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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ozone changes under solar geoengineering: implications for UV exposure and air quality" *by* P. J. Nowack et al.

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We thank the reviewer very much for his/her comments. We hope that our answers will help to clarify some of the key messages of our manuscript.

The referee's main concern relates to the experimental setup, which is further reflected in some of the specific questions raised. We have grouped these together and answer them first, before addressing the other specific questions. Our replies to the referee's comments are in bold and italic. Any references to pages and line numbers made by us refer to the published online version of the discussion paper and may have changed in the revised version.

OVERARCHING COMMENT ON THE EXPERIMENTAL SETUP:

The present study investigates the impact of solar geoengineering on climate, stratospheric ozone and surface UV-B using a global atmosphere-ocean-chemistry-climate model. The analysis is based on three model simulations: a pre-industrial control experiment, a 4xCO2 experiment and an experiment with reduced solar irradiance to offset the CO2 induced global warming. The simulation set-up follows the GeoMIP G1 experiment.
In general the manuscript is well written, the argumentation is easy to follow, and the figures are well prepared. There are some sections where more detailed information would be helpful. I added a couple of remarks and suggestions below.
My major concern is related to the experimental set-up, which is highly idealized and, in my view, not appropriate to address air quality issues under SRM. The applied scenario follows pre-industrial conditions. Although not directly mentioned in the manuscript, I assume this holds also for ozone depleting substances, tropospheric ozone precursors and aerosols. While such a scenario might be appropriate to investigate climate change under SRM, I have some doubts that this is also the case for tropospheric chemistry and air quality. I think we agree that SRM will never take place under clean air conditions. The authors state that the aim of the present study is not to provide a quantification of the effects, but to discuss principle changes of atmospheric chemistry and climate under SRM, but I think even a qualitative discussion is hardly possible, since the relative importance of the involved processes might change under a different atmospheric composition. I am aware that the authors simply follow the predefined set-up of the GeoMIP G1 experiment, but in my opinion this set-up is not appropriate to address tropospheric chemistry changes.

The G1 modelling set-up is indeed highly idealized; we have ourselves highlighted this fact in our manuscript (see page 31978, lines 17-23 and also page 31975, lines 2-3). However, we strongly disagree that this makes it inappropriate

for our study. We note the positive comments of Referee 2 in that regard. To address Referee 1's concerns we have redrafted the manuscript in several ways to clarify the logical progression. We now further emphasize that our focus is on stratospheric changes and how they impact the troposphere. Our stratospheric scheme is absolutely state-of-the-science and we have every confidence in the calculated changes. These changes then have an impact on the troposphere – by changing UV penetration to the surface (which again we feel very confident about) and by the UV impacting tropospheric composition, in combination with robust water vapour changes. We were very clear in our original manuscript to include appropriate caveats about the composition changes (next to the comment on page 31978 l.18-23 also on page 31983, lines 16-21 and page 31987 lines 13-28); we also emphasise that tropospheric composition impacts were summarised in a very brief discussion only, both in the introduction and as a section. We have reordered sections 3.3 and 3.4 to make the overall logic clearer and added the word 'stratospheric' to the title of the revised manuscript. In addition, we clarified the link between stratospheric ozone changes and tropospheric composition in the abstract. We have further emphasized these links in the introduction and extended our discussions on the scenario-dependency both in section 3.4 and section 4.

G1 is an experimental setup designed to be used widely by the GeoMIP community. We see our variant on that experiment as adding a further interesting dimension, helpful to define further studies. However, it is not the idea of the G1 set-up to make a realistic, quantitative analysis ('prediction') for a specific period in the history or future of the Earth's climate. This applies both to climate and air quality questions. Scenario uncertainty is, of course, inherent in any geoengineering study since it is unknown under which atmospheric conditions geoengineering would (if ever) be deployed. It is important however to highlight general principles; that changes in the stratosphere under SRM would impact

surface UV and tropospheric composition is one such principle.

Our model includes a sophisticated stratospheric scheme that allows us to study stratospheric ozone changes in great detail. As we show in our paper, the changes we find have implications for tropospheric ozone chemistry under solar geoengineering. Most importantly, stratospheric ozone increases, in combination with a generally reduced solar constant, lead to decreased UV fluxes into the troposphere. As a result, ozone photolysis and O(1D) production in the troposphere are reduced. In addition, solar geoengineering studies consistently show a weaker hydrological cycle, which gives rise to lower atmospheric specific humidity. This can also affect tropospheric chemistry. The tropospheric part of our study aims to highlight the significance of these important effects, as has been recognized by the second reviewer. This does not imply that the detailed results are directly transferable to other, less idealised, scenarios (although we note that the changes in tropospheric ozone, following a stratospheric ozone increase, are consistent with calculations using detailed tropospheric chemistry schemes (e.g. Banerjee et al, ACP, 2016, in press) . However, they do imply the importance of studying them in more detail. More generally, our study is designed to demonstrate that complex changes in the vertical structure of the atmosphere appear under solar geoengineering and that offsetting "global mean surface temperature" change will not suffice to reset the corresponding changes in atmospheric composition in the stratosphere and troposphere. We hope that our text amendments (see below and replies to the second reviewer) help to clarify this point further.

So, we conclude that the G1 experiment should be considered as an exercise to highlight key impacts on processes that merit further, more detailed studies. In our opinion, this is an important part of the incremental scientific process, in agreement with the view presented by the second reviewer. Here, we specifically highlight the significance of robust changes in key processes that are, inter alia,

expected to affect tropospheric chemistry under solar geoengineering. For this, the design of the G1 experiment provides a useful baseline on which follow-up studies can be built.

Banerjee et al. Drivers of changes in stratospheric and tropospheric ozone between year 2000 and 2100. Atmos. Chem. Phys. Discuss., 15, 30645–30691 (2015). Now in press in ACP.

What should I recommend now? It is a solid study, and I am not at all against idealized model experiments. They can be very useful, but the set-up must be appropriate. I would either suggest extending the discussion towards more realistic atmospheric conditions (the authors might have some further sensitivity studies available that could be added) or focusing more on climate change than on air quality.

We argue above that the experiment is 'appropriate' for our aim, which is to highlight important processes and interactions and not to make predictions. Idealized experiments are an essential part of the research armoury. Simple studies pointing to a mechanism (here, stratospheric change and its impact on the troposphere) are subsequently followed up by more comprehensive studies, so moving the science onwards. For example, using an earlier version of the MetOffice Unified Model, again with a simplified tropospheric chemistry, some of us published a study (Zeng et al., GRL, doi:10.1029/2004GL021353, 2005) showing the influence of the ENSO, via stratosphere to troposphere exchange, on tropospheric composition. This paper has been cited many times and prompted more detailed studies. Our paper was not the last word, but it did serve as a stimulus. Surely, that's how we all think things should work.

SOME SPECIFICS ON THE EXPERIMENTAL SET-UP:

- Section 3.3: In my opinion the whole discussion on tropospheric ozone changes is purely abstract. Here air quality issues under pre-industrial, i.e. clean, conditions are discussed. ... In my opinion the experimental set-up is not suited to investigate the impact of SRM on tropospheric chemistry and air quality, so this section could be skipped or revised by discussing more realistic scenarios, e.g. by comparing the idealized G1 experiment with other model simulations using present-day conditions.

The reviewer is restating his/her concern, to which we have already replied above. The scenario is idealized but our results are absolutely consistent: the change in stratospheric ozone would certainly produce the change in UV discussed; our calculated changes in tropospheric ozone are consistent with the changes calculated with more detailed tropospheric chemistry schemes (e.g., Banerjee et al., ACP, in press, 2016). While the detailed quantification would be scenario dependent, our study shows that the effect would be significant.

The referee might equally well ask why a present-day atmosphere should be realistic for a future time when geoengineering would be employed. Geoengineering is not discussed here as a realistic, immediate measure. Accordingly, it is unknown what the abundances of CFCs, NMVOCs and so on might be in the unlikely event of geoengineering. Even within this century, very different emission scenarios can be imagined, which would lead to very different surface ozone responses (e.g. Young et al., 2013) - with or without geoengineering. Our study simply aims to provide implications for robust mechanisms in which changes in stratospheric ozone, solar irradiance and tropospheric humidity could affect tropospheric chemistry under solar geoengineering.

We addressed this in the original manuscript dealing with emission, chemistry scheme and dynamical uncertainty (see page 31983 lines 16-24 and page 31987

lines 13-28). Reviewer 1 has now motivated us to go even further. So, for example, we have extended and clarified our discussion on this question in the last paragraph in (now) section 3.4 and in section 4.

Young, P. J. et al. Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP). Atmos. Chem. Phys. 13, 2063-2090 (2013).

- Section 2.2: Which scenario has been chosen for ozone depleting substances, tropospheric ozone precursors, etc.? Also pre-industrial? Section 2.2 describes only the CO2 scenarios and the solar irradiance change. I would like to see some more details about the experimental set-up.

