Author's Response

We thank the 2 anonymous reviewers and John Dykema for their very useful comments and suggestions for the paper. In our individualised replies, we have addressed each of the reviewer's comments separately and made the relevant changes to the manuscript where necessary. Below, we amalgamate the individual responses and changes to the manuscript, and then provide a revised manuscript. Note that significant changes have been made to the Supplement, including the addition of Section 2 – describing our method for conducting the simulations.

Sections

Anonymous Referee #1

1. Abstract - The reviewer notes that the following statement is too strong:
   As injection rates for titania are close to those for sulfate, there appears to be little benefit of using titania when compared to the injection of sulfur dioxide
   In response, we amend the statement to the following:
   As injection rates and climatic impacts for titania are close to those for sulfate, there appears to be little benefit in terms of climatic influence of using titania when compared to the injection of sulfur dioxide
   The amended statement takes into account that the conclusion is relevant with respect to the climatic impacts of titania and sulfate injection.

2. Introduction – The reviewer questions why we chose RCP8.5 as a baseline scenario. We have explained why we chose RCP8.5 in the following sentence, which is on P30046.
   RCP8.5 is selected to give a significant greenhouse effect against which to employ geoengineering, in order to distinguish the climatic impacts specific to each aerosol

We appreciate however that the choice of RCP8.5 over the more societally-optimistic RCP4.5 scenario is slightly-controversial (and novel) and will affect the findings of this report. In particular, the aerosol injection rates, deposition rates, mass concentrations and the magnitude of stratospheric changes would all be different with a baseline RCP4.5 scenario. We modify the introduction as follows.

RCP8.5, which is the high-end carbon-intensive CMIP5 scenario, is selected to give a significant greenhouse effect against which to employ geoengineering, in order to distinguish the climatic impacts specific to each aerosol. Observations have shown that the current global GHG emissions exceed the emissions inherent in RCP8.5 [Peters et al, 2013]; therefore our work could be considered as geoenineering against a business-as-usual scenario. Additionally, the next generation of GeoMIP simulations (GeoMIP6) will utilise a carbon-intensive scenario [Kravitz et al, 2015], hence our work will provide a useful supplement to those results.

Also, in section 5, we have added the following paragraph to discuss the possible differences if RCP4.5 had been used.

The climatic impacts described in section 4 are specific to geoengineering against a baseline RCP8.5 scenario. If instead we had used a middle-of-the-road GHG-concentrations scenario such as RCP4.5 [Taylor et al., 2012], as used in the first tier of GeoMIP scenarios [Kravitz et al., 2011], then less aerosol-injection would be needed to obtain TOA-Imb=0 and therefore the aerosol deposition rates and atmospheric mass concentrations would be less than those reported in section 4. One would expect that the magnitude of stratospheric temperature changes (fig. 8) and therefore zonal-mean zonal wind changes (fig. 12) would be much less for each of the aerosols, possibly confounding the conclusions giving here relating to their comparative efficacy. An estimate for the amount of SAI required for RCP4.5 can be garnered from integrating the temperature anomalies for RCP8.5 and RCP4.5 for the period 2020-2100. The ratio of the integrated temperature anomalies for RCP4.5 to RCP8.5 is 0.43, hence we can assume that the injection rates required for RCP4.5 are ~0.43 of those for RCP8.5, producing a climate perturbation ~0.43 times as great. A further set of simulations, which instead utilise RCP4.5 as the baseline scenario, would be required to test this hypothesis.

3. Figure 1 – The reviewer questions the discontinuities between the short wave and long wave points in figure 1. The discontinuities between the short wave and long wave points in figure 1 are not due to uncertainty in the optical properties. Rather, the wavelength at which the points are plotted is the average wavelength of the spectral waveband used in the model. There are 5 disparate wavebands in the short wave spectrum and 9 wavebands in the long wave. In the short wave, the wavebands used for this experiment are 0.2-0.32, 0.32-0.69, 0.69-1.19, 1.19-2.38, and 2.38-10 µm. In the long wave, the wavebands used are 3.34-6.67, 6.67-7.52, 7.52-8.33, 8.93-10.1, 8.33-12.5, 13.3-16.9, 12.5-18.1, 18.1-25, 25-10000 µm. It is then clear that the average-wavelengths of the wavebands of the two spectrums overlap (max SW = 6.19 µm, min LW = 5 µm), which explains the disparity in figure 1. The equivalent image from Ferraro et al (2011) – their figure 1 – shows the same discontinuities between the short wave and long wave bands, which is because both our models (their FDH code, our HadGEM2-CCS) use the Edwards and Slingo radiation code (ES96) and we both evaluate the optical constants at the same wavebands [Edwards and Slingo, 1996]. We appreciate that this point was not made clear in the article and could cause confusion to a reader. Therefore we amend the caption in figure 1 as follows.

   *"**Figure 1.** Optical properties as a function of wavelength for a) accumulation-mode sulfate, b) titania, c) black carbon. Points are plotted at the middle of each spectral waveband, as detailed in Bellouin et al (2007)"*

4. Figure 1 & figure 10 – The reviewer questions why we get different results to Ferraro et al (2011). There are many differences between the models and configurations used by ourselves and Ferraro et al (2011), hereafter F11, which could contribute to the different stratospheric temperature perturbations. We will concentrate on sulfate and titania, as black carbon is similar between the two, and give possible reasons for the temperature-change differences. We will then summarise these for addition to the article.
   a. The first point is that F11 use a fixed dynamical heating (FDH) code and not a dynamical-model; their code doesn't take into account feedback from changes in the atmospheric and oceanic circulation, sea surface temperatures, etc as HadGEM2-CCS will. Also their model has 38 vertical levels up to 40 km altitude rather than the 60 vertical levels up to 85 km in our model

b. The second point is that we use inherently different climatologies, which include different ozone, baseline stratospheric temperatures, greenhouse gases, clouds, etc. All of which will contribute to the overall temperature change

c. The third point is that we use different size distributions for titania and sulfate. We chose the titania size distribution (r_m=0.045 μm, σ=1.8) from considering results of Pope et al (2012), namely that this is an optimised size distribution for a scattering aerosol. F11's size distribution (r_m=0.1 μm, σ=2.0) was selected from observations of natural mineral dust aerosols of which titania is a minor component. The key difference between these size distributions is the median radius, which is much smaller in our case, and therefore the titania aerosols in our simulations absorb and scatter short wave radiation more efficiently, hence more solar radiation is absorbed in the aerosol layer resulting in greater stratospheric heating.

   For sulfate, we took the *volc2* size distribution from Rasch et al (2008) (r_m=0.376 μm, σ=1.25) which is based on stratospheric aerosol observations in a post-volcanic period. Our Pinatubo simulations in section 3 show close conformity to observations. F11's sulfate size distribution (r_m=0.1 μm, σ=2.0) is much wider (i.e. their σ is greater). Therefore, while the effective radii of the distributions are comparable (0.43 and 0.33 respectively), F11 has more coarse particles that absorb efficiently in the long wave spectrum. Hence F11's sulfate has a greater stratospheric heating efficacy (i.e. heating per burden) than our sulfate, from more long wave absorption

d. The fourth point is that our aerosol layer is much higher than F11's idealised aerosol layer. Whereas our aerosol concentrations peak at ~30 km altitude, F11's are evenly spaced between the tropopause and 22km altitude. Again, we have the advantage of accurate dynamical transport as we are using a 3-D global model

e. For titania, F11 use the parallel refractive indices, from Ribarsky (1977) whilst we use an average of the parallel and perpendicular (or ordinary and extraordinary – denoting the polarisation of the scattered field). Angus Ferraro (personal communication) reports that, when re-running their FDH code with the perpendicular refractive indices instead of parallel, "the perpendicular refractive indices produce slightly stronger tropical heating"

Although points (a), (b), (d), and (e) will contribute to the difference in stratospheric temperatures (i.e. that our titania produces more warming, whilst their sulfate produces more warming), it is point (c) that is likely most important. We believe that our chosen size-distributions are appropriate for the study here, as Pope et al (2012) try and maximise the impact of a specifically tailored TiO_2 aerosol. We therefore add the following paragraph to the discussion.

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 30% 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 further highlights the sensitivity of climatic effects to the specified aerosol size distribution.

While we are comparing our work with Ferraro et al (2011), it is necessary to highlight major errors within their work that should be taken into account when comparing our results. In a personal communication, Angus Ferraro informed me that the aerosol density was not modified/ considered in either the calculation of the optical properties or the specification of the aerosol layer mass in their work. This is important - the specific absorption/ scattering coefficients plotted in their figure 1 are intrinsic properties; they depend on the density of the aerosol. The assumed density of 1000 kg/m^3 as used by Ferraro et al (2011) for calculation of all their aerosol's optical properties is not appropriate for either sulfate or titania, which should be represented by densities of ~1600 kg/m^3 and ~4000 kg/m^3. Consequently, the coefficients plotted for sulfate and titania in their figure 1 should be multiplied by 1/1.6 and 1/4 respectively to obtain realistic values. Similarly, the mass burdens given in their table 1 for sulfate and titania should be multiplied by 1.6 and 4 respectively to obtain appropriate values. This correction gives forcing efficiencies comparable to ours (see the table below). Should the disparity between the optical coefficients in our figure 1

and theirs be scrutinised, then this is the explanation. We have added the following to the above paragraph.

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.

Table T1	**Sulfate**		**Titania**	
	F11	**This work**	**F11**	**This work**
Burden (Tg)	14.5	49.5	3	20.2
Estimated Forcing (Wm^{-2})	3.5	8.5	3.5	8.5
Forcing efficiency ($Wm^{-2}Tg^{-1}$)	0.24	0.17	1.17	0.42
Corrected burden (Tg)	23.2	49.5	12	20.2
Corrected forcing efficiency ($Wm^{-2}Tg^{-1}$)	0.15	0.17	0.29	0.42

5. P30050 – The reviewer notes that the description we provide for our methods is insufficient. We appreciate that this aspect of the report is under-developed and would benefit from further detail. We therefore describe the exact methods used to obtain injection rates and conduct the simulations, in order that a reader would be able to reproduce these results.

Firstly, as noted by another reviewer, the top of the atmosphere radiative flux imbalance (hereafter *TOA-Imb*) is loosely defined in the report and requires further elaboration. To calculate the TOA-Imb for a certain simulation, we calculate the TOA net radiation (incoming SW minus outgoing LW+SW) and average this annually and globally (denote this value *R(t)* where *t* refers to the year). Next we do the same for each year of the 240-year perpetual pre-industrial control simulation. We then average these values to obtain the net radiative imbalance of the pre-industrial control simulation (denote this *C*). The TOA-Imb for year *t* is calculated as *R(t)* – *C*. The following has been added to the supplement to explain the method.

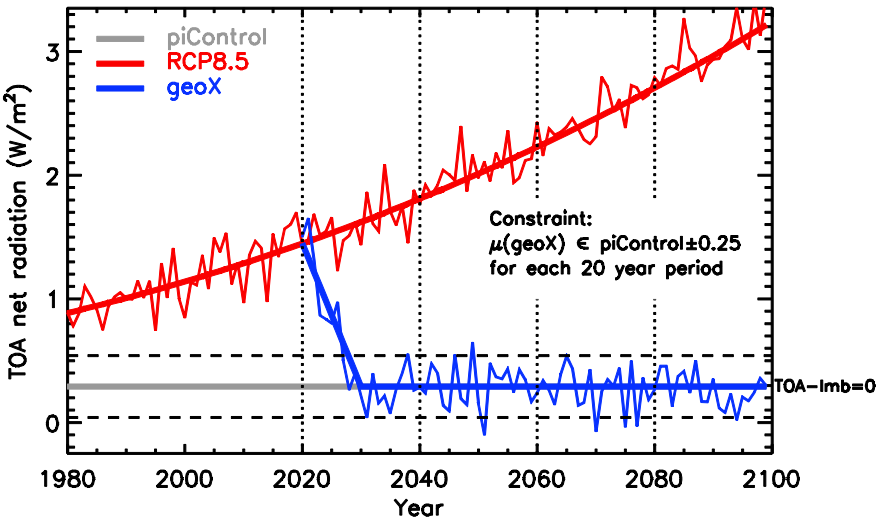


Fig. S2 *Schematic of the geoengineering experiment outline*

We now describe the simulation timeline. The RCP8.5 simulations had already been conducted prior to this investigation. The geoengineering simulations took place in 3 distinct phases: (a) we performed atmosphere-only simulations of 1Tg/yr aerosol injection to determine the aerosol TOA radiative effect; (b) we used the aerosol radiative effect to calculate initial injection rate estimates; (c) we began the 80-year GCM integrations, calibrating the injection rates *en route*.

a. We performed atmosphere-only simulations with a constant 1 Tg/yr aerosol injection rate using historical background-conditions (1990-2005). We then determined the steady-state annual/global-mean aerosol radiative effect (the difference in TOA net radiation between the aerosol simulation and the control), which is given in the following table. For sulfate, because the radiative effect was small, we performed an additional simulation with 5Tg[SO_2]/yr and then divided the results by 5 for precision. Similarly, the

Table T2	Sulfate	Titania	Black Carbon
TOA radiative effect (Wm^{-2} / Tg yr^{-1})	0.46	1.1	7.4

black carbon simulation failed to converge to steady state within 15 years and was therefore run for a further 15 years.

b. Rather than use the TOA-Imb from the RCP8.5 simulations to estimate the required aerosol injection rates, we instead used the Anthropogenic Radiative Forcing (ARF), which was acquired from http://www.pik-potsdam.de/~mmalte/rcps/ (see Meinshausen et al, 2011). Specifically, we deducted the 1860 ARF (0.17 Wm^{-2}) from the ARFs for 2020, 2040, 2060, 2080, and 2100, and then calculated the injection rates required to offset these adjusted ARFs by dividing by the TOA aerosol radiative effect. Because each model will have an ARF which is different from Meinshausen et al (2011) it is possible that our initial estimate is in error. However, our method uses this only as an initial 1st guess for the injection rates, which are iteratively adjusted as described in c). The model then linearly interpolates the injection rates between these years.

