

Responses to referees for manuscript “*Linkages between ozone depleting substances, tropospheric oxidation and aerosols*” by Voulgarakis et al.

Response to Referee #1

We thank the 1st referee for the useful comments. Below we respond to the reviewer’s comments and describe the changes that we made, following this review.

MAJOR COMMENTS

The language is rather vague in many places, so the manuscript can be stronger with careful editing. Most importantly, more information is needed to describe the methods.

We believe that the modifications that we have now made based on the reviewer’s Specific Comments address this concern.

Some evaluation of the model aerosol distribution, and the stratospheric ozone columns should be included. This may be as simple as pointing to figures in other publications, but as of now, the paper refers to evaluation of different versions of the model so, without more information as to the differences in versions, it is unclear whether these apply to the version used here.

We agree with the reviewer that the Koch et al. (2006) evaluation is a bit too old a reference for the current version of the model. We now include a reference to a manuscript that is currently in press (Shindell et al., 2012b), which showed comparisons of the model’s aerosols to a variety of datasets (AeroNet, MODIS, MISR, OMI). The corresponding part of the text in Sect. 2.1 has now been modified to discuss these comparisons and how they inform our study. Regarding stratospheric ozone, we believe that the citation of Shindell et al. (2012a) and the related text is sufficient.

How does the model inter-annual variability in OH compare to that determined from methyl chloroform observations?

The simulations performed here (or in Shindell et al., 2012a) are not suitable for examining interannual variability, as they are equilibrium simulations with repeating conditions every year. Furthermore, this study focuses on long-term effects of specific perturbations, thus interannual variability is not relevant for the analysis.

The abstract and conclusions emphasize the coupled composition-climate model yet the GCM does not seem crucial here...

The reviewer is right that since our composition-climate model’s meteorology is not directly linked to the model’s radiation scheme, the changes in composition studied

here are not allowed to fully impact the simulated climate response. However, the benefit of using a composition climate-model as opposed to a CTM is that the radiative forcing of the perturbations applied can be diagnosed immediately, without the need of extra radiative transfer calculations. We now clarify this in the first paragraph of Sect. 2.1.

We believe that our paper does underline the importance of composition-climate interactions, as the radiative forcing calculated can be translated to a potentially sizeable climate response. We agree though with the reviewer that since the full climate response is not examined, the text should be slightly modified. We add a parenthesis in the Conclusions (last paragraph) to amend this.

It is mentioned at the end that allowing SSTs to respond should reinforce the changes modeled here, since N₂O and CFCs are greenhouse gases, but this point can be made earlier.

We agree and now make this point in the last paragraph of Sect. 2.

What is the lifetime of stratospheric ozone in the model? Is the two-year spinup sufficient to allow the stratospheric ozone columns to respond fully to changes in CFCs and N₂O?

The lifetime of modeled ozone in the stratosphere is of the order of months or less. We have checked that stratospheric ozone is fully stabilized following spin-up. We now mention this in the end of Sec. 2.

Are only sulfate aerosols included in the model? Are indirect effects considered? The details of the radiative forcing calculations should be described somewhere. Are the aerosol RF calculations online?

Other aerosols tracers and indirect effects that are simulated are now mentioned (with references) in Sect. 2.1. We now briefly describe how the radiative transfer calculations are done in the footnotes of Table 2.

Throughout the text, phrases like, “significant responses” should be backed up with numbers and/or explain how the significance was determined.

We maintain the use of the word “significant” in the abstract, as both the effects it refers to are indeed significant (see Table 2). The same is true in most of the other occurrences in the text. We replace “significant” with “important” in the first paragraph of Sect. 3.

P25553 L7-13. Clarify whether the increased STE and the photolysis changes act in the same direction.

Done.

P25555 L15-20. Unclear. How much more sensitive? What are liquid tracer budgets?

Quantitative global metrics on the higher sensitivity are not available, but we can say with good confidence that our handling is likely more realistic (as mentioned in the text). The “liquid tracer budget” means that gas phase and dissolved tracers are tracked separately, so that the gas-liquid partitioning is not lost from one point in time to the next (other models tend to have only one tracer per species). We now clarify this in the text.

P25555 L25-27 Give the global annual emissions (or range since they are varying with meteorology) for consistency with the other emissions reported.

We now give lightning NO_x emissions in the text.

P25556 Earlier work also shows the importance of overhead ozone columns for OH, e.g., Spivakovsky et al, JGR, 2000.