By design, the G1 background atmosphere is based on pre-industrial conditions. Atmospheric CO2 is quadrupled and insolation reduced from this starting point. At the same time changes in CFCs, for example, are by design not included in the model. Other chemical species which impact ozone (such as NOx, HOx species) are, as explained, included in the chemistry model - with the emissions treated as described in the paper section 2.1. We have added the following sentence to section 2.2 for further clarification:

"By design, the G1 experimental set-up does not include pre-defined changes in surface emissions of ozone depleting substances from anthropogenic sources (e.g. CFCs whose abundance is equal to zero in this set-up), or tropospheric ozone precursors."

- P14, L4-6: Which scenario did you assume for ODS and ozone precursors?

CFCs and other anthropogenic ODSs are not included. As mentioned in section 2.1, some ozone precursors are included, e.g. methane, surface and lightning

NOx, etc.

- P11, L23-25: Same as above, tropospheric aerosols and their impact on the UV-I are neglected although it's very likely that they will play an important role under SRM in a future climate. In my opinion the scenario is too much idealized.

As we have already stated, we focus here on stratospheric changes and their impact on the troposphere. We agree that uncertain changes in clouds and aerosols would also have an impact on UV; however, not including them does not negate the central importance of the stratospheric changes.

- P12, L26-30: For such a general statement is it not necessary to run a fully coupled AOCCM. Some basic physical and chemical considerations would lead to the same conclusion. For a proper evaluation of risks and benefits of SRM one would need a quantification of these effects, which is not given here.

Again, we do not agree. Without explicit model simulations at the level of sophistication presented here, it would not be clear whether stratospheric and other climatic changes under solar geoengineering have the potential to affect tropospheric chemistry and surface UV fluxes in significant ways. Here, we do provide quantification for this particular scenario to demonstrate its significance. At the risk of repeating ourselves, we demonstrate that large, robust changes in the stratosphere have a significant impact on the troposphere. We reiterate that the G1 experiment is ideally suited to ask such questions and to point out some of the principal processes - without claiming completeness.

SPECIFIC COMMENTS:

- P2, L24ff: I would suggest to rewrite this paragraph or to split it into 2 sections. First, it describes the potential impact of SRM by particle injection on ozone depletion and, hence, increasing surface UV-B, and then it suddenly jumps to negative effects of decreasing surface UV-B on human health. This is not very intuitive and needs some more explanation.

Thanks for this suggestion. We have added a line break here to make the transition between the different effects of ozone changes clearer and have rewritten the section on the impacts of surface UV-B reductions, now linking them more explicitly to changes in ozone.

- Section 2.1: In line 24-26 you mention a simple tropospheric chemistry scheme that has been implemented to your model. How 'good' is your tropospheric chemistry, especially in terms of ozone? Since you discuss tropospheric ozone changes in Sect. 3.3 it would be very interesting to see how realistic your tropospheric chemistry is. Please provide some more details, e.g. references to previous model studies if available, or even a short evaluation of your tropospheric chemistry scheme. Otherwise it is hard to judge how reliable the simulated ozone changes are.

The chemistry scheme used here is a standard configuration of the UK Chemistry and Aerosol (UKCA) atmospheric chemistry model, which has been used for many chemistry-climate modelling studies. The tropospheric and stratospheric chemistry scheme used here are not separate, but part of the same model. It includes 41 key chemical species, especially those important for ozone chemistry such as HOx, CO, CH4, N2O, NOx (including lightning emissions etc.), which are involved in 169 chemical reactions. It is simplified, with respect to the inclusion of, e.g., isoprene as compared to the higher complexity tropospheric chemistry model version used in Banerjee et al. (2015).

With regard to validation, our pre-industrial global mean surface ozone values (12.0 ppbv, stated in Table 2) agree well with expectations for pre-industrial times (e.g. Marenco et al., 1994; Hauglustaine and Brasseur, 2001; Cooper et al., 2014). Our baseline pre-industrial value for STE of ozone of 456 Tg/yr (again Table 2) is also in very good agreement with estimates (see for example Banerjee et al., 2015 and references therein). A re-evaluation of the chemistry model (documented in Morgenstern et al. (2009) as cited in the manuscript) is not the purpose of this study. Indeed, a detailed validation would hardly be possible in this paper due to the lack of observational data with respect to pre-industrial, 4xCO2 or G1 conditions, i.e. with respect to the simulations discussed her. After all, our idealised study simply aims to demonstrate how solar geoengineering could affect some of the key mechanisms involved in determining surface ozone concentrations significantly, from a very general perspective.

We have added a few more details about the chemistry and photolysis models in the revised manuscript at the end of section 2.1, in response to both reviewers.

Banerjee et al. Drivers of changes in stratospheric and tropospheric ozone between year 2000 and 2100. Atmos. Chem. Phys. Discuss., 15, 30645–30691 (2015). Now in press in ACP.

Cooper, O. R. et al. Global distribution and trends of tropospheric ozone: An observation-based review. Elementa Sci. Anthropocene, 2, 000029, 2014, doi: 10.12952/journal.elementa.000029.

Hauglustaine, D. A. and Brasseur, G. P. Evolution of tropospheric ozone under anthropogenic activities and associated radiative forcing of climate. J. Geophys. Res. Atmos., 106, 32337–32360 (2001).

Marenco, A. Evidence of long-term increase in tropospheric ozone from Pic du

Midi data series: Consequences: Positive radiative forcing. J. Geophys. Res. Atmos. 99, 16617–16632 (1994).

- P5, L15: The solar irradiance reduction of 49 W/m2 – where does this value come from? Specified by the G1 experimental set-up or calculated by the authors to compensate the surface temperature increase under 4xCO2, taking into account the model specific climate sensitivity? In case the 49 W/m2 are a model-dependent value, it would be interesting to see a short comment about the climate sensitivity of the applied model. How does it compare to other models?

The solar irradiance reduction of 49 W/m2 was found by checking the radiative imbalance at the TOA after the simultaneous solar and CO2 forcings were imposed, combined with the aim to offset the overall effect on the global mean surface temperature, a simple and easy to calculate climate change metric. The final value was the result of trial-and-error testing to optimize the value with respect to these two goals. To a certain degree, the necessary solar dimming is expected to be model-dependent. To put our results into perspective: a multi-model study of the G1 experiment by Schmidt et al. (2012) found values between 48 and 53 W/m2 for three models and one model for which 64 W/m2 were needed. Consequently, 49.0 W/m2 for the model used here lies within the range found for previous climate modelling studies, which have typically been carried out without interactive chemistry. As stated in the original manuscript we found a value of 50.1 W/m2 for the non-interactive case, which equally lies within the range of the previous modelling results. We have added two sentences about this in section 2.2:

"This value lies well within the range found in previous G1 modelling studies (e.g. Schmidt et al., 2012). It was obtained by iterating the radiative imbalance at the top of the atmosphere and the global mean surface temperature response to various values of solar dimming, thereby optimizing the latter towards a zero

offset from the pre-industrial simulation."

Schmidt, H. et al. Solar irradiance reduction to counteract radiative forcing from a quadrupling of CO2: Climate responses simulated by four earth system models, Earth Syst. Dyn., 3, 63–78, doi:10.5194/esd-3-63-2012, 2012.

- P5, L20/21: Are 75 years enough with a coupled ocean? From Fig. 1 I got the impression that the 4xCO2 experiment is not yet in equilibrium after 75 years.

The original atmosphere-ocean coupled design of the G1 experiment was intended for 50 year long simulations as defined by Kravitz et al. (2011). Here, we chose a rather careful approach and ran for 75 years and discarded the first 25 years of each simulation in the analysis. For the 4xCO2 experiment, we chose the same run length even though small transient effects remain. In general, an atmosphere-ocean coupled model will not be absolutely in equilibrium after 75 years in response to a 4xCO2 forcing. As shown by Li et al. (2013) this takes several thousand years of simulation even for models much simpler than the one used here. Running the model to equilibrium would thus be disproportionate and not necessary for the questions addressed in this study. In fact, we extended the 4xCO2 simulation to 200 years before the submission of the manuscript, but decided to stick to the same run-length. For the G1 run this has no significant effect on the conclusions presented in our paper. After 25 years the transient changes become small enough for a fair analysis with respect to timescales of interest for the present study.

Kravitz, B. et al. The Geoengineering Model Intercomparison Project (GeoMIP). Atmos. Sci. Lett., 12, 162–167, doi:10.1002/asl.316, 2011.

Li, C. et al. Deep-ocean heat uptake and equilibrium climate response. Clim. Dyn., 40, 1071–1086, 2013.

- P6, L9-11: It seems that the authors performed some additional sensitivity runs that are not further discussed in the manuscript. For me this is a bit unsatisfying. How does the fixed ozone field differ from the interactive ozone? Which other chemical species were kept fixed at PI levels? How large is the RF of those species?

We originally mentioned the non-interactive runs to allow the dimming needed to be compared with the results of previous G1 studies, which did not consider composition feedbacks (see our reply immediately above). However, the non-interactive runs are not central to the arguments made in our paper and we have thus deleted this reference in the revised manuscript.

- P6/7, discussion of Fig. 3: I think this part needs some revision. The discussion of temperature and ozone changes in the 4xCO2 and G1 experiments is a bit unstructured. From what is written in L21-25 (P6) I got the impression that the ozone changes in G1 are also related to a colder stratosphere, although Fig. 3d shows a warming for large parts of the stratosphere. I would first show the temperature changes in 3a and b, and then the ozone changes in 3c and d.