Table T3	Anthro-RF (Wm^{-2})		SO_2 injection rate (Tg/yr)		Titania injection rate (Tg/yr)		BC injection rate (Tg/yr)	
Year	Actual	Adjusted	Initial	Final	Initial	Final	Initial	Final
2020	2.56	2.39	5.2	4.4	2.2	1.4	0.32	0.21
2040	3.83	3.66	8.0	7.4	3.3	2.5	0.49	0.29

2060	5.34	5.17	11.2	11.6	4.7	4.1	0.70	0.45
2080	6.79	6.62	14.4	13.6	6.0	4.8	0.89	0.62
2100	8.15	7.98	17.4	14.2	7.3	6.2	1.08	0.88

c. A single simulation was then initiated for each aerosol, with initial injection rates as specified in table T3. After every 20 year interval, the simulation was stopped and the TOA-Imb was calculated for that time period. If there was significant deviation from zero (we adopted |mean(TOA-Imb)|> 0.25 Wm^{-2} as the criterion), then we recalculated the amount of injection required. The recalibration was conducted as follows: the TOA-RF at the end of the 20 year period (time = t_{20}) was calculated for the mean of the RCP8.5 ensemble, denote this R_{rcp}. The injection of aerosol at time t_{20} at rate I_{geo} produced TOA-Imb R_{geo} which we wish to be zero. Therefore an improved injection rate at t_{20} would be $I'_{geo} = I_{geo} \cdot R_{rcp}/(R_{rcp} - R_{geo})$. Additionally, at all specified timesteps after t_{20} ($t_n = t_{20}+20n$, $n = 1,...$), we modify the injection rate as such: $I'_{geo}(t_n) = I_{geo}(t_n) \cdot R_{rcp}/(R_{rcp} - R_{geo})$. After resetting the injection rates, we restarted the simulation from the start of the last time period. Final injection rates are given in table T3. We then used the final injection rates to run two more ensemble members for each aerosol

We agree with the reviewer that it is important to include all of this information in our work so that our results are readily reproducible; therefore we have added this to the supplementary material. We will also the following sentence to section 3, to point the reader to the supplementary material for a detailed methodology.

A detailed description of our methods is provided in the supplementary material (section S2).

We add the following to our Methods section.

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 only signal/noise characteristic.

6. P30057 – The reviewer notes that our analysis pertaining to the hydrological cycle is too inadequate. We have altered the sentences concerning the hydrological cycle to add clarity to our argument.
   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-scattering aerosol such as sulfate, induces a comparatively greater SW forcing at the surface. Bala et al (2008) showed that reduced 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 greater reduction in precipitation exhibited by BC in figures 6-8.

7. P30058 – This query forms two separate parts: (a) what is the likelihood of the BC and titania aerosol being/ becoming hygroscopic? (b) And would this impact the results given here? We assess these questions and summarise our findings for addition to the manuscript.

   a. Firstly, for this analysis we will assume that mineral dust represents titania as this is the most apposite natural proxy [Ndour et al, 2008]. Mineral dust

consists of 1-10% titania by mass depending on location [Ndour et al, 2008] and therefore is arguably suitable for this approximation. The chemical and physical attributes of atmospheric (or at least tropospheric) BC are comparatively well-known [Liu et al, 2013]. Koehler et al (2009) show that most atmospheric mineral dust exhibits low hygroscopicity ($\kappa \sim 0.03$) although larger particles ($r_m > 0.1$ µm) exhibit a similar hygroscopicity to similar sized sea-salt and ammonium sulfate aerosols. Liu et al (2013) show that atmospheric BC generally exhibits low hygroscopicity in its pure form ($g_F \sim 1.05$) but the inclusion of soluble contaminants such as secondary organics increases the hygroscopity ($g_F \sim 1.25$). Therefore, fresh BC and titania particles are likely to exhibit low hygroscopicity and cloud condensation activity, but in an aged form and when internally mixed with hygroscopic species their hygroscopic activity will increase.

b. We apply the hygroscopic growth scheme used for sulfate [d' Almeida et al, 1991] to titania and BC, using the same size-distributions and properties as used in the article, in order to test the sensitivity of the optical properties to hygroscopicity. The 550nm absorption and scattering coefficients for BC are approximately similar for dry aerosol and at < 30% Relative Humidity (RH), but increase by 10% and 40% respectively at 50% RH. For titania, the 550 nm absorption and scattering coefficients are also similar for dry mode and at <30% RH, but increase by 55% and 32% respectively at 50% RH.

To summarise, the hygroscopicity of the BC and titania aerosol would increase over time due to internal mixing with hygroscopic constituents and coagulation, the latter process resulting in larger particles that can act as cloud condensation nuclei (CCN). However, the low relative humidity of the stratosphere (~0.3% [SPARC, 2006]) would result in minimal hygroscopic growth, and therefore our results should not be significantly affected.

Observations have shown that fresh BC aerosol is predominantly hydrophobic, but the uptake of soluble contaminants (e.g. secondary organics) results in increased hygroscopicity [Liu et al, 2013]. Mineral dust, which contains 1-10% titania by mass [Ndour et al, 2008], exhibits low hygroscopicity for radii < 0.1 µm and similar growth to equivalently-sized sulfate aerosol thereafter [Koehler et al, 2009]. Although the historical stratospheric water vapor content is low

(~4.2 ppmv in the tropical lower stratosphere during the HIST period), aerosol-induced stratospheric warming in the TTL would increase the specific humidity of air entering the stratosphere, therefore impacting hygroscopic growth.

8. P30059 – Thanks for the comment, we have added the Tang et al reference to this section.
   This work has already been started for titania by Tang et al [2014].

9. P30059 – We agree with the reviewer that this section is under-developed and with questionable applicability. The NIOSH recommendations refer to particles less than radius < 0.05 µm as 'ultrafine' and < 1.5 µm as 'fine'. For titania, NIOSH recommends time-weighted-average exposure limits of 0.3 mg/m^3 and 2.4 mg/m^3 respectively, allowing for a 10 hour day and 40 hour week. It is highly unlikely that titania injected in the ultrafine mode in the stratosphere will remain in the ultrafine mode until deposition. Therefore, we add the second NIOSH exposure recommendation for fine particles and a caveat.
   ... the USA's National Institute for Occupational Safety and Health (NIOSH) recommends maximum exposure limits of 0.3 mg m^{-3} for ultrafine titania particles (radius <0.05 µm) and 2.4 mg m^{-3} for fine particles (radius < 1.5 µm) [Dankovic et al, 2011]. After undergoing coagulation and ageing in the atmosphere, it is likely that the second exposure limit is more applicable to this work. In our simulations, the maximum 2090's near-surface air concentration of titania (e.g. Fig. 4) for land regions between 60^{o}S-60^{o}N is 254 ng/m^3, which is of the order of 10^2 less than the NIOSH 'fine-particle' exposure limit.

10. P30048 – The reviewer notes that the units for the scattering/absorption coefficients are incorrect. Thanks for the comment - the units have been corrected.

11. P30052 – The reviewer suggests that we add the titania deposition rate (the sulfate and BC deposition rates are already given).Thanks for the comment, the TiO_2 deposition rate has been added (11 mg/m^2/yr).

Anonymous Reviewer #2

1. 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*.

2. 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*.

3. 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. *See response 6 to Anonymous Referee #1*

4. 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.

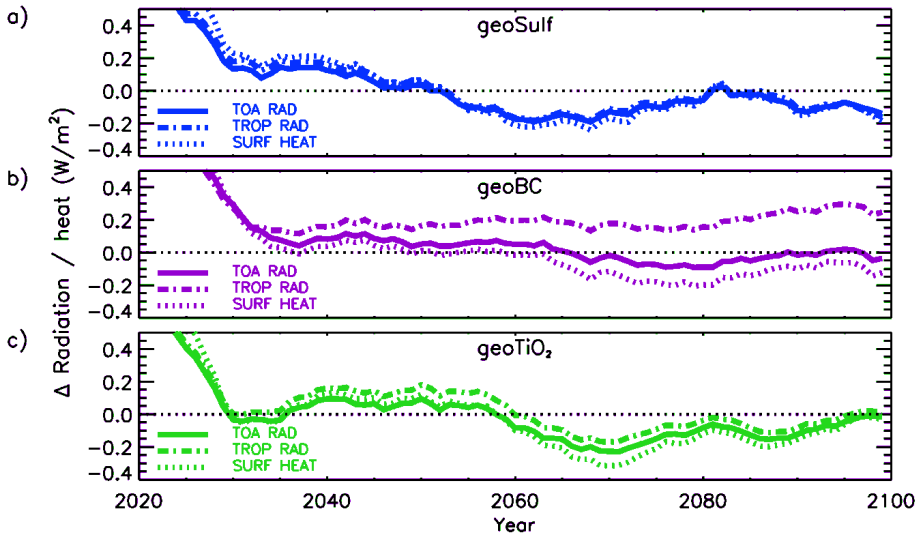


Fig. S3 *10-year running-average global/annual-mean net radiation anomaly at the tropopause and TOA, and net-downward heat flux anomaly at the surface, with respect to piControl. Positive values indicate an increase in net downward flux.*

a. 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.

5. 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 .

6. 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. *See response 4 to Anonymous Referee #1*

7. 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^oS-20^oN 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.

8. 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.

9. 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

10. 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

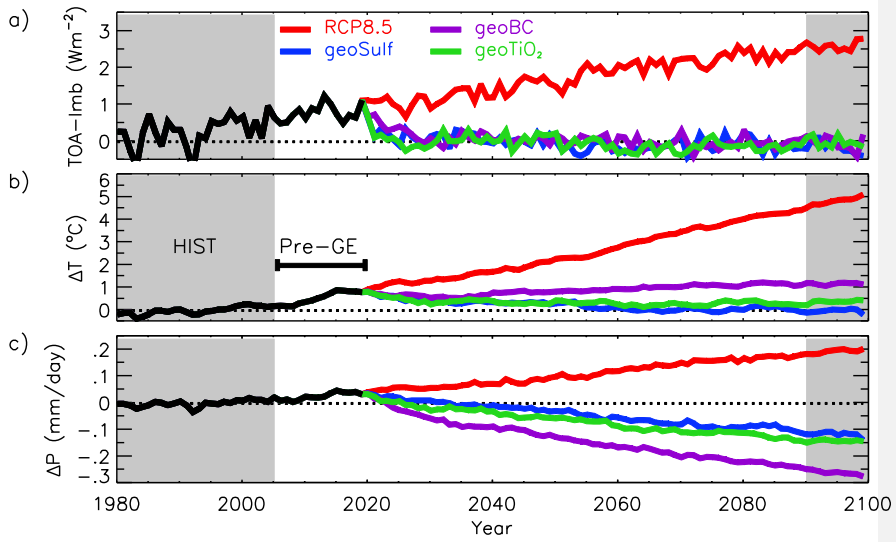
waveband. Further detail is provided in our reply to the anonymous reader #1 (specific response 3). The caption has been altered to include this detail.

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

12. 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).

13. 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].

14. 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%/^{o}C for sulfate, 2.5%/^{o}C for titania, and 4.6%/^{o}C for BC. The hydrological sensitivity for RCP8.5 is 1.32 %/^{o}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%/^{o}C for solar irradiance reduction and 1.4%/^{o}C for CO_2 increase.

15. 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).

16. 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.