It is not clear which part of the paper this comment refers to. The Spivakovsky et al. paper has been cited in the Introduction, where we also list several of the papers that have discussed stratospheric ozone effects on tropospheric OH.

P25557-8 The text here focuses on the sulfate response at northern mid-latitudes, which is where SO₂ emissions are presently highest. Could this shift towards tropical locations in the future? Similarly, if NO_x declines here but increases in tropical regions, would this alter the conclusions?

We have now added a sentence in the discussion (just before the last paragraph of Sect. 3) that mentions this interesting possibility.

The discussion could be more balanced between the WA scenario and the RCPs and the text should note that the WA induces responses ~10x larger (Table 2).

We now mention in the text (discussion of Table 2) that changes in the WACFCs scenario are roughly an order of magnitude larger. We believe that the more extensive discussion of the WACFCs scenario is fair, since the signals are much stronger.

Please provide the % global mean sulfate aerosol production via the gas-phase in order to compare the 11% given for aqueous-phase production.

We now provide this figure (38%) in the text.

Please clarify why Fig 3 is needed; since OH changes in these simulations, it follows that the changes in sulfate distribution follow the changes in sulfate production by OH.

We believe that this figure is a key one, as it is a very nice illustration of how well the chemical production changes are correlated with the actual sulfate changes. Since a study of this kind has not been pursued in the past, as both reviewers recognize, we think it is useful to underline the processes involved by using such figures.

What is the sulfate lifetime in the model?

The sulfate lifetime in the model is 3 days. We now mention this in Sect. 2.1.

P25558 L16. Are these values occurring over similar areas?

Yes, there is a fairly good agreement. We now mention this in the sentence.

L20-23. Another useful comparison is for 2050 or 2100 RCPs where the CFC and N2O effects offset each other (Table 2), noting that these are probably small relative to the large decreases in SO2 emissions under the RCP8.5 scenario.

We now make this interesting comment in the text.

L25-26. Could document the relative balance for this model here.

We now document this at the end of the corresponding paragraph.

P25559 L8-9. Because of accounting for indirect effects or because future projected changes are larger than what occurred historically?

We now add “due to the larger increases in 2000-2100 compared to 1850-2000”.

L13 Clarify, opposite in sign.

We believe this is explained clearly in the subsequent sentence.

L19-20. Unclear, is this transport of NOx from the stratosphere to the troposphere?

We modify to “modifications of transport from higher altitudes” to clarify.

L20-23. Were the minimal changes to climate by design, not allowing SSTs to respond?

Yes, and we now clarify this in the text.

L28. Future emissions of what?

Of gases that affect oxidation chemistry. We now clarify this in the text.

P255560. L2-6. Recent scenarios (RCPs) suggest large decreases in NOx.

We already discuss the possibility of more optimistic scenarios in the following sentence. We now specify that the RCPs are among those optimistic scenarios. Arguably, the RCPs are overly optimistic, but we agree that this is worth mentioning as they form an important part of the current scientific discussion on composition-climate interactions.

15-16. This cancellation seems important and should probably be emphasized in the conclusions.

We agree and now mention this in the first paragraph of the conclusions.

L19-20. Can you estimate how much stronger?

We now provide a rough estimate in the text.

P25561. L4 Why not also during the 21st century?

We mention the 22nd century since the concentrations may grow even more important if the trend persists.

L9-10 Unclear, try rephrasing.

We have now rephrased to “We suggest that the Montreal Protocol may have been a mediating factor for sulfate aerosol concentrations in the 21th century.”

Table 1 would be more useful as percentage changes in CFCs and N2O from year 2000 since the naming gives the year. The last 3 columns are unnecessary.

We agree with the reviewer and make the changes suggested.

Table 2. Why does the sign of the sulfate response for GL and NM differ in the 1960 CFCs case?

The differences are small and insignificant, so we feel that it is not worth commenting on this in the text. This is possibly a result of compensation of noisy features.

Why does the sign change for GL CH4RF and GLsulfate RF for 1850 N2O whereas the sign is consistent for these 2 variables across all the other simulations?

This is indeed an unexpected feature. It occurs because even though OH is lower than BASE in the 1850N2O simulation, sulfate is higher. We will mention potential reasons in the revised version of the manuscript.

More information is needed to understand how the methane change was inferred so that this can be compared with other models, e.g., give the sensitivity or feedback factor.

We now provide further information in the Table's footnotes.

Figure 1. Why are the patterns of change different for b) and c) where the maximum change occurs in the southern hemisphere versus d) where the maximum change is in the northern hemisphere?

