Response to Reviewers' Comments

Assessing the consequences of including aerosol absorption in potential Stratospheric Aerosol Injection Climate Intervention Strategies

Jim Haywood et al.

We would like to thank the reviewers for taking the time to provide reviews on the draft manuscript. The reviewers are divided. Reviewer #1 provided a particularly helpful review, where the assertions were backed up by useful and appropriate references at all times. Reviewer #2 appears not to have appreciated the careful and balanced text which details that we are not directly attacking the Gao et al. (2021) paper, but rather bringing to the attention of the scientific community some of the issues and impacts of including absorption in SAI. Our responses to specific comments are given in red:-

Reviewer #1:

I very much appreciate this study. I reviewed Gao et al. (2021), and while I thought it was a clever idea and worth discussion in the literature, it was easy to see some potential problems with adding absorbing aerosols to the stratosphere. The present study provides much of that context and does a nice job with it. Furthering the discourse is exactly what needs to happen. I am recommending revisions, some of which could take a bit of work.

We are glad that the reviewer appreciates the purpose of the study and thinks that we have made a nice job in providing context to potential problems in adding absorption to stratospheric aerosols. We are glad that the reviewer thinks that discourse in this area is needed. We couldn't agree more……

We also appreciate the revisions that are suggested by the reviewer. The reviewer is right that accommodating them has taken a bit of work (in particular analysis of stratospheric water vapour and re-doing the plots to present statistical significance), but we have adopted and incorporated all of the suggestions made by the reviewer.

General comment: None of the figures has statistical significance calculations, so it is difficult for me to understand whether the values I'm looking at are important. (I suspect they are, but I'd rather not guess.)

We have now included appropriate stippling. We have tried to be consistent in our choice of stippling throughout the manuscript. We chose to stipple areas of insignificance rather than areas of significance as this methods worked best across the document and does not interfere with interpreting the details in areas where there are significant changes.

Introduction: Some discussion of the work of Wake Smith is warranted.

OK – we've added the text underlined below to the text:-

"However, these studies assume that the technological solution for delivery exists while, to date, it does not in any suitably scalable form <u>although development of a fleet of aircraft capable of delivering payloads at 20km altitude using current technologies appears feasible (e.g. Smith, 2020)</u>."

Lines 154-155: I wonder if you're missing a few mechanisms here. Ozone is a greenhouse gas, and it will certainly change in G6abs. Stratospheric water vapor is too, and G6abs has strong heating of the tropopause cold point. And now that I'm looking through the paper, I don't see any mention of stratospheric water vapor, which seems like an important oversight.

Agreed. We now include a plot of the change in water vapour in section 3.5 which is retitled to include impacts on ozone and water vapour. We also include reference to Tilmes et al. (2022), which was not available at the time that the paper was submitted. For ozone:-

"Tilmes et al. (2022) provide the most comprehensive multi-model assessment of the impacts of SAI from the G6sulfur simulations on stratospheric ozone, finding significant ozone depletion during spring-time over polar regions, but general increases in total column ozone over mid-latitudes and the tropics."

and for water vapour:-

"The predominant source of stratospheric water vapour is from troposphere-stratosphere transport across the tropopause at tropical latitudes (Holton et al., 1995). This transport is limited by the tropopause acting as a "cold trap" (Sherwood and Dessler, 2001) that dries air to the local saturation vapour pressure as it crosses into the stratosphere leading to condensation of water vapour and dehydration of stratospheric air. A secondary source of water vapour is the oxidation of methane (e.g. Le Texier et al., 1988). One of consequences of the significant stratospheric heating from G6abs is a very significant increase in stratospheric water vapour by an order of magnitude owing to the significant increase in the tropical tropopause temperature, which becomes a less effective cold trap. Figure 7 presents the stratospheric humidity (ppmv). Under present day (PD) conditions, water vapour concentrations are typically in the range 5-10ppmv. By 2081-2100, under SSP5-8.5, stratospheric water vapour typically increases by around 5ppmv presumably owing to increases in the oxidation of atmospheric methane. Little change from PD is seen in SSP2-4.5 by 2081-2100. Under G6sulfur, water vapour concentrations are higher than both SSP2-4.5 and SSP5-8.5 despite the fact that the global mean surface temperature is (by design) the same as that of SSP2-4.5. In G6abs, the amount of water vapour in the stratosphere is increased by almost an order of magnitude owing to the reduced efficiency of the tropopause cold trap caused by the strong stratospheric heating at tropical latitudes (Figure 5). Note that even stronger increases in stratospheric water vapour concentrations have been noted in simulations of nuclear winter scenarios through the same mechanistic route (Coupe et al., 2019). As noted by Mills et al. (2014), the photolysis of water vapour in the stratosphere exacerbates stratospheric ozone destruction, thus contributing to the ozone loss shown in Figure 6. Our simulations are not able to assess the relative contribution of this process to the ozone loss."

