"Regional and global climate response to anthropogenic SO2 emissions from China in three climate models" by M. Kasoar et al.

Author response to anonymous referee #2

The authors are extremely grateful to the reviewer for their extremely helpful and positive comments. We very much appreciate the time taken to do provide these comments, which have helped highlight some areas of the paper where we were unclear or not precise enough.

Below we detail our responses to each minor comment in turn. We hope that these responses will satisfactorily address all the points raised. The referee's comments are included in italics, with our response to them and relevant changes to the manuscript in normal font.

Minor comment 1:

"Only temperature (and no other climate) responses are addressed, and this should be reflected in also in the title."

Modified.

Changes made:

1) 'Climate' changed to 'temperature' in the title

Minor comment 2:

"The descriptions of the three models in section 2.1 should be harmonized. It is especially important to provide the readers with a detailed enough summary of the aerosol and sulfur cycle treatments in each model – currently quite little is told about CESM1 and GISS-E2 aerosol/sulphur. The treatment of aerosol-cloud interactions within each model should also be briefly summarized."

We have attempted to harmonise the descriptions of the models through providing some additional details on CESM1 and GISS-E2, while slightly cutting down unnecessary text in the HadGEM3 description (we note that another reviewer actually thought our description of HadGEM3 was already too long, and so providing the right level of detail without hurting the flow and main message of the paper is a difficult balance). We have also, at the suggestion of the third reviewer, collated key details of the models into a table for easier reference.

Changes made:

1) In HadGEM3-GA4 description, removed:

"..., dynamically resolving the stratosphere"

"..., which includes 4 soil layers and 5 plant functions types. Although in principle this can be run in a fully interactive 'Earth System' mode with dynamic vegetation and a carbon cycle,..."

"More detailed description and evaluation of the atmosphere and land surface schemes can be found in Walters et al. (2014)."

"Critical to our study is the representation of aerosols; we..."

"..., which is described and evaluated in..."

"The remaining aerosol species are emitted directly in the particulate phase, and..."

"...can then undergo advection, wet and dry deposition, and..."

2) In HadGEM3-GA4 description, inserted:

"(Walters et al., 2014)" in first and second sentences.

"HadGEM3-GA4 can be run with a choice of two aerosol schemes of differing complexity – CLASSIC (Bellouin et al., 2011), and GLOMAP (Mann et al., 2010). Here we use the simpler CLASSIC scheme, which is less computationally expensive, and is also the aerosol scheme that was used for CMIP5 simulations with the predecessor of this model (HadGEM2). CLASSIC is a mass-based scheme, meaning that only aerosol mass (and not particle number) is tracked, and therefore all aerosol species are assumed to be externally mixed."

"...mass..." in the sentence: "Cloud droplet number concentration and effective radius are determined from the mass_concentration of these aerosols..."

plus minor connecting words so that sentences still read correctly after the phrases removed above.

- 3) In CESM1 description, removed:
 - "...modal aerosol scheme ... "
 - "...from anthropogenic and natural..."
- 4) In CESM1 description, added:

"CAM5-Chem uses the MAM3 modal aerosol scheme (Liu et al., 2012), which is the same as used for the CESM1 submission to CMIP5. Both aerosol mass and particle number are prognostic, and the scheme simulates sulfate, black carbon, primary organic matter, secondary organic aerosol, dust, and sea salt aerosol species as an internal mixture in Aitken, accumulation, and coarse modes."

"The model includes emissions of natural and anthropogenic SO2 and natural DMS as sulfate precursors, and..."

"Aerosols-cloud interactions allow for the effect of aerosols on both cloud droplet number and mass concentrations (Tilmes et al, 2015)."

5) In GISS-E2 description, split second paragraph in to two and moved "nitrate, elemental and organic carbon along with secondary organic aerosols and natural sea-salt and mineral dust"

from the last paragraph to the new third paragraph.

- 6) In GISS-E2 description, replaced "SO₂ from anthropogenic and natural sources..." with "SO₂ from these sources..."
- 7) In GISS-E2 description, added:

"GISS-E2 has a choice of three aerosol schemes of varying complexity – OMA (Koch et al., 2011; 2006), MATRIX (Bauer et al, 2008), and TOMAS (Lee and Adams, 2012). Following the GISS-E2 CMIP5 configuration, we use here simpler mass-based OMA scheme, which includes sulfate, ..."

"Aerosols are parameterised as an external mixture of dry and dissolved aerosol, with particle size parameterised as a function of relative humidity (Schmidt et al., 2006)."

"includes natural emissions of DMS, and natural and anthropogenic emissions of SO2."

"..., such that cloud droplet number concentration and autoconversion rate depend on the local concentration of aerosol."

8) Added a new table (Table 1; previous Table 1 is now Table 2) with key model details, as described in response to Referee #3 Minor Comment 3.

Minor comment 3:

"P5L12: What does 'mass based' scheme mean in this context when modes and bins are also treated? P5: Is aerosol microphysics (condensation, coagulation, etc.) treated in CLASSIC?"

We mean that only the mass concentration of each aerosol species (as opposed to number concentration) is tracked within each of the Aitken, accumulation, and dissolved modes. We have clarified this part of the description. Because only the mass of aerosol within each mode is tracked, microphysics is parameterised to allow transfer of mass between the different modes, based on the mass concentrations of each mode.

Changes made (also included in response to Minor comment 2 above):

1) Added "CLASSIC is a mass-based scheme, meaning that only aerosol mass (and not particle number) is tracked, and therefore all aerosol species are assumed to be externally mixed" in the description of CLASSIC

Minor comment 4:

"P6L9-10: Does this mean that chemistry is solved online? The formulation here seems overly complicated."

