their representation of precipitation over high orography."

8) Page 3834, line 22: why are HadGEM and ECHAM similar when their SO4 burdens are so different?

The change in SO2 emissions is the same in each case, so the change in SO4 will be roughly the same, regardless of the different absolute burdens. Since we are looking at a difference in temperature between the control and perturbed runs, the baseline SO4 concentration should not impact the results too much.

9) Page 3836, line 2: include references discussing the shift in ITCZ

Added references: Broccoli et al., 2006; Kang et al., 2008; Ceppi et al., 2013

10)Page 3836, line 23-25: how do we know that this is the causal link?

We have changed the text as follows. This is only hypothesis.

"There are broad regions over Russia and North America with increased precipitation. These are collocated with regions of increased surface temperature, which would provide more available moisture through evaporation. The increased precipitation could also be due to the reduced aerosol concentrations in these regions."

11)Page 3836, line 26: the discussion of run-off might be much more useful if it is recast in terms of river flows, maybe for the largest rivers.

We agree that the discussion of the run-off changes is not very useful. We have therefore decided to remove plots and discussion of run-off since differences are very small and mostly not significant.

Figures of run-off and corresponding discussion have been removed from the paper.

12)Page 3838, lines 19-21: how do the authors know that they are not simply looking at noise?

We have included stippling in Figures S1-S3 to show significance. This shows that the temperature responses and sea-ice changes in individual models are significant.

We have also rewritten the corresponding paragraphs in the text (pg 3838 line 19 to pg 3839 line

20) to aid clarity of this discussion.

13)Page 3842, lines 5-9: it is not that clear that the ITCZ shift is related to climate sensitivity. Please substantiate!

This sentence does not refer to the ITCZ shift. Reworded to "NorESM gives a weaker temperature response than the other two models."

14)Page 3842, lines 21-25: as the papers by Sand et al have indicated (among others) the location of the BC forcing is quite important.

Added sentence:

"...BC deposition on snow. As shown by Sand et al (2013b), this has a relatively large impact on surface temperature in the Arctic."

15)Page 3843, lines 25-26: I would expect that concentrations are available from those simulations. Therefore a statement other than "probably have" should be made.

The difficulty is actually in determining what the true BC distribution should be, since there are no observations that give a global, 3D picture of the present-day BC distribution.

Rewritten the paragraph on pg 3843:

"...partly to the different atmospheric BC distributions in the models, as shown in Fig. 1. Accurately representing the correct BC distribution in GCMs is very difficult (Samset et al. 2015). Recent studies (e.g. Schwarz et al. 2013, Hodnebrog et al. 2014, Samset et al. 2015) have compared BC distributions in GCMs with data from observational studies such as the HIPPO campaign, which provided observations from a large spatial area over the Pacific (Wofsy, 2011). They found that the models had too much BC at high altitudes in these regions, and that the BC lifetime was generally too long. Recent modifications to the convective scavenging scheme in HadGEM (which are included in the model set-up used here) were designed to reduce the amount of high-level BC to give better agreement with the HIPPO observations. The result of these changes is that HadGEM has less BC at high levels globally than the other models (except ECHAM-HAM), and a much shorter BC and OC lifetime (Table 1). ECHAM-HAM also has less BC at high levels, and a short BC lifetime. In contrast, NorESM and CESM-CAM4 have much more high-level BC and longer BC lifetimes, which may overestimate the direct forcing from anthropogenic BC (consistent with Samset et al. 2015) and may therefore exaggerate the impact of removing anthropogenic BC emissions.

Response to Reviewer 2

• Page 3824, Line 17: There are four models in the study, but here the authors refer to "all three models".

Removed the word "three" to avoid confusion.

• Page 3824, Lines 17-18: Perhaps rephrase to "northern hemisphere mid and (especially) high latitudes".

Done

• Page 3825, Line 7: Typo: SCLPs -> SLCPs

Done

• Page 3825, Lines 9-10: According to the UNEP definition, methane is also included in SLCPs, so the authors could include all species up to methane here in their definition, but mention that they restrict their focus to the constituents with lifetimes of days to months, which therefore have a particularly inhomogeneous distribution.

Reworded as follows:

SLCPs have relatively short atmospheric lifetimes compared with well-mixed greenhouse gases (WMGHGs) such as CO2, **with most** remaining in the atmosphere for only days to months. **The exception is methane, which has a lifetime of around a decade, but here we focus on the shorter-lived species.** The impacts of SLCP emissions on climate therefore occur on relatively short timescales of less than 30 yr (Collins et al., 2013). The short atmospheric lifetime of **non-methane** SLCPs ...

• Page 3826, Lines 2-3: Need to also mention explicitly the cloud lifetime effect here.

•

Modified this sentence:

Hydrophilic aerosols also provide cloud condensation nuclei (CCN), allowing more smaller cloud droplets to form, which increases the cloud albedo and the cloud amount, and prolongs the cloud lifetime by inhibiting precipitation. This further contributes to the negative forcing (Boucher et al., 2013).

• Page 3826, Lines 6-7: Please mention why BC warms the surface when near it (reemission in thermal wavelengths).

Added:

low-level BC can warm the surface **by re-emitting radiation in the thermal wavelengths**, whereas ...

• Page 3826, Line 25: Please add "global" before "temperature".

Done

• Page 3828, Line 12: Circulation changes are not really assessed in the paper, so I

would remove this word from here.

Done

• Page 3828, Lines 23-25: Please clarify whether photolysis is affected by the aerosol tracers in the models.

No it is not.

Added sentence to pg 3828 line 25: Photolysis is not affected by the aerosols in these models.

• Page 3829, Lines 11-13: Please mention whether stratospheric chemistry is simulated too.

No it is not

Added text to pg 3829 line 11:

"The UKCA TropIsop scheme is used to model gas-phase chemistry in the troposphere."

• Page 3829, Lines 21-26: I would suggest mentioning how aerosol effects on clouds are simulated.

These are modelled using an aerosol activation parameterisation.

Added sentence on pg 3829 line 25:

"The effects of aerosols on clouds are modelled using an aerosol activation parameterization (Abdul-Razzak and Ghan, 2002)."

• Page 3830, Line 8: It is mentioned earlier that the gas-phase chemistry is not modelled online in ECHAM6-HAM2. Where do the oxidants fields used for aerosol production come from? Worth mentioning.

•

Oxidant fields for the sulphate aerosol production calculation were a 2003-2010 average from the MACC reanalysis (Inness et al. 2013)

Added the above sentence to the text on pg 3038, end of line 11.

• Page 3831, Lines 5-6: Is the BC/dust deposition effect on surface albedo accounted for in the two models mentioned earlier in the text? Please clarify.

No, this is not represented by the other models.

Added to sentence:

In the fully coupled NorESM1-M, albedo-effects of BC and mineral dust aerosols deposited on snow and sea-ice are also taken into account; this process is not represented in the other three models.

• Page 3831, Line 7: Please clarify again here that the NCAR CESM model is only used

for the BC analysis. Added to start of line 7: "An additional model, NCAR CESM 1.0.4/CAM4, was used for the BC analysis only."

• Page 3831, Line 27: Please add "globally" after "removed".

Done

• Page 3832, Line 2: Was the spin-up performed for the control and the perturbation runs of equal length?

The same spin-up was used for all simulations. The control and perturbations were branched off at exactly the same point in the spin-up.

Expanded and reworded this sentence (pg 3832, line 1):

The control simulations were first run for several decades using an initial ocean state based on present-day CMIP5 conditions for all models except for ECHAM-HAM, which used a preindustrial control state (see below). The control and perturbed simulations were then run from this spun-up state for 50 yr, in order to separate a robust signal from the interannual variability.

• Page 3832, Line 26: Please mention explicitly what you mean by "other natural emissions", as this currently sounds a bit vague.

Added text to this sentence:

"Other natural emissions, including DMS and volcanic emissions, are included,..."

• Page 3833, Lines 1-2: What about methane in CESM-CAM4 – how is it treated?

It is prescribed at present-day levels as in HadGEM and NorESM.

Added:

... present-day levels in HadGEM, NorESM and CESM-CAM4 and at ...

• Page 3833, Line 12: It is a bit counter-intuitive that Africa has such strong anthropogenic emissions. Does this include agricultural biomass burning? If so, the distinction should be made clearer (i.e. whether there is absolutely no BB component in the anthropogenic emissions removed or whether there are exceptions).

Thank you for pointing this out. Agricultural BB is removed with the anthropogenic emissions.

Amended pg 3832 line 21, to clarify this:

Non-anthropogenic biomass burning emissions are from the GFED3 emissions dataset (http://www.globalfiredata.org) for the year 2005 (in ECHAM-HAM and NorESM) and 2008 (in HadGEM and CESM-CAM4), and are not perturbed. **Agricultural biomass burning emissions are included in the anthropogenic component of emissions which are perturbed.**

Also amended the caption for Fig. 1 to read "...non-anthropogenic biomass burning..."

• Table 1: Sulphate is not mentioned in the caption. Also, the caption says "three models",

whereas burdens for four models are shown.

Corrected

Caption changed to:

Summary of BC, OC and SO4 burdens in the control simulation for the four models.

• Page 3833, Lines 20-25: It would be useful to briefly mention here which model may be closer to reality when it comes to vertical BC distribution. Any ideas?

Added further discussion to this paragraph. In particular:

The larger amount of high-level BC in NorESM and CESM-CAM4 is consistent with the AeroCom models discussed by Schwarz et al (2013) and Samset et al (2014), who found that these models have too much BC at high altitudes when compared with observations over the Pacific in the HIPPO campaign (Wofsy et al., 2011), and overestimate the BC lifetime. In HadGEM, the lower concentrations of BC at high altitudes and shorter BC lifetimes are likely due to recent modifications to the convective scavenging scheme, which were implemented in order to improve the correspondence with these observations. However, the BC lifetime of 3.4 days is rather short, so in this case the model is underestimating the high-level BC concentration. The true BC distribution is therefore somewhere in between that of HadGEM and NorESM/CESM-CAM4.

• Figure 3: Please mention in the caption that these means are for the surface.

Added the word "surface" before "temperature"

• Page 3834, Lines 8-11: Are similar drifts also present in the perturbation simulations (so that they cancel out and do not affect the differences between perturbation and control runs)?

The perturbation experiments are started as perturbations from the control runs (at the beginning of the 50-year period we analyse). The drifts, are therefore still present in the perturbation experiments.

Added sentence to pg 3834 line 11:

These drifts are also present in the perturbation experiments, since these start from the control simulation at the beginning of the 50 yr period analysed.

• Page 3834, Line 15: Please add "in" after "interested".

Done

• Figures 4 & 6: Please mention in the caption whether the SW TOA fluxes are calculated for clear-sky cases only.

These figures show the all-sky SW TOA fluxes.

Added "all-sky" before "TOA SW flux" in both figure captions.

• Figures 5-8: Making the fonts of some of the labels (e.g. on the colour bar, or above the panels) somewhat larger would help with the readability of the figures.

We will consider this point at a later stage as the figures may be larger in the final (ACP) format than in the current (ACPD) format.

• Page 3835, Line 25: Please amend typo ("smilar").

Done

• Page 3835, Lines 25-28: Please clarify why it is more likely to be the aerosol indirect effect rather than direct (from pollution outflow and associated radiative effects).

Good point.

Added text and amended text on lines 22-28:

"This is consistent with the decreased aerosol concentrations in this region due to the reduced emissions in China. As well as the direct radiative effects, the reduced aerosols would also cause changes in cloud cover. It was shown by Wang et al. (2014) that Chinese aerosols increased cloud cover over the North Pacific, so removing these aerosols would reduce cloud cover. A similar region of positive TOA SW flux change also occurs over the North Atlantic, which is similarly due to aerosol-radiation and aerosol-cloud effects over this region resulting from the aerosol emissions reductions over the eastern USA."

• Page 3836, Lines 1-2: Please support this with an example reference (there are plenty).

Added:

(Rotstayn et al., 2000, Broccoli et al., 2006)

• Page 3836, Lines 16-19: The reduction in precipitation is seen south of the equator, whereas the reduction in SW TOA flux is maximum just north of the Equator. How do the authors explain this inconsistency?

This is consistent with the ITCZ moving northwards. As well as the precipitation shift there will also be a northward shift in cloud related to the ITCZ, which leads to the decrease in TOA SW to the north (where the cloud increases). This was already discussed in pg 3835 line 28- pg 3836 line 2.

Reworded this sentence to clarify this:

In the tropics there are regions of decreased TOA SW flux just north of the equator and increased TOA SW flux just to the south. These relate to a northward shift in the ITCZ, which increases the cloud cover north of the equator and decreases it to the south.

• Page 3836, Lines 20-23: Worth highlighting the Sahel wettening as well. And mentioning a few key references, as done for the South Asian case.

Added sentence to line 23:

There is a large increase in precipitation over the Sahel. This is consistent with the results of Rotstayn and Lohmann (2002) who found that present-day anthropogenic sulphate aerosol had contributed to reduced precipitation in the Sahel.

