Reply to Anonymous Referee #2 for Climatic Impacts of Stratospheric Geoengineering with Sulfate, Black Carbon and Titania Injection

We thank the anonymous reviewer for their thorough critique and many useful comments and suggestions. In our response, we aim to address each of the reviewer's comments and make the corresponding changes to the manuscript where necessary (red indicates sentences removed, blue indicates sentences added).

Specific Responses

1. Radiative forcing

    a. The reviewer notes that the acronym 'RF' is used for both anthropogenic *radiative forcing* (ARF) and top-of-the-atmosphere *radiative flux imbalance* (TOA-RF). Radiative forcing and radiative fluxes are different concepts; therefore the use of RF for both might confuse a reader. We therefore use the acronym *TOA-Imb* instead of *TOA-RF*.

    b. The reviewer highlights that the GeoMIP 'G3' specifications are significantly different from our specifications, both in the baseline GHG-concentrations scenario (we use RCP8.5, GeoMIP uses RCP4.5) and in goals (we target TOA radiative fluxes, GeoMIP targets radiative forcing). Therefore the reviewer recommends that we use a different nomenclature for our geoengineering simulations, which we agree to. Therefore *G3S*, *G3TiO$_2$* and *G3BC* are changed to *geoSulf*, *geoTiO$_2$*, and *geoBC*.

    c. The reviewer questions the goal of the investigation; what does "maintain TOA-Imb balance" entail? The reviewer also queries how the TOA-Imb relates to the radiative forcing. We provide the following text in the Methods section, but also include an entirely new section S2 in the Supplementary Material in which we describe how the simulations were conducted.
    We inject aerosol at such a rate as to maintain the top-of-the-atmosphere (TOA) net radiation at piControl levels. Specifically, we define the TOA radiative flux Imbalance (TOA-Imb) as the annual/global-mean TOA net radiation (incoming SW minus outgoing LW+SW) minus the average TOA net radiation of the piControl period. By sufficient aerosol injection, we aim to maintain TOA-Imb=0. This scenario represents our interpretation of 'equal amount of geoengineering' for each aerosol. The advantage of returning net radiation to piControl levels (rather than completely equilibrating TOA fluxes) is that piControl had already been simulated comprehensively for CMIP5 (240 model-years), hence permitting robust statistics to be calculated. The TOA radiative imbalance is a metric that satellites are able to measure (e.g. CERES [L'Ecuyer et al, 2015] and

EarthCare [Illingworth et al, 2015]), albeit with +/- 3 W/m^2 accuracy at present [Priestley et al, 2011; von Schuckmann et al., 2016]. Therefore our target could be applicable to an actual SAI scenario. In contrast, Radiative Forcing (RF) (the net radiation perturbation at the tropopause from some external forcing, after stratospheric adjustment), cannot be directly measured by satellites and therefore it would be difficult to obtain a specified radiative forcing in an actual SAI scenario. Of course, other metrics could be chosen (e.g. MacMartin et al., 2013), with each metric having its own signal/noise characteristic.

d. The reviewer notes that the IPCC report citations concerning temperature trends are only applicable to *radiative forcing* and not *radiative fluxes*. The reviewer questions whether the temperature trends in fig. 3 are instead related to the consistently non-adjusted stratosphere. The reviewer notes that the surface temperature disparities might occur for an equal tropopause radiative forcing, if energy is distributed differently in the climate system. In answer to these questions, we have assessed the net radiative fluxes at the top of the atmosphere and the tropopause, and the net heat flux (radiation + sensible + latent) at the surface. We firstly remove this analysis from section 4.1:
This is due to the absorption of radiation by BC (and a lesser extent the absorption by titania) heating the stratosphere which then increases the terrestrial longwave radiation entering the troposphere reducing the tropopause-RF. As noted in several Intergovernmental Panel on Climate Change reports [e.g. Ramaswamy et al., 2001; Forster et al., 2007], it is the global mean tropopause-RF rather than the TOA-RFI that is proportional to global mean surface temperature changes. Further analysis of stratospheric temperature changes will be provided in section 4.4.