Yes – this has been clarified.

Changes made:

 In CESM1 description, added "...online..." in "...we use a<u>n online</u> representation of tropospheric and stratospheric chemistry..."

Minor comment 5:

"P7L1: 'aerosol-coating of dust': Dust is an aerosol particle itself; do you mean (secondary) coating of dust?"

Yes – have amended to clarify.

Changes made:

1) Changed "aerosol-coating of dust" to "secondary coating of dust"

Minor comment 6:

"How different are the control climates between the different models? Would you expect this to impact your results?"

The control climates are fairly similar between the models – an annual mean climatology is plotted below for comparison. If compared with observations, all three models have similar magnitude temperature biases. GISS is a bit too warm in the tropical oceans, CESM is a bit too warm over the northern mid-latitude land, all three - although especially HadGEM - are too warm in the Southern Ocean, and possibly too cold over the polar regions, by a few degrees in each case. On average, GISS is about ½ a degree warmer than HadGEM, which is about ½ a degree warmer than CESM.

In terms of whether this would impact our results – we do not think it could have a substantial difference to the models' responses to an aerosol emissions perturbation. Firstly, because the changes in SW flux themselves explain much of the diversity in the models' temperature responses. The effect of climate feedbacks and climate sensitivity may play a role in setting the exact magnitude of the final response, but these vary between models anyway, unrelated to the climate state, and so this is part of the structural uncertainty we wish to explore. To our knowledge, studies that have looked for example at the time-dependence of climate sensitivity and feedbacks in transient warming scenarios generally find that it varies slowly, and so inter-model variations in climate sensitivity are likely much more important than the base climate state, unless this were to be very different.



Minor comment 7:

"P7L27: Are the runs restarted from an earlier simulation? 50-year spin-up by itself doesn't seem sufficient for a coupled model."

Yes, the runs were restarted from previous coupled simulations that had already been run for present-day conditions, though not necessarily with the exact model set-up that was used here. The 50 years is not intended to spin-up the control runs, but rather to allow the response to the perturbation to establish itself. We have expanded the experimental setup section to clarify this. Previous studies which apply an abrupt forcing (e.g. Andrews et al. (GRL, 2012, doi:10.1029/2012GL051942) have generally seen that most of the global surface temperature response is realised within this timeframe, and from inspection of the time series of global temperature changes, this seemed to be the case here as well.

Changes made:

- 1) In Section 2.2 (Experimental Setup), added ", initialised from a present-day state," to the description of the control simulations, and "from the same initial state," to the description of the perturbation simulations.
- 2) In first sentence of Section 3, replaced bracketed phrase "the first 50 years were discarded as spin-up" with "the first 50 years are discarded to allow the response to the perturbation to establish itself".

Minor comment 8:

"P10: Both HadGEM and CESM1 simulate H2O2 and O3 oxidation pathways in the aqueous phase, so including both pathways cannot be an explanation to fast conversion to SO4 in HadGEM. This should be explicitly stated."

We agree that including both pathways cannot explain any differences in the SO₂ oxidation rates between HadGEM3 and CESM1, only for HadGEM3 and GISS-E2. We have added a sentence explicitly stating this.

Changes made:

1) Added additional sentence at the start of fourth paragraph of Section 4.1:

"CESM1 includes the same oxidation pathways as HadGEM3-GA4, and in fact has a slightly shorter SO2 lifetime still, and so the differences between these two models have different origins."

Minor comment 9:

"P16L6-8: Do you refer to sulphate aerosol above cloud top here? Simulated cloud distributions can have large impacts also in other ways, e.g. the background aerosol amount (clean/polluted) has large impacts on indirect effects, which start to saturate at high aerosol concentrations."

Yes – as suggested we've rephrased this sentence to add a mention here of other ways cloud distribution has a potential impact via the saturation of indirect effects (plus reference).

Changes made:

1) Added to first paragraph of Section 4.2 (section in square brackets was already there):

"For instance, [the radiative effect of sulfate aerosol is modulated by the reflectivity of the underlying surface in the radiation scheme (Chýlek and Coakley, 1974; Chand et al., 2009), which may often be a cloud-top.] The low contrast with a highly reflective cloud surface means that sulfate aerosol above a cloud top will have a reduced direct radiative forcing. Blocking of radiation by clouds will also reduce the direct radiative effects of any aerosols within or below them (e.g. Keil and Haywood, 2003). Additionally, aerosol indirect effects can saturate in regions with a high level of background aerosol (e.g. Verheggen et al., 2007; Carslaw et al., 2013), meaning that the potential for indirect radiative forcing can also vary with the location of clouds. On top of diversity in indirect effects, and in the climatological distribution of clouds, different dynamical changes in cloud cover could also alter the all-sky flux."

2) Inserted additional references (Keil and Haywood, 2013, Verheggen et al., 2007, and Carslaw et al., 2013) in bibliography

Minor comment 10:

"P16L20-21: Can you speculate which dynamical processes cause the increase in cloudiness when sulphate is removed? Based on 2nd indirect effect one would assume decreased cloudiness."

Indeed, we also expected decreased cloudiness from the 2nd indirect effect, and so the observed increases in GISS are presumably dynamical in origin. Dynamical feedbacks can be complex and chaotic, and cause and effect hard to untangle. Moreover, despite all having local warming at the surface in east China, all three models have quite different regional cloud changes, and so whatever dynamical processes are at play are not robust. Therefore, we do not wish to speculate further here.