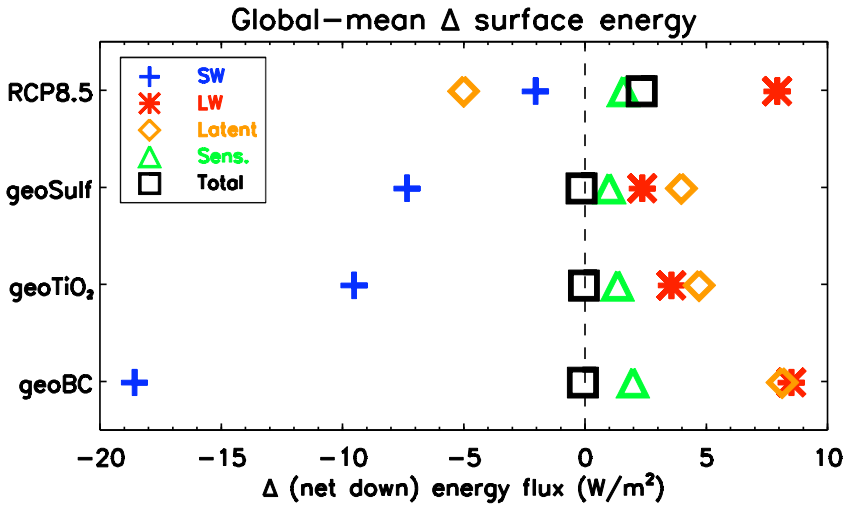


Fig. S6 *2090s global/annual-mean net downward energy flux anomalies at the surface (W/m^2). Calculated with respect to piControl*

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.

17. 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].

18. 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.

19. P56L7 – Tropospheric has been added to tropical circulation

20. 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.

21. 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.

22. 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.

John Dykema

We refer the reader to our specific reply to John Dykema for additional discussion. Below, we highlight the sole change to the manuscript.

1. Precipitation – The reviewerr proposes that we add the model sensitivity into the text for comparison with other GCMs. Thanks for the suggestion, we have added the model sensitivity to the text (section 4.1).

   "The precipitation sensitivity for the model is 1.32 %/^{o}C, which is close to the CMIP5 ensemble-mean [Fig. 12.7 from Collins et al, 2013]."

Climatic Impacts of Stratospheric Geoengineering with Sulfate, Black Carbon and Titania Injection

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Abstract

In this paper, we examine the potential climatic effects of geoengineering by sulfate, black carbon and titania injection against a baseline RCP8.5 scenario. We use the HadGEM2-CCS model to simulate scenarios in which the top-of-the-atmosphere radiative imbalance due to rising greenhouse gas concentrations is offset by sufficient aerosol injection throughout the 2020-2100 period. We find that the global-mean temperature is effectively maintained at historical levels for the entirety of the period for all 3 aerosol-injection scenarios, though there are a wide range of side-effects which are discussed in detail. The most prominent conclusion is that although the BC injection rate necessary to produce an equivalent global mean temperature-response is much lower, the severity of stratospheric temperature changes (> +70 ^{o}C) and precipitation impacts effectively exclude BC from being a viable option for geoengineering. Additionally, while it has been suggested that titania would be an effective particle because of its high scattering efficiency, it also efficiently absorbs solar ultraviolet radiation producing a significant stratospheric warming (> +20 ^{o}C). As injection rates and climatic impacts for titania are close to those for sulfate, there appears to be little benefit in terms of climatic influence of using titania when compared to the injection of sulfur dioxide, which has the added benefit of being well modelled through extensive research that has been carried out on naturally occurring explosive volcanic eruptions.

1. Introduction

The climatic impacts of continued greenhouse gas (GHG) emissions are likely to be severe which has prompted countenance of new strategies for tackling GHG-induced global warming [e.g Collins et al., ~~2014~~2013]. Geoengineering strategies, or large-scale climate interventions that aim to reduce global warming, include strategies to sequester atmospheric carbon dioxide – Carbon Dioxide Removal (CDR) methods, and strategies to reduce solar irradiance at Earth's surface – Solar Radiation Management (SRM) methods [Shepherd et al., 2009]. Stratospheric Aerosol Injection (SAI), an SRM scheme which has received significant attention, involves the enhancement of the stratospheric aerosol layer in order to reflect more sunlight back to space. This scheme mimics large volcanic eruptions such as Mt Pinatubo in 1991, which injected approximately 15-20 Tg of sulfur dioxide (SO_2) into the tropical stratosphere and induced a globally averaged surface cooling of around -0.3 ^{o}C for the following two years [Stenchikov et al., 2002].

Sulfate (SO_4) aerosols have featured predominantly in SAI research because of the volcanic analogue (e.g. in the Geoengineering Model Intercomparison Project, GeoMIP [Kravitz et al., 2013]). General Circulation Model (GCM) simulations suggest that, while sufficient sulfate injection could effectively reduce global-mean temperature, possible side effects include changes to regional precipitation [e.g. Bala et al., 2008; Tilmes et al., 2013], ozone [e.g. Tilmes et al., 2009; Pitari et al., 2014], stratospheric dynamics [Aquila et al., 2014] and sea-ice extent [Berdahl et al., 2014]. Precipitation changes could result from changes to the moist static stability of the atmosphere and a concomitant weakening of the hydrological cycle [Bala et al., 2008], and the regional precipitation changes under GeoMIP simulations have been shown to be reasonably consistent across a range of climate models [Tilmes et al., 2013]. Ozone concentrations could change as a result of enhanced heterogeneous chemistry on the surface of sulfate aerosols or indirectly by changes to the stratospheric dynamics and chemistry [e.g. Tilmes et al., 2009]. Stratospheric dynamical changes could occur as the result of tropical heating in the sulfate layer and by changes to wave propagation from the troposphere [e.g. Aquila et al., 2014].

In order to ameliorate the known side-effects of sulfate injection, some authors have proposed alternative aerosols to sulfate [e.g. Teller et al., 1997]. Crutzen (2006) suggested the possible injection of black carbon (BC), which would mimic hypothetical nuclear winter scenarios. One advantage of BC over sulfate is that less mass would be needed for an equivalent radiative forcing [Crutzen, 2006]. BC particles efficiently absorb solar radiation, unlike

sulfate which primarily reflects solar radiation [Ferraro et al., 2011]. Alternatively, minerals such as titania (TiO_2), silica (SiO_2) and alumina (Al_2O_3), which have a high refractive index at wavelengths of peak solar radiative flux (~550 nm), have also been suggested [Pope et al., 2012]. Although the use of alternative aerosols is not a new suggestion [e.g. Teller et al., 1997], comparatively little research has been conducted on their potential utility. Kravitz et al (2012) simulated a constant BC injection scenario of 1 Tg/yr in the tropics for small radius (0.03 μm) and large radius (0.15 μm) aerosols. They found that the small particle BC aerosol scenario produced a global surface cooling of -9.45 ^{o}C, but also induced stratospheric warming > +60 ^{o}C and global ozone loss of 50%. The large particle BC aerosol scenario had a neglible climatic impact. Using a fixed dynamical heating (FDH) code, Ferraro et al (2011) compared the stratospheric heating of sulfate, titania, and BC layers for an equivalent instantaneous radiative forcing. Their results showed a tropical stratospheric warming signal for all the aerosols, though much greater in the case of BC. To date, no work has used a comprehensive fully coupled atmosphere-ocean GCM to directly compare the possible climatic impacts of SAI with alternative aerosols to sulfate, which is the motivation for this research.

In this work, we simulate the stratospheric injection of sulfate, titania and BC against a baseline RCP8.5 concentrations scenario using a fully-coupled GCM. Titania is selected to represent an efficient light-scattering aerosol and BC is selected as a light-absorbing aerosol. RCP8.5, which is the high-end carbon-intensive CMIP5 scenario, ~~RCP8.5~~ is selected to give a significant greenhouse effect against which to employ geoengineering, in order to distinguish the climatic impacts specific to each aerosol. Observations have shown that the current global GHG emissions exceed the emissions inherent in RCP8.5 [Peters et al., 2013]; therefore our work could be considered as geoengineering against a business-as-usual scenario. Additionally, the next generation of GeoMIP simulations (GeoMIP6) will utilise a carbon-intensive scenario [Kravitz et al., 2015], hence our work will provide a useful supplement to those results. We chose to inject aerosol at a sufficient rate to counterbalance the Top Of the Atmosphere (TOA) global/annual-mean radiative flux~~Radiative Flux (TOA-RF)~~ imbalance caused by increasing atmospheric GHGs. Our simulation design is similar to the G3 scenario of the Geoengineering Model Intercomparison Project (GeoMIP), which instead used the RCP4.5 concentrations scenario as its baseline and injected sulfate at a sufficient rate to counterbalance GHG radiative forcing [Kravitz et al., 2011]. We analyse the

climate changes in the 2090s with respect to a simulated historical period and discuss impacts on a wide range of meteorological parameters.

Model

2.1. The HadGEM2-CCS model

For this investigation, we use the HadGEM2-CCS climate model in a fully coupled atmosphere-ocean mode. HadGEM2-CCS is the high-top configuration of the HadGEM2 family of models, and includes a well-resolved stratosphere. The atmosphere component comprises 60 vertical levels extending to 84km and a horizontal resolution of 1.25^{o} x 1.875^{o} latitude by longitude respectively. The 40-level ocean component has a horizontal resolution of 1^{o} by 1^{o} from the poles to 30^{o}N/S, with the latitudinal resolution then increasing smoothly to 0.33^{o} at the equator [The HadGEM2 Development Team, 2011]. For this investigation, GHG concentrations, stratospheric ozone, anthropogenic aerosols and aerosol precursor gases are prescribed following the Coupled Model Intercomparison Project phase 5 (CMIP5) [Taylor et al., 2012] protocol, with historical data from 1860-2005 and RCP8.5 concentrations from 2005-2100. HadGEM2-CCS contains the aerosol module Coupled Large-scale Aerosol Simulator for Studies in Climate (CLASSIC). The module's sulfur cycle is described in detail in Bellouin et al (2011). Briefly, it includes the oxidation of sulfur dioxide (SO_2) to sulfate aerosol in aqueous and gas phase reactions. Sulfate is represented by Aitken, accumulation and dissolved modes, with hygroscopic growth in the accumulation mode following d'Almeida et al (1991). Aerosol size modes are represented by lognormal size-distributions with a prescribed dry-mode median radius (r_m) and geometric standard deviation (σ).

2.2 Stratospheric aerosol microphysical and optical properties

For this investigation, stratospheric sulfate is modelled using the *volc2* size-distribution from Rasch et al (2008) for the sulfate accumulation mode, with r_m = 0.376 μm and σ = 1.25; the relatively large r_m is chosen to reflect the high concentrations of SO_2 injected in this experiment. ~~Hygroscopic growth is parameterised following Deepak and Gerber (1983).~~

CLASSIC includes a tropospheric BC scheme with fresh, aged and in-cloud modes [Bellouin et al, 2011]. We introduce an additional non-hygroscopic stratospheric BC component and prescribe a lognormal size-distribution with r_m = 0.0118 μm and σ = 2.0, which is taken from tropospheric BC observations [Deepak and Gerber, 1983]. We prescribe a density for BC of 1000 kg/m^3 and take refractive indices from a World Meteorological Organisation report [Deepak and Gerber, 1983].

For stratospheric titania, we assume the non-hygroscopic lognormal size distribution of Pope et al. (2012) with r_m = 0.045 μm and σ = 1.8. This size-distribution was selected to give the titania aerosol a high scattering efficiency, as shown by Pope et al (2012). We prescribe a density for titania of 4230 kg/m^3 [Pope et al, 2012], and for the refractive indices we follow Ferraro et al (2011) and use the average of the extra-ordinary and ordinary values from Ribarsky (~~1984~~1985).

The specific absorption (k_{abs}) and scattering (k_{sca}) coefficients for sulfate (accumulation/dry-mode), titania and BC are plotted in Fig. 1 as a function of wavelength. For sulfate, the specific extinction coefficient (k_{ext}) at 500nm of 3200 m^2/kg ~~kg/m^3~~ and single scattering albedo (ω_o) of 1 reflects the non-absorbing properties of sulfate. Although titania's 500nm scattering efficiency (k_{sca} = 3850 m^2/kg~~kg/m^3~~) is greater than sulfate's in this instance, titania additionally absorbs SW radiation (k_{abs} = 2000 m^2/kg~~kg/m^3~~ at 250 nm, and k_{abs} = 600 m^2/kg~~kg/m^3~~ at 500 nm) which can be explained by the band-theory of solids [Yang et al, 2003]. Thus titania is partially absorbing. Our modelled BC efficiently absorbs SW radiation (k_{abs} = 8300 m^2/kg~~kg/m^3~~ at 500nm) but also produces a non-negligible SW scattering effect (k_{sca} = 2500 m^2/kg~~kg/m^3~~ at 500nm) which is comparable in magnitude to the equivalent scattering efficiency of both titania and sulfate. Therefore, to describe titania as an efficient light-scatterer and/or BC as an efficient light-absorber is an over-simplification.