• Page 3836, Lines 23-25: Any ideas why much of Europe and parts of the US become

drier? Possibly circulation adjustments? Or a northern expansion of the NH subtropical regions?

The drying in these regions is not statistically significant so we don't want to put too much emphasis on it. We hypothesize that it could be linked to the northward shift of the ITCZ and a corresponding adjustment of the Hadley Circulation.

Added to text on page 3836 line 25:

"Over much of Europe and the USA there is a decrease in precipitation. While this is not statistically significant, we hypothesize that this is linked to the northward ITCZ shift and corresponding changes in circulation."

• Page 3837, Lines 9-10: Not in temperature, it seems, as in Fig. 4a HadGEM seems a bit higher than ECHAM.

Corrected:

"...despite having the largest increase in precipitation (Fig. 4 c and d)."

• Page 3837, Line 13: I would say, "qualitatively agree", as the agreement on the magnitude is not apparent, with one model showing half the response. You could then add that two of the models show very good quantitative agreement too.

Added "qualitatively" before "agree" in this sentence. Added a final sentence: "HadGEM and ECHAM-HAM show very good quantitative agreement in the response."

• Page 3837, Lines 24-25: It would be appropriate to refer to Table 2 here. And generally to mention Table 2 a bit more often in the text, as it can help the reader make linkages.

Added reference to Table 2 in the following places:

Pg 3834 line 24 (after "Fig 4a") Pg 3837 line 22 (after "Fig 4") Pg 3840 line 22 (after "Fig 4a")

• Page 3838, Lines 4-5: However, it is worth mentioning here the findings of the (very)recent paper by Myhre and Samset (2015), which claims that current models tend to underestimate BC forcing.

Agreed.

Added: However, we note the results of Myhre and Samset (2015) which indicate that climate models may underestimate the forcing from BC by around 10%.

• Page 3838, Line 18: I think the authors meant to write "high-latitudes" instead of "highaltitudes".

Yes – corrected.

• Page 3838, Lines 15-18: Was the methodology for generating the different ensemble members in CESM-CAM4 different to that in NorESM?Were the initial conditions in any

way more drastically perturbed in the extra member of CESM-CAM4? Or do the authors believe that this disagreement/agreement between members is a totally random non-linear feature?

In both cases there were two different control runs each with a corresponding perturbation run, and the method of generating the two different control runs was essentially the same (in both cases a different model start dump was used, but everything else was the same). The surface temperature in the control simulations (Fig 3) shows that the CAM4 simulations seem to be generally more noisy/have more variability between years than the NorESM simulations, which could explain why the CAM4 members diverge more than the NorESM members. So yes, we believe it is a random non-linear feature.

Added to pg 3844 line 16:

"It is also interesting to note the very similar behaviour of the two NorESM members compared to the quite different responses between the two CAM4 members. In both cases the two members were generated by initializing with a different atmospheric state but keeping everything else the same. This further emphasizes the importance of using more than model, since some models are more sensitive to small perturbations in the initial conditions than others."

• Page 3839, Lines 5-7: Yes, but what about the widespread warming over Eurasia? This does not seem related to sea ice.

The warming over Europe in HadGEM is discussed separately. However, we appreciate that this is a bit confusing and unclear.

We have rewritten this whole paragraph (pg 3838 line 19 to pg 3839 line 20) to aid clarity, and have take this comment into account.

• Page 3839, Lines 7-10: It is not as clear as what the authors claim. Sea-ice decreases in some parts of the Arctic, but the changes are fairly localised, and there are even areas of increased sea ice.

We agree that this section was not very clearly written.

We have rewritten this whole paragraph (pg 3838 line 19 to pg 3839 line 20) to aid clarity, and have take this comment into account.

• Page 3840, Line 2: Preferably rephrase "model-mean" to "multi-model mean".

Done

• Page 3840, Lines 17-19: It is a bit counterintuitive that precipitation decreases over land, where most of the BC exists, and where most of the de-stabilisation of the atmosphere is expected due to the BC removal. It would be useful here to briefly discuss possible explanations.

Good point. The precipitation changes are driven by circulation changes, rather than local effects of BC.

Added discussion to pg 3840 line 3: "It is interesting to note that over India, where the anthropogenic BC emissions are large, the removal of the BC emissions results in a decrease, rather than an increase, in precipitation. These precipitation changes are driven by circulation changes (e.g. the southward shift in the ITCZ) which dominate over the local effects on precipitation due to BC removal causing destabilization of the atmosphere."

• Page 3841, Line 2: I would add "qualitatively" before "agree", given the very much smaller positive changes in ECHAM, as seen in Fig. 5f.

Done

• Page 3841, Line 15: Please mention that comparisons of the short-term instantaneous or effective forcing are "(not shown)", as otherwise the reader may be misled to think that you are referring to the SW flux comparisons pursued in the manuscript, which I presume is not the case (as the latter are the effect and not the cause of what is discussed here).

Agreed this is not entirely clear.

Amended sentence at end of line 13 to read: "The **TOA SW flux change** from the OC emissions perturbation ..."

• Page 3842, Line 4: Not all is caused by the ITCZ shift. The higher latitude changes probably have to do with the thermodynamic effect of temperature increases.

Yes.

Added to end of line 4:

"Further precipitation increases are seen in the northern hemisphere due to the increased temperature, and in the Indian monsoon region, linked to the reduced aerosol emissions."

• Page 3843, Lines 14-16: I presume the authors mean that there are fluctuations that happen at frequencies lower than 50 years (otherwise the average effect of natural variability would be negligible). This needs to be made clearer here.

Amended lines 14-16:

"...may therefore be driven by **changes in circulation leading to, for example, the change in cloud and snow cover over Europe,** which overwhelm the relatively weak forcing from the BC emissions perturbation."

• Page 3843, Line 17: I would suggest changing "some" to "large".

•

Done

• Page 3843, Line 28: It would perhaps make sense to add a sentence at the end of this paragraph speculating that possibly the expected effect is somewhere in the middle.

Added as suggested:

"The true BC distribution is most likely somewhere in between these model estimates."

• Page 3844, Lines 14-15: I suggest rephrasing to: "where natural variability is a relatively large contributor" (as internal variability is what it is and does not have special features in this study).

Done

• Page 3844, Line 20: The scenarios are idealised, but are they really "extreme", given the drastic decreases expected for the aerosols examined here in the future? It might be worth comparing these reductions with changes between present-day and e.g. 2100 in a typical future emissions scenario, to put things in perspective. Also, it is worth discussing here or in the Discussion section the possible future role of nitrate aerosols, and whether their inclusion in the models could have led to different conclusions or not (both regarding the future role of aerosols in general and regarding the effects of the aerosol types examined here, e.g. sulphate).

Since we are simulating 100% emissions reductions, and these could never realistically be achieved, we believe that these are extreme scenarios, since future emissions could never be less than this (i.e. negative).

Regarding the nitrate discussion, we have added the following to pg 3845 line 2: "We note that the models used in this study do not respresent nitrate chemistry. This means that they may be overestimating the climate responses to removal of SO2 emissions, since reducing SO4 would increase the potential amount of ammonium nitrate aerosol formation, counteracting some of the effects of the reduced SO4 aerosol."

• Page 3844, Line 22: Perhaps add "mainly" before "using".

Done

• Page 3844, Lines 23-24: Suggested rephrasing: ": : : to capture the fast and slow responses due to these emissions perturbations, as well as the uncertainties in these responses."

Done

• Page 3845, Line 10: Suggested rephrasing: ": : : AOGCMs due to responses in ocean temperature and circulation, sea-ice, and atmospheric circulation and cloud responses that are realised on long timescales."

Done

Manuscript prepared for Atmos. Chem. Phys. Discuss. with version 2014/05/30 6.91 Copernicus papers of the LATEX class copernicus.cls. Date: 2 June 2015

Climate responses to anthropogenic emissions of short-lived climate pollutants

L. H. Baker¹, W. J. Collins¹, D. J. L. Olivié², R. Cherian³, Ø. Hodnebrog⁴, G. Myhre⁴, and J. Quaas³

¹Department of Meteorology, University of Reading, P.O. Box 243, Reading, RG6 6BB, UK
 ²Norwegian Meteorological Institute, Oslo, Norway
 ³Institute for Meteorology, University of Leipzig, Leipzig, Germany
 ⁴Center for International Climate and Environmental Research – Oslo (CICERO), Oslo, Norway

Correspondence to: L. H. Baker (l.h.baker@reading.ac.uk)

Abstract

Policies to control air quality focus on mitigating emissions of aerosols and their precursors, and other short-lived climate pollutants (SLCPs). On a local scale, these policies will have beneficial impacts on health and crop yields, by reducing particulate matter (PM) and surface ozone concentrations; however, the climate impacts of reducing emissions of SLCPs are less straightforward to predict. In this paper we consider a set of idealised, extreme mitigation strategies, in which the total anthropogenic emissions of individual SLCP emissions species are removed. This provides an upper bound on the potential climate impacts of such air quality strategies.

We focus on evaluating the climate responses to changes in anthropogenic emissions of aerosol precursor species: black carbon (BC), organic carbon (OC) and sulphur dioxide (SO_2). We perform climate integrations with four fully coupled atmosphere-ocean global climate models (AOGCMs), and examine the effects on global and regional climate of removing the total land-based anthropogenic emissions of each of the three aerosol precursor species.

We find that the SO_2 emissions reductions lead to the strongest response, with all three models showing an increase in surface temperature focussed in the northern hemisphere mid and (especially) high latitudes, and a corresponding increase in global mean precipitationand run-off. Changes in precipitation and run-off patterns are driven mostly by a northward shift in the ITCZ, consistent with the hemispherically asymmetric warming pattern driven by the emissions changes. The BC and OC emissions reductions give a much weaker forcing signal response, and there is some disagreement between models in the sign of the climate responses to these perturbations. These differences between models are due largely to natural variability in sea-ice extent, circulation patterns and cloud changes. This large natural variability component to the signal when the ocean circulation and sea-ice are free-running means that the BC and OC mitigation measures do not necessarily lead to a discernible climate response.

Discussion Paper

Discussion Paper

Introduction 1

Anthropogenic emissions of short-lived climate pollutants (SLCPs), such as aerosols and tropospheric ozone precursors, contribute to poor air quality by increasing particulate matter (PM) and surface ozone concentrations. These are damaging to both human health and agriculture (HTAP; Amann et al., 2013; Tai et al., 2014). Air quality policies therefore aim to reduce emissions of SLCPs. While these policies will have a -beneficial impact on air quality, the climate impacts of reducing emissions of SCLPs SLCPs are less clear.

SLCPs have relatively short atmospheric lifetimes compared with well-mixed greenhouse gases (WMGHGs) such as CO₂, with most remaining in the atmosphere for only days to months. The exception is methane, which has a lifetime of around a decade, but here we focus on the shorter-lived species. The impacts of SLCP emissions on climate therefore occur on relatively short timescales of less than 30 yr (Collins et al., 2013). The short atmospheric lifetime of non-methane SLCPs means that their distribution is not homogeneous as in the case of WMGHGs, and concentrations tend to be highest nearer to source regions. Therefore the resulting forcing patterns are also inhomogeneous, and diagnosing the regional and global climate impacts is much more complex than for WMGHGs (Shindell et al., 2009; Shindell and Faluvegi, 2009). In particular the majority of anthropogenic emissions of SLCPs are in the northern hemisphere, so the forcing is much stronger in the northern hemisphere than the southern hemisphere (Shindell, 2014). The direct effects and the indirect and semi-direct effects of aerosols on clouds aerosol-radiation interactions and aerosol-cloud interactions bring further inhomogeneities, so the resulting impacts of SLCPs on regional and global climate are quite different to those for the WMGHGs.

In this paper we focus on aerosol and aerosol precursor emissions, namely black carbon (BC), organic carbon (OC) and sulphur dioxide (SO_2), which is a -precursor to sulphate (SO_4) aerosol formation.