Our original wording was evidently not sufficiently clear. Figure 3d shows the temperature differences between G1 and 4xCO2 (see page 31980, lines 12-13 and also the label in Figure 3d) and not between G1 and piControl. As (initially) correctly understood by the reviewer, we say that both stratospheres are much cooler under increased atmospheric CO2 than under pre-industrial conditions and that a large part of the stratospheric ozone changes in G1 is indeed due to these cooler stratospheric conditions, see p. 31979 lines 20-25 to p. 31980 lines 1-12. However, the overall ozone increases are larger in G1 than in 4xCO2. This difference is what we explain in more detail in the manuscript. We show that these additional ozone increases are not mainly driven by temperature changes, but rather by changes in the abundance of oxygen atoms and OH radicals in the stratosphere.

In the revised manuscript, we have added the following sentence to the text before equations (R1.1) and (R1.2):

"Note that this cooling effect largely persists in G1; the stratosphere is warmer in some areas than in 4xCO2, but remains much colder than in piControl (compare Fig. 3c and 3d)."

and also added a clarification to the caption of Figure 3. We now hope that this point is sufficiently clear.

- P7, L2/3: Why does the stratospheric cooling shift the ratio between atomic oxygen and ozone towards ozone? Why does the atomic oxygen in R1.2 come from? Photolysis?

It is the temperature dependence of the reaction O + O2 + M -> O3 + M that is of prime importance for the partitioning of odd oxygen (i.e. the ratio between O and O3) in the middle-upper stratosphere. We have added a few words to clarify this. The atomic oxygen is, of course, produced by photolysis. This is basic stratospheric chemistry and a more detailed explanation within an atmospheric chemistry journal would surely be otiose.

- P7, 16-20: Why is the decrease in atomic oxygen only visible on pressure levels and not on model levels? Please provide at least a short explanation. Referring to another paper is not very reader-friendly.

This is due to pressure changes in the stratosphere. In the vertical, such pressure changes can lead to an offset between the fixed altitude coordinate and the adaptable pressure coordinate. This is, perhaps, of interest to atmospheric chemists from a purely modelling point of view and was discussed in detail in the cited paper by Jonsson et al. (2004). However, it is a deviation from the story

of how the stratosphere affects the troposphere and we have decided to omit this comment.

- P7, L32/22: What is the reason for the increased upper stratospheric NOx abundances under 4xCO2?

The increased upper stratospheric NOx abundances are a net result of increased transport of the precursor species nitrous oxide (N2O) into the upper stratosphere from the troposphere under 4xCO2 due to the strengthening of the Brewer-Dobson circulation, a ubiquitous climate modelling feature, and differences in chemical NOx production and loss, e.g. due to the reaction N2O+O(1D) (where O(1D) also changes significantly as highlighted in the manuscript), the coupling with the HOx cycle as well as temperature dependencies of the chemical reactions, see for example Revell et al. (2012) for an overview of the effects involved. We do not want to distract the reader with a detailed description of these changes; we would like to keep the message (and thus the discussion on detailed chemical changes) as simple and short as possible. Therefore, we focus on the main effects, i.e. HOx, oxygen radicals and temperature changes here.

Revell, L. E. et al. The effectiveness of N2O in depleting stratospheric ozone, Geophys. Res. Lett., 39(15), 1–6, doi:10.1029/2012GL052143, 2012.

- Table 2: Are the shown changes all statistically significant?

As noted in the text (and as expected), not all changes are statistically significant. We give the standard deviation for the annual mean data of the last 50 years of each experiment in Table 2 of the revised manuscript.

- Figure 2, right: Since the shown temperature differences range between -4 K and +4 K, I would adjust the color bar.

We chose the non-linear colour bar because of the very different changes in temperature between the two scenarios. The alternative would have been to use different colour scales for the two subplots. However, this could also lead to confusion, as evident in the misinterpretation of Figure 3 in this review. We think that the chosen colour scale is a good compromise to point out the main features in surface temperature change of either scenario without changing the colour scale between the two plots.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 31973, 2015.

Atmos. Chem. Phys. Discuss., 15, C12595–C12603, 2016
www.atmos-chem-phys-discuss.net/15/C12595/2016/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ozone changes under solar geoengineering: implications for UV exposure and air quality" *by* P. J. Nowack et al.

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We thank the reviewer very much for the positive and constructive comments, which we found very helpful to improve our manuscript.

Below, we reply point-by-point to the referee's comments (normal font) in bold, italic. Any references to pages and line numbers made by us refer to the published online version of the discussion paper and may have changed in the revised version.

This global modeling study investigates the mechanisms and processes where solar radiation management (SRM) geoengineering techniques can impact surface UV and

tropospheric chemistry. The implied SRM technique is that of space mirrors, where the solar constant is turned downwards to emulate a blocking out of the sun's rays, the magnitude of which tuned to match the radiative forcing from 4xCO2 (this is the so called G1 experiment).

The authors present some "standard results" (e.g. spatial pattern of temperature changes) before discussing chemical and related impacts more thoroughly. Key results include how a cooling stratosphere couples with reduced water vapor to drive ozone increases in the G1 simulations. The G1 simulations also increase tropospheric ozone, driven mainly by reduced water vapor (reduced OH production) and UV penetration (due to higher stratospheric ozone). The authors also highlight that the reduced UV could be important for human health through reduced vitamin D production.

Overall, I feel that this study positively adds to the growing literature analyzing the impacts of geoengineering techniques. Composition and UV impacts have not been studied in detail, and, while the scenarios are not necessarily ideal (e.g. using preindustrial levels of ozone precursors; as also noted by another reviewer), the authors note these weaknesses, and I think that it provides a good basis to compare future work against. In summary, I would be happy to recommend this for publication after the authors have considered my minor comments/corrections below (mostly very minor).

Specific comments (page and line numbers refer to the original Word version)

- P1, L14: Why italicize geoengineering?

This was thought to put some emphasis on the central idea under study. We do not feel strongly about this emphasis, so we leave it out in the revised version.

- P2, L1: "However, despite..." – I feel this sentence rather trivializes an extremely complex issue. It is not possible to just turn off CO2 emissions without all kinds of

(nonatmospheric) consequences!

We certainly did not want to trivialise. We have changed the text slightly to read: "It is recognized that reducing greenhouse gas emissions is difficult so that, under these circumstances, there is discussion on alternative measures to counteract the effects of climate change".

- P2, L2: It's not just researchers talking about GE

The sentence given in the previous reply circumvents this specification as well.

- P2, L16: Suggest: "The central problem..." -> "A major issue..."

Done.

- P2, L18: Full stop after "Earth system" and then start a new sentence.

Done.

- P3, L11: Might want to be clear how UV (or other GE-related factors) can influence surface ozone

In the revised manuscript, we now have a separate short introduction for the potential tropospheric ozone changes driven by changes in the stratosphere. It reads:

"Surface ozone is a pollutant, which has been associated both with diseases of the respiratory system and crop damage (Avnery et al., 2011; Silva et al., 2013). Many countries have introduced emission controls aimed at reducing emissions of tropospheric ozone precursors. However, tropospheric surface ozone depends not just on in situ emissions but also on processes in the stratosphere.

For example, changes in stratospheric ozone will impact tropospheric chemistry by altering the photolysis environment in the troposphere (Madronich et al., 2015). Similarly, the transport of ozone from the stratosphere is an important component of the tropospheric ozone budget (e.g. Holton et al., 1995; Neu et al., 2014). Any SRM scheme which affects the stratosphere could therefore also impact tropospheric composition."

- P3, L26: "Finally, section 4..."

Done.

- P4, L7: Ref for MetUM?

The atmosphere, ocean and sea-ice model versions and their coupling were described together in the HadGEM3 reference to Hewitt et al. (2011). For the atmosphere model, this paper is definitely the most appropriate one to cite. In order to avoid confusion, we add the citation a second time in the revised manuscript.

- P4, L12: comma after MetUM

Done.

- P4, L15-: Refs for these models?

In the revised version we also include some more original citations for the standalone ocean and sea-ice models even though the actual atmosphere-ocean-sea-ice coupled model used here is described in Hewitt et al. (2011).

- P4, L21-: Is aerosol chemistry included? Are there composition/climate feedbacks with CH4 and N2O? Clarify whether the photolysis scheme respond to clouds, ozone

and solar flux?

The CLASSIC aerosol scheme is included in the model (Bellouin et al., 2011), which is not coupled to UKCA. Concerning composition-climate feedbacks with CH4 and N2O the model is fully interactive. The FastJX photolysis scheme is interactive with respect to clouds, ozone and solar flux, we have added the following sentences to clarify this:

"Ozone, nitrous oxide and methane are fully interactive in the model so that their changes feedback onto changes in radiation. [...] Photolysis in FastJX responds, inter alia, to ozone and solar flux as well as to multiple layers of clouds of varying degrees of thickness."

Bellouin, N. et al.: Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate, J. Geophys. Res., 116, D20206, doi:10.1029/2011JD016074, 2011.

- P5, Sect 2.2: I would be explicit that the CFC levels are (presumably) zero in the simulations.