Our choice of particle size and density will impact the aerosol's gravitational sedimentation rate and therefore its atmospheric residence time (the sedimentation rate is also a property of the local atmospheric conditions) [Rasch et al., 2008]. To determine the importance of our choice of aerosol properties, we have calculated the respective gravitational sedimentation rates by using the method of Pruppacher and Klett (1979) (which utilises Stoke's law) and incorporating temperature and pressure values from the International Standard Atmosphere [ICAO, 1993] (~~Supplementary~~ Fig. S1 in the Supplement~~1~~). We find that the average sedimentation rates between 18-26 km altitude for our prescribed sulfate, titania, and BC are

23, 9.5 and 0.75 m/day respectively, and the equivalent rates between 26-30 km are 52, 22, and 1.8 m/day. Therefore, one would expect BC to be advected to much higher altitudes than sulfate in these simulations. For perspective, Schoeberl et al (2008) deduced from observations that the atmospheric tropical vertical velocity between 18-26 km has an upper limit of 35 m/day, and the equivalent velocity between 26-30 km is below 61 m/day.

Method

We first validated the model's stratospheric sulfate scheme by simulating the Mt Pinatubo eruption and then comparing the results with observations. These simulations comprised a 10-member ensemble in which 20 Tg[SO_2] is injected between 16-18 km over a single day in June 1991, following the method of Aquila et al (2012). Figure 2a shows the global/annual-mean sulfate aerosol optical depth (AOD) anomaly for the HadGEM2-ensemble and for AVHRR and SAGE-II observations. The model clearly captures the peak AOD from the AVHRR data, and the exponential decline thereafter. Figures 2b-d show the zonal-mean AOD anomaly for the same time period. The agreement between the model and observed AOD is reasonable. Some differences in the temporal evolution of the AODs in the model and the observations are due to the almost concurrent eruption of Cerro Hudson which injected approximately 3.3Tg[SO_2] into the southern hemisphere [Deshler and Anderson-Sprecher, 2006]. This relatively close agreement between observations and HadGEM2 estimates, together with other modelling studies of other volcanic eruptions [Haywood et al., 2010] suggests that the model is a useful tool for stratospheric geoengineering simulations.

The geoengineering investigation was based on a 240-year Pre-Industrial Control simulation (forced by constant 1860's GHGs and aerosol emissions) and historical simulations for the period 1860-2005 following CMIP5 [Taylor et al., 2012] protocol followed by RCP8.5 emission specified from 2005-2019. Leading on from these simulations, we performed 3-member ensembles for the period 2020-2100 for: RCP8.5 only, RCP8.5 with SO_2 injection (~~G3S~~geoSulf), RCP8.5 with TiO_2 injection (geoTiO_2~~G3TiO_2~~), and RCP8.5 with BC injection (~~G3BC~~geoBC). ~~The G3 nomenclature is adopted because of the similarity of our simulations to the G3 experiments of GeoMIP although the original G3 experiments were compared against RCP4.5 and the geoengineering period was terminated at 2070 [Kravitz et al., 2011].~~ Aerosol (or gaseous SO_2 for the ~~G3S~~ geoSulf scenario) was injected at a constant rate between 23-28 km altitude in a single vertical column at the equator. The injection altitude

and location were chosen to ~~prolong~~ maximise the stratospheric lifetime of the aerosol, which is transported poleward by the upper branch of the Brewer-Dobson circulation [Niemeier et al., 2011], and therefore make the geoengineering approach reasonably efficient.

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.

To determine the injection rates required to maintain TOA-Imb~~RF~~ balance, we first conducted ~~10~~15-year atmosphere-only simulations of 1 Tg aerosol (or SO_2 for sulfate) injection per year to calculate the specific radiative effect for each aerosol. We then used the radiative effect to calculate the injection rate necessary to offset the RCP8.5 anthropogenic radiative forcing (ARF) for the 2020-2100 period (with ARF values from Meinshausen et al (2011)). We used the ARF to estimate the injection rates required to produce TOA-Imb=0 as this produces reasonable initial injection rates. As the geoengineering simulations progressed, we altered the injection rate when necessary to ensure that TOA-~~RF~~ Imb balance was maintained~~.~~ (Fig. S2 in the Supplement). A detailed description of our methods is provided in the supplementary material (Section S2).

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Our analysis focuses initially on the temporal evolution of the TOA ~~RF~~ Imb and global mean temperature changes to show that our simulations provide plausible counterbalances to global mean temperature changes under RCP8.5. However, our main focus is on the differences between ~~the~~ a recent historical period (1980-2005) (hereafter denoted HIST) and the geoengineering experiments during the period 2090-2100, with an emphasis on different geographical patterns. 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 (1986-2005) [e.g. Fig. 12.10 from Collins et al., 2013] which facilitates comparison of our RCP8.5 climate changes with the CMIP5 multi-model means.

Results

4.1 Effectiveness at maintaining global mean TOA ~~RF~~ Imb and near surface temperature

Figure 3 shows the global/annual-mean TOA ~~RF~~ Imb ~~imbalance~~ and near-surface air temperature anomaly for the geoengineering and RCP8.5 simulations, with respect to the HIST period. For all of the geoengineering simulations we were able to maintain TOA Imb≈0 ~~RF balance~~ for the entirety of the 80-year period (Fig. 3a). For geoSulf, geoTiO$_2$ ~~G3S, G3TiO$_2$~~ and ~~G3BC~~geoBC, the TOA ~~RF~~ Imb was maintained within +/-0.21, +/-0.18 and +/-0.20 Wm^{-2}, respectively (1 standard deviation throughout the 2020-2100 period).

~~However, the near-surface global temperature response differs between the aerosols with a greater cooling effect for sulfate than for titania or BC. 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 RF that is proportional to global mean surface temperature changes. Further analysis of stratospheric temperature changes will be provided in section 4.4.~~

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

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anomalous warming in geoBC, we assess the net radiation 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. 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.

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 unit temperature change, is 2%/^{o}C for sulfate, 2.5%/^{o}C for titania, and 4.6%/^{o}C for BC. The hydrological sensitivity for RCP8.5 is 1.32 %/^{o}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.

4.2 Aerosol distribution

The time-averaged injection rates for the 2090s period are 14 Tg[SO_2]/yr, 5.8 Tg/yr and 0.81 Tg/yr for geoSulf, geoTiO_2 ~~G3S, G3TiO_2~~ and ~~G3BC~~geoBC, respectively. This SO_2 injection rate is approximately equivalent to 1 Mt Pinatubo eruption per year [Dhomse et al, 2014]. These injection rates equate to global aerosol mass-burden anomalies of 49.5, 20.2, and 5.1 Tg for geoSulf, geoTiO_2 ~~G3S, G3TiO_2~~ and ~~G3BC~~geoBC, respectively. The ~~G3BC~~ geoBC mass burden is comparable to the equilibrium burdens of the high-altitude (HA) and small-radius (SmR) experiments from Kravitz et al (2012), although they injected BC at a constant rate of 1 Tg/yr, around 20% higher than in our study. Figure 4 shows the 2090s annual, June-July-August (JJA) and December-January-February (DJF) aerosol mass concentration anomalies (annual mean aerosol optical depths are shown in ~~Supplementary~~ Fig. S4 in the Supplement ~~2~~). Peak sulfate concentrations are found at the injection region at the equator (Figs. 4a,d,g) and over the winter pole. Titania and BC reach greater altitudes than sulfate (>50 km), which is due to their smaller size-distributions and self-lofting from SW-absorption [Kravitz et al, 2012]. While sulfate aerosol concentrations are highest at the equator, the highest concentrations of BC are found in the polar stratosphere. This is because the larger particle size of the sulfate aerosol is subject to a larger sedimentation velocity (~~see Supplementary~~ Fig. S1 in the Supplement ~~1~~) and thus a greater fraction of aerosol is removed close to the source region. The results from titania suggest a spatial distribution intermediate between sulfate and BC owing to the intermediate size distribution.

Figure 5 shows the total annual, JJA and DJF aerosol deposition anomalies averaged over the 2090s (the seasonal cycle of the deposition anomalies are shown in ~~Supplementary~~ Fig. S5 in the Supplement ~~3~~). Sulfate is predominantly deposited in the Northern Hemisphere (NH) extratropics in the boreal spring and summer (Fig. 5d) which is likely attributable to tropopause fold events in the lower branch of the Brewer-Dobson circulation (BDC) [Kravitz et al., 2012]. In contrast, Titania and BC are primarily deposited at high latitudes in the polar winter, which is attributable to the diabatic descent of air in the deep branch of the BDC [e.g. Tegtmeier et al., 2008]. Kravitz et al (2012) also found in their SmR experiment that BC deposition was limited to the polar regions, but their maximum deposition was during polar

summer rather than polar winter. The global/annual-mean deposition rates of sulfate and BC from geoengineering are 37 and 1.5 mg/m^2/yr, respectively. These amounts may be compared with 231 and 12.7 mg/m^2/yr from non-geoengineering sources, amounting to increases of 16 % and 12 % respectively. The global/annual-mean deposition rate for titania is 11 mg/m^2/yr.

4.3 Temperature and precipitation

Figure 6 shows the annual mean near-surface air temperature (Figs. 6a-d) and precipitation anomalies (Figs. 6e-h) with respect to HIST. 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 ~~positive snow~~surface-albedo feedback [e.g. Kharin et al., 2013]. This results in an annual mean warming of +11.3 ^{o}C averaged over the Arctic region (> 60 ^{o}N) and an average NH land warming of +7.3 ^{o}C. This figure provides an alarming picture of the change in global mean temperature by the end of this century should global society follow the RCP8.5 (essentially a business as usual) pathway. All 3 SAI experiments produce a surface-cooling with respect to RCP8.5, with ~~G3S~~ geoSulf exhibiting the greatest global-mean cooling effect of -4.85 ^{o}C, considering TOA-Imb is balanced for each geoengineering experiment. The latitudinal distribution of cooling varies markedly between the SAI experiments, with relative tropical cooling for ~~G3S~~ geoSulf and geoTiO$_2$~~G3TiO$_2$~~ (Figs. 6b,d) and polar cooling for ~~G3BC~~ geoBC (Fig. 6c). Defining the 'SAI cooling effect' as the temperature difference between SAI and RCP8.5, the ratio of cooling effect at high latitudes (> 60^o) between ~~G3BC~~ geoBC and ~~G3S~~ geoSulf is 1.19 and between ~~G3BC~~ geoBC and ~~G3TiO$_2$~~ geoTiO$_2$ is 1.23. In the tropics and mid-latitudes (< 60^o) the equivalent ratios are 0.64 and 0.71 respectively. The high-latitude cooling in the case of ~~G3BC~~ geoBC is attributable to the zonal distribution of BC (Figs. 4c,f,i) which is more evenly spread over the stratosphere than for ~~G3S~~ geoSulf and ~~G3TiO$_2$~~geoTiO$_2$. The result is a greater surface SW forcing at high-latitudes in the summer hemisphere for ~~G3BC~~geoBC. For instance, in the Arctic (>60^{o}N) in JJA, the surface SW forcing is -25.65 Wm^{-2} in ~~G3BC~~ geoBC and -3.3 and -6.55 Wm^{-2} in ~~G3S~~ geoSulf and ~~G3TiO$_2$~~ geoTiO$_2$ respectively.

Although the global-mean precipitation rate increases for the RCP8.5 scenario (Fig. 6e), certain regions such as the Amazon basin exhibit a drying trend. This is in line with the CMIP5 multi-model projections documented in the Intergovernmental Panel on Climate Change 5th assessment report (IPCC AR5) [e.g. Fig. 12.~~7~~10 from Collins et al., 2013]. All of

the SAI experiments show a global-mean precipitation reduction with respect to both HIST and RCP8.5 (Figs. 6f-h), which is due to the deceleration of the hydrological cycle and is a robust model response to SAI [e.g. Yu et al., 2015; Tilmes et al., 2013; Bala et al., 2008]. The magnitude of the precipitation changes are greater for ~~G3BC~~ geoBC than for ~~G3S~~ geoSulf or ~~G3TiO2~~geoTiO$_2$; for instance, the global mean precipitation anomaly is -0.26 mm/day for ~~G3BC~~ geoBC compared to -0.12 mm/day for ~~G3S~~ geoSulf and -0.14 mm/day for ~~G3TiO2~~geoTiO$_2$. ~~This is because the stratospheric heating in G3BC applies an additional LW forcing at the tropopause and TOA which must be ameliorated by additional SW absorption in order to maintain radiative balance [Ferraro et al, 2011]~~. In order to maintain TOA-Imb=0, BC must produce a greater SW perturbation at the tropopause and at the TOA than sulfate or titania, which is compensated by the increased LW perturbation resulting from stratospheric warming. The troposphere is relatively transparent to SW radiation but absorbs efficiently in the LW spectrum, therefore the annual-mean surface radiative forcing in the ~~G3BC~~ geoBC experiment is greater (−18.6 ~~-10.2~~ Wm^{-2}) than for ~~G3S~~ geoSulf or geoTiO$_2$~~G3TiO2~~ (~~-5.1~~ -7.4 and ~~-6.06~~ -9.6 Wm^{-2} respectively - see ~~Supplementary~~ Fig. S6 in the Supplement ~~4~~). Bala et al (2008) showed that the magnitude of the precipitation response is dependent on the surface radiative imbalance; therefore the precipitation reduction is amplified in ~~G3BC.~~geoBC. 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$_2$. From Fig. S6 in the Supplement, 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 W/m^2 while our study is consistent at a value of -5.4 W/m^2 (not plotted). However, in all geoengineering cases, this reduction is comprehensively overwhelmed by aerosol direct effects.