Lines 182ff: How do these results compare with the hydrological cycle impacts of tropospheric BC and sulfate? Are the mechanisms related? Also, you don't provide a mechanism –Bala et al. (2008, PNAS) seems be relevant here.

Yes, it is interesting how the BC has a stronger hydrological sensitivity than sulphate, results that are consistent with tropospheric BC and sulphate aerosol perturbations. We have added some discussion of this point and an explanation of the mechanism:-

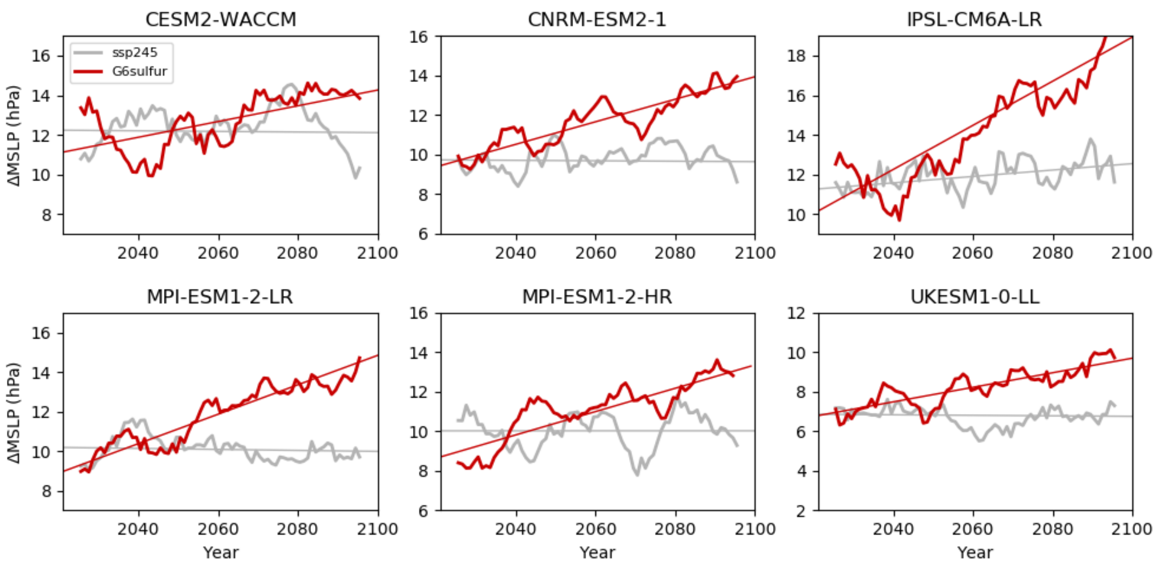
"The results show interesting parallels with results for perturbations to tropospheric BC and sulphate aerosols, though through different mechanisms. In the troposphere, warming in the mid and upper troposphere is known to reduce global precipitation by countering the balance between longwave cooling and latent heat release through precipitation (Smith et al., 2016). This mechanism is particularly strong for tropospheric BC (e.g. Johnson et al., 2019) and leads to a much greater hydrological sensitivity compared to tropospheric sulphate perturbations or reductions in the solar constant (Samset et al., 2018)."

We do already reference the Bala et al. (2008) study. However, we include more detail via the following sentence:

"As stated in Bala et al (2008), for same surface temperature change, forcings acting in the solar spectrum result in relatively larger changes in net radiative fluxes at the surface than those from CO_2 that act in the terrestrial spectrum. These larger changes are compensated by larger changes in the sum of latent and sensible heat fluxes."

