

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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Received and published: 19 February 2016

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.

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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

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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

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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,

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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.

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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

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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 NO_x, HO_x 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

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NO_x, 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.

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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).

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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. Marengo 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 here. 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).

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

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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

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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.

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- 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.

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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 $O + O_2 + M \rightarrow O_3 + M$ 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

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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 NO_x abundances under 4xCO₂?

The increased upper stratospheric NO_x 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 NO_x 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 HO_x 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. HO_x, 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.

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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.

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