The effects of anthropogenic aerosols on climate are complex. Scattering aerosols (such as SO₄ and OC) reflect downwelling solar radiation back out of the atmosphere, resulting in a -negative top-of-atmosphere (TOA) short-wave (SW) forcing. This reduction in the solar ra-

face temperature. Hydrophilic aerosols also provide cloud condensation nuclei (CCN), which increases allowing more smaller cloud droplets to form, which increases the cloud albedo and alters other properties including the cloud amount, and prolongs the cloud lifetime by inhibiting precipitation. This further contributes to the negative forcing (Boucher et al., 2013). In contrast, BC aerosol absorbs incoming solar radiation, which means it has a -net warming effect on the atmosphere and gives a -positive TOA SW forcing. The local impact of BC on the surface temperature is dependent on the altitude of the BC: low-level BC can warm the surface by re-emitting radiation in the thermal wavelengths, whereas higher-level BC can reduce the surface temperature by absorbing part of the downwelling solar radiation before it reaches the surface (Ramanathan and Carmichael, 2008). Even in cases where the surface is cooled locally, the additional solar radiation absorbed by the BC results in a -warming effect on the higher atmosphere. BC located near to clouds can cause evaporation of clouds, known as the semidirect effects (Koch and Del Genio, 2010). However, depending on the exact location of the BC and type of cloud, BC can either increase or decrease cloud cover via various different mechanisms (Ban-Weiss et al., 2012), so the net impact on clouds of a -given atmospheric distribution of BC is highly complex. BC aloft causes stabilisation of the atmosphere, which can lead to increased stratocumulous clouds (Koch and Del Genio, 2010). BC also has important impacts at high latitudes when it is deposited on snow, as it decreases the albedo of the snow surface (Ramanathan and Carmichael, 2008), and can enhance snow melt by absorbing solar radiation after it is deposited (Flanner et al., 2007). However, the impacts of BC forcing in the Arctic on surface temperature are complex, as the result is highly dependent on the altitude and location of the forcing (Sand et al., 2013a, b; Flanner, 2013). Aerosols also affect precipitation (e.g. Kristjánsson et al., 2005; Ming et al., 2010;

diation absorbed by the atmosphere climate system results in a -decrease in global mean sur-

Aerosols also affect precipitation (e.g. Kristjánsson et al., 2005; Ming et al., 2010; Boucher et al., 2013; Osborne and Lambert, 2014). On a –global scale, we might expect the precipitation to change in proportion to a <u>given_given_global</u> temperature change driven by aerosol forcing, due to the increased amount of water vapour that the atmosphere can hold (Lambert and Webb, 2008). However, since the direct, semi-direct and indirect effects of aerosols will change precipitation patterns, this does not necessarily hold locally. Hydrophilic

aerosol species can reduce precipitation locally, by enhancing cloud droplet nucleation, which allows more smaller cloud droplets to form but inhibits the amount of droplets that become large enough to form precipitation. Other effects such as convective invigoration that might also affect precipitation (Rosenfeld et al., 2008) are not parameterised in the models assessed here. BC has more complex effects on precipitation patterns since it warms the atmosphere (Andrews et al., 2010) but can either warm or cool the surface, which will increase or reduce the amount of surface evaporation and resulting precipitation (Ming et al., 2010). The net effect on precipitation is therefore dependent on the region and vertical profile of the BC aerosol (Andrews et al., 2010; Ban-Weiss et al., 2012; Kvalevåg et al., 2013). Furthermore the hemispherically asymmetric forcing from anthropogenic aerosol emissions impacts the temperature in the northern hemisphere more than in the southern hemisphere, leading to a -meridional shift in the Intertropical Convergence Zone (ITCZ) towards the warmer hemisphere (e.g. Kang et al., 2008; Ceppi et al., 2013; Hwang et al., 2013), which will impact local precipitation in the tropics and the monsoon regions (Ming and Ramaswamy, 2009). Several studies have shown that anthropogenic aerosol emissions in recent decades have contributed to the weakening of the northern hemisphere monsoon (e.g. Bollasina et al., 2011; Polson et al., 2014). Aerosols also impact the hydrological cycle by reducing the amount of solar radiation reaching the surface, a -process known as solar dimming (Gedney et al., 2014). Solar dimming acts to reduce evaporation, and results in increased run-off and suppressed evapotranspiration.

Policies to reduce anthropogenic aerosol emissions are generally designed to have positive impacts on air quality by reducing PM concentrations; however they can have mixed effects on climate. Reducing SO_2 and OC emissions is expected to have a -detrimental effect on climate in the sense that such measures would be contributing to an increase in global temperature; however the impacts on precipitation patterns could be beneficial, for example by preventing further reduction in monsoon precipitation. In contrast, mitigating BC emissions is expected to reduce global temperature, while the resulting impacts on precipitation are less clear. It is therefore important to evaluate the climate impacts of individual aerosol species in order to evaluate these effects.

Here we assess the climate impacts of removing the total land-based anthropogenic emissions of each of SO_2 , OC and BC in three coupled climate models (four models for the BC experiments) with interactive chemistry and aerosols. The multi-model nature of this work gives greater confidence in the results since we are not drawing conclusions based on results from just one model. The 100%-100% perturbations were used in order to achieve a -strong enough forcing signal. Results from atmosphere-only simulations (e.g. Bellouin et al., 2015) suggest that the removal of anthropogenic SO_2 and OC emissions will lead to a -positive forcing and a -global temperature increase, while removing anthropogenic BC emissions will lead to a -negative forcing and a -global temperature decrease. Using coupled models allows the ocean circulation and heat uptake, and sea-ice extent, to respond to the atmospheric changes from the emissions perturbations. We assess the resulting changes in temperature , circulation patterns, precipitation and run-off and precipitation both globally and regionally.

In Sect. -2, the climate models, experimental setup and emissions datasets are described. In Sect. -3 the climate impacts of removing the emissions of individual anthropogenic aerosol species are presented. These results are discussed further in Sect. -4, and conclusions are given in Sect. -5.

2 Methodology

2.1 Description of models

The three main models used are HadGEM3, ECHAM6-HAM2 and NorESM1-M. HadGEM3 and NorESM1-M have interactive aerosols and chemistry; ECHAM6-HAM2 has interactive aerosols but does not include interactive chemistry. Therefore in HadGEM3 and NorESM1-M, changes in the aerosols can affect the chemistry via changes in oxidation of SO_2 and changing the available surface for heterogeneous chemistry; these processes will directly and indirectly affect O_3 and OH. Photolysis is not affected by the aerosols in these models. The fact that ECHAM6-HAM2 does not include interactive chemistry is expected to lead to only minor differences from the other two models with interactive chemistry with regard to the radiative and climate effects of aerosol and aerosol precursor emissions. For the BC perturbation experiments some additional simulations were performed: one extra ensemble member was run by each of HadGEM3 and NorESM1-M, and two three ensemble members were run by NCAR CESM 1.0.4/CAM4. The extra BC simulations were included in order to explore the BC results further, as this work was part of a larger project of which BC was a key focus.

HadGEM3 is the Hadley Centre Global Environment Model version 3 (Hewitt et al., 2011). The atmosphere component has a -horizontal resolution of $1.875^{\circ} \times 1.25^{\circ}$ and 85 vertical levels extending to 85 km in height (of which 50 are below 18 km). The atmosphere is coupled to the NEMO ocean modelling framework with a -horizontal resolution of 1.0° and 75 vertical levels, and to the CICE sea-ice model (Hunke and Lipscomb, 2008). The UKCA TropIsop scheme is used to model gas-phase chemistry. This treats 55 chemical species (37 of which are transported) including hydrocarbons up to propane, and isoprene and its degradation products (O'Connor et al., 2014). Atmospheric gas and aerosol tracers are advected using the same semi-Lagrangian advection scheme as used for the physical climate variables. Parameterized transport such as boundary layer mixing and convection is also as used for the physical climate variables. Aerosols are modelled by the UKCA GLOMAP-mode aerosol scheme (Mann et al., 2010; Abdul-Razzak and Ghan, 2000). This models the internal mixing of SO₄, OC, BC, dust and sea-salt using a -two-moment modal approach and dynamically evolving particle size distributions. There are seven modes: four soluble (nucleation to coarse) and three insoluble (Aitken to coarse). Aerosol processes are simulated in a -size-resolved manner, including primary emissions, secondary particle formation by binary homogeneous nucleation of sulphuric acid and water, particle growth by coagulation, condensation, and cloud-processing, and removal by dry deposition, in-cloud and below-cloud scavenging. The effects of aerosols on clouds are modelled using an aerosol activation parameterization scheme (Abdul-Razzak and Ghan, 2002). The radiative impact from aerosols is calculated using the Edwards-Slingo radiation scheme (Edwards and Slingo, 1996).

ECHAM6-HAM2 is the European Centre for Medium-Range Weather Forecasts Hamburg model version 6 (Stevens et al., 2013). The atmospheric simulations were made using the ECHAM6 GCM with a -horizontal resolution of T63 (about $\frac{1.8^{\circ} \times 1.8^{\circ}1.8^{\circ} \times 1.8^{\circ}}{1.8^{\circ} \times 1.8^{\circ}}$) and a -ver-

tical resolution of 47 levels (extending from the surface to 0.01 hPa). The atmospheric model is coupled to the Max Planck Institute Global Ocean/Sea-Ice Model (MPIOM) with a -bipolar grid with 1.5° resolution (near the equator) and 40 vertical levels (Jungclaus et al., 2013). The atmospheric model is extended with the Hamburg aerosol model (HAM2) version 2 (Zhang et al., 2012). The main components of HAM are the microphysical module M7, which predicts the evolution of an ensemble of seven internally mixed lognormal aerosol modes (Vignati et al., 2004), an emission module, a –sulfate chemistry scheme (Feichter et al., 1996), a -deposition module, and a -radiative transfer module (Stier et al., 2005) to account for sources, transport, and sinks of aerosols as well as their radiative impact. Five aerosol components, namely SO₄, OC, BC, sea-salt, and mineral dust, are considered in this model. Aerosol effects on liquid-water and ice clouds are considered following Lohmann et al. (2007). Oxidant fields for the sulphate aerosol production were a 2003–2010 average from the MACC reanalysis (Inness et al., 2013).

NorESM1-M is the Norwegian Earth System Model version 1 (Bentsen et al., 2013; Iversen et al., 2013), with horizontal atmospheric resolution of $\frac{1.9 \times 2.5^{\circ}}{1.9^{\circ} \times 2.5^{\circ}}$, and 26 levels in the vertical with a -hybrid sigma pressure coordinate and model top at 2.19 hPa. The ocean module is an updated version of the isopycnic ocean model MICOM (with a -1.1° resolution near the equator and 53 layers), while the sea-ice (CICE4) and land (CLM4) models and the coupler (CPL7) are basically the same as in CCSM4 (Gent et al., 2011). The atmosphere module CAM4-Oslo (Kirkevåg et al., 2013) is a -version of CAM4 (Neale et al., 2011, 2013) with advanced representation of aerosols, aerosol-radiation and aerosol-cloud interactions. It uses the finite volume dynamical core for transport calculations. CAM4-Oslo calculates massconcentrations of aerosol species that are tagged according to production mechanisms in clear and cloudy air and four size-classes (nucleation, aitken, accumulation, and coarse modes). These processes are primary emission, gaseous and aqueous chemistry (cloud processing), nucleation, condensation, and coagulation. Loss terms are dry deposition, in-cloud and below-cloud scavenging. The aerosol components included are SO₄, BC, organic matter (OM), sea-salt, and mineral dust, and are described by 20 tracers. In the model version used in this study, the aerosol module of CAM4-Oslo is coupled with the tropospheric gas-phase chemistry from MOZART (Emmons et al., 2010), which treats around 80 gaseous species. This coupling allows for a -more

explicit description of the formation of secondary aerosol (SO₄ and secondary OM). The radiative forcing from aerosols is calculated using the Collins (2001) radiation scheme. In the fully coupled NorESM1-M, albedo-effects of BC and mineral dust aerosols deposited on snow and sea-ice are also taken into account; this process is not represented in the other three models.

An additional model, NCAR CESM 1.0.4/CAM4, was used for the BC analysis only. NCAR CESM 1.0.4/CAM4 is the National Center for Atmospheric Research Community Earth System Model (Gent et al., 2011) run with the Community Atmosphere Model version 4 (Neale et al., 2011). The atmospheric component is set up here with a -horizontal resolution of $1.9^{\circ} \times 2.5^{\circ}$, and 26 vertical layers (extending from the surface to 2.19 hPa). CAM4 is coupled to a -full ocean model (Danabasoglu et al., 2012), which is based on the Parallel Ocean Program version 2 (Smith et al., 2010), to the CICE4 sea ice model (Hunke and Lipscomb, 2008), and the CLM4 land model (Lawrence et al., 2011). Here, the model has been run without interactive chemistry and aerosols, and instead used prescribed 3-D monthly mean concentrations of ozone and aerosols (BC, OC and SO₄) from the Oslo Chemistry-Transport model version 2 (OsloCTM2) (Søvde et al., 2008; Myhre et al., 2009). OsloCTM2 is driven by meteorological data from the ECMWF-IFS model, and has been run with T42 (approximately $2.8^{\circ} \times 2.8^{\circ}$) horizontal resolution and 60 vertical layers (extending from the surface to 0.1 hPa). In CAM4, the direct and semi-direct aerosol effects of BC are included, while indirect aerosol effects and the effect of BC deposited on snow and ice are not included.

Hereafter we refer to the four models discussed above as HadGEM, ECHAM-HAM, NorESM and CESM-CAM4, respectively.