We have added the following sentence to be more explicit about the settings:

"By design, the G1 experimental set-up does not include pre-defined changes in surface emissions of ozone depleting substances from anthropogenic sources (e.g. CFCs whose abundance is equal to zero in this set-up), or tropospheric ozone precursors."

- Also, would any putative space mirrors be uniformly efficient at all wavelengths?

Indeed, this is another idealised assumption. Possibly, mirrors could even be designed to dim some wavelengths more than others, similar to the wavelength-

dependent reflective properties of aerosols. However, in practice, a uniformly dimming mirror would be most effective in terms of the radiative energy reflected per unit area (assuming the very hypothetical case of an actual implementation of space-mirror geoengineering). An only partially reflective mirror would let through certain wavelengths of light and thus a part of the potentially reflected energy incident on the mirror's surface. Space-mirror geoengineering would likely be strongly limited by (financial) resources to bring sufficient mirror surface area into space. Therefore, the mirror surface area would likely be the 'bottleneck' in any such operation, thus making uniformly (totally) reflective mirrors the most effective ones.

- P7, L6: Delete "rather"

Done.

- P7, L8: "heating by higher ozone levels"

Done.

- P7, L11: "in G1, as discussed below."

Done.

- P7, L33: why is NOx higher? Temperature effects?

The increased upper stratospheric NOx abundances are a net result of increased transport of the precursor species nitrous oxide (N2O) into the upper stratosphere from the troposphere under 4xCO2 due to changes in the strength of the Brewer-Dobson circulation (which are effectively reset in G1) and differences in chemical NOx production and loss, e.g. due to the reaction N2O+O(1D) (where

O(1D) also changes significantly as highlighted in the manuscript), the coupling with the HOx cycle and last but not least temperature dependencies of the chemical reactions, see for example Revell et al. (2012) for an overview of the effects involved. We added the Revell et al. (2012) citation to the paper to highlight the issue.

Revell, L. E. et al. The effectiveness of N2O in depleting stratospheric ozone, Geophys. Res. Lett., 39(15), 1–6, doi:10.1029/2012GL052143, 2012.

- P8, L6-8: Is this because the BDC slows?

Yes, this is what we meant to say by "the residual circulation (not shown) and thus ozone (Fig. 3b) in the tropical lower stratosphere is almost brought back to pre-industrial levels". However, the Brewer-Dobson circulation does not slow down relative to pre-industrial levels in our simulations, i.e. it is effectively reset by the solar dimming.

- P8, L21: “As discussed in section 1, tropospheric ozone...affecting human health and air quality”

Done.

- P8, L31: The photolysis reaction has a temperature dependence too (vibrational excitation), which further complicates things.

Yes, there is also a cross section temperature dependence for wavelengths longer than about 300nm, both of which are included in the photolysis scheme. The offsets are rather small and likely not as significant as the changes in UV fluxes and humidity. In any case they are indeed included in the reaction fluxes now given in the revised manuscript (see next question).

- P9, L13-22: Do you have tropospheric ozone budget data to help with this analysis?

We have added reaction fluxes for the two discussed reactions to our manuscript.

- P9, L24: “in G1, as shown by the data in Table 2.” (A comma and then “see X” does not read well – there are other examples that could be addressed.)

Thank you, we replaced such forms in this and any other occasion where we found this possible.

- P10, L3-14: I would remind the reader that the conclusions are based on simulations with PI conditions. Some of the impacts (ΔT, Δhumidty) will be important for trop chem in general.

We have added the sentences

"Here, we assume pre-industrial conditions by following the G1 scenario, which only allows for low, natural background pollution. Under different forcing scenarios other aspects of tropospheric chemistry could change the surface ozone response."

to our discussion in section 3.4 in the revised manuscript. We have also added the sentences

"Nevertheless, changes in humidity and photolysis as described here are robust modelling features that could occur under a range of geoengineering scenarios and these changes would impact tropospheric chemistry. These mechanisms will be key to tropospheric chemistry considerations under geoengineering in general."

later in the same section.

- P11, L16-19: You might be able to use data in Madronich (2007) to estimate the impact on vitamin D (he has empirical values for ozone/weighted-UV derived for lots of different action spectra). ...However, if the simulations have PI ODS levels, is it really worth talking up the health impacts too much?

We agree and really just calculate the UVI to illustrate the well-known point about unforeseen consequences. We would thus prefer to stick to the UVI-index calculations already made, mainly because the empirical formula used for the cloud UV changes is also designed to yield the UVI. This way, we are able to compare the results directly.

- P14, L3: I would emphasize this weakness first. It's not terminal, but it is important. [You might be able to point to other studies that have investigated UV-tropospheric chemistry links to infer potential impacts if the ozone precursors were not at PI levels]

We have moved this point to the top of the paragraph and reference other studies (Young et al., 2013 and Squire et al., 2015), which have looked at such effects.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 31973, 2015.

Stratospheric ozone changes under solar geoengineering: implications for UV exposure and air quality

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Abstract

Various forms of *geoengineering* have been proposed to counter anthropogenic climate change. Methods which aim to modify the Earth's energy balance by reducing insolation are often subsumed under the term Solar Radiation Management (SRM). Here, we present results of a standard SRM modelling experiment in which the incoming solar irradiance is reduced to offset the global mean warming induced by a quadrupling of atmospheric carbon dioxide. For the first time in an atmosphere-ocean coupled climate model, we include atmospheric composition feedbacks for ~~such as ozone changes under~~ this experiment~~scenario~~. While the SRM scheme considered here could offset greenhouse gas induced global mean surface warming, it leads to important changes in ~~Including the~~ atmospheric composition ~~changes.~~., W~~w~~e find large stratospheric ozone increases that induce significant reductions in surface UV-B irradiance, which would have ~~with~~ implications for vitamin D production. In addition, the higher stratospheric ozone levels lead to decreased ozone photolysis in the troposphere. In combination with lower atmospheric humidity under SRM, this results in overall ~~, and increases in~~ surface ozone ~~concentrations~~increases in the idealised G1 experiment. ~~, b~~Both UV-B and surface ozone changes are ~~of which could be~~ important for human health. We therefore highlight that both tropospheric and

stratospheric ozone changes ~~should~~ must be considered in the assessment of any SRM scheme, due to their important roles in regulating UV exposure and air quality.

1. Introduction

The scientific consensus (Stocker et al., 2013) is that man-made climate change caused by anthropogenic emissions of greenhouse gases such as carbon dioxide is taking place. It is recognized that reducing greenhouse gas emissions is difficult so that, ~~However, despite this knowledge, atmospheric carbon dioxide levels are still rising rapidly. U~~under these circumstances, there is ~~researchers have reopened the~~ discussion on alternative measures to counteract the effects of climate change (e.g. Bala and Caldeira, 2000; Cicerone, 2006; Crutzen, 2006). Such interventions ~~manipulative measures~~ are commonly referred to as geoengineering, “the intentional large-scale manipulation of the environment that is intended to reduce undesired anthropogenic climate change” (Keith, 2000).

Here, we use an atmosphere-ocean chemistry-climate model to study atmospheric composition changes for one of the most common geoengineering modelling experiments: the reflection of solar energy before it can enter the Earth's atmosphere, an idea often depicted by the use of space mirrors (Early, 1989; Seifritz, 1989). This idealised geoengineering experiment belongs to methods subsumed under the term Solar Radiation Management (SRM). SRM methods aim to offset the additional radiative forcing due to increases in atmospheric greenhouse gas concentrations by reflecting solar radiation before it can reach the Earth's surface. A major issue ~~The central problem~~ with any SRM scheme is that they are not designed to directly address the cause of change, namely the elevated levels of carbon dioxide and other greenhouse gases in the Earth system. Instead, they ~~but, rather, to~~ affect other processes whose changes counteract those due to the greenhouse gases (Shepherd, 2009). This has been demonstrated in numerous SRM modelling studies (e.g. Bala and Caldeira, 2000; Bala et al., 2002, 2003, 2008; Jones et al., 2011; Kravitz et al., 2012, 2013b; Lunt et al., 2008; Matthews and Caldeira, 2007; Niemeier et al., 2013; Ricke et al., 2010; Schmidt et al., 2012; Tilmes et al., 2013).

Atmospheric composition changes under SRM have received much attention in the context of stratospheric particle injection schemes (Budyko, 1977; Crutzen,

2006) as increased particle loadings could enhance the heterogeneous catalysis of reactions that eventually lead to ozone depletion (e.g. Heckendorn et al., 2011; Pitari et al., 2014; Pope et al., 2012; Rasch et al., 2008; Tilmes et al., 2008, 2009, 2012; Weisenstein and Keith, 2015). This would have important implications for human health since ozone is the major absorber of solar UV-B radiation. UV-B radiation ~~,~~ ~~which~~ interacts with the human DNA and ~~which~~ has been connected to many acute and chronic illnesses of the eye, immune system and skin and, *inter alia,* to various forms of skin cancer (e.g. Norval et al., 2011; Slaper et al., 1996).

