Authors response

Reviewer 1

The first suggestion is essential in my opinion, whereas the remaining comments would improve the quality and readability of the manuscript.

We thank the reviewer for their positive review. We address the individual points below in blue.

Please make the used data available via zenodo or other means to ensure reproducibility.

As requested, we have deposited the data to Zenodo under DOI: 10.5281/zenodo.21382844.

l. 81: I meant quotations for "middle-of-the-road".

We apologize for the confusion – now added.

l. 83: I meant quotations for "business-as-usual".

We apologize for the confusion – now added.

ll. 121-122: Please indicate the CESM model (sub-) version used, e.g., 2.1.5. Bugs or similar issues may be found in the respective version which are easier to identify that way.

Added.

Figure 2: My point here is that it is possible to get/more less ozone w/o decreasing surface temperatures, and vice versa. See GHGs, halogens, ... I understand the motivation to connect the "cooling efficiency" of the respective method to the extent of ozone depletion. But in its current form, the different contributing processes are omitted from the reader. Ideally, the motivation for this metric would be more clearly justified and introduced in the beginning to avoid misunderstanding. The same holds for my previous comment to ll. 186-188. Here, a clear statement (as in the response to my comment) would improve the understanding of the manuscript and clarify the employed metrics. My point is, that I would like to see a justification of the used metric, being along the lines of comparing the "desired" reduction in Tsurf to the impact on O3. Not because they are physically justifyably connected but because the paper focusses on SAI and comparison of different approaches. Otherwise, the underlying physics may be understood blurred.

As requested, we have added a statement introducing the motivation of this metric in Section 3: “Such approach allows comparison of the impact of SAI aerosol on ozone in the prism of the ‘intended’ aerosol-induced surface cooling, and can thus aid discussion of trade-offs of different SAI choices (scenarios and strategies).”

Previous comment to ll. 416-419:
This metric is focused on estimating how much O3 depletion one has to take into account per "achieved" degree of cooling, which is more of a policy/trade-off metric and has less physical meaning beyond that. Since the connection between Tsurf and O3 is not direct, differences in models (chemistry, radiation, resolution, ...) could also lead to very different results. The metric (if it would be as straightforward as mentioned in the paper) would be great to have, but I find it more confusing/ambiguous than useful to understand the physics. I would rather just e.g., normalize the TO3 with respect to climatology.

We believe there is a physical connection between the SAI-induced ozone depletion and SAI-induced surface cooling, in that both of these effects are driven by sulfate aerosols from SAI. Hence, both are expected to scale with the magnitude of the intervention (SO2 injection or amount of global warming to be offset). While we agree that two important uses of this metric is for informing policy as well as discussing trade-offs of different SAI strategies and scenarios (as we now explain in Section 3 in response to the previous comment), we believe this is also a useful metric to compare different models and thus quantify structural uncertainty (since the reviewer is correct that differences in models will also contribute to differences in that metric), as we do in the manuscript. We now state that explicitly: "highlighting substantial structural uncertainty in the magnitude of this SAI 'side-effect' on Antarctic ozone."

Previous comment to ll.478-480:
I would imagine that the main difference comes from RH due to different cold point temperatures at the tropopause which affects stratospheric heterogeneous chemistry. So simply pointing to surface temperature normalization may not be sufficient. Stratospheric meridional temperature differences, notably tropical lower stratospheric warming are previously found to show significant effects on stratospheric circulation and resulting changes in the hydrological cycle and ozone.

We agree with the reviewer that impacts of aerosol induced tropical lower stratospheric heating are important for driving changes in stratospheric circulation and chemistry that directly impact ozone concentration. However, as discussed in Bednarz et al. (2023, https://doi.org/10.1029/2023GL104726), changes in tropical lower stratospheric heating - and the associated dynamical and chemical changes in the tropics - tend to scale linearly with the amount of SAI-induced cooling (and hence aerosol burden). A similar relationship can also be expected in the mid-latitudes. As such normalizing the responses here with the associated global cooling value should account for any such differences. While the previous study did report potential non-linearities in the high-latitude ozone responses, we expect that impacts of any such non-linearities – if present here under relatively small differences in injection rates and temperature – on ozone would be by far outweighed by the influence of interannual variability. We now report the magnitude of the difference in the injection rates and cooling values between the two sets of experiments to put the results into context.

Reviewer 2

The manuscript by Bednarz et al. analyzes a stratospheric aerosol injection (SAI) experiment, using SSP2-4.5 as the control without SAI, for three chemistry-climate models that participated. Important differences from earlier SAI experiments include a more moderate greenhouse gas scenario as the control and specified sulfur injection at subtropical latitudes. The simulations are analyzed with a focus on the impacts on stratospheric ozone, looking at how chemical and dynamical changes induced by the SAI change the evolution of ozone over the rest of the 21st century.

I will note that I have been asked to review the article after it has already been through one round of reviews with a quite extensive set of comments and responses from the authors. I find the manuscript in its current form to be generally well written and a clear presentation of the model results and the analysis. I believe the article should be published after addressing a couple of relatively minor aspects related to the presentation or interpretation of the results.

We thank the reviewer for their positive review. We address the individual points below in blue.