The following is added in its place.
The near-surface global temperature response differs between the aerosols with a greater cooling trend for sulfate than for titania or BC (Fig. 3b). To determine the cause of the anomalous warming in geoBC, we assess the net energy fluxes at the top of the atmosphere for 2020-2100. Fig. S3 in the Supplement shows the global-mean net-downward radiation anomaly for the geoengineering experiments, evaluated at the TOA and the tropopause; and the global-mean net-downward heat flux anomaly at the surface. The radiation changes at the TOA and tropopause, and the heat flux anomaly at the surface, are comparable for the geoSulf and geoTiO_2 experiments for the duration of 2020-2100. In contrast, geoBC exhibits an increasingly positive net radiation anomaly at the tropopause (+0.2 W/m^2 averaged over 2020-2100) despite the negligible TOA radiation anomaly. After stratospheric temperature adjustment, radiative perturbations at the TOA and tropopause are equal for a given climate forcing, which implies that the consistently non-adjusted stratosphere (due primarily to

increasing aerosol injection rates) is responsible for the differences in TOA and tropopause radiative perturbations in geoBC. This implies that if we had injected aerosol sufficiently to produce an equal radiative effect at the tropopause, the temperature trends for the geoengineering experiments in Fig. 3 would have been more comparable. If we were to choose stabilisation of temperature as our basic metric, then one could approximate the results by simply scaling the results by the ratio of the temperature perturbation relative to 1980-2005 to that for geoSulf. The scaling would be 1 (by design) for geoSulf, 1.1 for geoTiO_2 and 1.28 for geoBC. If the metric chosen were instead to keep the global mean precipitation the same, then the scaling would be 1 (by design) for geoSulf, 0.91 geoTiO_2 and 0.68 for geoBC. However, we shall see that the changes in many of the variables we consider are dominated by large scale changes in the spatial patterns of response rather than the 10-30% changes in magnitude of the response that applying such a scaling would induce. We therefore choose to present un-scaled results here but caveat that such a scaling could be applied should we wish to apply a different metric.