However, UV-B radiation is also needed in beneficial biological processes such as in the photobiological production of vitamin D (Holick, 1981). Consequently, a large future increase in the total column amount of ozone, and thus decreased surface UV-B radiation, could itself have severe adverse effects on life on Earth (McKenzie et al., 2009). Vitamin D deficiency, for example, has been related to an increased likelihood of occurrence of internal cancers, autoimmune diseases, mental illnesses, lower bone density ~~and many more~~ (e.g. Mora et al., 2008; Norval et al., 2011; Ross et al., 2011; Williamson et al., 2014). ~~Therefore, significantly lower surface UV-B could also have considerable adverse effects on human health or could make lifestyle changes necessary (McKenzie et al., 2009). Similarly, o~~Other organisms in~~of~~ the biosphere also depend on UV radiation including certain types of plants whose defence mechanisms against pests and pathogenic micro-organisms are regulated by UV-B radiation (Williamson et al., 2014).

~~Another important factor is changes in~~Surface ozone is ~~at the surface, where ozone acts as~~ a pollutant, which has been associated both with diseases of the respiratory system and crop damage (Avnery et al., 2011; Silva et al., 2013). Many countries have introduced emission controls aimed at reducing emissions of tropospheric ozone precursors. However, tropospheric surface ozone depends not just on in situ emissions but also on processes in the stratosphere. For example, changes in stratospheric ozone will impact tropospheric chemistry by altering the photolysis environment in the troposphere (Madronich et al., 2015). Similarly, the transport of ozone from the stratosphere is an important component of the tropospheric ozone budget (e.g. Holton et al., 1995; Neu et al., 2014). Any SRM

scheme which affects the stratosphere could therefore also impact tropospheric composition.

In contrast to the often studied case of particle injection schemes, stratospheric composition changes and their potential tropospheric ~~health~~ impacts in a “space-mirror” geoengineered climate have not yet been included in a 3D atmosphere-ocean modelling study. ~~Here, w~~We investigate changes in ozone, and consequently in biologically active ultraviolet surface radiation (in particular UV-B), contrasting our results with composition changes under pure greenhouse gas forcing. Changes in UV-B fluxes by changes in clouds and surface albedo are also considered. ~~In addition,~~Finally, we briefly discuss potential surface ozone, and thus air quality, changes as a result of SRM.

This paper is organised as follows: sections 2.1 and 2.2 introduce the model used to run the simulations and the experimental setup. Section 3.1 introduces the global and regional surface temperature response. Changes in atmospheric composition and their impact on surface UV and air quality are explained in sections 3.2. to 3.4. ~~The final~~Finally, section 4 puts our results into context, also regarding other SRM schemes and health implications.

2. Experimental Setup

2.1 Model Description

A version of the recently developed atmosphere-ocean coupled configuration of the Hadley Centre Global Environment Model version 3, additionally coupled to an atmospheric chemistry scheme, has been employed here (Hewitt et al., 2011; Nowack et al., 2015).

For the atmosphere, the UK Met Office's Unified Model (MetUM) version 7.3 is used (Hewitt et al., 2011). The configuration is based on a regular grid with a horizontal resolution of 3.75° longitude by 2.5° latitude and comprises 60 vertical

levels up to a height of ~84 km, and so includes a full representation of the stratosphere. Its dynamical core is non-hydrostatic and employs a semi-Lagrangian advection scheme. The radiation scheme by Edwards and Slingo (1996) is used in the MetUM, with 9 bands in the longwave and 6 bands in the shortwave part of the spectrum, extended by the *k*-distribution method by (Cusack, (1999). Subgridscale features such as clouds and gravity waves are parameterised.

The For ocean dynamics and thermodynamics an updated version of the OPA component (Hewitt et al., 2011; Madec et al., 1998) of is the Nucleus for European Modelling of the Ocean (NEMO) framework model version 3.0, coupled to the Los Alamos sea ice model CICE version 4.0 (Hunke and Lipscomb, 2008) is used. It contains 31 vertical levels reaching down to a depth of 5 km. The NEMO configuration used in this study deploys a tripolar, locally anisotropic grid which has 2° resolution in longitude everywhere, but an increased latitudinal resolution in certain regions with up to 0.5° in the tropics.

Atmospheric chemistry is represented by the United Kingdom Chemistry and Aerosols (UKCA) model in an updated version of the stratospheric chemistry configuration (Morgenstern et al., 2009) which is coupled to the MetUM. A relatively simple tropospheric chemistry scheme that simulates hydrocarbon oxidation is also included, which provides for emissions of 3 chemical species (NO (surface, lightning), CO (surface), HCHO (surface)). In addition, surface mixing ratios of 4 further species (N_2O, CH_3Br, H_2, CH_4) are constrained by calculating the effective emission required to maintain their surface mixing ratios, e.g. for nitrous oxide 280 ppbv and for methane 790 ppbv. This keeps their tropospheric mixing ratios approximately constant at pre-industrial levels in all simulations. Nitrogen oxide emissions from lightning are parameterized according to Price and Rind (1992, 1994). Ozone, nitrous oxide and methane are fully interactive in the model so that their changes in composition feedback onto changes in the radiation. Changes in photolysis rates in the troposphere and the stratosphere are calculated interactively using the Fast-JX photolysis scheme (Bian and Prather, 2002; Neu et al., 2007; Telford et al., 2013; Wild et al., 2000). Photolysis in FastJX responds, *inter alia*, to ozone and solar flux as well as to multiple layers of clouds of varying degrees of thickness.

2.2 The Simulations – The GeoMIP G1 Experiment

Our simulations follow standards set for the *G1* experiment (see Table 1), which was defined as part of the Geoengineering Model Intercomparison Project (GeoMIP) (Kravitz et al., 2011, 2013a). In the *G1* experiment the effect of an abrupt quadrupling of atmospheric carbon dioxide (CO_2) on the global mean surface temperature is approximately offset by reducing the model's solar constant. This can be thought of as an experiment in which space-mirrors reflect sunlight before it enters the Earth's atmosphere (Early, 1989; Seifritz, 1989). Starting from approximately pre-industrial concentrations with atmospheric CO_2 at ~285 ppmv (*piControl*), we thus carried out, firstly, an abrupt *4xCO2* experiment, in which atmospheric CO_2 is instantaneously quadrupled to ~1140 ppmv and, secondly, a *G1* type experiment in which the global warming caused by $4xCO_2$ was offset by a solar irradiance reduction of 49.0 Wm^{-2} (~3.6%). This value lies well within the range found in previous G1 modelling studies (e.g. Schmidt et al., 2012). It was obtained by iterating the radiative imbalance at the top of the atmosphere and the global mean surface temperature response to various values of solar dimming, thereby optimizing the latter towards a zero offset from the pre-industrial simulation. The radiative forcing in the *4xCO2* experiment roughly matches the levels attained by the end of the 21st century under the transient RCP8.5 forcing scenario defined for the Coupled Model Intercomparison Project phase 5 (Moss et al., 2010; Taylor et al., 2012). Both experiments were run for 75 years after the CO_2 and solar forcings were imposed. For analysis, we use the last 50 years of each experiment in the following. By design, the G1 experimental set-up does not include pre-defined changes in surface emissions of ozone depleting substances from anthropogenic sources (e.g. CFCs whose abundance is equal to zero in this set-up), or tropospheric ozone precursors.

The highly idealised nature and theoretical simplicity of the *G1* experiment allows us to discuss possible unintended consequences of solar geoengineering in an intuitive way. Our stratospheric chemistry scheme allows a detailed analysis of possible changes in UV penetration into the troposphere as well as of stratosphere-troposphere exchange of ozone. Our tropospheric chemistry scheme, while simplified, then allows a simple, first-order quantification of the impact of these on

tropospheric composition. ~~These include changes in composition, UV transmission as well as air quality.~~ While the exact quantification of any changes would be strongly dependent on both forcing scenario and SRM scheme, this study aims to demonstrate ~~in a qualitative way~~ why changes in these metrics are to be expected for any SRM scheme.

3. Results

3.1 Surface Temperature Response

The temporal evolution of the global mean surface temperature for all simulations is shown in Fig. 1. As expected, a rapid warming is found in *4xCO2* relative to *piControl* in response to the abrupt forcing whereas *G1* remains (by design) at effectively the same average surface temperature (Table 2) Although surface temperatures are offset globally, there are important regional differences between *4xCO2* and *G1*. ~~Changes in atmospheric composition (e.g. ozone) exert an additional radiative forcing which can alter the magnitude of the surface warming response to CO_2 (Nowack et al., 2015) and the amount of solar dimming needed to offset it. For example, we needed an additional solar constant reduction of 1.1 Wm^{-2} (50.1 Wm^{-2} instead of 49.0 Wm^{-2}) to offset the global mean surface warming in experiments where we kept ozone and other chemical species fixed at pre-industrial levels. However, we focus on the interactive chemistry model results here. For the pattern of remaining surface temperature anomalies (Fig. 2)~~As shown in Fig. 2, the ~~our~~ model yields the characteristic distribution of overcooling in the tropics and warming at high latitudes in *G1* (Kravitz et al., 2013b), an effect which can be explained by the proportionally larger impact of reducing insolation on the tropics than on high latitudes (Bala and Caldeira, 2000; Lunt et al., 2008).