This is due to the NO_x effects which have already been mentioned in the text (see comment on P25559 L19-20). Due to the increased tropospheric ozone production, we have compensation between stratospheric ozone and tropospheric ozone changes in the Southern Hemisphere column, and thus a smaller impact on photolysis at low altitudes. We now comment on this in a parenthesis: "(also see Fig. 1d for effect of this feature on photolysis)".

Figure 2. Showing the global map could be useful for understanding the CH₄ response.

The reason why we show the northern midlatitude maps is because we prefer to concentrate on sulfate responses (the central focus of this study), which are important mainly in these areas. High relative differences in other regions that would be associated with low sulfate concentrations would distract the reader from the main messages.

Figure 4. Panel a) looks noisy. Are these changes statistically significant with only 7 years of simulation and the 10 times smaller responses as compared to panel b)? Why not show global maps?

We now test statistical significance for panel (a) and comment in the text. The reason why we concentrate on northern midlatitude maps is explained above.

Response to Referee #2

We thank the 2nd referee for the useful comments. Below we respond to the reviewer's comments and describe the changes that we made, following this review.

MAJOR COMMENTS

1) An evaluation of the aerosol distribution in the newest model version (Shindell et al., 2012) has evidently not been published. Have the authors evaluated the aerosols in this new model version? Is the aerosol evaluation similar to that published by Koch et al (2006) and Shindell et al. (2006). The authors should show in a supplement or otherwise convince the reader that their aerosol distribution in the new model formulation is reasonable. It is not clear the extent to which the evaluations by Koch and Shindell published in 2006 apply to the newest version of the model.

As we also mentioned in response to Referee #1's comments, we agree with the reviewer that the Koch et al. (2006) evaluation is a bit too old a reference for the current version of the model. We now include a reference to a manuscript that is currently in press (Shindell et al., 2012b), which showed comparisons of the model's aerosols to a variety of datasets (AeroNet, MODIS, MISR, OMI). The corresponding part of the text in Sect. 2.1 has now been modified to discuss these comparisons and how they inform our study.

MINOR COMMENTS

1) p2555,114: *Please give tropospheric methane lifetime.*

We now give the lifetime to the text.

2) p2555,125: *Please give Lightning NO_x emissions averaged over present day.*

We now give lightning NO_x emissions in the text.

3) p2555,126: *Please give isoprene emissions averaged over the present day.*

We now provide isoprene emissions in the text.

4) p2555,118: *The soil NO_x emissions seem very low, at least compared with the Jaegle estimates. Please provide a reference for the 2.7 TgNyr⁻¹.*

Emissions from soils are as in Shindell et al. (2012a). See reference in the text.

5) p2555,19: *Please state at the onset of this paragraph that the simulations assume present day climate.*

We now specify this in the onset of the paragraph.

6) P2555,118: *I believe the additional experiment discussed here included the WACFC scenario for ODS. Please make this explicit when initially describing the experiment.*

No, actually ODSs were kept to present-day levels in this simulation. We now modify the sentence to reflect this.

7) P2555, 13: *"and to more". Please clarify.*

We now specify the exact percentage.

8) P2555,122: *"climate changes due to CFCs". Maybe better to say circulation changes rather than climate changes?*

This sentence does not refer to circulation changes, so we would prefer avoid this term.

9) P25559: *“changing future emissions and climate change: :” This is a rather complex subject involving changes in stratospheric ozone, changes in STE and changes in tropospheric chemistry. Some of these other changes are discussed in the following paragraph. Nevertheless, the statement that changes in future emissions and climate augment the changes in ODS could be made more coherently.*

We now apply various modifications to this discussion to make it more solid. See also responses to Referee #1.

REFERENCES

Shindell, D. T., Lamarque, J.-F., Schulz, M., Flanner, M., Jiao, C., Chin, M., Young, P., Lee, Y. H., Rotstayn, L., Milly, G., Faluvegi, G., Balkanski, Y., Collins, W. J., Conley, A. J., Dalsøren, S., Easter, R., Ghan, S., Horowitz, L., Liu, X., Myhre, G., Nagashima, T., Naik, V., Rumbold, S., Skeie, R., Sudo, K., Szopa, S., Takemura, T., Voulgarakis, A., and Yoon, J.-H.: Radiative forcing in the ACCMIP historical and future climate simulations, *Atmos. Chem. Phys. Discuss.*, 12, 21105–21210, doi:10.5194/acpd-12-21105-2012, 2012b.