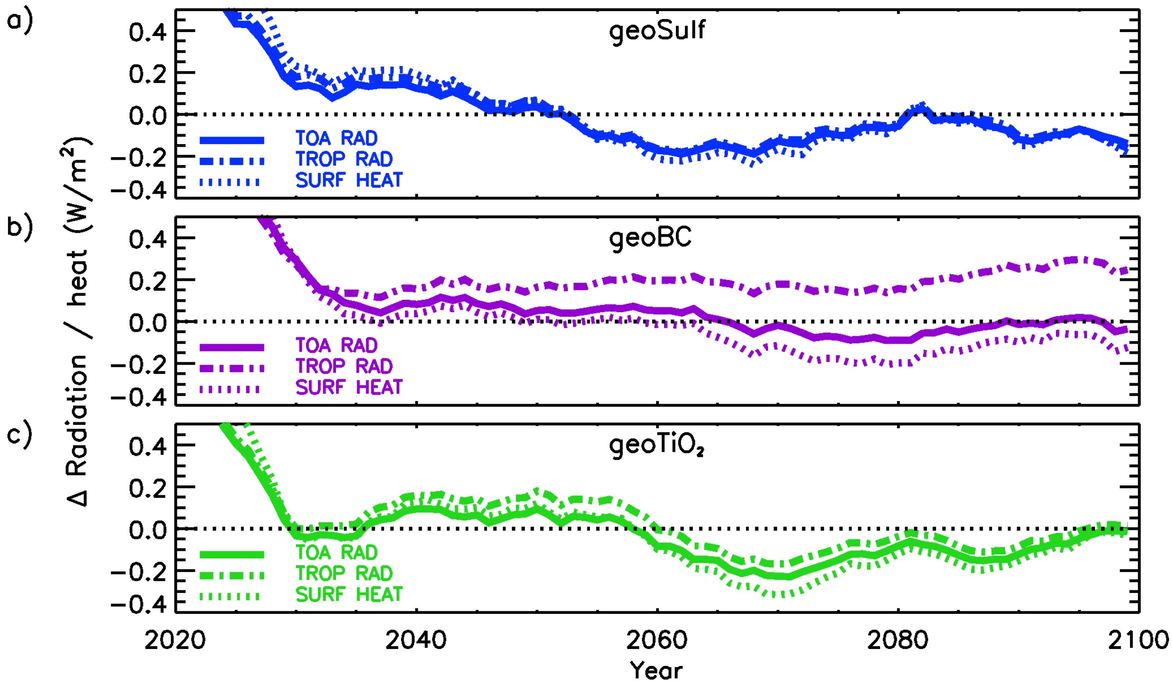


Fig. S3 *10-year running-average global/annual-mean net radiation anomaly at the tropopause and TOA, and net-downward heat flux anomaly at the surface, with respect to piControl. Positive values indicate an increase in net downward flux.*

e. The reviewer questions where the additional energy into the climate system in the geoSulf experiment goes, considering that temperatures decrease over time despite a net flux of energy into the system. Additionally the reviewer asks whether energy is conserved in the model. HadGEM2-CCS's dynamical core, 'New-Dynamics', does not conserve energy [Davies et al., 2005]. Instead, an energy correction flux is applied at the end of each model day as a globally homogeneous heating-rate perturbation at all levels and grid-points. However, the fact that the temperature trend in fig. 3b is negative for geoSulf is more likely due to an uneven vertical distribution of this energy gain. The following explanation is added to the text.

From Fig. 3b, geoSulf exhibits a near-surface air cooling trend with respect to 2020 despite a net gain of atmospheric energy, which is likely due to an uneven vertical distribution of this energy gain.

2. HIST period – The reviewer questions the motivations behind our choice of control period (HIST: 1980-2005) and how this relates to the goal of our experiment. We accept that our choice of control period is arbitrary, we chose this period as the temperature change for geoSulf was approximately 0. We add the following explanation for the choice of HIST period to the text.
   As we were not explicitly attempting to reach a specific global mean temperature, the choice of reference period was left until after the geoengineering simulations had been completed. We then selected a recent historical period from which the 2090s global-mean temperature anomaly for geoSulf was negligible (fig. 3b). The HIST period selected is close to the historical control period used in the IPCC AR5 report (1860-2005) [e.g. Fig. 12.10 from Collins et al, 2013] which permits comparison of our RCP8.5 results with the CMIP5 multi-model mean.

   We also add the following caveat to section 4.3.
   It is important to note that if the RCP8.5 warming relative to HIST was completely offset in the geoBC and geoTiO_2 experiments, the hydrological response would be greater than in fig. 6. Using the hydrological sensitivities calculated in section 4.1, the precipitation changes relative to HIST would be -0.34 mm/day for geoBC and -0.16 mm/day for geoTiO_2 .

3. Aerosol representation – The reviewer notes that we do not discuss the sensitivity of our results to the choice of size distribution. We have addressed the same issue in our reply to anonymous referee #1, which we repeat below. Specifically, we have compared our results to Ferraro et al (2011), and discussed the likely reasons for the difference in temperature perturbations. The following is added to the Discussions section of this report.
   We find that sulfate induces less stratospheric warming than titania. In contrast, Ferraro et al (2011) found that the peak stratospheric warming for titania was approximately a third of that from sulfate. Although the different climatologies, model configurations, and aerosol spatial distributions will contribute to the difference in stratospheric temperature adjustment between our and Ferraro's work, the primary reason for the disparity is likely to be the aerosol size distributions. Our titania is smaller (median radius = 0.045 μm compared to 0.1 μm for Ferraro et al (2011)) and therefore scatters and absorbs SW more efficiently, producing a greater localised 'solar' warming. Their sulfate distribution contains a larger spread (σ = 2.0 for