Line 281: This is a bit of an overstep. I agree that if the models get all of the dynamics right, they should all show this response. I have to imagine there are some models that have problems here.

We do not believe that this oversteps. All models that participate in the G6sulfur simulations show a pretty large increase in the DJF NAO. This means that none of the models have problems in representing the dynamical response and they all show a consistent positive trend, but the magnitude of this trend may differ. Given that we understand the driving mechanism that forces the positive phase of the DJF NAO (i.e. differential stratospheric heating across the polar night, which increases zonal wind through thermal wind balance) it is reasonable to assume that any increase in the differential stratospheric heating from absorbing aerosols will lead to a more positive phase of the DJF NAO. Indeed, the fact that the NAO increases with time in each of the models as the sulfate loading increases reflects the fact that this differential stratospheric heating is getting larger and larger as time goes on. We include the relevant plot from Jones et al (2022) here:-



We therefore believe it is reasonable to assume that absorbing aerosols will increase the positive DJF NAO in all the models, but make a concession at the end of the sentence:-

"although dedicated multi-model assessments would be needed to prove this assumption."

Figure 8: Can you report slopes and R2 values?

Now included.

Section 5: Your results for the QBO are dependent upon these being equatorial injections – see Richter et al. (2017, 2018) and Kravitz et al. (2019). I'm not disputing your results, but I do think your description needs this qualification.
Citation: https://doi.org/10.5194/acp-2021-1032-RC1

We agree and have now included reference to these papers:

"We emphasise again here that the simulations for both G6sulfur and G6abs inject aerosol only at tropical latitudes (10°N-10°S). Simulations using SO2 injection positioned in the sub-tropics (e.g. 15°N, 30°N, 15°S, and 30°S) in other models appear to prevent significant locking of the QBO into the westerly phase (e.g. Richter et al., 2017, 2018; Kravitz et al., 2019). "

One other paper that was brought to our attention during the review process was that of Brenna et al. (2021) who model the response of the QBO to a 'super-eruption' where over 1000MtSO2 are injected into the stratosphere in the CESM2-WACCM model. The response bears a striking similarity, so we have included the following text in the revised paper:-

"Simulations with the CESM2-WACCM6 climate model of volcanic 'super-eruptions' (Brenna et al., 2021), where a pulse of more than 1000MtSO2 was injected into the tropical stratosphere, cause a peak tropical lower stratospheric warming of around 30K (see also our Figure 5). Brenna et al. (2021) report a similar breakdown of the QBO to that reported here, with persistent easterlies that switch to persistent westerlies that evolve into a more recognisable QBO structure as the stratospheric temperatures reduce. That the CESM2-WACCM6 model performs in a similar way to UKESM1 for these extreme stratospheric temperature changes lends confidence to our results."

Reviewer #2:

This study is examining impacts of absorbing aerosol injected with sulfate (which also partially absorbed) for stratospheric geoengineering. The responses noted are not new, in that some of these have been noted in papers using much more absorbing aerosol than employed here. One example (which is referenced here) is the paper by Ben Kravitz (Kravitz, B., A. Robock, D. T. Shindell, and M. A. Miller, 2012. Sensitivity of stratospheric geoengineering with black carbon to aerosol size and altitude of injection. J. Geophys. Res. 117, D09203 (2012).) that discussed how using large amounts of black carbon for SAI purposes had potentially detrimental impacts on climate. However, Kravitz et al. used black carbon as the SAI material, and used a large amount, 1 Tg BC per year. As noted by references in this paper, it has been previouslyl established that using BC as the SAI material causes detrimental effects, and, the Gao et al. proposal being criticized here also notes that issue.

We were a little disappointed with this review, which seemed rather partisan.

We are surprised that the reviewer cites only two peer reviewed papers: Gao et al (2021) and Kravitz et al (2012), and suggests that we present a completely different piece of research using a kind of toxicity/sensitivity approach rather than the approach we took of demonstrating the potential detrimental impacts of absorbing aerosol on climate and climate dynamics using a single assumed BC:sulfate ratio. The problem here is that this ratio is very ill-defined: Gao et al. (2021) themselves suggest that more SO2 could theoretically be lifted using the same BC emission. So, and as we make clear in the text, we do not directly attack the Gao et al. (2021) study, but rather bring readers' attention to issues with absorbing aerosols in the stratosphere more generically.