Figure 7 shows the JJA temperature (Figs. 7a-d) and precipitation (Figs. 7e-h) anomalies. In the ~~G3S~~ geoSulf and geoTiO$_2$~~G3TiO2~~ scenarios, the temperature is effectively maintained at HIST levels (Figs. 7b,d). However, a slight bias towards high-latitude NH warming in ~~G3S~~ geoSulf and geoTiO$_2$~~G3TiO2~~ results in a northward displacement of the Inter-Tropical Convergence Zone (ITCZ), which is exemplified by the Sahelian precipitation increase in Figs. 7f,h. This phenomenon was noted by Haywood et al (2013) and has been observed after large hemispherically asymmetric volcanic eruptions [~~(~~Oman et al., 2006]~~)~~. Although the

general pattern of precipitation change is similar for the 3 SAI scenarios, ~~G3BC~~ geoBC again displays a greater drying signal, with 80% of the total land area experiencing a JJA precipitation reduction in ~~G3BC~~ geoBC compared to 70% for $geoTiO_2$ ~~$G3TiO_2$~~, 57% for ~~G3S~~ geoSulf and 52% for RCP8.5.

Figure 8 shows the DJF temperature (Figs. 8a-d) and precipitation (Figs. 8e-h) anomalies. The temperature reduction over Greenland in ~~G3BC~~ geoBC (Fig. 8c) is due to the significant decrease in downwelling SW radiation at the surface during the Arctic sea-ice formation season (September-October-November), which leads to a positive sea-ice albedo feedback and further localised cooling. This inference is corroborated by Fig. 9, which shows the Arctic DJF sea-ice extent in terms of the average DJF sea-ice boundary (the Antarctic DJF sea-ice extent is shown in ~~Supplementary~~ Fig. S7 in the Supplement ~~5~~). The sea-ice boundary in ~~G3BC~~ geoBC (Fig. 9c) extends to well below Greenland, and the total sea-ice extent anomaly is +1.72 million km^2 which vastly exceeds the HIST standard deviation of +/- 0.52 million km^2. In comparison, the sea-ice extent anomaly of -11 million km^2 for RCP8.5 (Fig. 9a) marks a reduction by 43% of the total HIST sea-ice extent. Returning to Fig. 8, the poleward shift in the NH extratropical rain-belt over the Atlantic in RCP8.5 (Fig. 8e) is a robust result of GHG-induced global warming and is related to storm track displacement [Lombardo et al, 2015]. This same response is evident in the geoengineering simulations (Figs. 8f-h), although to a much lesser extent in ~~G3S~~ geoSulf and $geoTiO_2$ ~~$G3TiO_2$~~.

4.4 Stratospheric changes

Figure 10 shows the zonal-mean temperature change as a function of latitude and altitude for the JJA and DJF seasons. The stratospheric cooling in conjunction with tropospheric warming in RCP8.5 (Figs. 10a,e) is a robust result of increasing GHG-concentrations [e.g. Schmidt et al., 2013]. Aerosols directly affect temperature by absorbing radiation, and indirectly by scattering radiation and by ambient dynamical and chemical changes [Carslaw and Kärcher., 2006]. ~~-~~ Sulfate predominantly absorbs in the LW and near-infrared spectrum (Fig. 1a)~~, therefore the stratospheric radiative heating in G3S is mostly confined to the tropical region, where the stratosphere is significantly colder than the underlying warm troposphere [Ferraro et al, 2011].~~ . 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]. In contrast, titania and BC absorb in both the SW and LW spectrum (Figs. 1b,c), and therefore preferentially warm the summer-hemisphere and tropical stratosphere, where solar radiation is most prevalent. ~~G3BC~~ geoBC produces the most significant warming effect, with an average stratospheric (15-50 km altitude) temperature increase of +33 ^{o}C and a maximum temperature increase of +68 ^{o}C, which occurs in JJA (Figs. 10c,g). The maximum BC-induced heating relative to the baseline RCP8.5 scenario is +76 ^{o}C (~~Supplementary~~ Fig. S8 in the Supplement~~6~~), which is comparable to the ~80 ^{o}C temperature change Kravitz et al (2012) found in their SmR scenario. For comparison, the maximum sulfate-induced and titania-induced heating relative to RCP8.5 are far more modest at +7 ^{o}C and +22 ^{o}C, respectively.

A warming of the lower tropical stratosphere could have multiple climatic repercussions such as a weakening of the tropospheric tropical circulation [Ferraro et al., 2014], strengthening of the polar vortex [Driscoll et al., 2012] and modification of the Quasi-Biennial Oscillation (QBO) [Aquila et al., 2014]. 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.

A strengthening of the polar vortex could be instigated by an increased temperature gradient between the tropical/mid-latitude and polar stratospheres, a phenomenon which was observed after the Pinatubo eruption [Stenchikov et al., 2002]. We concentrate on the Arctic wintertime (DJF) response to SAI, and adopt a similar metric to that used by Ferraro et al (2011) to determine the stratospheric temperature gradient. Explicitly, we determine the difference in temperature between 20^{o}N-20^{o}S (Tropics) and 50^{o}N-90^{o}N (North Pole) at 17-22 km altitude in the DJF season. Using this metric, the change in temperature gradients for ~~G3BC~~geoBC, ~~G3S~~ geoSulf and geoTiO_2~~G3TiO_2~~ are +10.4 ^{o}C, +7 ^{o}C, and +10.1 ^{o}C, respectively, indicating a steeper temperature gradient between the tropics and poles. Additionally, Fig. 11 shows the

50hPa DJF geopotential height anomalies over the Arctic for RCP8.5 and the 3 SAI experiments. The negative geopotential height anomaly centered over the North Pole in all the SAI experiments is indicative of a strengthened polar night jet and a positive Arctic Oscillation phase [Stenchikov et al, 2002]. The DJF zonal-mean zonal-wind anomaly (~~Supplementary~~ Fig. S9 in the Supplement ~~7~~) substantiates our inference of a strengthened polar-night jet under SAI, with increased zonal windspeeds at 65^{o}N / 40km altitude of 62 m/s, 17 m/s, and 37 m/s for geoBC, geoSulf~~G3BC, G3S~~, and geoTiO$_2$~~G3TiO$_2$~~ respectively.

The Quasi-Biennial Oscillation (QBO) is a periodic change in the equatorial zonal wind pattern in the stratosphere, which fluctuates between easterly and westerly-shear phases [Baldwin et al., 2001]. Aquila et al (2014) showed that radiative heating in the aerosol layer could prolong the westerly-phase of the QBO (where the phase is defined at 40 hPa) by enhancing the residual-mean upwelling motion and strengthening the westerly winds. HadGEM2-CCS includes a non-orographic gravity wave scheme that permits the model to internally generate a QBO and is therefore capable of assessing QBO changes [The HadGEM2 Development Team, 2011]. The average QBO period for the HIST-era ensemble is 27 months (~~Supplementary~~ Fig. S10 in the Supplement~~8~~) which agrees closely with observations [e.g. Baldwin et al., 2001]. Figure 12 shows the 2090s QBO timeseries for one ensemble member of the RCP8.5 and SAI experiments (~~Supplementary~~ Figs. S11a,~~9a,~~b in the Supplement ~~-~~show the QBO timeseries for the other 2 ensemble members). The average QBO periods for this timespan, which are determined using all 3-ensemble members, are 20 months for RCP8.5, 31 months for ~~G3S~~ geoSulf and 36 months for geoTiO$_2$~~G3TiO$_2$~~. For ~~G3BC~~geoBC, the no QBO-like oscillation can be detected in the 10-year time span,~~periodicity of the QBO extends beyond the 10-year span considered here,~~ suggesting a persistent westerly-phase such as observed by Aquila et al (2014) in their $G_5^{22\text{-}25km}$ scenario. In their HadGEM2-CC simulations, Kawatani and Hamilton (2013) also observed a decline in the QBO period for the RCP8.5 scenario, although they were unable to provide a reason for this. A robust inference from this work is that the magnitude of SAI's impact on stratospheric zonal winds correlates with the magnitude of the stratospheric warming.

Discussion

In this work, we have assessed the climatic impacts of sulfate, black carbon and titania-injection against a baseline RCP8.5 scenario, by comparing the 2090s climate with a

simulated historical period. We have shown that, although the distribution of climate changes are similar for the 3 SAI scenarios, the magnitude 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. Additionally, we have shown that ~~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-scattering aerosol ~~scatterer~~ such as sulfate, induces a comparatively ~~produces a~~ greater SW forcing at the surface. Bala et al (2008) showed that reduced 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.~~which could further disrupt the hydrological cycle beyond the hydrological perturbation expected for sulfate injection~~. The ~~G3BC~~ geoBC scenario displays a greater cooling at high-latitudes than the ~~G3S~~ geoSulf and geoTiO$_2$ ~~G3TiO$_2$~~ scenarios (Figs. 6-8), which comparatively exhibit a net tropical cooling. This raises the question of whether a combination of aerosols could potentially be injected to produce a zonally-homogeneous cooling if necessary. Although SAI with sulfate and titania effectively maintains the regional distribution of temperature at HIST levels, with a slight residual warming at high latitudes, the hydrological cycle decelerates substantially in all SAI scenarios which is exemplified by a global-mean reduction in precipitation. However, annual-minimum sea-ice extent in both hemispheres and global-mean thermosteric sea-level (~~Supplementary~~ Fig. S12 in the Supplement) are ~~10) is~~ almost entirely maintained at HIST levels for all SAI scenarios.

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 effects 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.

It is important to note that the climate impacts described in section 4 ~~above~~ are dependent on the optical properties of the aerosol, which are further dependent on the aerosol particle's size, shape, and composition [e.g. Kravitz et al., 2012]. In this investigation, the dry-mode size distribution of the aerosol species is held constant~~,~~ and hygroscopic growth is not represented in the BC and titania schemes, nor are the effects of internal mixing represented. Observations have shown that fresh BC aerosol is predominantly hydrophobic, but the uptake of soluble particulates (e.g. secondary organics) results in increased hygroscopicity [Liu et al., 2013]. Mineral dust, which contains 1-10% titania by mass [Ndour et al., 2008], exhibits low hygroscopicity for radii < 0.1 μm and similar growth to equivalently-sized sulfate aerosol thereafter [Koehler et al., 2009]. Although the historical stratospheric water vapor content is low (~4.2 ppmv in the tropical lower stratosphere during the HIST period), aerosol-induced stratospheric warming in the TTL would increase the specific humidity of air entering the stratosphere, therefore impacting hygroscopic growth. The injection of aerosol into pre-existing aerosol layers would lead to larger particles through coagulation and condensation, which would further ~~alters~~ alter the aerosol's optical and physical properties. The actual size of the aerosol in an SAI scheme would therefore depend on the injection strategy (e.g. location/ season) and the size and composition of the injected species [e.g. Carslaw and Kärcher, 2006; Heckendorn et al., 2009]. Recent research from Heckendorn et al (2009), Pierce et al (2010), English et al (2012), and Weisenstein et al (2015) have highlighted the importance of representing aerosol growth in SAI simulations. A detailed assessment of the aerosol microphysics for sulfate, BC, and titania injection is not within the scope of this paper, but presents an important subject for future work.

The climatic impacts described in section 4 are specific to geoengineering against a baseline RCP8.5 scenario. If instead we had used a middle-of-the-road GHG-concentrations scenario such as RCP4.5 [Taylor et al., 2012], as used in the first tier of GeoMIP scenarios [Kravitz et al., 2011], then less aerosol-injection would be needed to obtain TOA-Imb=0 and therefore the aerosol deposition rates and atmospheric mass concentrations would be less than those reported in section 4. One would expect that the magnitude of stratospheric temperature changes (Fig. 8) and therefore zonal-mean zonal wind changes (Fig. 12) would be much less for each of the aerosols, possibly confounding the conclusions giving here relating to their comparative efficacy. An estimate for the amount of SAI required for RCP4.5 can be garnered from integrating the temperature anomalies for RCP8.5 and RCP4.5 for the period 2020-2100. The ratio of the integrated temperature anomalies for RCP4.5 to RCP8.5 is 0.43, hence we can assume that the injection rates required for RCP4.5 are ~0.43 of those for RCP8.5, producing a climate perturbation ~0.43 times as great. A further set of simulations, which instead utilise RCP4.5 as the baseline scenario, would be required to test this hypothesis.