Ferraro et al (2011) compared to σ = 1.25 here), resulting in more coarse-mode particles and greater LW absorption. This disparity highlights the sensitivity of climatic impacts to the specified aerosol size distribution. On a separate note, Ferraro et al (2011) neglected to alter the aerosol density component in the calculation of their aerosol masses and specific optical properties [A. Ferraro, personal communication]. The density that they used for all the aerosols of 1000 kg/m^3 is arguably applicable to black carbon, but not to sulfate and titania (which instead are ~1600 and ~4000 kg/m^3). Therefore, their aerosol burdens for sulfate and titania should be multiplied by 1.6 and 4 respectively, and their optical coefficients divided by 1.6 and 4, to obtain appropriate values.

4. Stratospheric water vapor – The reviewer notes that stratospheric temperature and dynamical changes could perturb the stratospheric water vapor content, with resultant impacts on radiation, chemistry and dynamics. We add the following text to section 4.4.
   Additionally, an increase in the Tropical Tropopause Layer (TTL) temperature would increase the specific humidity of air entering the stratosphere [Dessler et al., 2013]. Changes to the stratospheric water vapor content could have significant chemical and radiative impacts, contributing to ozone depletion via the HO_x cycle and stratospheric warming via LW-absorption [Kravitz et al., 2012]. To assess the effects of geoengineering on stratospheric water vapor, we calculate the time-averaged H_2O mixing ratio averaged between 20^{o}S-20^{o}N and 16-20 km altitude. In the HIST era, the H_2O MMR is 4.2 ppmv, in close agreement with HALOE observations [Gettelman et al., 2010]. In the 2090s, the average H_2O MMR is 6.3 ppmv for RCP8.5, 4.8 ppmv for geoSulf, 7.1 ppmv for geoTiO_2, and 32.7 ppmv for geoBC. The stratospheric water vapor feedback is therefore greater for geoBC and geoTiO2 than for geoSulf.

5. Abstract – The reviewer notes that a conclusion that we offer in the abstract is not present in the main text. Specifically, we conclude that the stratospheric heating invoked by BC is so severe as to exclude BC from being a viable candidate particle for SAI. We agree that this is a strong conclusion that should also be in the manuscript. The following has been added to the discussion.
   We have shown that, although the distributions of climate changes are similar for the 3 SAI scenarios, the magnitudes of the changes differ, for instance, BC produces a substantially greater stratospheric warming signal with concomitantly greater changes to stratospheric dynamics. The severity of the stratospheric temperature changes effectively excludes BC from being a viable option for geoengineering.

6. P44L23 – The reviewer informs us of missing citations. We thank the reviewer for highlighting the missing citations which have been added to the references list (the additional citations are below). Additionally, 'Collins et al (2014)' has been changed in the text.

Ramaswamy et al, 2001; Peters et al, 2013; Kravitz et al., 2015; Dhomse et al., 2014; Pithan and Mauritsen, 2014; Schmidt et al., 2013; Dessler et al, 2013; Gettelman et al., 2010; Niemeier et al., 2013; Liu et al., 2013; Ndour et al., 2008; Koehler et al., 2009; Weisenstein et al., 2015; Tang et al., 2014; Davies et al., 2005; Bellouin et al., 2007; Priestley et al., 2011; L'Ecuyer et al., 2015; Illingworth et al., 2015; Haywood et al., 2011; MacMartin et al., 2013; von Schuckmann et al., 2016