2.2 Experimental setup and emissions

Each of the three main models (HadGEM, ECHAM-HAM and NorESM) ran a -control simulation and a -set of three perturbation experiments in which the total land-based anthropogenic component of a -single aerosol emission species was removed globally. In addition, HadGEM and NorESM ran a -second control and perturbed BC experiment, and CESM-CAM4 ran two control and two three control and three perturbed BC experiments. The control and perturbed simulations were run for

The control simulations were first run for several decades using an initial ocean state based on present-day CMIP5 conditions for all models except for ECHAM-HAM, which used a pre-industrial control state (see below). The control and perturbed simulations were then run from the same point in this spun-up state for 50 (after an initial spin-up period of several decades)yr, in order to separate a -robust signal from the interannual variability. The climate is not necessarily expected to be stationary after the spin up, but any underlying climate trends are expected to be present in the control and perturbations. By taking the difference between the control and the perturbations we are therefore removing any underlying trends not associated with the changes in aerosol emission. The 50-year integration length was deemed sufficient based on previous studies, e.g. Kristjánsson et al. (2005) performed integrations of length 40 yr after 10 yr of spin-up, and Pausata et al. (2012) showed that most of the temperature response to a -step CO₂ perturbation in AOGCMs is achieved within around the first 10 yr or so (the Cx2 case in their Fig. -1), after which the temperature remains relatively constant, with only a -very gradual continued increase towards the equilibrium response temperature.

We focus on global mean and zonal mean values of the following climate variables: surface temperature, precipitation, and run-offsurface temperature and precipitation. We also examine the top-of-atmosphere (TOA) short-wave (SW) fluxes to aid understanding of these results. This is not the same as the TOA SW forcing in prescribed-SST simulations since in the coupled simulations it includes the fast and slow cloud and sea-ice responses and feedbacksfeedbacks from snow and ice albedo changes and cloud responses to surface temperature, so it is a combination of SW radiative forcing and these feedback changes on the SW flux. It is useful in understanding the causes of changes in climate variables, particularly on regional scales.

The control simulations have present-day anthropogenic emissions of SLCP species from the ECLIPSE emission dataset V4.0a (for the year 2008 (Klimont et al., 2013, 2015), for all models except CESM-CAM4 which used 2000). Biomass ECLIPSE V5.0 emissions for the year 2000. Non-anthropogenic biomass burning emissions are from the GFED3 emissions dataset (http://www.globalfiredata.org) for the year 2005 (in ECHAM-HAM and NorESM) and 2008 (in HadGEM and CESM-CAM4), and are not perturbed. Agricultural biomass burning

emissions are included in the anthropogenic component of emissions which are perturbed. Seasalt and dust aerosol emissions are interactive in HadGEM and ECHAM-HAM; in NorESM, dust emissions are prescribed from a -climatology but sea-salt emissions are interactive; and in CESM-CAM4 both dust and sea-salt emissions concentrations are prescribed from a -climatology. Other natural emissionsare included, , including DMS and volcano emissions, are included and are not perturbed. The concentrations of WMGHGs are also kept fixed at presentday levels in HadGEM, NorESM and CESM-CAM4, and in ECHAM-HAM are fixed at preindustrial (1850) levels. The surface methane concentration is also prescribed at present-day levels in HadGEMand NorESM and _, NorESM and CESM-CAM4, and at pre-industrial levels in ECHAM-HAM. For ECHAM-HAM, the pre-industrial greenhouse gas concentrations were chosen since the model was spun up to equilibrium for this case, and a -new spin-up for increased levels of greenhouse gas concentrations would have been computationally too costly. Since only differences between experiments and control simulations are considered here, no large effect caused by the differences in greenhouse gas concentrations is expected.

Figure –1 shows the emissions of BC, OC and SO₂, divided into the anthropogenic emissions that are perturbed in the experiments (left column) and other emissions that are input to the model (natural, biomass burning and shipping; right column). The strongest anthropogenic emissions of all three species are mostly concentrated over China, India, Europe, the eastern US and parts of Africa and South America.

2.3 Description of the control simulations

Despite all the models having the same emissions input, there is a -large discrepancy between models in the vertical distribution of aerosols in the atmosphere, and in the total aerosol burden, which is typical for current global aerosol models (Textor et al., 2007). HadGEM and ECHAM-HAM have relatively low total burdens of BC, and short atmospheric lifetimes, compared with NorESM and CESM-CAM4 (Table 1). In contrast, NorESM and CESM-CAM4 have relatively low burdens of compared with HadGEM and ECHAM-HAM. The OC burden in NorESM is considerably higher than in the other three models. Figure _Figure 2 shows vertical sections of the annual average, zonal mean BC mass mixing ratio in the control simulation for each

of the models considered. HadGEM and ECHAM-HAM (Fig. -2a and b) have low concentrations of BC at high altitude, which means there is less BC above clouds. In contrast, NorESM and CESM-CAM4 show high BC concentrations extending to above 200 hPa throughout most of the northern hemisphere and southern hemisphere tropics (Fig. -2c and d). This has implications for the impact that removing anthropogenic BC emissions may have. BC at high altitude can have very strong direct effects if it is located above high-albedo cloud surfaces. In the models with higher concentrations of BC at high levels in the control simulations, more of this high-level BC can be removed in the BC perturbation experiment, leading to a -larger change in BC direct forcing. The larger amount of high-level BC in NorESM and CESM-CAM4 (which uses aerosol input from OsloCTM2) is consistent with the AeroCom models discussed by Schwarz et al. (2013) and Samset et al. (2014) who found that these models have too much BC at high altitudes when compared with observations over the Pacific in the HIPPO campaign (Wofsy, 2011), and overestimate the BC lifetime. At lower levels the models underestimate BC concentrations due to the emissions being too low: Hodnebrog et al. (2014) found that increasing emissions of BC and decreasing the BC lifetime in models gave a better agreement with observations. In HadGEM, the lower concentrations of BC at high altitudes and shorter BC lifetimes are likely due to recent modifications to the convective scavenging scheme, which were implemented in order to improve the correspondence with these observations. However, the BC lifetime of 3.4 days is shorter than the AeroCom average. The true BC distribution may therefore lie somewhere in between that of HadGEM and NorESM/CESM-CAM4. The OC burden in NorESM is considerably higher than in the other three models, and its lifetime is correspondingly longer. The range of OC burdens between models is expected due to differences in OA burdens and OA/OC ratios between models (Tsigaridis et al., 2014). NorESM and CESM-CAM4 have relatively low burdens of SO_4 , and short lifetimes, compared with HadGEM and ECHAM-HAM. There are also differences in the vertical distribution of OC and SO_4 between models (not shown) but as these are scattering, rather than absorbing, aerosols the impact of the vertical distribution of the aerosol will have less of an impact on the results. More detailed evaluations of the models used here against observations are given in Eckhardt et al. (2015), Quennehen et al. (2015) and Stohl et al. (2015).

Figure -3 shows the annual average global mean surface temperature in the control simulations for each of the models. ECHAM-HAM has a -lower mean temperature than the other models due to its pre-industrial WMGHG and methane concentrations. CESM-CAM4 has a -higher mean temperature than the others. ECHAM-HAM has a -slight negative drift in surface temperature over the integration period, while both NorESM ensemble members have a -slight positive drift; the other two models remain relatively stable.-, although the second HadGEM member has a decrease in temperature over the first 10 years or so. These drifts in the global mean surface temperature are also present in the perturbation experiments since these start from the control simulations at the beginning of the 50-year period analysed. Therefore we do not expect any drift in the signal, i.e. in the difference between the perturbed and control simulations.

There are some differences between models in the precipitation patterns, particularly in the tropics (Fig. S1). All models suffer from the 'double ITCZ' problem (i.e. there is an overly strong band of precipitation to the south of the equator) which is a known problem in CMIP5 AOGCMs (Li and Xie, 2014). This is most pronounced in ECHAM-HAM (Fig. S1d). ECHAM-HAM and HadGEM also have regions of very low precipitation around the Equator in the Pacific (Figs. S1c and d). There is some variation in the north-eastward extent of the North Atlantic storm track: in NorESM it extends too far north-east, while in CAM4 it does not extend far enough (Figs. S1e and f); in HadGEM and ECHAM-HAM it matches the observations well. All models have too much precipitation over the Himalayas and the Andes, which is probably due to inaccuracies in their representation of precipitation over high orography.

3 Results

In this section we examine the climate responses to perturbing each of the emissions species. The results shown are annual means averaged over the 50-year integration period for each model. Note that since we are interested in the impacts that removing anthropogenic emissions would have, the plots show the perturbation run (i.e. the run with emissions removed) minus the control run. This is different from most other studies, which in general tend to show, e.g.

for example, the forcing of the present-day aerosol compared with a -pre-industrial background state.

3.1 Response to perturbing SO₂ emissions

All three models show an increase in global mean surface temperature as a -result of removing anthropogenic SO₂ emissions: HadGEM and ECHAM-HAM show almost equal temperature increases while NorESM warms by approximately half this value (Fig. <u>4a</u> <u>4a</u> and <u>Table 2</u>). The multi-model mean global mean surface temperature increases by 0.69 K. The zonal mean temperature change is positive at all latitudes, and increases with increasing latitude, with a -multi-model mean, zonal mean temperature increase of around 2.5 K at the North Pole (Fig. <u>-5b</u>). Figure <u>-5a</u> shows warming over almost all areas of the globe, including all land areas. The three models are in agreement on the sign of this temperature response As shown by the stippling, these temperature responses are significant throughout almost all the Northern Hemisphere northern hemisphere land shows warming of at least 1 K, with some northern regions exceeding 2 K.

These temperature responses can be understood further by comparison with the TOA SW flux changes. The global mean TOA SW flux change is positive for all three model simulations (Fig. -4b4c). HadGEM, which has the strongest temperature response, also has the largest change in TOA SW flux, while NorESM, which has the weakest temperature response, has the smallest change in TOA SW flux. The ratio of temperature change to SW flux change is similar between the models $(0.33-0.40 \text{ K} (W \text{ m}^{-2})^{-1}, \text{ Table 2})$. The strongest increase in TOA SW flux change occurs in the Northern Hemisphere northern hemisphere mid-latitudes, where the anthropogenic emissions are largest (Fig. -6b). There is good agreement between the three models in the zonal distribution of TOA SW flux change, although NorESM shows smaller values in the Northern Hemisphere, which may explain the weaker temperature increase in this model compared to the others. There is agreement between the three models in the positive sign of the TOA SW flux change. The multi-model mean changes are significant throughout most of the Northern Hemisphere northern hemisphere (Fig. -6a). There are regions of strong

TOA SW flux change over Europe, the eastern USA and China, which correspond to locations with the largest anthropogenic emissions. Over Europe and the eastern USA, this explains the relatively strong warming in these regions (Fig. -5a). The positive TOA SW flux change over China also extends in a -band over the North Pacific. This is likely to be caused by changes in cloud cover consistent with the decreased aerosol concentrations in this region due to the reduction in aerosol reduced emissions in China, in agreement with the results of . As well as the direct radiative effects, the reduced aerosols would also cause changes in cloud cover. It was shown by Wang et al. (2014) which showed that Chinese aerosols that Chinese aerosol emissions increased cloud cover over the North Pacific. A smilar, so removing these aerosols would reduce cloud cover. A similar region of positive TOA SW flux change also occurs over the North Atlantic, which could similarly be due to aerosol-induced cloud changes is similarly due to aerosol-radiation and aerosol-cloud effects over this region resulting from the aerosol emissions reductions over the eastern USA. The regions of negative TOA SW flux change in the Pacific and Atlantic just north of the equator relate to a -northward shift in the ITCZ, which increases the cloud cover north of the equator and decreases it to the south. This northward ITCZ shift is expected due to the hemispherically asymmetric warming (Rotstayn et al., 2000; Broccoli et al., 2006).

At high northern hemisphere latitudes there are regions of enhanced warming and corresponding increased TOA SW flux (Figs. -5a and 6a), the most pronounced being over the ocean north of Europe. These correspond to regions with large reductions in sea-ice (not shown). All three models agree on a –large loss of Arctic sea-ice, due to the strong northern hemisphere warming. In the southern hemispere, all three models actually show a –region of increased seaice east of the Antarctic Peninsula, which explains the reduced temperatures and decreased TOA SW flux there.

The removal of anthropogenic SO₂ emissions results in an increase in global mean precipitation (Fig. -4e4e). This increase is expected due to the increased surface temperature. The multi-model mean percentage precipitation change per unit warming can be calculated from Table 2 as $2.50\% \text{ K}^{-1} 2.50\% \text{ K}^{-1}$, which is consistent with the value for SO₄ found by $(2.46 \pm 0.11\% \text{ K}^{-1} \text{ Andrews et al.} (2010) (2.46 \pm 0.11\% \text{ K}^{-1})$. While there is a -global increase in precipitation, the southern hemisphere actually shows an overall decrease in precipitation (Fig. -7b). This is mostly due to the northward shift in the ITCZ (discussed above), which can be seen as a -clear dipole in precipitation change about the equator -(Fig. 7b). All three models agree on the northward shift in tropical precipitation over the ITCZ regions and the corresponding pattern of precipitation change is significant in much of the tropics (Fig. -7a). There is a -relatively strong increase in precipitation over India and China, collocated with regions of high anthropogenic emissions of SO₂. There is a -clear increase in precipitation in the Indian monsoon region, which is consistent with the findings that anthropogenic aerosol has caused a -weakening of the summer monsoon (Bollasina et al., 2011; Polson et al., 2014). There is a large increase in precipitation over the Sahel. This is consistent with the results of Rotstayn and Lohmann (2002) who found that present-day anthropogenic sulphate aerosol had contributed to reduced precipitation in the Sahel. There are broad regions over Russia and northern America Canada with increased precipitation corresponding to collocated with regions of increased surface temperature(and therefore. The increased temperature will provide more available moisture through evaporation).