Regarding any detrimental impacts of absorption, this is what is written in Gao et al. (2021) – referred to as G21 hereafter - regarding continuous SAI with absorbing aerosols:-

“For example, a modelling scenario using 1 Tg year-1 of BC produced a stratospheric warming of 60°C (Kravitz et al., 2012)”

That is it. So G21 only mention the stratospheric heating and do not mention the knock on detrimental impacts on climate at all. We therefore consider it necessary to remind the scientific community of these detrimental impacts.

Kravitz et al. (2012, hereafter K12) focus on the sensitivity of SAI effectiveness to the black carbon size distribution and the altitude of injection, rather than on the impacts of this stratospheric warming on climate impacts. Our results suggest that inclusion of absorption:-

i) reduces the cooling efficiency per unit mass of aerosol injected
ii) increases deficits in global precipitation
iii) delays the recovery of the stratospheric ozone hole
iv) disrupts the Quasi Biennial Oscillation when applying moderately absorbing aerosols to combat a warming of just 0.1K
v) enhances the positive phase of the wintertime North Atlantic Oscillation, associated with floods in North Europe and droughts in Southern Europe.

Taking each of the above in turn:

i) K12 use a fixed SST model and state “showing results for globally averaged surface air temperature anomalies would not be particularly useful, as our simulations were conducted with fixed sea surface temperatures, which precludes the ability of the ocean to respond to radiative forcing.” They have to satisfy themselves with multiplying the radiative forcing by a fixed climate sensitivity, which I’m sure that the reviewer will admit is hardly ideal. They do not provide any quantitative assessment of how the temperature change using a partially absorbing aerosol compares to strategies including sulfate aerosol only.

ii) K12 do not investigate any impacts on global mean precipitation beyond wet and dry aerosol deposition rates.

iii) K12 investigate ozone loss sensitivity to aerosol size and injection altitude. They do cover this in some detail, but at no point do they compare against ozone loss from sulfate aerosol alone.

iv) QBO – not considered in K12.

v) NAO – not considered in K12.

The above demonstrates that the objections that the reviewer raises i.e. that the detrimental impacts have been documented before by Kravitz et al (2012) are incorrect.

The crux of the matter appears to be that the reviewer thinks we present modelling results that attack/criticise the results of G21. We go to great lengths to caveat our results and emphasize that this paper is not a direct attack on the study of G21, but a cautionary note that explains that the use of absorbing aerosol is not a magic bullet for the climate problem. Examples of our carefully caveated language are given here, beginning with the Introduction:

1. “We investigate the impacts of including a moderate amount of aerosol absorption by adjusting the single scattering albedo of the stratospheric aerosol at 550nm from 1 to 0.95 which is equivalent to assuming a stratospheric BC:sulfate mass ratio of around 2%. As such, this is a significantly higher fraction than that assumed in the plume rise modelling of Gao et al. (2021) who perform simulations with injections of BC and 2Tg of SO2 over a ten-day period in the CESM2 model and then downscale the minimum BC injection rate to produce the same lifting impact within a plume model, finding a minimum injection of 0.01TgBC. Thus Gao et al. (2021) effectively assumes a BC:sulfate ratio of just 0.3% while we assume ratios almost ten times higher.”

So, we are very clear that we are utilising a much higher BC ration than that of G21.

2. “We note not only technological challenges in the plume deployment procedure documented in Gao et al (2021) which might increase the BC:sulfate ratio, but also that Gao et al. (2021) suggest that more SO2 could theoretically be lifted which might decrease the BC:sulfate ratio.”

So our paper is very balanced here. We acknowledge that the ratio of BC:sulfate might be higher owing to the assumptions employed by G21, but equally, that the BC:sulfate ratio could be lower if more SO2 were deployed.

3. “Thus our simulations are not meant to directly follow, nor challenge the injection scenario of Gao et al. (2021), but rather to establish with current state-of-the-art model simulations what the impacts of including a moderate amount of absorption would be upon resulting climate impacts.”