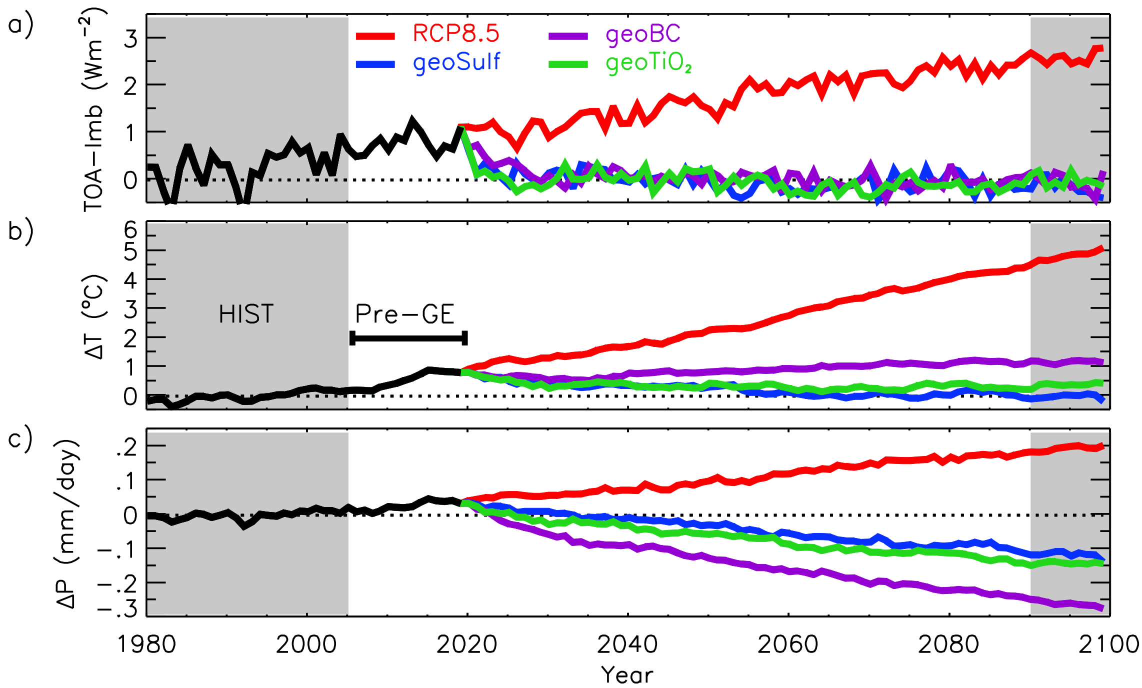
7. Figure 1 – The reviewer questions why the LW and SW coefficients do not agree in figure 1. This is because the points are plotted at the middle of each spectral waveband. Further detail is provided in our reply to the anonymous reader #1 (specific response 3). The caption has been altered to include this detail.

8. P50L14 – The reviewer contends with our use of 'prolong', as in 'prolong the stratospheric lifetime', which was misused here. We replace *prolong* with *maximise*.

9. P50L23 – The reviewer notes that our method for conducting the simulations would benefit from further discussion, which we agree. We did not calculate the injection rate alterations online, this was done in stages. For detail, we simulated 20 model-years at a time and calculated the average TOA_RFI of that period. If the average TOA_RFI exceeded a threshold (0.25 Wm^{-2}), we recalculated the injection rates for that segment of time and restarted the simulation at the start of that time-period. A single ensemble member was used to obtain injection rates for each aerosol; the other 2 ensemble members were conducted later. Whether this method is applicable to a real geoengineering scenario is less certain, an 'online' algorithm would certainly be a more realistic representation of an actual geoengineering strategy. We go into further detail in specific response 5 to anonymous referee #1, which is then added to the supplement along with a schematic (Section S2 in the Supplement).