There is an increase in global run-off, which is consistent with the increased global mean surface temperature and precipitation (Fig. 4d). Spatially these changes are strongly linked to the changes in precipitation patterns (Fig. **??**a compared with Fig. 7a). The most coherent changes in run-off occur in the tropics, due to the shift in the ITCZ, notably–. The reduced aerosols in these regions may also cause an increase in run-off over India and a band of increased run-off over the Sahel, both of which are due to collocated increases in precipitation. Over Europe and much of North America much of Europe and the USA there is a decrease in run-off, which is a result of increased surface temperature, increased solar radiation reaching the surface, but decreased precipitation these regions, which will increase evaporation but reduce available moisture reaching the surface. This is consistent with the work of , who attributed increased run-off in heavily polluted parts of Europe to high aerosol concentrations. It is interesting to note that ECHAM-HAM has a much smaller global change in run-off than the other two models, despite having the largest increases in precipitation and temperature (Fig. 4a, c and d). Inspection of spatial maps of run-off for ECHAM-HAM (not shown) shows that this is due

Overall the models agree qualitatively on the climate response to removing anthropogenic SO_2 emissions, showing northern hemisphere warming and a –northward shift in the ITCZ. HadGEM and ECHAM-HAM show very good quantitative agreement in the response.

3.2 Response to perturbing black carbon emissions

For the BC perturbation experiments, we consider, in addition to the original simulations from HadGEM, ECHAM-HAM and NorESM, one extra ensemble member from each of HadGEM and NorESM, and two-three ensemble members from CESM-CAM4. For the calculations of multi-model means, each of these additional members is weighted equally with the other modelsimulations we take the mean of the mean values for each model.

The temperature response to removing anthropogenic BC emissions is much smaller overall than the response to perturbing SO₂ emissions (Fig. <u>44 and Table 2</u>). All the models except HadGEM show a -net decrease in global mean surface temperature <u>, although CESM-CAM4</u> member 2 shows only a very small decrease (Fig. 4a). This results in a -small negative multi-model mean value for the global surface temperature response. A However, we note the results of Myhre and Samset (2015) which indicate that climate models may underestimate the forcing from BC by around 10%. Figure 4b shows the temperature response in the individual model members. This shows that HadGEM member 1 has a significant increase in global mean surface temperature, while the other simulations all show a decrease, although the sign of this response is uncertain in the cases of HadGEM member 2 and CESM-CAM4 member 2.

A similar pattern is seen for the change in TOA SW flux (Fig. -4b), although it is interesting to note that CESM-CAM4 member 2 has a relatively strong negative TOA 4c). For the majority of the simulations the ratio of temperature change to SW flux change compared to its very small temperature response between 0.21 and 0.28 K (W m⁻²)⁻¹ (Table 2) which is smaller than for

SO₂; however the HadGEM member 1 and CESM-CAM4 member 2 simulations are outliers with ratios of 0.78 and 0.03 K (W m⁻²)⁻¹ respectively.

The multi-model mean temperature response is within ± 0.5 K everywhere (Fig. -5c). There are stippled regions (where at least five of the six ensemble members agree on the sign) These temperature changes are significant in large parts of the southern hemisphere ocean and the tropical Pacific , but much less stippling but less so in the northern hemisphere. The TOA SW flux change is also relatively small everywhere, with the strongest TOA SW flux decrease over northern India. (Fig. -6c). There are stippled regions over areas However, the changes in TOA SW flux are significant over large areas of land in the northern hemisphere, in general over areas with high anthropogenic BC emissions, with the strongest TOA SW flux decrease over northern India.

The small multi-model mean temperature and TOA SW flux responses are the result of conflicting regional responses in the different models, rather than weak responses in each model. This can be seen in Fig. -5d, which shows the range of zonal mean temperature responses between models. Both NorESM members show NorESM shows relatively strong cooling, which is stronger towards high latitudes, reaching around -0.4-0.4 K at the north pole. In contrast, HadGEM shows warming of a similar magnitude, again increasing towards high latitudes and in the northern hemisphere, but to different degrees in the two ensemble members: in member 1 the temperature increases towards the north pole, reaching 0.4 at the north K; in contrast member 2 shows only slight warming, and a decrease in temperature at the pole. ECHAM-HAM shows a -weak response in general but a -small increase towards the north pole. The two-three CESM-CAM4 members show different behaviour: member 1 shows all three show weak cooling at most latitudes, peaking at but north of around $60^{\circ}N$ and $70^{\circ}S$; member 2 shows warmingincreasing throughout the northern hemisphere mid- to high-altitudes and reaching, which increases towards the pole and reaches 0.6 at the north pole K. The zonal mean TOA SW flux change also shows large differences between models (Fig. 6d), which helps to explain the range of temperature responses in each model in the northern hemisphere.

The spatial responses in each of the model simulations can be seen in Figs. S1–S3 in the Supplement. HadGEM supplementary Figures S2–S6, and can explain some of the differences

between models discussed above. HadGEM member 1 shows warming in the Arctic and over most of the northern hemisphere mid-latitudes, including Europe, which is unexpected since anthropogenic BC emissions are relatively large there (Fig. <u>S1a</u>). CESM-CAM4 member 2 S2a). This is due to increased TOA SW flux over Europe (Fig. <u>S3b</u>)also shows warming over the Arctic, but not over the rest of the northern hemisphere. In contrast, both NorESM members S2c). Inspection of cloud and snow cover fields (not shown) shows that this is in fact a result of a combination of reduced cloud cover and reduced snow cover over Northern Europe; these changes are likely due to circulation changes, and their combined effect is enough to more than balance the negative forcing from local removal of BC. The warming in the Arctic is linked to decreases in sea-ice (Fig. <u>S2a and b</u>) show robust cooling over these regions, while ECHAM-HAM-S2e) and collocated increases in TOA SW flux (Fig. <u>S1a</u>)shows cooling over some areas of the mid-latitudes but S2c). HadGEM member 2 shows warming over much of the Arctic. CESM-CAM4 member 1 shows weak cooling over some mid- and high-latitude regions Russia but cooling over North America (Fig. <u>S3a</u>).-

The zonal mean S2b). There is also strong warming along the south-eastern edge of Greenland and in the Barents Sea, which is linked to increased TOA SW flux change also shows large differences between models (Fig. S2d) and large decreases in sea-ice (Fig. -6d), which helps to explain the range of temperature responses in each model in the northern hemisphere. The three simulations that show S2f). Both HadGEM members show strong decreases in TOA SW flux over India due to the emissions reductions, but these do not translate to strong temperature decreases. ECHAM-HAM also shows some localised warming in the northern hemisphere high latitudes (HadGEM, ECHAM-HAM and CESM-CAM4 member 2) all show positive TOA SW flux changes in the northern hemisphere, peaking between around 60 and 70°N-Arctic, but cooling in much of the rest of the northern hemisphere (Fig. -6d). These high-latitude regions of S3a), although most of this is not significant. As in HadGEM, the regions of Arctic warming are collocated with increased TOA SW flux can be seen in Figs. S1c and d, and S3d. Comparison with the respective (Fig. S3b) and decreased sea-ice changes (Figs. S1e and f, and S3f)shows a strong correlation between increased (Fig. S3c). There are regions with decreased TOA SW flux and decreased sea-ice. over India, China and the Eastern

USA, which correspond to large reductions in BC emissions (Fig. S3b). In contrast, for both the NorESM members, which both NorESM members show cooling in the northern hemisphere, there is a clear decrease in Arctic and significant cooling over much of the globe (Figs. S4a and b). In both members this corresponds to decreased TOA SW flux over much of the Arctic and most of the northern hemisphere high-latitudes hemispere land area (Figs. S2c S4c and d), and collocated. There are regions with increases in sea-ice, such as in the Barents Sea in Fig. S4f, but also small regions where the sea-ice decreases, although these decreases are generally not significant (Figs. S2e S4e and f). The three CESM-CAM4 members show different temperature responses in the Arctic: member 1 shows only relatively small changes in TOA SW fluxes very little temperature response in the Arctic (Fig. S3c), and smaller changes in S5a), while member 2 shows significant warming over much of the Arctic (Fig. S5b); member 3 shows cooling of a similar magnitude over the Arctic (Fig. S6a). Corresponding to the warmer Arctic temperatures in member 2, there are also widespread decreases in Arctic sea-ice (Fig. -S3e). All the models show decreased TOA SW flux over India and China, consistent with the location of the strongest BC emissions reductions (middle panels of Figs. S1-S3). All models except HadGEM also show a decrease in TOA SW flux over Europe, which is expected since the anthropogenic emissions of BC are relatively strong here. The region of positive S5f), while member 1 shows more mixed sea-ice changes (Fig. S5e) and member 3 shows some regions with increase sea-ice (Fig. S6c). Member 1 shows significant cooling in much of the southern hemisphere ocean, while members 2 and 3 show only weak temperature responses. Both members show significant cooling in the North Pacific, linked to the reduction in aerosol emissions from China. Compared to members 2 and 3, member 1 shows stronger decreases in TOA SW flux change over Europe in HadGEM is in fact a result of a combination of reduced cloud cover and reduced snow cover over Northern Europe (not shown); these changes are likely due to circulation changes, and their combined effect is enough to more than balance the negative forcing from local removal of BC over China and Europe in response to the emissions reductions, which could explain the stronger overall temperature reduction in member 1 (Figs. S5c, 5d and 6b).

The global mean precipitation response to removing anthropogenic BC emissions is relatively small (Fig. 4cFigs. 4e and f). Despite the different signs of temperature response, the global

precipitation increases in all the modelsmodel simulations. This is not surprising since the removal of BC from the atmosphere will lead to a -negative atmospheric forcing, which in turn is expected to lead to increased precipitation (Andrews et al., 2010). Both NorESM members show a <u>NorESM shows a</u> pronounced southward shift in the position of the ITCZ, which is consistent with the cooling in the northern hemisphere in these simulations (Fig. -7d). HadGEM shows a member 1 shows a weak northward shift in the ITCZ, while the other models model simulations do not show a -coherent shift in its position. The opposing direction of the ITCZ shift in HadGEM member 1 and NorESM partly explains why there are so few regions where all the models agree on the sign of precipitation change, and the the model-mean responses are generally relatively weak everywhere (Fig. -7c).

There is a decrease in the multi-model mean global run-off response (Fig. 4d). All models except HadGEM show a decrease in run-off, while HadGEM shows a small increase. However, in all models there is large interannual variability in these values, so there is considerable uncertainty in these values. It is interesting to note that this decrease in run-off occurs despite a global increase over India, where the anthropogenic BC emissions are large, the removal of the BC emissions results in a decrease, rather than an increase, in precipitation. However, the increase in precipitation occurs mostly over the ocean; regions of reduced run-off, which are mostly in the tropics and mid-latitudes, correspond to regions with reduced precipitation over land (Figs. ??b and 7c). These precipitation changes are driven by circulation changes (e.g. the southward shift in the ITCZ) which dominate over the local effects on precipitation due to BC removal causing destabilization of the atmosphere.

Overall, the climate response to removing anthropogenic BC emissions is weaker than the response to removing SO_2 emissions. Although there is a -mean global temperature decrease, there is a -large variation between models in the temperature response, particularly in the northern hemisphere high latitudes. All models agree on an increase in precipitation globally, although there is some variation between models in the patterns of precipitation response. There is an overall decrease in run-off, which is due to a decrease in precipitation over land, despite an increase in precipitation globally.

Discussion Paper

3.3 Response to perturbing organic carbon emissions

The multi-model mean response to removing anthropogenic OC emissions is an increase in global mean surface temperature (Fig. -4a 4a and Table 2). HadGEM and NorESM show a -clear increase in surface temperature, with the largest response in HadGEM; ECHAM-HAM shows a -weak reduction in global mean surface temperature, although the error bars indicate some uncertainty in the sign of this response. HadGEM and NorESM show an increase in the zonal mean surface temperature throughout the northern hemisphere, increasing towards the pole; ECHAM-HAM shows almost no change in the zonal mean surface temperature (Fig. -5f). Despite the different behaviour in ECHAM-HAM compared with the other models, there are broad areas where all three models agree on an increase in surface temperature , including much of the northern hemisphere mid-latitudes and some regions further north in the northern hemisphere where the temperature changes are significant, including over much of Europe (Fig. -5e).

The TOA SW flux change is weakly positive over most of the northern hemisphere, with only a few regions where all three models agree on the sign of the change and is mostly not significant (Fig. -6e). HadGEM and NorESM show an increase in zonal mean TOA SW flux over the northern hemisphere (Fig. -6f), and in particular show increased TOA SW flux over the mid-latitudes, which have the largest anthropogenic OC emissions (Fig. -1e). In contrast, ECHAM-HAM shows a -decrease in TOA SW flux over the northern hemisphere mid-latitudes (Fig. -6f). Inspection of spatial maps (not shown) indicate that this is due to decreased SW flux over Europe and the eastern USA, despite the reduced OC emissions in these regions. This may be due to natural variability in cloud cover over these regions driven by changes in atmospheric circulation patterns. The forcing signal TOA SW flux change from the OC emissions perturbation seems to be much weaker in ECHAM-HAM than in the other models, so natural variability may dominate.

