

We thank both reviewers for their very constructive and helpful comments. We have carried out some additional simulations and data analysis and also revised the MS following the reviewers' suggestions. Our responses to the reviewers' comments are provided below, with the reviewer's comments italicized and our responses in regular fonts.

Referee #1

Major comments: "Further isolating the impact of thicker stratospheric ozone columns on the tropospheric ozone lifetime from the impacts via strat-to-trop exchange is interesting and worth pursuing. However, more work is needed to bring the paper to a level appropriate for ACP. Most notably, the interpretation of results is quite thin, e.g., with four figures discussed in one sentence in Section 3.3. The figures are repetitive and careful thought should be given to what is needed to communicate the key findings."

Response:

Point well taken. We have carried out further analysis and updated the MS.

1. The methodology needs clarification. Given the stratospheric residence time, one year is not sufficient for spinning up if the stratospheric ozone distribution depends on transporting the ozone produced by LINOZ. It is stated that stratosphere-troposphere exchange is held constant but it is not explained how this is accomplished when the stratospheric ozone distribution is changing. Also, this information belongs in Section 2 rather than in the final paragraph of the conclusions.

Response:

The stratospheric ozone recovery can affect tropospheric composition and chemistry through two different channels: (a) changing STE; (b) changing UV radiation and therefore photolysis rates. Since there have been multiple literature studies on (a), in this study we intend to focus on the latter effect and isolate it from the former one. In the GEOS-Chem model, the STE and photolysis rates are calculated separately. Therefore we keep the STE unchanged from control run to the sensitivity run but only changes the ozone column when calculating radiation and photolysis rates. We have updated the title of the MS to "Effects of stratospheric ozone recovery on photochemistry in the troposphere".

We have also added clarification in Section 2 –

"To isolate the potential impact of stratospheric ozone recovery on tropospheric chemistry through changes in UV radiation and therefore photolysis rates from other effects such as the changes in stratospheric-tropospheric-exchange (STE) associated with transport (Zeng et al., 2010), we only apply the perturbations to stratospheric ozone column in calculating the actinic flux and photolysis rates of various species. The STE of ozone remains unchanged from the control run to the sensitivity run."

For the 1 year model spin-up, that was only for atmospheric composition in the troposphere, we have added clarification in the text –

“For both simulations, a whole year spin-up for the tropospheric chemistry and composition using 2005 meteorology was done ...”

2. The model should be evaluated, at minimum to show that the present-day stratospheric ozone distribution simulated with LINOZ is sufficiently accurate to diagnose the impacts from the relatively small changes found here, especially in the tropics where the changes are a few percent at most according to Table 1, and yet this is where we'd expect to have the largest impact on tropospheric OH.

Response:

As discussed above, we purposely keep STE the same in the control run and the sensitivity run; this way the differences in results from the control run to sensitivity run only reflect the changes in actinic flux and photolysis.

3. The authors should place their results in the broader context of findings in several recent papers addressing topics related to findings here.

Lang, C., D. W. Waugh, M. A. Olsen, A. R. Douglass, Q. Liang, J. E. Nielsen, L. D. Oman, S. Pawson, and R. S. Stolarski (2012), *The impact of greenhouse gases on past changes in tropospheric ozone*, *J. Geophys. Res.*

Hegglin, M. I., and Shepherd, T. G.: *Large climate-induced changes in ultraviolet index and stratosphere-to-troposphere ozone flux*, *Nature Geosci*, 2, 687-691, http://www.nature.com/ngeo/journal/v2/n10/supinfo/ngeo604_S1.html, 2009.

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P. J., Prather, M. J., Wild, O., Field, R. D., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Folberth, G. A., Horowitz, L. W., Josse, B., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: *Analysis of present day and future OH and methane lifetime in the ACCMIP simulations*, *Atmos. Chem. Phys.*, 13, 2563-2587, doi:10.5194/acp-13-2563-2013, 2013.

Response:

We have added in the MS discussion and comparison to all the papers suggested by the reviewer (in section 3).

4. The sensitivity of OH to changes in stratospheric ozone columns could be compared with e.g. Table 6 of Spivakovsky, C. M., et al. (2000), Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, J. Geophys. Res., 105(D7), 8931–8980, doi:10.1029/1999JD901006.

Response:

We have added in the MS discussion and comparison to the above paper (in section 3.2).

5.Impacts on air quality are emphasized but the largest changes in surface ozone appear to be over the oceans, so is this really important? An interesting point is raised with respect to impacts on intercontinental transport but is not explored. This could be expanded upon and placed in the context of a recent paper looking at climate change impacts on intercontinental pollution: Doherty, R. M., et al. (2013), Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study, J. Geophys. Res. Atmos., 118, 3744–3763, doi:10.1002/jgrd.50266.