3.2 Stratospheric Ozone and Temperature Changes

Fig. 3a to Fig. 3d show latitude-height cross sections of changes in zonal mean ozone mass mixing ratio and zonal mean temperature. We find large increases in ozone in the middle-upper stratosphere (~30-50 km altitude, Fig. 3a and 3b) under both *4xCO2* and *G1*, a ubiquitous feature in chemistry-climate modelling studies (e.g. Oman et al., 2010) with a cooler stratosphere under increased atmospheric CO_2 concentrations (Fels et al., 1980). Note that this cooling effect largely persists in *G1*; the stratosphere is warmer in some areas than in *4xCO2*, but remains much colder than in *piControl* (compare ~~, see~~ Fig. 3c and 3d). ~~The~~ CO_2-driven ozone increases in the middle-upper stratosphere are well understood and are mainly caused by a slowing of temperature-dependent catalytic ozone (O_3) loss reactions

$$X + O_3 \rightarrow XO + O_2 \quad \text{(R1.1)}$$

$$XO + O \rightarrow X + O_2 \quad \text{(R1.2)}$$

$$\text{Net: } O + O_3 \rightarrow 2\,O_2$$

under cooler stratospheric conditions (Haigh and Pyle, 1982), with the radical species X typically being NO, OH, Cl or Br. In addition, the cooling also shifts the thermal partitioning ~~ratio~~ between atomic oxygen and ozone towards the latter, which further slows down the rate-determining step (R1.2) in the catalytic cycles (Jonsson et al., 2004). As already mentioned, ~~S~~stratospheric cooling due to increased CO_2 persists in *G1*. *In fact,* ~~ and~~ the solar irradiance reduction would, as a single effect, ~~rather~~ be expected to ~~-~~further cool the stratosphere (Bala et al., 2003; Braesicke et al., 2011). However, some regions in the stratosphere are actually warmer in *G1* than in *4xCO2* (Fig. 3d). Increased shortwave heating by more ozone, local tropopause height shifts and changes in dynamical heating certainly contribute to this, and importantly so does less longwave cooling as a result of the much lower stratospheric water vapour concentrations (Maycock et al., 2011) in *G1*, as discussed ~~see~~ below.

~~In spite of the partly warmer stratospheric conditions, t~~The ozone increases in the upper stratosphere are larger in *G1* than under *4xCO2* (compare Fig. 3a to Fig. 3b), see also Jackman and Fleming (2014). In our simulations, there are two main drivers behind this additional ozone increase. Firstly, less ozone is photolysed (O_3 + hv -> O_2 + O) as a consequence of the reduced insolation in *G1*, which happens at the expense of ~~are a significant reduction of stratospheric specific humidity in *G1* in combination with reduced abundances of~~ atomic oxygen ~~species~~ abundances: in *G1*

both ground state O(^{3}P) and excited state O(^{1}D) at a given atmospheric ~~constant~~ pressure ~~levels. Atomic oxygen abundances are decreased in *G1* for both ground state O(^{3}P) and the excited state O(^{1}D) by~~ ~3-8% less abundant than in ~~compared to~~ *4xCO2* (not shown). ~~These decreases are only observable when pressure levels are used as vertical coordinates instead of height coordinates, see Jonsson et al. (2004).~~ Less abundant atomic oxygen in turn ~~at a given atmospheric pressure~~ implies a slowing of reaction (R1.2) and thus further reduced ozone loss. Secondly, ~~In addition,~~we find a significant decrease in stratospheric specific humidity in *G1*, which reduces HO_x (OH, HO_2, H) formation and therefore ozone loss via, for example, (R1.1) and (R1.2). Specifically, the stratosphere ~~our model yields an~~is ~10-20% drier ~~atmosphere for~~in *G1* than in~~for~~ *piControl*~~, as compared to a much more humid atmosphere in *4xCO2* (stratosphere wetter by ~30% than pre-industrial)~~. This is related to ~~e drier atmosphere under SRM is part of~~ a weaker hydrological cycle under SRM (e.g. Bala et al., 2008; Govindasamy et al., 2003; Kravitz et al., 2013b; Lunt et al., 2008; Matthews and Caldeira, 2007; Ricke et al., 2010; Schmidt et al., 2012; Tilmes et al., 2013, 2009), which gives rise to characteristic reductions in global mean precipitation (~~see~~ Table 2) and evaporation. In contrast, ~~T~~the more humid stratosphere found under *4xCO2* (~30% wetter than pre-industrial) results in greater production of HO_x species, which is additionally coupled to the above mentioned changes in O(^{1}D) via the HO_x-producing reaction H_2O + O(^{1}D) -> 2 OH. As O(^{1}D) concentrations are lower in *G1* than in *4xCO2*, this further enhances the differences in HO_x; overall ~~from water vapour and thus more ozone loss via (R1.1) and (R1.2) than in *G1* (~~the abundance of OH and HO_2 is ~15-25% smaller in the middle-upper stratosphere in *G1*~~).~~ ~~Similarly~~Finally, higher ~~abundances~~ levels of nitrogen oxides (NO_x = NO, NO_2; ~5-13%) in the upper stratosphere under *4xCO2* *will* also contribute to the differences in ozone. They are mainly driven by changes in stratospheric temperature, photolysis, transport of the NOx precursor nitrous oxide as well as its reaction with O(^{1}D); a discussion of various factors involved is for example given in ~~(~~Revell et al.~~,~~ (2012). Changes in other radical species play secondary roles in this experiment (Jackman and Fleming, 2014).

In the tropical lower stratosphere, we find ozone decreases under *4xCO2*, which is characteristic for an acceleration of the Brewer-Dobson circulation under CO_2 driven tropospheric warming (Nowack et al., 2015; Shepherd and McLandress,

2011). In response to solar geoengineering, the residual circulation (not shown) and thus ozone (Fig. 3b) in the tropical lower stratosphere is almost brought back to pre-industrial levels. The remaining ozone decreases mainly result from an effect often referred to as "inverse self-healing" of the ozone column (e.g. Haigh and Pyle, 1982; Jonsson et al., 2004; Portmann and Solomon, 2007), in which the increased ozone concentrations in the upper stratosphere allow less shortwave radiation to propagate to lower altitudes. Relative to pre-industrial conditions, this mechanism acts in concert with the (by design) reduced insolation to leave fewer photons of relevant wavelengths to produce ozone in the lower stratosphere. However, these effects are partly compensated by coincident decreases in ozone losses in *G1*, mainly due to the lower temperatures and lower HO_x concentrations than in *piControl*. Overall, the significant changes in stratospheric ozone have important implications for UV fluxes into the troposphere and to the surface, see sections 3.3 and 3.4.

Swapped section 3.3 and 3.4 in response to Referee 1

3.43 The effect of column ozone and cloud changes on surface UV-B

UV-B surface fluxes can change for a variety of reasons (Bais et al., 2015; McKenzie et al., 2011). Changes in column ozone have the potential to provide particularly strong contributions since ozone is the only major absorber of UV-B radiation in the atmosphere. As discussed above, SRM could lead to changes in column ozone; in *G1*, we find that relative to *piControl* the global mean column ozone increased by ~8% compared to only ~4% under *4xCO2*~~,~~ (~~see~~ Fig. 4~~5~~ and Table 2).

The harmful effect of UV exposure on human skin is commonly measured using the UV-Index (UVI), starting at 0 and with higher UVI equalling greater skin-damaging potential (WHO, 2002). Here, we use the approximate formula of Madronich (2007) to estimate UVI changes in response to the changes in column ozone in *4xCO2* and *G1* under clear-sky, unpolluted conditions

$$UVI \sim 12.5\mu^{2.42}(\Omega/300)^{-1.23} \quad (2~~3~~)$$

where μ is the cosine of the solar zenith angle and Ω the total vertical ozone column in Dobson Units (DU). As a further approximation, we use monthly and zonal mean values for column ozone, but have updated the solar zenith angle on a daily basis

according to the changing solar declination. The resulting UVI is therefore both a function of the changing angle of incidence of the Sun's radiation to the Earth's surface and the seasonally varying column ozone (Fig. 45c and 45d) at a given location. The UVI found for *piControl* at noon and relative changes (ΔUVI) for *G1* and *4xCO2* in percentages, are shown in Fig. 45e and 45f, (see also Table 2 for global mean differences). In *G1*, the UVI decreases everywhere during the whole year due to both changes in column ozone and the 3.6% reduced intensity of the solar radiation. However, the effect of the changes in ozone generally dominates. In particular, during Northern Hemisphere (NH) spring and summer average decreases of 10-20% are found at NH mid and high latitudes in *G1*. We caution that althoug the percentage changes at high latitudes may be larger, but they are relative to occur on much lower background UVI levels. In addition, formula (23) is expected to perform less well in areas of high surface albedo, as well as is the case in those regions with widespread occurrences of sea and land-ice (Madronich, 2007). Nevertheless, a Still, we highlight that further lowered reduction in UV irradiance in already light-poor seasons and regions could aggravate medical conditions connected to vitamin D deficiency. We note that vitamin D production exhibits a slightly different sensitivity to certain wavelengths of solar radiation than is assumed in the calculation of the UVI (Fioletov et al., 2009; McKenzie et al., 2009) so that our calculations should be considered as qualitative.