The global mean precipitation changes in each model are consistent with their respective temperature responses: HadGEM and NorESM show an increase in global precipitation, while ECHAM-HAM shows a -decrease (Fig. -4e4e). Despite the variation in temperature responses,

all three models show a –northward shift in the ITCZ (Fig. –7f). The changes in precipitation patterns are similar to those for the SO_2 experiments but with weaker magnitude (compare Fig. –Figs. 7c and 7e). Run-off changes are generally small, and are driven by the change in precipitation patterns, particularly the ITCZ shift (Fig. ??c).

Overall the response to removal of anthropogenic OC emissions is an increase in surface temperature and precipitation, primarily in the northern hemisphere. The spatial patterns of changes in these quantities are broadly similar to those for the SO_2 emissions perturbation, but with smaller magnitude.

4 Discussion

The three models are in good agreement about the impacts of removing anthropogenic SO_2 emissions, all showing a -warming concentrated in the northern hemisphere and a -northward shift in the ITCZ, bringing more precipitation to the northern hemisphere. Further precipitation increases are seen in the northern hemisphere due to the increased temperature. NorESM gives a -weaker overall response than the other two models. This is not surprising since NorESM has a lower SO_4 burden than the other models, so the SO_2 emissions changes will have less impact. Furthermore, NorESM is known to have a -relatively low climate sensitivity (Andrews et al., 2012), which Iversen et al. (2013) attribute to a -strong Atlantic Meridional Overturning Circulation in NorESM. This may explain the smaller changes in Arctic sea-ice extent changes in NorESM than in the other two models in the SO_2 experiment, reducing the impact of the additional positive feedback on temperature of the melting ice.

The response to removing anthropogenic OC emissions is similar to the that for removing SO_2 , but much weaker overall. in terms of temperature change per unit SW flux change (Table 2). The absolute magnitude of the response is about 5 times smaller for OC. ECHAM-HAM appears to have a -weaker response to the removal of OC than the other models, and this is within the range of natural variability between individual years. The other models show similar patterns of response to those in the SO₂ experiment, but with weaker magnitude.

In contrast, there are differences between models in their response to removing anthropogenic BC emissions: both NorESM members show a NorESM shows a clear cooling, particularly in the Northern Hemisphere; ECHAM-HAM shows an overall cooling but some warming in the Arctic; HadGEM shows an overall warming, which is most pronounced in the northern hemisphere; and the two CAM4 members show an overall cooling but very different temperature responses in the Arcticnorthern hemisphere; the other models show weaker responses, and HadGEM member 1 actually shows a global mean warming, with the largest temperature increases in the northern hemisphere high latitudes. The stronger effects of BC removal in NorESM compared with the other models may be due in part to the fact that this model includes representation of the albedo effect of BC deposition on snow. As shown by Sand et al. (2013b), this has a relatively large impact on surface temperature in the Arctic. This provides a -mechanism to explain the stronger cooling over the Arctic in the BC experiments in this model than in the other models. When the BC emissions are reduced, less BC would be deposited on snow at high latitudes, leading to higher-albedo snow. This hypothesis is supported by the decrease in TOA SW flux over the Arctic in both NorESM members, which is consistent with an increased surface albedo, while the other models show mostly positive TOA SW flux change here. However, we note that the variability is large at high northern latitudes as shown by the variation between models and between the two HadGEM members and the three CESM-CAM4 ensemble members. Furthermore, NorESM has a -high BC abundance at mid- and high-latitudes as shown in Fig. -2. The different, and somewhat surprising, climate responses to the BC perturbations in HadGEM the two HadGEM members, and the weaker responses in ECHAM-HAM, may be due to the fact that HadGEM has these models have smaller amounts of BC at high altitudes in the control run than NorESM and CAM4. ECHAM-HAM also has lower amounts of high-level BC, and has a weak temperature and TOA SW flux response compared to NorESM and CAM4. CESM-CAM4. The lack of high-level BC is important since the strongest direct effects of BC are from BC above clouds or other high albedo surfaces, so these effects will be much weaker in the control simulation in HadGEM and ECHAM-HAM-ECHAM than in the other models. Removal of anthropogenic BC emissions will therefore have a -smaller impact in the models with less high-level BC since the BC

forcing in the control simulation is weak to begin with. The climate responses in HadGEM may therefore be driven by natural variability (changes in circulation, leading to, for example, the change in cloud and snow cover over Europe), which overwhelms in HadGEM member 1. These circulation changes overwhelm the relatively weak forcing from the BC emissions perturbation. Apart from the HadGEM simulation the models suggest a lower ratio of temperature change to SW flux change for BC than for OC and SO₂.

The results from this study show that there is some-large uncertainty as to the climate response to removing anthropogenic BC emissions. The different behaviour between models is due partly to the different atmospheric BC distributions in the models, as shown in Fig. 1. Accurately representing the correct BC distribution in GCMs is very difficult .- For example, recent (Samset et al., 2014). Recent studies (e.g. Schwarz et al., 2013; Hodnebrog et al., 2014; Samset et al., 2014) have compared BC distributions in GCMs and CTMs with data from observational studies such as the HIPPO campaign (Wofsy, 2011), which provided observations from a large spatial area over the Pacific. They found that the models had too much BC at high altitudes in these regions, and that the BC lifetime was generally too long. Recent modifications to the convective scavenging scheme in HadGEM (which are included in the model set-up used here) were designed to reduce the amount of high-level BC, which was previously too large, and this model setup gives good agreement with data from the HIPPO field campaigns in the Pacific ; however, in other areas the results compare less well with observations, and the amount of high-level BC is probably too low in generalto give better agreement with the HIPPO observations. The result of these changes is that HadGEM has less BC at high levels globally than the other models (except ECHAM-HAM), and a much shorter BC and OC lifetime (Table 1). ECHAM-HAM also has less BC at high levels, and a short BC lifetime. In contrast, NorESM and CAM4 probably have too much CESM-CAM4 have much more high-level BC compared to the HIPPO campaign observations, which overestimates and longer BC lifetimes, which may overestimate the direct forcing from anthropogenic BC, and therefore exaggerates (consistent with Samset et al. (2014)) and may therefore exaggerate the impact of removing anthropogenic BC emissions. The true BC distribution at high levels is most likely somewhere in

between these model estimates, while at lower levels the emissions are likely underestimated (Hodnebrog et al., 2014).

A -further feature influencing the results in this study is the contribution of changes in sea-ice extent. Particularly for the OC and BC emissions perturbations, which give a -weaker forcing than the SO_2 emissions perturbations, these sea-ice changes appear to be due to natural variability, rather than a -forced response. However, they do contribute a reasonable amount to the total SW flux changes and surface temperature changes. This adds an extra element of natural variability that is not an issue in atmosphere-only simulations, which have fixed SSTs and prescribed sea-ice. This motivated our decision to perform three additional simulations, in order to increase our sample size. It can be seen from these simulations that the sea-ice responds quite differently to the BC perturbation in different simulations, even in two simulations from the same model.

It is interesting to note the range of climate responses between models, and even between different simulations run by the same model. This highlights the importance of using an ensemble of simulations in studies such as this, where natural variability is relatively large a relatively large contributor, and differences in the formulation of individual models can have a -large impact on the results. It is also interesting to note the very similar behaviour of the two NorESM members compared to the quite different responses between the individual members in the other models. In all cases the different members were generated by initializing with a different atmospheric state but keeping everything else the same. This further emphasizes the importance of using more than one model, since different models have different sensitivity to small perturbations in the initial conditions.

5 Conclusions

Air quality policies now and in the future will lead to reduced emissions of aerosols and other SLCPs. This study aims to evaluate the possible climate impacts of these emissions reductions, by considering a -set of extreme idealised scenarios in which 100% of the land-based anthropogenic emissions of individual aerosol precursor species (BC, OC and SO₂) are removed. The

experiments were performed <u>mainly</u> using three AOGCMs with interactive aerosols and chemistry, in order to capture the fast and slow responses to these emissions perturbations <u>as well</u> as the <u>uncertainties in these responses</u>. We also included additional simulations from another AOGCM (without interactive aerosols) for the BC experiments.

The results show strong impacts on climate of removing SO_2 emissions, with an increase in global mean surface temperature, focussed mainly in the northern hemisphere, and a -northward shift in the ITCZ, driving changes in precipitation and run-off patterns, patterns particularly in tropical regions. We note that the models used in this study do not respresent nitrate chemistry. This means that they may be overestimating the climate responses to removal of SO_2 emissions, since reducing SO_4 would increase the potential amount of ammonium nitrate aerosol formation, counteracting some of the effects of the reduced SO_4 aerosol (West et al., 1999; Bellouin et al., 2011).

The OC and BC emissions perturbations produced a much weaker signal much weaker climate responses. In both cases the models were not all in agreement on the sign of the global mean TOA SW flux change or surface temperature response. These results are different from those obtained in other studies using prescribed-SST, atmosphere-only simulations (e.g.)(e.g. Bellouin et al., 2015), where the forcing response to such emissions perturbations is more likely to have the same sign in all models, since. This is because the design of these such experiments removes much of the variability that we see in fully-coupled AOGCMs in-due to responses in temperature and in ocean circulation, sea-ice, atmospheric circulation changes and slow cloud responses and cloud responses that are realised on long timescales. Overall the removal of OC emissions leads to similar patterns of response to the SO_2 experiments, but with much weaker magnitude. There is a -weak northward shift in the ITCZ, and corresponding changes in run-offprecipitation. The BC response is more complex, and due to the large disagreement in response between two of the models, we included three-five additional ensemble members. Even between two ensemble members from the same model there are large differences in the surface temperature and precipitation responses. From this study we conclude that, while BC mitigation is unlikely to be detrimental to climate -(like in the case of SO₂ and OC mitigation), the climate benefits are likely to be very small, and may not be discernable above natural variability in the climate.

The Supplement related to this article is available online at doi:10.5194/acpd-0-1-2015-supplement.

Model simulations were performed by L. H. Baker (HadGEM), D. J. L.Olivié (NorESM), R. Cherian (ECHAM-HAM) and Ø. Hodnebrog (CESM-CAM4). Analysis of the results was performed by L. H. Baker with contributions from D. J. L.Olivié. L. H. Baker prepared the manuscript with contributions from all co-authors.

Acknowledgements. The research leading to these results has received funding from the European Union Seventh Framework Programme (FP7/2007-2013) under grant agreement no 282688 – ECLIPSE. The ECHAM-HAM model is developed by a -consortium composed of ETH Zurich, Max Planck Institut fr-für Meteorologie, Forschungszentrum JlichJülich, University of Oxford, the Finnish Meteorological Institute and the Leibniz Institute for Tropospheric Research, and managed by the Center for Climate Systems Modeling (C2SM) at ETH Zurich. R. Cherian and J. Quaas acknowledge the computing time provided by the German High Performance Computing Centre for Climate and Earth System Research (Deutsches Klimarechenzentrum, DKRZ). L. Baker and W. Collins acknowledge the-use of the MON-SooN supercomputing facility for running the HadGEM3 simulations. system, a collaborative facility supplied under the Joint Weather and Climate Research Programme, which is a strategic partnership between the Met Office and the Natural Environment Research Council. Ø. Hodnebrog and G. Myhre acknowledge additional funding from the Research Council of Norway through the SLAC project.

References

Abdul-Razzak, H. and Ghan, S. J.: A parameterization of aerosol activation: 2. Multiple aerosol types, J. Geophys. Res.-Atmos., 105, 6837–6844, 2000.