We have used prescribed ozone fields in these simulations because representing stratospheric chemistry is prohibitively computationally expensive for the multiple centennial simulations performed here [The HadGEM2 development team, 2011]. Kravitz et al (2012) showed that BC injection could potentially result in global ozone depletion of >50%, therefore the chemistry changes in SAI could potentially exceed the importance of the physical changes in terms of climatic impacts (e.g. UV radiation at the surface). Tilmes et al (2012) showed that SW-scattering by geoengineered sulfate could potentially compensate for ozone-loss by back-scattering UV radiation in the tropics, but that this effect was insufficiently compensatory at high latitudes. Their result was scenario-dependent; ozone loss due to heterogeneous chemistry is enhanced for smaller particles and in the presence of higher free-radical concentrations. Therefore, additional research is needed in order to understand the effects on atmospheric chemistry of injecting alternative aerosols. This work has already been started for titania by Tang et al (2014).

Another important aspect of SAI which is comparatively under-researched is the potential for impacts on human health. Aerosol concentrations in the air near the surface are of interest because of potential human respiratory impacts [Robock, 2008]. For instance, the USA's National Institute for Occupational Safety and Health (NIOSH) recommends ~~a~~ maximum

exposure limits of 0.3 mg/m^3 for ultrafine titania particles (radius <0.05 μm) and 2.4 mg m^{-3} for fine particles (radius < 1.5 μm) [Dankovic et al., 2011]. After undergoing coagulation and ageing in the atmosphere, it is likely that the second exposure limit is more applicable to this work. In our simulations, the maximum 2090's near-surface air concentration of titania (e.g. Fig. 4) for land regions between 60^{o}S-60^{o}N is 254 ng/m^3, which is of the order of 10^2~~10^3~~ less than the NIOSH 'fine-particle' exposure limit. The equivalent maximum concentration anomalies of BC in ~~G3BC~~ geoBC and SO_4 in ~~G3S~~ geoSulf are 10 ng/m^3 and 1851 ng/m^3 respectively. More work is needed to assess the potential impacts of SAI on air quality and human health.

Another thus far unmentioned aspect of this research is the potential for surface albedo modification by aerosol deposition. In particular, BC deposition on snow reduces the snow albedo through enhanced snow-melt and the coarsening of snow grains, which results in amplified high-latitude warming [Marks and King, 2013]. HadGEM2-CCS does not include the BC-on-snow feedback; therefore we estimate it by comparing the deposition rates for 2090s ~~G3BC~~ geoBC with the historical period. Jiao et al (2014) report that the simulated annual mean Arctic (>60^{o}N) BC deposition for the 2006-2009 period ranges from 13-35x10^7 kg/yr for the AEROCOM Phase II models. The annual mean Arctic BC deposition for the 2006-2009 period from our HadGEM2-CCS simulations is 23x10^7 kg/yr, which is within the AEROCOM range. The annual mean Arctic BC deposition anomaly for the 2090s period in ~~G3BC~~ geoBC is 19.6x10^7 kg/yr. Therefore, the effects of dirty snow in such an SAI scenario would likely be significant, which would have impacts on the distribution of temperature, particularly at high latitudes, potentially confounding some of our conclusions.

This research has highlighted potential climate impacts of injecting various stratospheric aerosols in order to ameliorate global warming. However, further research is needed to further assess the climatic impacts of stratospheric aerosol injection such as the impacts on ozone. Whilst research ~~has shown~~ indicates that SAI ~~to be~~is capable of averting certain climate changes such as surface-warming, SAI provides no amelioration for other climate impacts, such as ocean acidification. It is therefore important to note that the safest possible solution to avoiding the sort of climate change instantiated by (e.g.) Fig. 6a of this report is to effectively mitigate greenhouse-gas emissions.

Author contribution

ACJ designed the experiments, performed the simulations, analysed the data, and wrote the manuscript with guidance and advice from JMH and AJ.

Data sets

Data used to generate figures, graphs, plots and tables are freely available via contacting the lead author: aj247@exeter.ac.uk.

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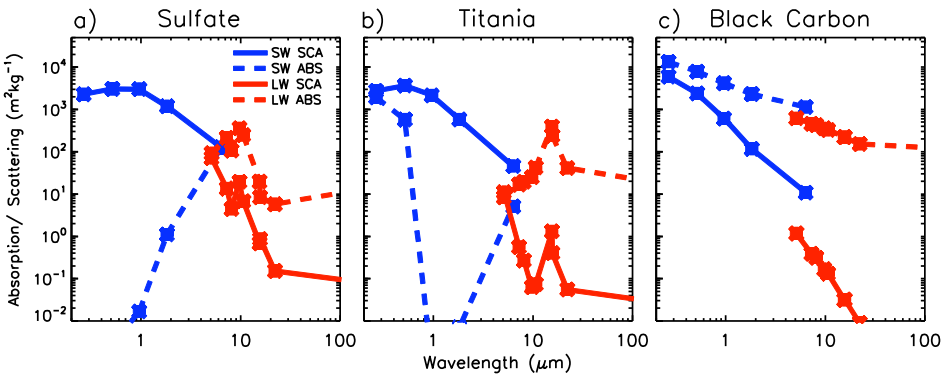

Figure 1. *Optical properties as a function of wavelength for a) accumulation-mode sulfate, b) titania, c) black carbon. Points are plotted at the middle of each spectral waveband, as detailed in Bellouin et al (2007)*

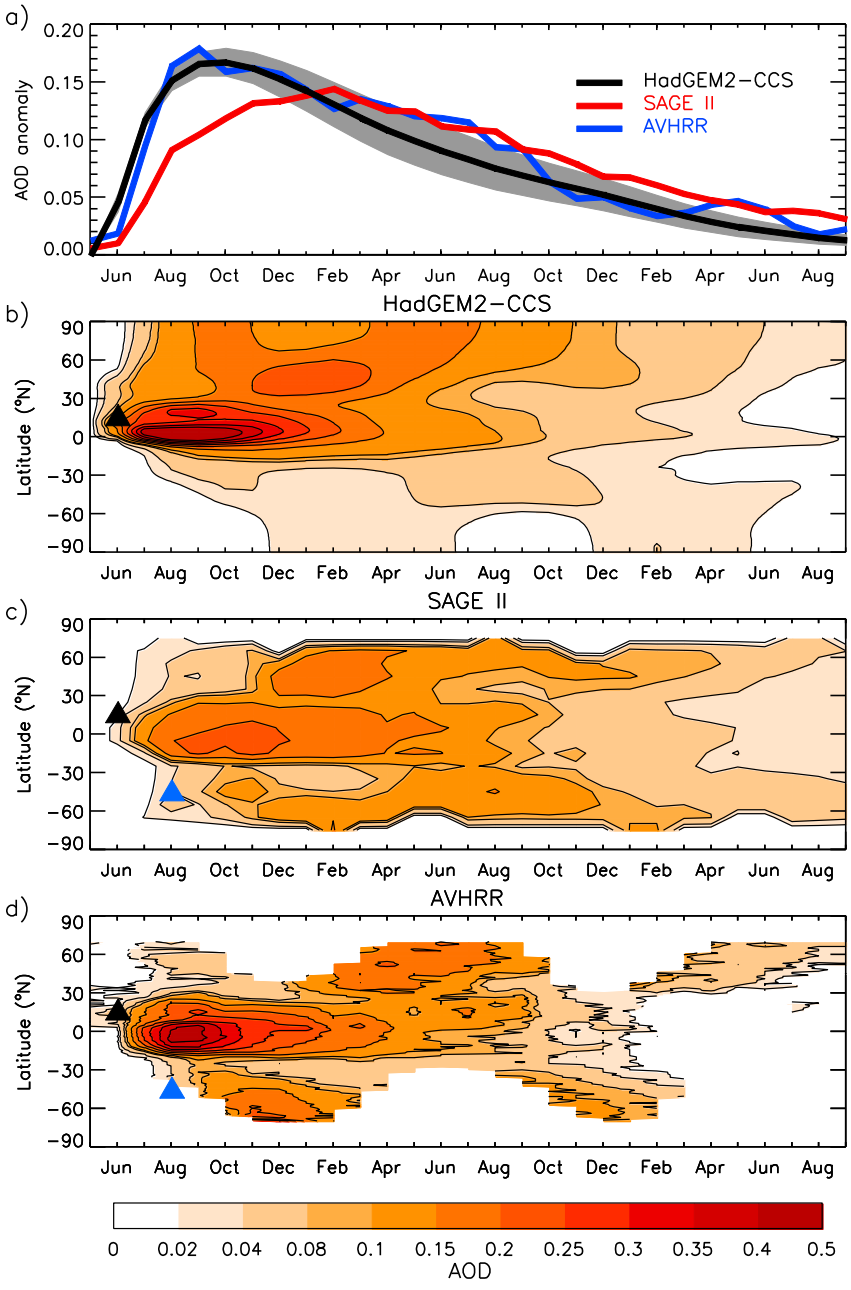

Figure 2. *a) 75°S-75°N-mean 550nm sulfate AOD anomaly for the Pinatubo simulations and observations, b-d) timeseries of zonal-mean 550nm sulfate AOD anomaly*

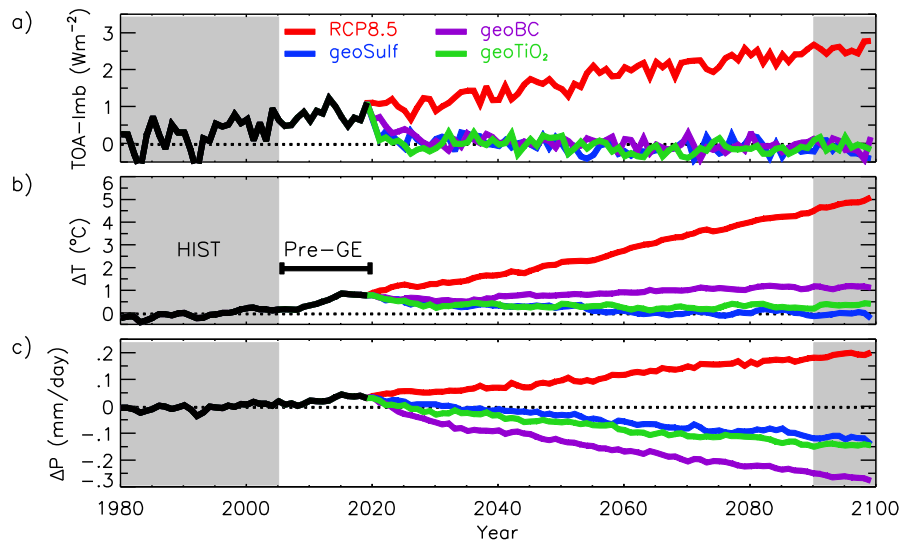

Figure 3. *Timeseries of annual/global-mean a) top-of-the-atmosphere radiative flux anomaly with respect to the pre-industrial control simulation b) near-surface air temperature anomaly with respect to the HIST period c) global mean precipitation anomaly with respect to HIST*