Thus we explicitly state that we do not follow or challenge the G21 results. We return to this aspect again in the Conclusions:-

“We stress again that in the suggestion by Gao et al. (2021) a smaller amount of BC aerosol was proposed to lift sulfate into the stratosphere, which assumed a ratio of BC to sulfate around ten times less than that modelled here. It would be critical to assess whether the assumed BC amounts used in such proposals could achieve the efficacy of lofting that is stated.”

And again:-

“Where the main uncertainties arise are not in the climatic response of global mean temperatures, precipitation patterns, or the impacts on dynamical features such as the QBO and NAO and their subsequent impacts on regional climate, but in uncertainties around the effectiveness of the physical deployment of such strategies (Gao et al., 2021). A quantitative uncertainty analysis of physical and logistical factors that could reduce (or enhance) the efficiency of such deployment strategies would seem like an essential first step in assessing whether such technologies could theoretically be used to combat global climate change.”

The Gao et al. paper uses .01 Tg/yr. The authors of this current paper note that they are using a factor of 10 more BC material in their assessment than Gao et al. did, and effectively introduce this study as a criticism of the Gao et al. proposal. However, because of that factor of 10, it’s really not a fair criticism of the method.

As noted above, we’ve gone to great lengths to show that we are not specifically criticizing G21 but rather pointing out potential problems associated with the suggested use of absorbing aerosol.

What would be a useful addition to this study is something akin to a toxicity study, where one determines at what concentration a substance is toxic. I recommend examining consequences at 1X, 3X and 10Z the .01 Tg/yr case. This study is not including quite enough for BC to be the SAI material (like the 1 Tg/yr used by Kravity), but it used way too much to be considered as a lofting material as discussed in Gao et al. Consider something like a medical supplement; a small amount of a vitamin is beneficial for health, but a large amount is detrimental. You would not then recommend not taking the vitamin at all because a large dosage is detrimental.

The issues here mainly surround the effectiveness of the deployment strategy. There are many, many ways in which things can go wrong in practical deployment which could reduce the effectiveness of a 'perfect' deployment, but very few ways in which things can get better. As we state in the final paragraph of the Conclusions, the main uncertainties are not in the modelling of the climate response but in uncertainties surrounding deployment: "A quantitative uncertainty analysis of physical and logistical factors that could reduce (or enhance) the efficiency of such deployment strategies would seem like an essential first step in assessing whether such technologies could theoretically be used to combat global climate change"

The other point that these authors missed in regards to the Gao et al. paper is that they used BC as an example, but also suggested using Brown Carbon for the initial lofting, which would then break down in the stratosphere, and the heating effects would be different.

Many of the authors have worked for decades on assessing the physical and optical properties of biomass burning which includes organic (brown) carbon as well as black carbon (e.g. Haywood et al., 2003; 2008; 2021; Capes et al., 2008; Morgan et al., 2020, Wu et al., 2020, Taylor et al., 2020). Even for tropospheric aerosols that contain brown carbon, that have been relatively well documented, the microphysical and optical properties and how they age remain extremely difficult to quantify. Saying that you could use brown carbon that would bleach in the stratosphere is far easier said than done.

A sensitivity study looking at varying amount from very small (.01 Tg/yr) to the values assumed in this study would be useful. Are the feedbacks and consequences really linear with the forcing? A sensitivity study could examine whether you get 10X the response using 10X the amount of BC. It could be smaller, or larger, and that would be useful to know.

The problem with performing such a sensitivity study at the present time is that the BC:sulfate ratios needed are far from being established, as we note in our work evaluating "logistical factors that could reduce (or enhance) the efficiency of such deployment strategies would seem like an essential first step". We have established that, if any potential deployment was not as successful as the perfect deployment scenario suggested in G21, and the BC levels needed were around ten times higher, then significant detrimental climate impacts would occur. Then again – as we state – the BC:sulfate could theoretically be enhanced by lifting more SO2. Re-running a three member UKESM1 ensemble over centennial timescales several times to examine linearity of the climate response does not really add to the main thrust of the current paper which is to provide an up-to-date assessment of the impacts of absorbing aerosols on key climate indicators.

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