10. P53L7 – The reviewer informs us of recent research suggesting that temperature feedbacks contribute to Arctic amplification more than surface-albedo feedbacks. We thank the reviewer for this information and modify the sentence accordingly. *RCP8.5 (Fig. 6a) shows the typical global warming signal of amplified warming at high-latitudes due to temperature feedbacks [Pithan and Mauritsen, 2014] and the surface-albedo feedback [e.g. Kharin et al., 2013].*

11. P54L7 – The reviewer suggests that we include the global-mean precipitation anomaly time-series in figure 3, which we think is a good idea (the revised figure is plotted below). Additionally the reviewer suggests that we provide normalised values for the precipitation in terms of the temperature anomaly (%/^{o}C). We add this to section 4.1.



In section 4.1 we have added the following paragraph.

Fig. 3c shows the global mean precipitation anomaly with respect to the HIST period. The precipitation reduction is greater for BC than for sulfate and titania, despite the positive temperature trend in geoBC (fig. 3b). The hydrological sensitivity to geoengineering, defined as the global mean precipitation change per temperature change, is 2%/°C for sulfate, 2.5%/°C for titania, and 4.6%/°C for BC. The hydrological sensitivity for RCP8.5 is 1.32 %/°C, which is close to the CMIP5 ensemble-mean [Fig. 12.7 from Collins et al, 2013]. For comparison, Bala et al (2008) found a hydrological sensitivity of 2.4%/°C for solar irradiance reduction and 1.4%/°C for CO_2 increase.

12. P54L9 – The reviewer questions the meaning of the following sentence: "must be ameliorated by additional SW absorption". We appreciate that this statement is ambiguous and requires elaboration. By this we mean that the SW-absorption for BC exceeds the SW-backscatter for sulfate and titania. The SW radiative perturbation at the tropopause and TOA are therefore greater in geoBC than in geoSulf and geoTiO_2. We have modified the text accordingly.
In order to maintain TOA-Imb=0, geoBC produces a greater SW perturbation at the tropopause and TOA than geoSulf and geoTiO_2, which is compensated by the increased LW perturbation resulting from stratospheric warming in geoBC. The troposphere is relatively transparent to SW radiation but absorbs efficiently in the LW spectrum, therefore the annual-mean surface radiative forcing in the geoBC experiment is greater (−18.6 W m^{-2}) than for geoSulf or geoTiO_2 (−7.4 and −9.6 W m^{-2} respectively – see Fig. S6 in the Supplement).

13. P54L14 – The reviewer is unsure as to how to interpret Fig. S4 in the supplement. We agree that fig. S4 is perhaps confusing, and have decided to swap it for the following

plot. Please note that when calculating the global-mean surface flux anomalies in the new plot, we found that the original values given were in error, and have now been corrected in the text (p 30054). Our analysis is not affected by these changes.