However, cColumn ozone changes are not the only factor with the potential to change surface UV as a result of climate engineering. Changes in clouds, surface reflectivity (due to surface albedo changes), or aerosols could all significantly affect UV transmission, reflection and scattering. Here, we focus on the impact of ozone and cloud changes, assuming that other changes are small under pre-industrial background conditions. The residual high-latitude warming in *G1* (Fig. 2b) implies that albedo changes could play a role, e.g. due to decreases in snow and sea-ice. However, in our model, the higher temperatures do not suffice to trigger statistically significant ice or snow loss under SRM, in agreement with multi-model studies of the *G1* experiment (Kravitz et al., 2013b; Moore et al., 2014).

A common way to estimate the average effect of clouds on shortwave (SW) surface radiation is the cloud modification factor (CMF_{SW}). The CMF_{SW} is the total

solar irradiance (Wm^{-2}) reaching the Earth's surface at any point (all-sky) divided by its idealised clear-sky value in which any cloud effects are ignored (den Outer et al., 2005). A CMF_{SW} of 1 thus implies that the net cloud effect on surface SW radiation is zero, values larger than 1 imply SW amplification by clouds, values smaller than 1 net reflection of SW radiation by clouds. Fig. 56a and 56b show differences in the CMF_{SW} for *4xCO2* and *G1* relative to *piControl*. Under *4xCO2*, the overall pattern of CMF_{SW} changes is in agreement with previous (chemistry-)climate modelling results (Bais et al., 2011, 2015) under greenhouse gas forcing. In *G1* (Fig. 56b), the CMF_{SW} is predicted to increase in many regions while decreases are virtually non-existent. Similar cloud changes have been found in previous *G1* modelling studies and have been attributed to reductions in the highly reflective cloud cover at low altitudes (Kravitz et al., 2013b; Schmidt et al., 2012). Consequently, an increase in surface SW radiation from cloud changes is expected in *G1*, in contrast to the decrease in UVI which would follow ~~could partly diminish~~ the ~~ΔUVI due to~~ column ozone changes.

In order to compare the UV effects of changes in the CMF_{SW} and changes in ozone, we use an empirical relationship established by den Outer et al. (2005) and modified by Staiger et al. (2008) to estimate the effect of the CMF_{SW} changes in terms of the UVI at noon. The results are presented in Fig. 56c and ~~6~~5d. In *G1*, the UVI changes by clouds are overall positive. As expected, this is the opposite sign response to the UVI changes induced by ozone. However, the cloud effect is much smaller with percentage increases of only ~1-2% for most latitudes and times. Only during NH summer, between around 40N-60N, are UVI increases of comparable size (~5%) to the decreases driven by changes in the ozone column ~~ozone attained~~. ~~Indeed, o~~Our calculations show that cloud effects are generally small and do not offset ozone-induced UV changes in light-poor seasons, ~~i.e. at~~which are the times when major problems connected to vitamin D deficiency primarily occur.

In summary, our results imply that differences in column ozone and ~~thus~~ hence surface UV fluxes represent an important ~~other example of a~~ change to the climate system, which could arise following a SRM scheme and which ~~that~~ is of potential importance for human health. ~~and lifestyle, but which cannot be offset in a simple manner by proposed SRM methods. Such considerations have~~These changes would need to be taken into account when evaluating benefits and risks of

any possible geoengineering scheme in which elevated atmospheric CO_2 concentrations persists.

3.43 Tropospheric Ozone Changes

As mentioned in section 1, Ttropospheric ozone is an important factor inaffects air quality,; it affects human health and ecology, see section 1. Ozone concentrations in the troposphere are controlled by a variety of processes which could be affected by SRM. These include

i) photochemical processes influenced by changing UV-B (280-315 nm) and UV-A (315-400 nm) fluxes into the troposphere (Madronich et al., 2015; Williamson et al., 2014). High energy photons needed to produce ozone from molecular oxygen (λ < 240 nm) are absorbed at higher altitudes and tropospheric ozone levels are determined by other mechanisms of ozone production and loss processes. For example, under clean environmental background conditions, ozone loss and production of the hydroxyl radical OH via

$$O_3 + h\nu\ (\lambda < 328\ nm) \rightarrow O_2 + O(^1D)\ (R32.1)$$

$$O(^1D) + H_2O \rightarrow 2\ OH\ (R32.2)$$

$$Net:\ O_3 + H_2O + h\nu \rightarrow O_2 + 2\ OH\ (R32)$$

is of prime importance. This reaction pathway is non-linearly dependent on stratospheric ozone changes due to the photons needed in reaction (R32.1) (McKenzie et al., 2011).

ii) changes in tropospheric concentrations of chemical species involved in the formation of ozone or its depletion, for example due to changes in atmospheric humidity and thus in concentrations of a key reactant in loss reactions such as (R32).

iii) changes in Stratosphere-Troposphere Exchange (STE) (Holton et al., 1995; Lin et al., 2014, 2015; Morgenstern et al., 2009; Neu et al., 2014; Zeng et al., 2010), i.e. due to changes in the transport of ozone from the ozone-rich stratosphere into the troposphere. Such changes are strongly coupled to changes in atmospheric dynamics.

In our simulations, there is a global mean surface ozone increase in *G1* (+5.0%) and a decrease in *4xCO2* (-4.2%), see Table 2. The differences between the runs are to first order determined by processes *i* and *ii*. Firstly, UV fluxes into the troposphere decrease in *G1* ~~both~~ due both to the solar irradiance reduction and the increase in ~~greater~~ stratospheric ozone ~~concentrations and the solar irradiance reduction~~. The~~is~~ UV reductiones in *G1* relative to *piControl* leads to a ~5-10%~~ozone loss directly via a~~ reduction in the flux through ~~photolysis~~ reaction (R3~~2~~.1) in the tropical troposphere (and ~15% reduction at higher latitudes). These results contrast with the changes between *4xCO2* and *piControl* where the reaction flux increases in the tropical troposphere by ~15%. It is clear that changes in the stratosphere under both increased greenhouse gases, or under solar radiation management, would have important consequences for the UV fluxes into the troposphere and, hence, for surface irradiation and tropospheric chemistry. SRM does not avoid changes to the stratosphere (and hence to the troposphere) that increased CO2 would lead to.

~~which slows reaction (R2.2).~~ Secondly, ~~tropospheric ozone loss is further decreased as a result of the up to 20% lower~~ the tropospheric humidity ~~found~~ under SRM contrast significantly with those found under *4xCO2*. In the latter case, tropospheric humidity increases while for G1 we find, in common with many other studies mentioned above, a weakening of the hydrological cycle and reduced specific humidity. In our calculations tropospheric humidity is up to 20% lower in *G1* under SRM than in *piControl*~~,~~. In consequence, (R3.2) slows down by ~10-20% in the lower-middle troposphere and by up to ~25-30% in the upper troposphere in *G1*. a~~s compared to the much more humid conditions under *4xCO2*, which gives rise to the opposite sign response.~~

Changes in STE (*iii*) have a negligible effect on the global mean surface ozone change in *G1*~~, see~~ (Table 2). Nonetheless, STE can be regionally and seasonally important under *4xCO2*, where surface ozone increases at mid- and high latitudes in the Northern Hemisphere and Southern Hemisphere, ~~see~~ (Fig. 6~~4~~a). These annual mean changes result from increases during the respective winter and spring seasons ~~(not shown), and are thus likely driven by greater~~when STE increases (~~increased~~ by ~38% on the annual mean). ~~Similarly, tropospheric ozone and HO_x changes due to greater lightning NO_x emissions contribute under *4xCO2* (Banerjee et al., 2014; Zeng~~

et al., 2008), but are not a factor in *G1*, see Table 2. In any case they do not define the sign of the global mean response.

We emphasize that the effect of SRM on tropospheric chemistry is expected to be strongly dependent on the scenario, reference state and geoengineering method used. Here, we assume pre-industrial conditions by following the G1 scenario, which only allows for low, natural background pollution. Under different forcing scenarios other aspects of tropospheric chemistry could change the surface ozone response. For example, air pollution by nitrogen oxides could change the relative importance of different chemical mechanisms could be more important for SRM under more polluted conditions (Morgenstern et al., 2013; Squire et al., 2014; Tang et al., 2011). Nevertheless, changes in humidity and photolysis as described here are robust modelling features that could occur under a range of geoengineering scenarios. These mechanisms will be key to tropospheric chemistry considerations under geoengineering in general. In addition, our experimental setup does not allow us to assess the full impact of solar geoengineering on the complex chemical mechanisms happening in the troposphere (section 2.1). Nevertheless,Consequently, our results demonstrate the potential for substantial changes in tropospheric chemistry and thus air quality in the different climatic state created by SRM. Here, we find a particularly strong effect in the tropics, where model surface ozone increases under *G1* and decreases under *4xCO2*, amounting to annual mean differences of around 5 ppbv between these two simulations in some regions, compare Fig. 64a and 64b. As with the surface ozone response under a range of RCP scenarios (which can differ in sign, (Connor et al., 2014; Young et al., 2013), there is clearly a need to study surface ozone changes for a range of geoengineering forcing scenarios.