In the minor comments I point out I had trouble understanding (lines 140 – 157) how exactly the background SAD and SAI-enhanced SAD are treated in the UKESM experiments analysed here. If I have understood the setup correctly, heterogeneous chemistry on the specified SAD is turned off in both the SSP2-4.5 and G6-1.5K-SAI experiments for UKESM simulations used here? Given the dominance of the aerosol from SAI I am not concerned about the lack of heterogeneous chemistry on the background aerosols in the G6-1.5K-SAI simulation, but having zero aerosols in the SSP2-4.5 simulation seems to be a bit of an unrealistic case. With zero sulphate aerosol in the SSP2-4.5 simulation the attenuation of halogen effects on ozone by reactive nitrogen compounds will be stronger than they should be - and it is noted in the text that the SSP2-4.5 simulation used here has higher ozone than the standard SSP2-4.5 UKESM simulation with background SAD. As a consequence the impact of SAI on ozone estimated from the differences between G6-1.5K-SAI and SSP2-4.5 will be larger than it should be. I would argue this is likely part of the reason for the larger change in TCO in UKESM shown in Figure 2b even though the SAD from SAI in UKESM is not that large. This increased sensitivity to SAD in UKESM is then partly compensated for by the smaller increases in SAD to get the same cooling – Figure 1a and 1e – so that the impact on ozone per degree cooling comes out roughly the same as shown in Figure 2c. I would argue that this aspect of the UKESM results need to be put into context for the discussion.

We have added the following discussion of this to Section 2: “Finally, since the background SAD is not included in either SSP2-4.5 and SAI simulations, we assume the impact of this on ozone is the same in both simulations and thus cancels out when inferring the ozone response to SAI in the model (calculated as the difference between G6-1.5K-SAI and SSP2-

4.5). However, this might not necessarily be the case if the chemical ozone response is significantly non-linear to increasing SAD magnitudes. If so, the impact removing such background SAD would have on ozone may be larger in the SSP2-4.5 simulation than in the SAI simulation (where SAI SAD >> background SAD), thus potentially increasing the inferred chemical ozone response to SAI in the model."

In a few places the changes in the chemistry with SAI are attributed to chlorine activation on the increased sulphate (for example, at line 205 and at lines 307–310). I might be dated in my thinking, but I thought the explanation for Pinatubo effects on ozone was an increased sequestering of reactive nitrogen in nitric acid through N2O5 hydrolysis on sulphate aerosol, leaving less reactive nitrogen in forms that can tie up chlorine and bromine – at least in mid to low latitudes. Part of that mechanism is a result of the temperature dependence of reactions like HCl + ClONO2 on sulphate aerosols with very slow reaction rates (small gammas) much above 200K. See, for example, Kawa et al., Activation of chlorine in sulfate aerosols as inferred from aircraft observations, J. Geophys. Res, 102, 3921-3933, doi:10.1029/96JD01992, 1997. I would accept that certain features of the response are due to 'chlorine activation', like the larger response in TCO in the Southern Hemisphere (line 378) that is likely the hemispheric impact from increase Antarctic ozone depletion. But the more widespread low- to mid-stratosphere changes through the tropics and mid-latitudes I believe are more due to changes in chlorine as a result of changes to NOx.

Thank you for pointing this out – we have rephrased the interpretation to account for this.

Line 45: The phrase 'to reduce the incoming solar radiation' seems a bit imprecise if you consider incoming to be at the edge of the atmosphere.

We agree and have rephrased this to "reduce solar radiation reaching the troposphere".

Lines 140 – 157: The discussion of how UKESM treats heterogeneous reactions on background sulphate could use a single definitive statement of how sulphate reactions were treated in the two experiments used for this paper. I think I can find it, but it requires looking through the entire paragraph and parsing a bit of information from different sentences. From the text I understand that 'a second revised set of both SSP2-4.5 and G6-1.5K-SAI simulations was carried out' and that 'in these revised UKESM simulations no heterogeneous chemistry occurs on the background imposed CMIP6-recommended stratospheric aerosol surface area density (SAD)'. It just seems a bit counterintuitive that to turn on heterogenous chemistry on the SAI sulphate it was decided to turn it off in both SSP2-4.5 and G6-1.5K-SAI. Being an important point it would help to see that stated explicity here.

We apologize for the confusion. The reason for this set up is that in the current model configuration heterogenous chemistry can only occur on either the imposed SAD field or SAD simulated online by the GLOMAP aerosol scheme. Hence, to include both background and SAI aerosols one would need to include stratospheric SO2 emissions from both volcanic/natural (i.e. background) and SAI origins, but the former were not available to us at the time of the simulation.

As suggested, we have rephrased that paragraph to make this clearer.

Lines 524 – 527: In reference to the changes in TCO for the WITH_HET and NO_HET simulations shown in Figure 13, do the authors have any explanation for why the NO_HET TCO shows a clear increase between 2045-2064 and 2065-2084 in the Northern hemisphere, but the WITH_HET simulation shows almost identical changes for the two periods? This starts at about 30N and extends right to the North Pole.

We are not sure of the exact reason for the difference between 2045-2064 and 2065-2084, but we think the response itself is related to the ozone increase in the upper troposphere and lowermost stratosphere, which is likely related to the SAI-induced tropospheric cooling and concurrent lowering of the tropopause. Such response is much stronger in the NO_HET simulations because of the absence of the offsetting influence from the SAI-induced lower stratospheric ozone decrease (from heterogenous chemistry on SAI aerosol).