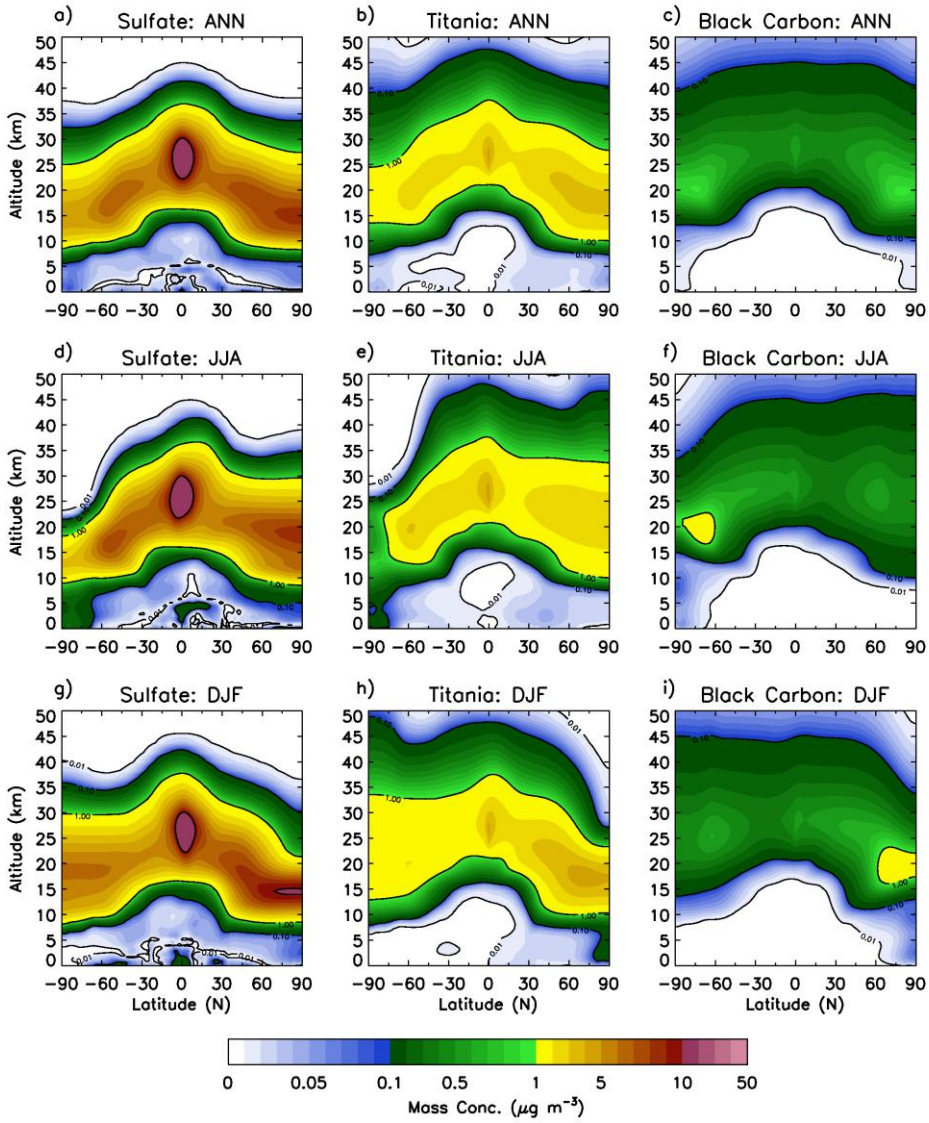

Figure 4. *Annual and seasonal zonal-mean mass concentration anomalies for sulfate (geoSulf - left), titania (geoTiO$_2$ - centre) and black carbon (geoBC - right)*

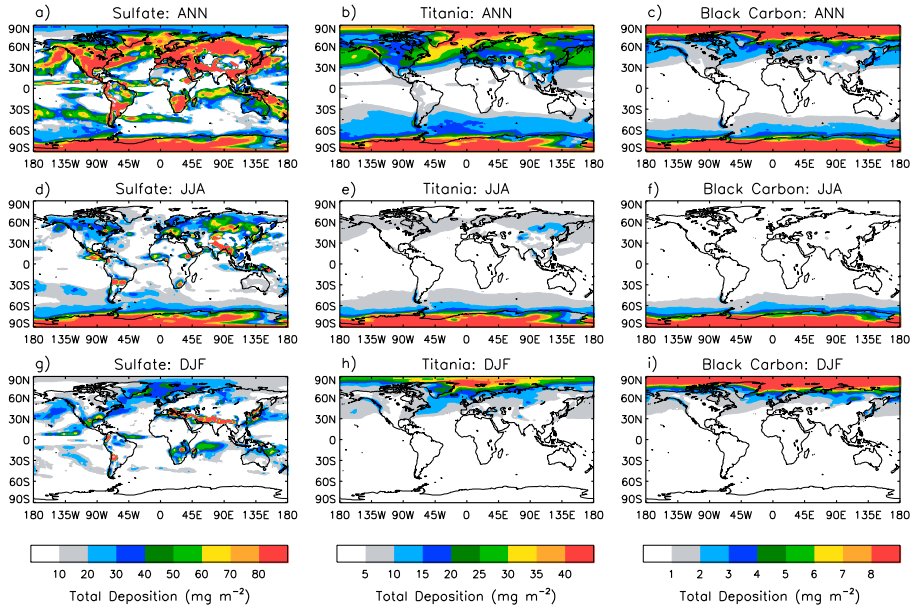

Figure 5. *Annual and seasonal total deposition anomalies (in units of mg m^{-2} yr^{-1} and 0.25x mg m^{-2} yr^{-1} respectively)*

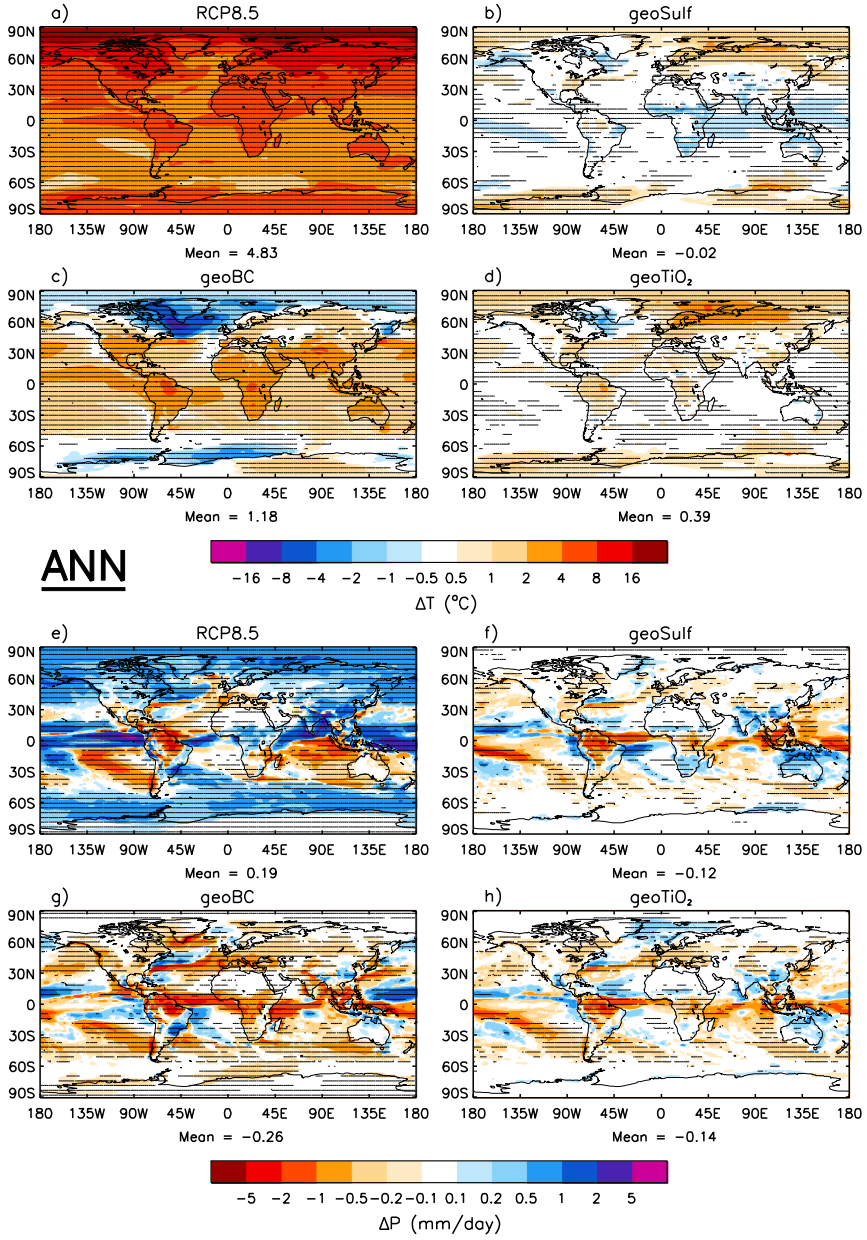

Figure 6. *Annual-mean near-surface air temperature (top) and precipitation rate (bottom) anomalies with respect to HIST. Stipling indicates where changes are significant at the 5% level using a two-tailed Student's t-test*

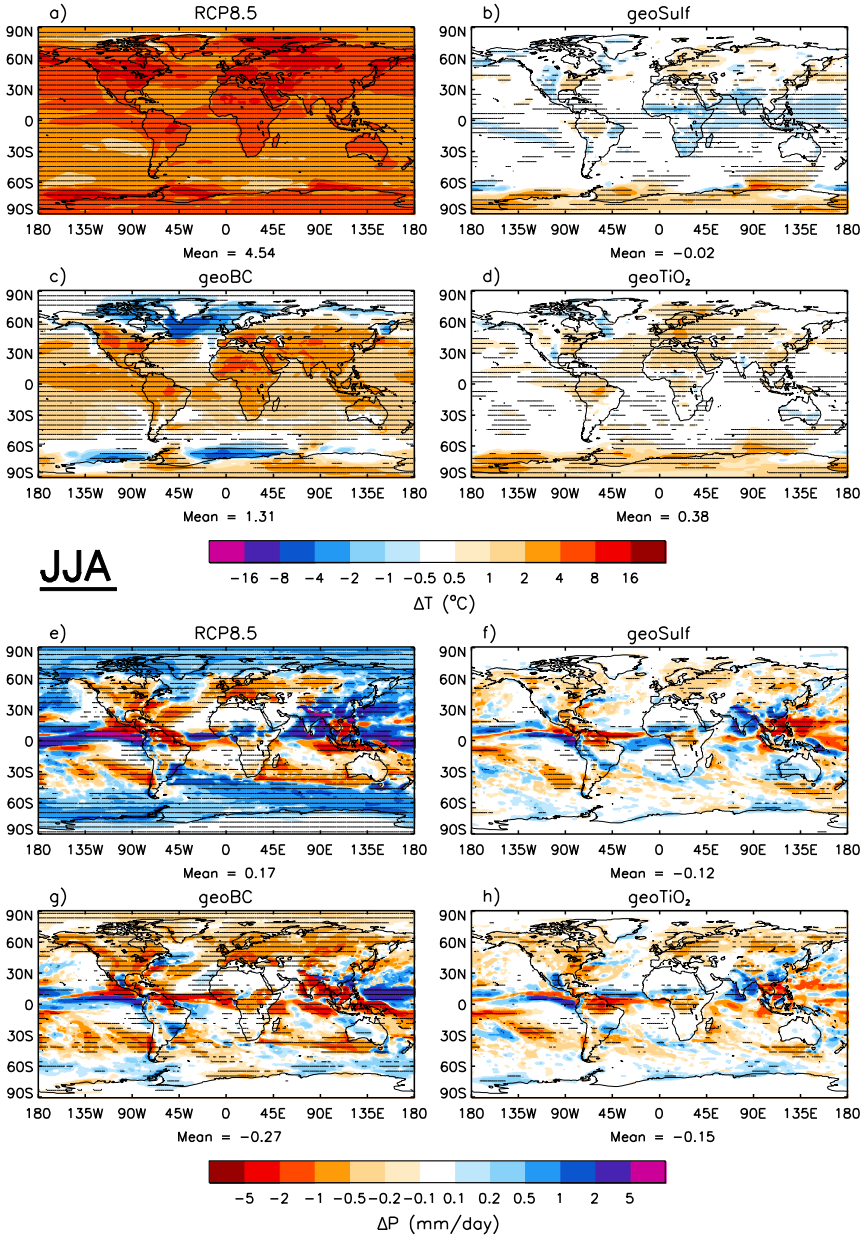

Figure 7. *JJA near-surface air temperature (top) and precipitation rate (bottom) anomalies with respect to HIST*