- Abdul-Razzak, H. and Ghan, S. J.: A parameterization of aerosol activation 3. Sectional representation, J. Geophys. Res.-Atmos., 107, doi:10.1029/2001JD000483, 2002.
- Amann, M., Klimont, Z., and Wagner, F.: Regional and Global Emissions of Air Pollutants: Recent Trends and Future Scenarios, Annual Review of Environment and Resources, 38, 31–55, 2013.
- Andrews, T., Forster, P. M., Boucher, O., Bellouin, N., and Jones, A.: Precipitation, radiative forcing and global temperature change, Geophys. Res. Lett., 37, L14701, doi:10.1029/2010GL043991, 2010.
- Andrews, T., Gregory, J. M., Webb, M. J., and Taylor, K. E.: Forcing, feedbacks and climate sensitivity in CMIP5 coupled atmosphere-ocean climate models, Geophys. Res. Lett., 39, L09712, doi:10.1029/2012GL051607, 2012.
- Ban-Weiss, G. A., Cao, L., Bala, G., and Caldeira, K.: Dependence of climate forcing and response on the altitude of black carbon aerosols, Clim. Dynam., 38, 897–911, 2012.
- Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J., and Boucher, O.: Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, J. Geophys. Res., 116, 2011.
- Bellouin, N., Baker, L., Cherian, R., Hodnebrog, O., Olivié, D., Samset, B., MacIntosh, C., Esteve-Martinez, A., and Myhre, G.: Regional and seasonal radiative forcing by perturbations to aerosol and ozone precursor emissions, in preparation, 2015.
- Bentsen, M., Bethke, I., Debernard, J. B., Iversen, T., Kirkevåg, A., Seland, Ø., Drange, H., Roelandt, C., Seierstad, I. A., Hoose, C., and Kristjánsson, J. E.: The Norwegian Earth System Model, NorESM1-M Part 1: Description and basic evaluation of the physical climate, Geosci. Model Dev., 6, 687–720, doi:10.5194/gmd-6-687-2013, 2013.
- Bollasina, M. A., Ming, Y., and Ramaswamy, V.: Anthropogenic aerosols and the weakening of the South Asian summer monsoon, Science, 334, 502–505, 2011.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X.-Y.: Clouds and aerosols, in: Climate Change 2013: The Physical Science Basis, Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change, 571–657, Cambridge University Press, Cambridge, UK and New York, NY, USA, 2013.
- Broccoli, A., Dahl, K., and Stouffer, R.: Response of the ITCZ to Northern Hemisphere cooling, Geophys. Res. Lett., 33, doi:10.1029/2005GL024546, 2006.
- Ceppi, P., Hwang, Y.-T., Liu, X., Frierson, D. M., and Hartmann, D. L.: The relationship between the ITCZ and the Southern Hemispheric eddy-driven jet, J. Geophys. Res.-Atmos., 118, 5136–5146, 2013.

- Collins, W. J., Fry, M. M., Yu, H., Fuglestvedt, J. S., Shindell, D. T., and West, J. J.: Global and regional temperature-change potentials for near-term climate forcers, Atmos. Chem. Phys., 13, 2471–2485, doi:10.5194/acp-13-2471-2013, 2013.
- Collins, W. D.: Parameterization of generalized cloud overlap for radiative calculations in general circulation models, J. Atmos. Sci., 58, 3224–3242, 2001.
- Danabasoglu, G., Bates, S. C., Briegleb, B. P., Jayne, S. R., Jochum, M., Large, W. G., Peacock, S., and Yeager, S. G.: The CCSM4 ocean component, J. Climate, 25, 1361–1389, 2012.
- Eckhardt, S., Quennehen, B., Olivié, D. J. L., Berntsen, T. K., Cherian, R., Christensen, J. H., Collins, W., Crepinsek, S., Daskalakis, N., Flanner, M., Herber, A., Heyes, C., Hodnebrog, Ø., Huang, L., Kanakidou, M., Klimont, Z., Langner, J., Law, K. S., Massling, A., Myriokefalitakis, S., Nielsen, I. E., Nøjgaard, J. K., Quaas, J., Quinn, P. K., Raut, J.-C., Rumbold, S. T., Schulz, M., Skeie, R. B., Skov, H., Lund, M. T., Uttal, T., von Salzen, K., Mahmood, R., and Stohl, A.: Current model capabilities for simulating black carbon and sulfate concentrations in the Arctic atmosphere: a multi-model evaluation using a comprehensive measurement data set, Atmos. Chem. Phys. Discuss., 15, 10425–10477, doi:10.5194/acpd-15-10425-2015, http://www.atmos-chem-phys-discuss.net/15/10425/2015/, 2015.
- Edwards, J. and Slingo, A.: Studies with a flexible new radiation code. I: Choosing a configuration for a large-scale model, Q. J. Roy. Meteor. Soc., 122, 689–719, 1996.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.
- Feichter, J., Kjellström, E., Rodhe, H., Dentener, F., Lelieveldi, J., and Roelofs, G.-J.: Simulation of the tropospheric sulfur cycle in a global climate model, Atmos. Environ., 30, 1693–1707, 1996.
- Flanner, M. G.: Arctic climate sensitivity to local black carbon, J. Geophys. Res.-Atmos., 118, 1840–1851, 2013.
- Flanner, M. G., Zender, C. S., Randerson, J. T., and Rasch, P. J.: Present-day climate forcing and response from black carbon in snow, J. Geophys. Res.-Atmos., 112, D11202, doi:10.1029/2006JD008003, 2007.
- Gedney, N., Huntingford, C., Weedon, G., Bellouin, N., Boucher, O., and Cox, P.: Detection of solar dimming and brightening effects on Northern Hemisphere river flow, Nat. Geosci., 7, 796–800, 2014.
- Gent, P. R., Danabasoglu, G., Donner, L. J., Holland, M. M., Hunke, E. C., Jayne, S. R., Lawrence, D. M., Neale, R. B., Rasch, P. J., Vertenstein, M., Worley, P. H., Yang, Z. L., and Zhang, M. H.: The community climate system model version 4, J. Climate, 24, 4973–4991, 2011.

- Hewitt, H. T., Copsey, D., Culverwell, I. D., Harris, C. M., Hill, R. S. R., Keen, A. B., McLaren, A. J., and Hunke, E. C.: Design and implementation of the infrastructure of HadGEM3: the next-generation Met Office climate modelling system, Geosci. Model Dev., 4, 223–253, doi:10.5194/gmd-4-223-2011, 2011.
- Hwang, Y. -T., Frierson, D. M. W., and Kang, S. M.: Anthropogenic sulfate aerosol and the southward shift of tropical precipitation in the late 20th century, Geophys. Res. Lett., 40, 2845-2850, 2013.
- Hodnebrog, Ø., Myhre, G., and Samset, B. H.: How shorter black carbon lifetime alters its climate effect, Nat. Comm., 5, 5065, 2014.
- HTAP: Hemispheric Transport of Air Pollution 2010 Part A: Ozone and Particulate Matter, Air Pollution Studies No. 17, edited by: Dentener, F., Keating, T., and Akimoto, H., United Nations, New York and Geneva, 2010.
- Hunke, E. C. and Lipscomb, W. H.: CICE: the Los Alamos Sea Ice Model Documentation and Software User's Manual Version 4.0 LA-CC-06-012, 2008.
- Inness, A., Baier, F., Benedetti, A., Bouarar, I., Chabrillat, S., Clark, H., Clerbaux, C., Coheur, P., Engelen, R. J., Errera, Q., Flemming, J., George, M., Granier, C., Hadji-Lazaro, J., Huijnen, V., Hurtmans, D., Jones, L., Kaiser, J. W., Kapsomenakis, J., Lefever, K., Leitão, J., Razinger, M., Richter, A., Schultz, M. G., Simmons, A. J., Suttie, M., Stein, O., Thépaut, J.-N., Thouret, V., Vrekoussis, M., Zerefos, C., and the MACC team.: The MACC reanalysis: an 8 yr data set of atmospheric composition, Atmos. Chem. Phys., 13, 4073–4109, 2013.
- Iversen, T., Bentsen, M., Bethke, I., Debernard, J. B., Kirkevåg, A., Seland, Ø., Drange, H., Kristjansson, J. E., Medhaug, I., Sand, M., and Seierstad, I. A.: The Norwegian Earth System Model, NorESM1-M – Part 2: Climate response and scenario projections, Geosci. Model Dev., 6, 389–415, doi:10.5194/gmd-6-389-2013, 2013.
- Jungclaus, J., Fischer, N., Haak, H., Lohmann, K., Marotzke, J., Matei, D., Mikolajewicz, U., Notz, D., and Storch, J.: Characteristics of the ocean simulations in the Max Planck Institute Ocean Model (MPIOM) the ocean component of the MPI-Earth system model, Journal of Advances in Modeling Earth Systems, 5, 422–446, 2013.
- Kang, S. M., Held, I. M., Frierson, D. M., and Zhao, M.: The response of the ITCZ to extratropical thermal forcing: idealized slab-ocean experiments with a GCM, J. Climate, 21, 3521–3532, 2008.
- Kirkevåg, A., Iversen, T., Seland, Ø., Hoose, C., Kristjánsson, J. E., Struthers, H., Ekman, A. M. L., Ghan, S., Griesfeller, J., Nilsson, E. D., and Schulz, M.: Aerosol–climate interactions in the Norwegian Earth System Model – NorESM1-M, Geosci. Model Dev., 6, 207–244, doi:10.5194/gmd-6-207-2013, 2013.

- Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., and Schoepp, W.: Global anthropogenic emissions of particulate matter, in preparation, 2015.
- Koch, D. and Del Genio, A. D.: Black carbon semi-direct effects on cloud cover: review and synthesis, Atmos. Chem. Phys., 10, 7685–7696, doi:10.5194/acp-10-7685-2010, 2010.
- Kristjánsson, J., Iversen, T., Kirkevåg, A., Seland, Ø., and Debernard, J.: Response of the climate system to aerosol direct and indirect forcing: role of cloud feedbacks, J. Geophys. Res.-Atmos., 110, D24206, doi:10.1029/2005JD006299, 2005.
- Kvalevåg, M. M., Samset, B. H., and Myhre, G.: Hydrological sensitivity to greenhouse gases and aerosols in a global climate model, Geophys. Res. Lett., 40, 1432–1438, 2013.
- Lambert, F. H. and Webb, M. J.: Dependency of global mean precipitation on surface temperature, Geophys. Res. Lett., 35, L16706, doi:10.1029/2008GL034838, 2008.
- Lawrence, D. M., Oleson, K. W., Flanner, M. G., Thornton, P. E., Swenson, S. C., Lawrence, P. J., Zeng, X., Yang, Z.-L., Levis, S., Sakaguchi, K., Bonan, G. B., and Slater, A. G.: Parameterization improvements and functional and structural advances in version 4 of the Community Land Model, Journal of Advances in Modeling Earth Systems, 3, M03001, doi:10.1029/2011MS00045, 2011.
- Li, G. and Xie, S.-P.: Tropical Biases in CMIP5 Multimodel Ensemble: The Excessive Equatorial Pacific Cold Tongue and Double ITCZ Problems, J. Climate, 27, 1765–1780, 2014.
- Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425–3446, doi:10.5194/acp-7-3425-2007, 2007.
- Mann, G. W., Carslaw, K. S., Spracklen, D. V., Ridley, D. A., Manktelow, P. T., Chipperfield, M. P., Pickering, S. J., and Johnson, C. E.: Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model, Geosci. Model Dev., 3, 519– 551, doi:10.5194/gmd-3-519-2010, 2010.
- Ming, Y. and Ramaswamy, V.: Nonlinear climate and hydrological responses to aerosol effects, J. Climate, 22, 1329–1339, 2009.
- Ming, Y., Ramaswamy, V., and Persad, G.: Two opposing effects of absorbing aerosols on global-mean precipitation, Geophys. Res. Lett., 37, L13701, doi:10.1029/2010GL042895, 2010.

Myhre, G. and Samset, B.: Standard climate models radiation codes underestimate black carbon radiative forcing, Atmos. Chem. Phys., 15, 2883–2888, 2015.

- Myhre, G., Berglen, T. F., Johnsrud, M., Hoyle, C. R., Berntsen, T. K., Christopher, S. A., Fahey, D. W., Isaksen, I. S. A., Jones, T. A., Kahn, R. A., Loeb, N., Quinn, P., Remer, L., Schwarz, J. P., and Yttri, K. E.: Modelled radiative forcing of the direct aerosol effect with multi-observation evaluation, Atmos. Chem. Phys., 9, 1365–1392, doi:10.5194/acp-9-1365-2009, 2009.
- Neale, R., Richter, J., Conley, A., Park, S., Lauritzen, P., Gettelm, A., Williamson, D., Rasch, P., Vavrus, S., Taylor, M., Collins, W., Zhang, M., and Lin, S.-J.: Description of the NCAR Community Atmosphere Model (CAM 4.0), Tech. rep., National Center for Atmospheric Research (NCAR), Boulder, Colorado, 2011.
- Neale, R. B., Richter, J., Park, S., Lauritzen, P. H., Vavrus, S. J., Rasch, P. J., and Zhang, M.: The mean climate of the Community Atmosphere Model (CAM4) in forced SST and fully coupled experiments, J. Climate, 26, 5150–5168, 2013.
- O'Connor, F. M., Johnson, C. E., Morgenstern, O., Abraham, N. L., Braesicke, P., Dalvi, M., Folberth, G. A., Sanderson, M. G., Telford, P. J., Voulgarakis, A., Young, P. J., Zeng, G., Collins, W. J., and Pyle, J. A.: Evaluation of the new UKCA climate-composition model Part 2: The Troposphere, Geosci. Model Dev., 7, 41–91, doi:10.5194/gmd-7-41-2014, 2014.
- Olivié, D., Peters, G., and Saint-Martin, D.: Atmosphere response time scales estimated from AOGCM experiments, J. Climate, 25, 7956–7972, 2012.
- Osborne, J. M. and Lambert, F. H.: The missing aerosol response in twentieth-century mid-latitude precipitation observations, Nature Climate Change, 4, 374–379, 2014.
- Pausata, F. S. R., Gaetani, M., Messori, G., Kloster, S., and Dentener, F. J.: The role of aerosol in altering North Atlantic atmospheric circulation in winter and air-quality feedbacks, Atmos. Chem. Phys. Discuss., 14, 22477–22506, doi:10.5194/acpd-14-22477-2014, 2014.
- Polson, D., Bollasina, M., Hegerl, G., and Wilcox, L.: Decreased monsoon precipitation in the Northern Hemisphere due to anthropogenic aerosols, Geophys. Res. Lett., 41, 6023–6029, doi:10.1002/2014GL060811, 2014.
- Quennehen, B., Raut, J.-C., Law, K. S., Ancellet, G., Clerbaux, C., Kim, S.-W., Lund, M. T., Myhre, G., Olivié, D. J. L., Safieddine, S., Skeie, R. B., Thomas, J. L., Tsyro, S., Bazureau, A., Bellouin, N., Daskalakis, N., Hu, M., Kanakidou, M., Klimont, Z., Kupiainen, K., Myriokefalitakis, S., Quaas, J., Rumbold, S. T., Schulz, M., Cherian, R., Shimizu, A., Wang, J., Yoon, S.-C., and Zhu, T.: Multi-model evaluation of short-lived pollutant distributions over East Asia during summer 2008, Atmos. Chem. Phys. Discuss., 15, 11049–11109, doi:10.5194/acpd-15-11049-2015, http://www.atmos-chem-phys-discuss.net/15/11049/2015/, 2015.

- Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nat. Geosci., 1, 221–227, 2008.
- Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A., and Andreae, M. O.: Flood or drought: how do aerosols affect precipitation?, Science, 321, 1309–1313, 2008.
- Rotstayn, L. D. and Lohmann, U.: Tropical Rainfall Trends and the Indirect Aerosol Effect, J. Climate, 15, 2103–2116, 2002.
- Rotstayn, L. D., Ryan, B. F., and Penner, J. E.: Precipitation changes in a GCM resulting from the indirect effects of anthropogenic aerosols, Geophys. Res. Lett., 27, 3045–3048, 2000.
- Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S.-M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., and Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, Atmos. Chem. Phys., 14, 12465–12477, 2014.
- Sand, M., Berntsen, T. K., Kay, J. E., Lamarque, J. F., Seland, Ø., and Kirkevåg, A.: The Arctic response to remote and local forcing of black carbon, Atmos. Chem. Phys., 13, 211–224, doi:10.5194/acp-13-211-2013, 2013a.
- Sand, M., Berntsen, T. K., Seland, Ø., and Kristjánsson, J. E.: Arctic surface temperature change to emissions of black carbon within Arctic or midlatitudes, J. Geophys. Res.-Atmos., 118, 7788–7798, 2013b.
- Schwarz, J. P., Samset, B. H., Perring, A. E., Spackman, J. R., Gao, R. S., Stier, P., Schulz, M., Moore, F. L., Ray, E. A., and Fahey, D. W.: Global-scale seasonally resolved black carbon vertical profiles over the Pacific, Geophys. Res. Lett., 40, 5542–5547, 2013.
- Shindell, D. and Faluvegi, G.: Climate response to regional radiative forcing during the twentieth century, Nat. Geosci., 2, 294–300, 2009.
- Shindell, D. T.: Inhomogeneous forcing and transient climate sensitivity, Nature Climate Change, 4, 274–277, doi:10.1038/nclimate2136, 2014.
- Shindell, D. T., Faluvegi, G., Koch, D. M., Schmidt, G. A., Unger, N., and Bauer, S. E.: Improved attribution of climate forcing to emissions, Science, 326, 716–718, 2009.
- Smith, R., Jones, P., Briegleb, B., Bryan, F., Danabasoglu, G., Dennis, J., Dukowicz, J., Eden, C., Fox-Kemper, B., Gent, P., Hecht, M., Jayne, S., Jochum, M., Large, W., Lindsay, K., Maltrud, M., Norton, N., Peacock, S., Vertenstein, M., and Yeager, S.: The Parallel Ocean Program (POP) reference manual:

Ocean component of the Community Climate System Model (CCSM), Los Alamos National Laboratory, LAUR-10-01853, 2010.

- Søvde, O. A., Gauss, M., Smyshlyaev, S. P., and Isaksen, I. S.: Evaluation of the chemical transport model Oslo CTM2 with focus on arctic winter ozone depletion, J. Geophys. Res.-Atmos., 113, D09304, doi:10.1029/2007JD009240, 2008.
- Stevens, B., Giorgetta, M., Esch, M., Mauritsen, T., Crueger, T., Rast, S., Salzmann, M., Schmidt, H., Bader, J., Block, K., Brokopf, R., Fast, I., Kinne, S., Kornblueh, L., Lohmann, U., Pincus, R., Reichler, T., and Roeckner, E.: Atmospheric component of the MPI-M Earth System Model: ECHAM6, Journal of Advances in Modeling Earth Systems, 5, 146–172, 2013.
- Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125–1156, doi:10.5194/acp-5-1125-2005, 2005.
- Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian, R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes, C., Hodnebrog, O., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund, M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J., Quennehen, B., Raut, J.-C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, O., Shine, K. P., Skeie, R. B., Wang, S., Yttri, K. E., and Zhu, T.: Evaluating the Climate and Air Quality Impacts of Short-1 Lived Pollutants, Atmos. Chem. Phys. Discuss., submitted, 2015.
- Tai, A., Martin, M., and Heald, C.: Threat to future global food security from climate change and ozone air pollution, Nature Climate Change, 4, 817–821, 2014.
- Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Feichter, J., Fillmore, D., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I. S. A., Iversen, T., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J. E., Pitari, G., Reddy, M. S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: The effect of harmonized emissions on aerosol properties in global models an AeroCom experiment, Atmos. Chem. Phys., 7, 4489–4501, doi:10.5194/acp-7-4489-2007, 2007.
- Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S. E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S., Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen, T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H., Lin, G., Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J.-J., Müller, J.-F.,

- Myhre, G., Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M., Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou, T., Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q., and Zhang, X.: The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem. Phys., 14, 10845–10895, 2014.
- Vignati, E., Wilson, J., and Stier, P.: M7: an efficient size-resolved aerosol microphysics module for largescale aerosol transport models, J. Geophys. Res.-Atmos., 109, D22202, doi:10.1029/2003JD004485, 2004.
- Wang, Y., Wang, M., Zhang, R., Ghan, S. J., Lin, Y., Hu, J., Pan, B., Levy, M., Jiang, J. H., and Molina, M. J.: Assessing the effects of anthropogenic aerosols on Pacific storm track using a multiscale global climate model, P. Natl. Acad. Sci. USA, 111, 6894–6899, 2014.
- West, J. J., Ansari, A. S., and Pandis, S. N.: Marginal PM2.5: Nonlinear aerosol mass response to sulfate reductions in the eastern United States, J. Air Waste Manage. Assoc., 49, 1415–1424, 1999.
- Wofsy, S.: HIAPER Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically important atmospheric gases and aerosols, Philos. T. R. Soc. A, 369, 2073–2086, 2011.
- Zhang, K., O'Donnell, D., Kazil, J., Stier, P., Kinne, S., Lohmann, U., Ferrachat, S., Croft, B., Quaas, J., Wan, H., Rast, S., and Feichter, J.: The global aerosol-climate model ECHAM-HAM, version 2: sensitivity to improvements in process representations, Atmos. Chem. Phys., 12, 8911–8949, doi:10.5194/acp-12-8911-2012, 2012.

Table 1. Summary of BCand., OC and SO_4 burdens (Tg) and lifetimes (days) in the control simulation for the three models each model.

	HadGEM	ECHAM-HAM	NorESM	CAM4
BC burden (Tg)-	0.080	0.102	0.163	0.144
OC burden (Tg)	0.734	0.769	1.047	0.601
SO_4 burden (Tg)-	3.355	5.345	1.813	1.918
BC lifetime	3.40	5.17	7.82	6.28
OC lifetime	3.02	4.95	7.44	4.83
SO_4 lifetime	5.23	4.02	4.12	3.51

Emission	Model	ΔT	Δ SW	ΔP	ΔP	$\Delta T / \Delta SW$	Δ P/ Δ SW
pert.		(K)	$(W m^{-2})$	$(mm d^{-1})$	(%)	(K/	$(mm d^{-1}/$
-						${ m W}~{ m m}^{-2})$	$\mathrm{W}\mathrm{m}^{-2}$)
SO_2	HadGEM	0.838	2.531	0.057	1.916	0.331	0.022
SO_2	ECHAM-HAM	0.831	2.244	0.062	2.141	0.370	0.028
SO_2	NorESM	0.396	1.001	0.029	1.047	0.396	0.029
SO_2	Mean	0.688	1.925	0.049	1.701	0.366	0.026
BC	HadGEM 1	0.085	0.108	0.013	0.431	0.781	0.118
BC	HadGEM 2	-0.008	-0.057	0.004	0.123	0.145	-0.065
BC	HadGEM mean	0.038	0.026	0.008	0.277	0.463	0.027
BC	ECHAM-HAM	-0.034	-0.164	0.003	0.097	0.209	-0.017
BC	NorESM 1	-0.129	-0.555	0.005	0.171	0.232	-0.009
BC	NorESM 2	-0.152	-0.548	0.004	0.135	0.277	-0.007
BC	NorESM mean	-0.141	-0.552	0.004	0.153	0.255	-0.008
BC	CESM-CAM4 1	-0.084	-0.354	0.005	0.157	0.236	-0.013
BC	CESM-CAM4 2	-0.008	-0.220	0.008	0.290	0.034	-0.039
BC	CESM-CAM4 3	-0.031	-0.192	0.007	0.237	0.163	-0.036
BC	CESM-CAM4 mean	-0.041	-0.255	0.007	0.228	0.145	-0.029
BC	Mean	-0.044	-0.236	0.005	0.189	0.268	-0.007
OC	HadGEM	0.250	0.572	0.019	0.653	0.438	0.034
OC	ECHAM-HAM	-0.025	-0.136	-0.004	-0.151	0.185	0.032
OC	NorESM	0.172	0.456	0.012	0.442	0.377	0.027
OC	Mean	0.132	0.297	0.009	0.315	0.333	0.031

 Table 2. Summary of global mean annual average climate responses.



Figure 1. Emissions of aerosol and aerosol precursor species. (a, b): SO₂; (c, d): BC; and (e, f): OC emissions. Left column: ECLIPSE V4.0a anthropogenic emissions, which are perturbed in the respective experiments. Right column: natural, non-anthropogenic biomass burning (for the year 2008) and shipping emissions, which are not perturbed in these experiments.



Figure 2. Annual average zonal mean BC mass mixing ratio ($\mu g k g^{-1}$) in the control simulation for each model. (a) HadGEM, (b) ECHAM-HAM, (c) NorESM and (d) CAM4.



Figure 3. Time evolution of global mean annual average temperature in the control <u>simulations</u>. Solid lines show the member 1 control simulation for each model -and, where present, dashed lines show member 2 and dotted lines show member 3.

Discussion Paper



Figure 4. Summary of global mean annual average changes in (a) (a–b) surface temperature, (b) (c–d) all-sky TOA SW flux , (c) precipitation and (d) run-off(e–f) precipitation. In the left panels the values shown for the BC simulations are the means for each model (where more than one simulation was run). The values for the individual BC simulations are shown in the right panels. The error bars indicate the 95% confidence interval on the error in the mean $(\frac{2\sigma}{\sqrt{n2}\sigma}/\sqrt{n})$, where *n* is 50 the number of years of data included in the mean; i.e. $n = 50 \times$ number of ensemble members).



Figure 5. Annual average change in surface temperature for $(a, b) (a, b) SO_2$, (c, d) (c, d) BC and (e, f) (e.f) OC perturbations. Left column: multi-model mean maps. Right column: zonal mean. In (a, e)(a, c, e), stippling shows points where all three models agree on the sign of the response . In (e) stippling shows points where is significant at least five of the six model simulations agree on the sign 95% level (determined by a Student's t-test using all years of all models).



Figure 6. Annual average change in <u>all-sky</u> TOA SW flux for (a, b) (a, b) $(SO_2, (c, d)$ (c, d) BC and (e, f) (e, f) OC perturbations. Left column: multi-model mean maps. Right column: zonal mean. In (a, e)(a, c, e), stippling shows points where all three models agree on the sign of the response . In (e) stippling shows points where is significant at least five of the six model simulations agree on the sign 95% level.



Figure 7. Annual average change in precipitation for (a, b) (a-b) SO₂, (c, d) (c-d) BC and (e, f) (e-f) OC perturbations. Left column: multi-model mean maps. Right column: zonal mean. In (a, e)(a, c, e), stippling shows points where all three models agree on the sign of the response . In (e) stippling shows points where is significant at least five of the six model simulations agree on the sign 95% level.

Multi-model mean maps of annual average change in run-off for (a), (b) BC and (c) OC perturbations. In (a, c), stippling shows points where all three models agree on the sign of the response. In (b) stippling shows points where at least five of the six model simulations agree on the sign.