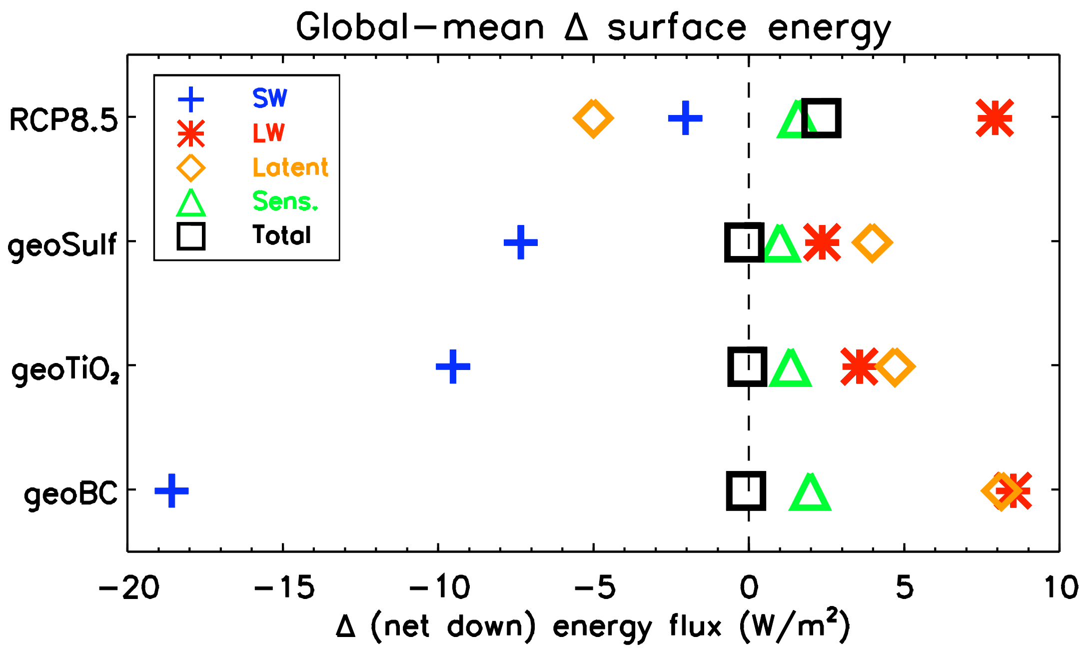


Fig. S6 *2090s global/annual-mean net downward energy flux anomalies at the surface (W/m^2). Calculated with respect to piControl*

The following additional analysis of Fig. S6 will also be added to the manuscript in section 4.3.

The reduction in surface SW flux in the RCP8.5 scenario is due to increases in water vapor [Haywood et al., 2011]. Haywood et al (2011) report a clear-sky reduction of -5.7 Wm^{-2} while our study is consistent at a value of -5.4 Wm^{-2} (not plotted). However, in all geoengineering cases, this reduction is comprehensively overwhelmed by aerosol direct effects.

14. P55L21 – The reviewer notes that the stratospheric warming under sulfate is the result of net absorption of LW radiation (less emission than absorption). We add the following detail for clarity.
Sulfate predominantly absorbs in the LW and near-infra-red spectrum (Fig. 1a). The stratospheric radiative heating in geoSulf is most pronounced in the tropical region, where sulfate absorbs outgoing LW radiation from the warm troposphere below, and then emits comparatively less radiation from the ambient cold stratosphere [Ferraro et al, 2011].

15. P56L5 – The reviewer questions why we give the maximum sulfate-induced warming as +7^{o}C, when Fig. 10 contravenes this. The maximum sulfate-induced warming is

calculated with respect to the RCP8.5 simulation and not to HIST, i.e. displayed in Fig. S6 in the supplement (now Fig. S8). We refer to Figure S6 when giving the maximum BC-induced warming.

16. P56L7 – Tropospheric has been added to tropical circulation

17. P57L11 – The reviewer notes that our analysis of the QBO modification for geoBC would benefit from a rewrite. We agree with this suggestion.
    No QBO-like oscillation can be detected in the 10-year time span.

18. P57L24 – The reviewer notes parallels between our work and Niemeier et al (2013). We thank the reviewer for this notification, and we have added the following to the discussion.
    Additionally, producing an equivalent top of the atmosphere radiative perturbation with a SW-absorbing aerosol such as BC (or to a lesser extent titania) compared to a SW-scatterer such as sulfate, induces a comparatively greater SW forcing at the surface. Bala et al (2008) showed that latent heat fluxes compensate for the SW reduction at the surface, instigating a deceleration of the hydrological cycle that is proportional to the magnitude of the SW reduction. This explains the comparatively greater precipitation reduction exhibited by geoBC in figures 6-8. Our results complement Niemeier et al (2013), who showed that a LW-absorbing sulfate layer would produce a greater hydrological perturbation per TOA SW forcing than a simple solar irradiance reduction scenario.

19. P60L10 – The reviewer notes that our conclusion pertaining to the general efficacy of SAI is too definitive. We agree that the statement is a little too strong. We therefore swap “has shown” with “indicates”.
    Whilst research indicates that SAI is capable of averting certain climate changes such as surface-warming, SAI provides no amelioration for other climate impacts, such as ocean acidification.