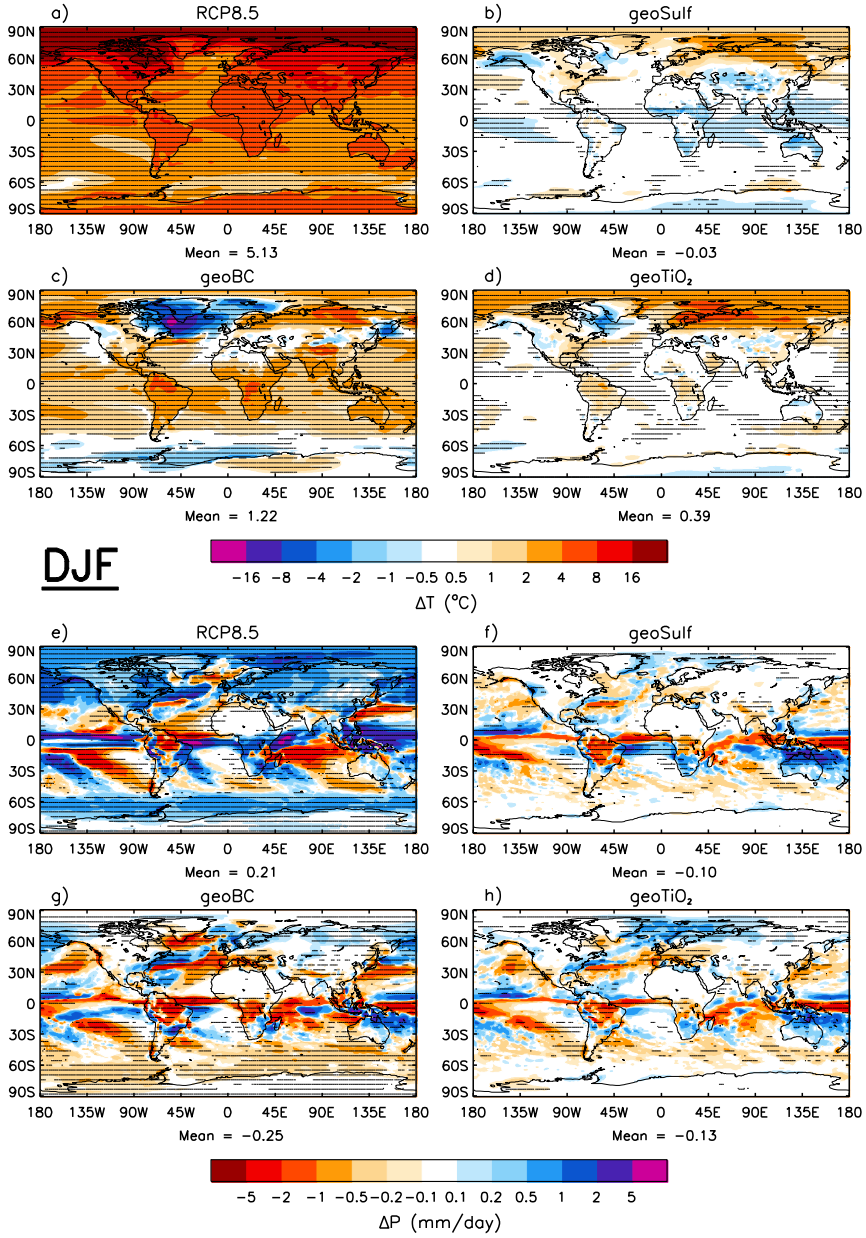

Figure 8. *DJF near-surface air temperature (top) and precipitation rate (bottom) anomalies with respect to HIST*

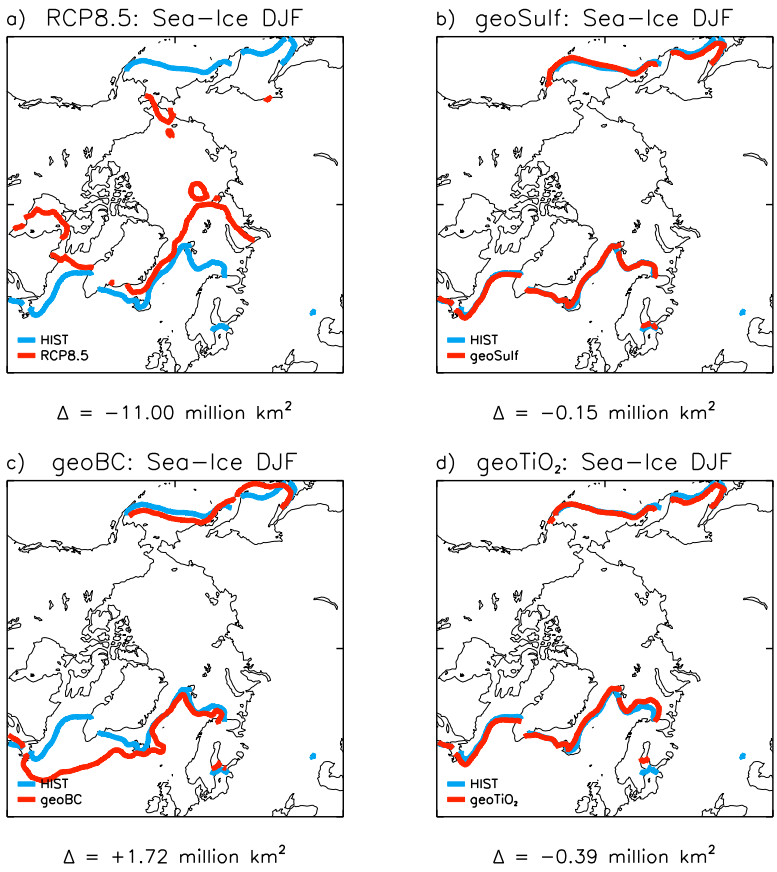

Figure 9. *DJF northern-hemisphere sea-ice edge plotted with the HIST extent*

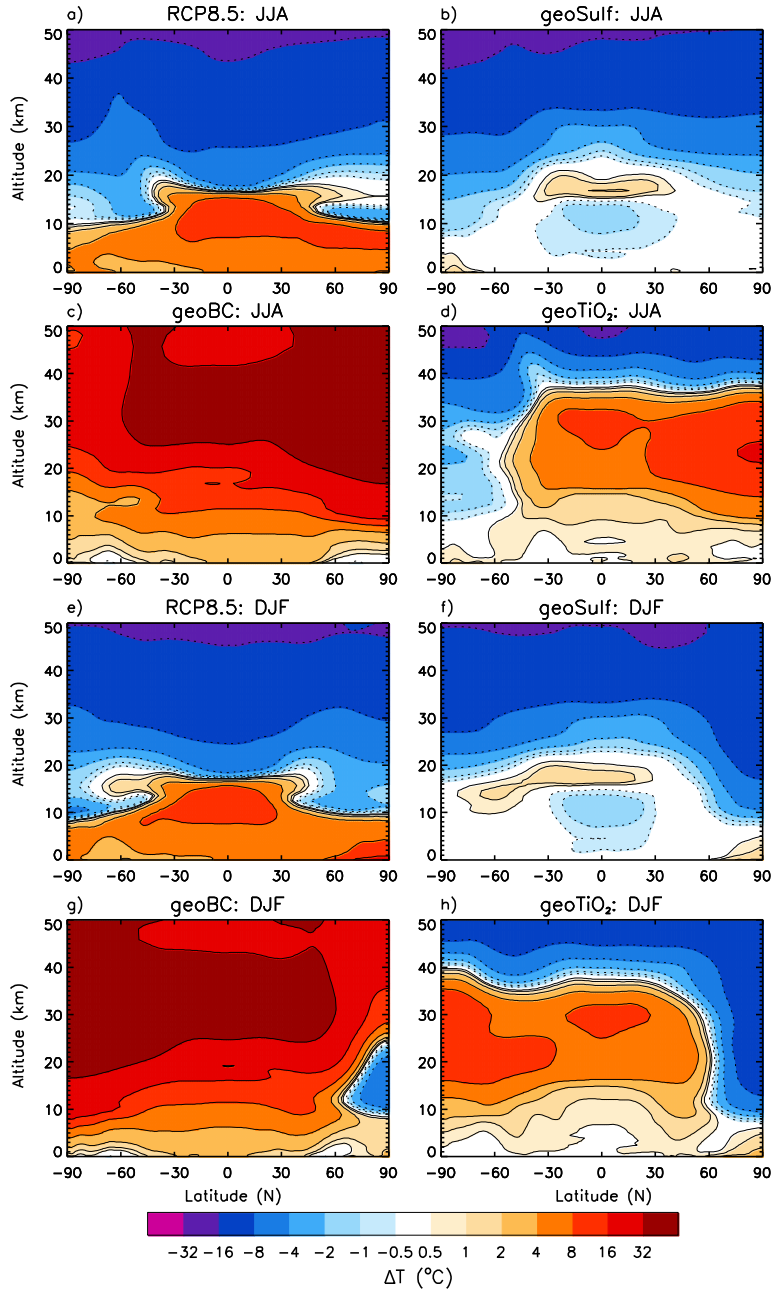

Figure 10. *JJA (top) and DJF (bottom) zonal-mean temperature anomaly with altitude, with respect to HIST*

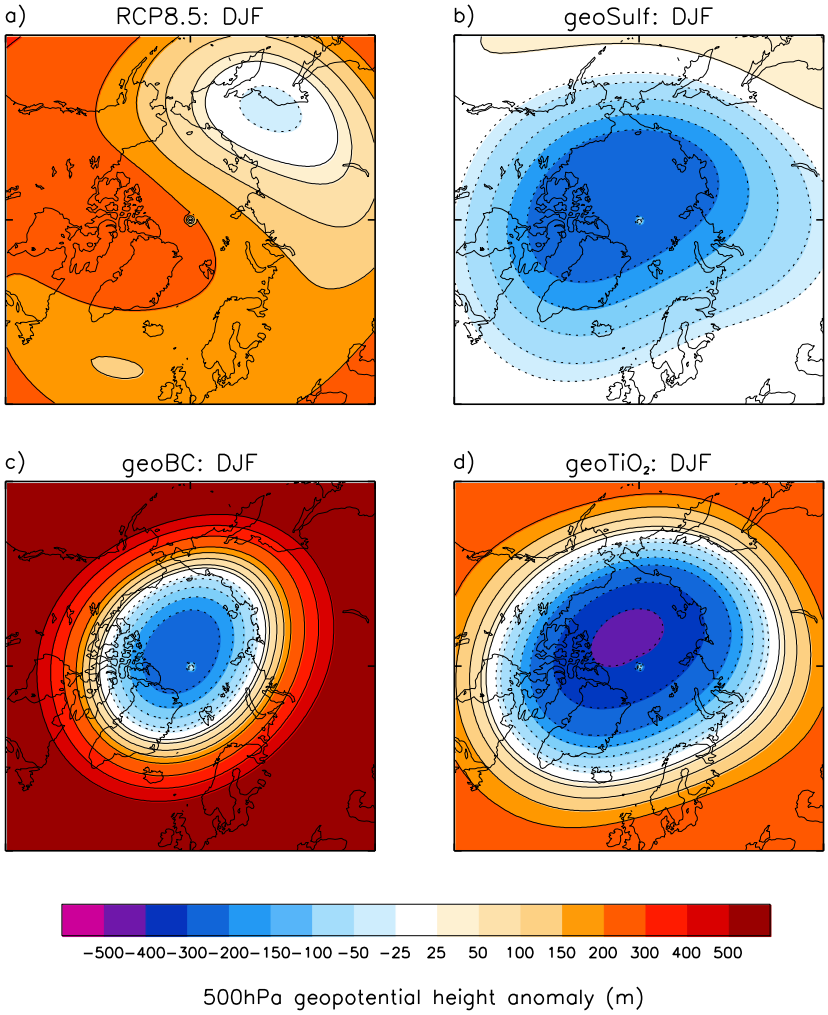

Figure 11. *DJF 50hPa geopotential height anomaly*

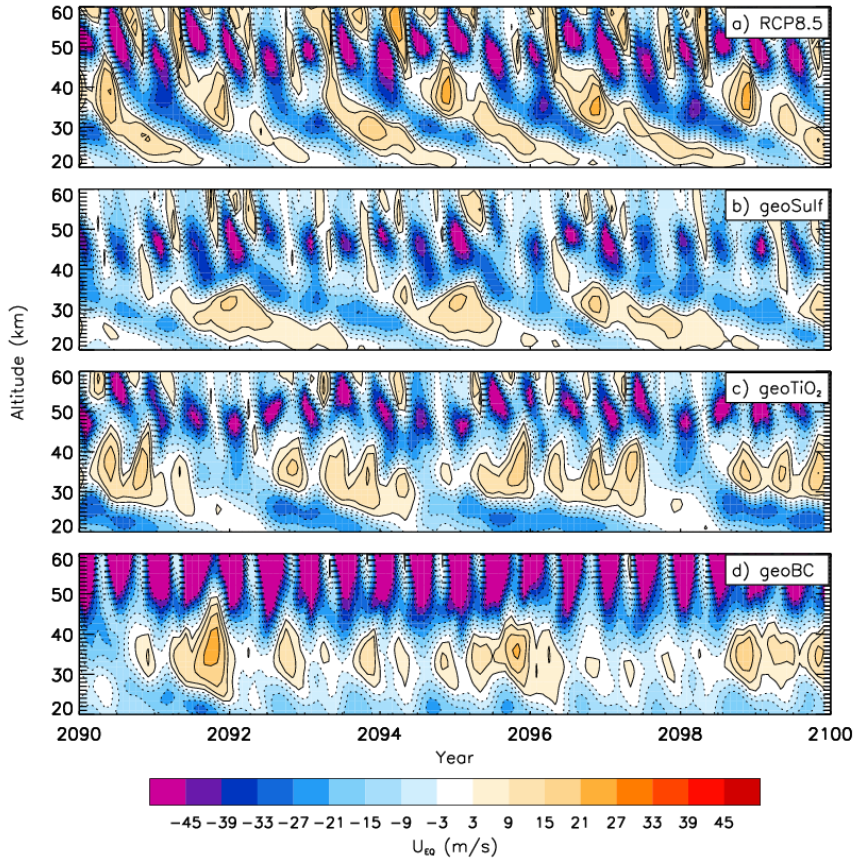

Figure 12. *Timeseries of equatorial (5°S-5°N) zonal-mean zonal wind profile*