4 Discussion and Conclusions

Using a coupled atmosphere-ocean chemistry-climate model, we have carried out an idealised SRM experiment in which we offset the effect of quadrupling atmospheric carbon dioxide on the global mean surface temperature by reducing the incoming solar radiation. Although the global mean surface temperature is, by design,

~~therefore~~ unchanged in this geoengineering experiment, other environmental factors change considerably. In particular, we find large ~~changes~~ increases in stratospheric ozone~~atmospheric composition~~, with an ~8% increase in global mean column ozone. Solar radiation management under *G1* fails to offset the cooling of the stratosphere resulting from increased CO_2, which leads to higher ozone concentrations there. The reduction in solar flux intensity in *G1* also plays a role in reducing ozone loss. In consequence, the stratospheric optical depth increases and leads to a reduction in tropospheric UV, with ~~R~~regional~~ly~~ and seasonal~~ly, those increases can be much larger and give rise to estimated~~ reductions of up to ~20% in local UV-indices. This ~~R~~reduced surface UV ~~in turn~~ could have adverse effects on medical conditions connected to vitamin D deficiency. In contrast, the general decrease in UV radiation is also expected to have beneficial effects such as a reduced likelihood in populations of developing skin cancer. We find that cloud-induced UV changes play a minor role compared with the change in ozone column.

A further unintended consequence of the SRM scheme considered here would be a change in tropospheric composition. The main drivers of change are decreases in tropospheric specific humidity as well as a reduced flux of UV-B and UV-A radiation into the troposphere. Relative to the pre-industrial control run, surface ozone increases in *G1* by about 5% (and decreases in *4xCO2*). Such an increase is qualitatively consistent with calculations, with detailed tropospheric chemistry schemes, of tropospheric ozone changes following an increase in stratospheric ozone (e.g. Banerjee et al., 2015). A major challenge in the 21st century will be to prevent large changes in tropospheric ozone, which would follow increased emissions of NO_x and volatile organic compounds. It is important that geoengineering schemes do not make this challenge more difficult. We note that the increase in ozone found here ~~is~~ could also lead to a change in the lifetime of the greenhouse gas methane in a geoengineered climate (Holmes et al., 2013; Morgenstern et al., 2013) and thus in the amount of solar geoengineering needed to offset the anthropogenic greenhouse gas forcing.

It is important to stress again that the modelled changes in atmospheric composition ~~and air quality~~ are strongly scenario- and SRM scheme-dependent. Important factors in other scenarios that would affect composition include the

reduction in ozone depleting substances by the Montreal Protocol, not considered here, or more detailed changes in tropospheric ozone precursors (Squire et al., 2015; Young et al., 2013) ~~For instance, f~~For stratospheric particle injection schemes, stratospheric ozone depletion would be a major concern (e.g. Pope et al., 2012), especially in the near future. In addition, UV considerations for aerosol schemes are further complicated by UV scattering and absorption by the aerosol particles (Tilmes et al., 2012) as well as aerosol indirect effects (Kuebbeler et al., 2012). ~~The relative importance of all of these factors would in turn be dependent on the geoengineering strategy, e.g. the injection methodology (Kravitz et al., 2012; Niemeier et al., 2011) as well as the amount and type of aerosol used (Ferraro et al., 2011; Pope et al., 2012; Tilmes et al., 2008).~~ Aerosol geoengineering might also affect the stratospheric circulation (Ferraro et al., 2015) with likely changes in STE different than found here for the *G1* experiment. Finally, it is also unclear how long-term injections of aerosols into the atmosphere would affect air quality at the surface due to potentially much increased particle pollution. ~~Other important factors that would affect composition include the reduction in ozone depleting substances by the Montreal Protocol, not considered here, or changes in tropospheric ozone precursors.~~

In conclusion, increases in CO_2 will increase the stratospheric ozone column and solar radiation management will not offset this increase. In the G1 experiment considered here, large increases in ~~that~~ stratospheric ozone are calculated leading to decreases in tropospheric UV. That surface UV and surface ozone ~~do~~ would change under solar geoengineering is a robust modelling result and~~;~~ their effects on human health and ecology could be considerable. ~~Similarly to the oft-cited problems of~~Just as with continued ocean acidification (Caldeira and Wickett, 2003) and changes in the hydrological cycle under SRM, ozone changes and their effect on surface UV and air quality would have to be expected in a solar geoengineered world. Consequently, we highlight this issue as an important factor to be accounted for in future discussions and evaluations of all SRM methods.

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Table 1 | Overview of the simulations.

Run	Carbon dioxide (ppmv)	Solar constant reduction (Wm^{-2})
piControl	285	-
4xCO2	1140	-
G1	1140	49.0

Table 2 | Global annual mean quantities. For *piControl* and corresponding differences under *4xCO2* and *G1* (highlighted in bold). The clear-sky, unpolluted UV index at noon is calculated using the formula by Madronich (2007), including only changes by column ozone and by the solar irradiance reduction. Standard deviations for the annual mean data are given in brackets, with the exception of the mean UVI indices, which were calculated from the climatological ozone fields without inter-annual variation.

	piControl	***4xCO2***	***G1***
Surface temperature (K)	**288.27** (0.13)	**+4.80** (0.16)	**-0.02** (0.14)
Precipitation (mm day^{-1})	**3.09** (0.01)	**+0.19** (0.01)	**-0.15** (0.01)
Surface ozone vmr (ppbv)	**12.0** (0.1)	**-0.5** (0.1)	**+0.6** (0.1)
~~LNO_x emissions (Tg N/yr)~~	~~**6.2**~~	~~**+3.6**~~	~~**-0.2**~~
STE O_3 (Tg/yr)	**456** (22)	**+172** (27)	**-7** (21)
Column ozone (DU)	**305.70** (1.2)	**+12.85** (1.7)	**+23.57** (1.6)
UV index	**7.93**	**-0.07**	**-0.79**

Figure 1 | Temporal evolution of the annual and global mean surface temperature anomalies. The anomalies (ºC) are shown relative to the average temperature of the pre-industrial experiment. The *piControl* and *G1* experiment are highlighted in the inset panel with the straight lines marking the average temperature anomalies. The grey and red shading give the ±2σ temperature interval for *piControl* and *G1* respectively.

Figure 2 | Annual mean surface temperature differences. The differences are based on the average temperatures of the last 50 years of each experiment. **a** *4xCO2* relative to preindustrial conditions. **b** *G1* relative to pre-industrial conditions. Note the non-linear colour scale. Non-significant changes (using a two-tailed Student's t-test at the 95% confidence level) are marked by stippling.

Figure 3 | Differences in zonal and annual mean ozone mass mixing ratio and temperature. a, **b** Percentage differences in ozone as labelled. **c**, **d** Temperature differences (K) as labelled. Note that **d** shows the difference between *G1* and 4xCO2, i.e. not the changes relative to piControl, in contrast to **a**-**c**, and that a different colour scale is used than in **c**. The ozone changes are given in percentages to highlight the in terms of absolute mass mixing ratios much smaller changes in the ozone-poor troposphere as compared to the larger absolute changes in the stratosphere, which in turn occur on much higher background ozone levels. The colour scale for ozone is adapted to changes in the middle-upper stratosphere; for the whole extent of the changes in the tropical upper troposphere and lower stratosphere under *4xCO2*, see Nowack et al. (2015). Differences are calculated on altitude levels, the pressure axis gives approximate values for pre-industrial conditions. Coloured lines in **a**, **b** mark the zonal and annual mean tropopause heights for each experiment. Non-significant differences (using a two-tailed Student's t-test at the 95% confidence level) are crossed out.

Figure 64 | Annual mean surface ozone changes. Absolute values (ppbv). Difference between **a** *4xCO2* and *piControl*, **b** *G1* and *piControl*. Non-significant changes (using a two-tailed Student's t-test at the 95% confidence level) are marked by stippling.

Figure 45 | Column ozone differences and their impact on the UV index. Relative to *piControl*: left for *4xCO2*, right for *G1*. Top row: annual mean ∆column ozone (colours, %). Non-significant changes (using a two-tailed Student's t-test at the 95% confidence level) are marked by stippling. Middle row: seasonal cycle of the column ozone changes as longitudinal and monthly means. Bottom row: seasonal cycle of the column ozone induced changes in the UV-index, and in **f** additionally by the solar constant reduction, at noon. Polar night regions in **e**, **f** are crossed out; both daily (solar declination) and monthly changes (ozone) are considered, giving rise to a less smooth appearance. Contour lines show pre-industrial column ozone in Dobson Units (DU) in the upper two rows and pre-industrial UV-indices in the last row.

Figure ~~6~~5 | Differences in the cloud modification factor and their impact on the UV index. a Annual mean ΔCMF (colours) under *4xCO2* and **b** under *G1* relative to *piControl* (contour lines). Non-significant changes (using a two-tailed Student's t-test at the 95% confidence level) are marked by stippling. Zonal mean percentage changes in the UV-index at noon induced by ΔCMF are shown for **c** *4xCO2* and **d** *G1* according to the formulas by den Outer et al. (2005) and Staiger et al. (2008). Polar night regions in **c**, **d** are crossed out; both daily (solar declination) and monthly changes (ozone) are considered, giving rise to a less smooth appearance.