

Interactive comment on “Linkages between ozone depleting substances, tropospheric oxidation and aerosols” by A. Voulgarakis et al.

Anonymous Referee #1

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The manuscript describes the response of tropospheric hydroxyl radicals, and the resulting radiative forcings associated with changes in methane and sulfate aerosol, to changes in stratospheric ozone columns driven by changing concentrations of CFCs or N₂O, both substances which deplete stratospheric ozone. Changes in stratospheric ozone columns primarily affect tropospheric OH in the model by changing photolysis rates (JO1D). This manuscript is the first to attempt to link all of these processes and quantify the sulfate aerosol response to changes in ODS.

After substantial revisions, the manuscript should be suitable for publication. 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. Some evaluation of the model aerosol distribution, and the stratospheric ozone columns

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

The abstract and conclusions emphasize the coupled composition-climate model yet the GCM does not seem crucial here since the SSTs/sea ice are fixed at year 2000 conditions and so the climate response is not fully included. Rather, it seems the essential pieces are the fully coupled stratosphere and troposphere chemistry, which could be done in a chemical transport model with fixed meteorology and thus avoid the problem of the inter-annual variability generated by the climate model. Please include some discussion as to why the chosen study design is most appropriate. 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.

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?

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?

Specific comments.

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

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

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

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

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

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? 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). Please provide the % global mean sulfate aerosol production via the gas-phase in order to compare the 11% given for aqueous-phase production. 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. What is the sulfate lifetime in the model?

P25558 L16. are these values occurring over similar areas? 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. L25-26. Could document the relative balance for this model here.

P25559 L8-9. Because of accounting for indirect effects or because future projected changes are larger than what occurred historically? L13 Clarify, opposite in sign. L19-20. Unclear, is this transport of NO_x from the stratosphere to the troposphere? L20-23. Were the minimal changes to climate by design, not allowing SSTs to respond? L28. Future emissions of what?

P255560. L2-6. Recent scenarios (RCPs) suggest large decreases in NO_x. L15-

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16. This cancellation seems important and should probably be emphasized in the conclusions. L19-20. Can you estimate how much stronger?

P25561. L4 Why not also during the 21st century? L9-10 Unclear, try rephrasing. 11-12 Clarify what aspects of the climate response were included here.

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.

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

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?

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

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?

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