Response:

We have conducted further model sensitivity simulations to examine the impacts of stratospheric ozone recovery on intercontinental transport of ozone.

We have added in the MS –

“Sensitivity model simulations are carried out to examine the impacts of stratospheric ozone recovery on intercontinental transport of ozone pollution from Asia to North America. Following (Wu et al., 2009), we shut off emissions of ozone precursors from Asia (the region of 63°E-150°E and 10°N-58°N) in the model. Four groups of simulations were performed for year 2006-2008 and the results refer to the 3-year average: (1) Control run; (2) Control run with Asian emission off; (3) Sensitivity run with stratospheric ozone recovery perturbations; (4) Sensitivity run with stratospheric ozone recovery perturbations and Asian Emission off.

Results from these 4 groups of simulations are compared with each other: [(1) - (2)] represents the intercontinental transport of ozone associated with Asian emissions and [(3) - (4)] the intercontinental transport of ozone associated with Asian emission under stratospheric ozone recovery scenario, while [(3) - (4)] - [(1) - (2)] indicates the potential impacts of stratospheric ozone recovery on intercontinental transport of ozone (Figure A1 and Figure A2).

The impacts of stratospheric ozone recovery on continental outflow of ozone pollution from Asia show a strong seasonal variation: weakest in the winter time but peaks in summer when there is abundant UV radiation. As we can see from Figure A2, the stratospheric ozone recovery could enhance the global background ozone attributed to Asian emissions by up to 20% in the Northern hemisphere.”

We have also added reference to (Doherty et al., 2013).

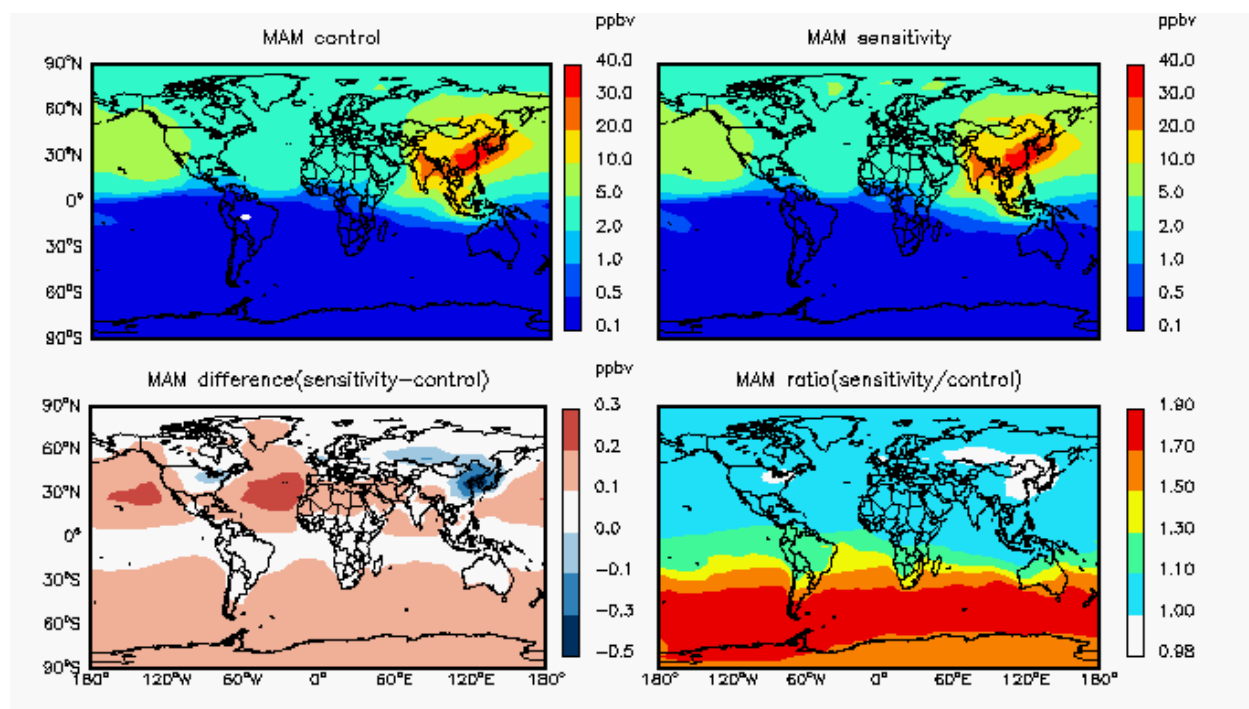


Figure A1. MAM intercontinental transport of ozone

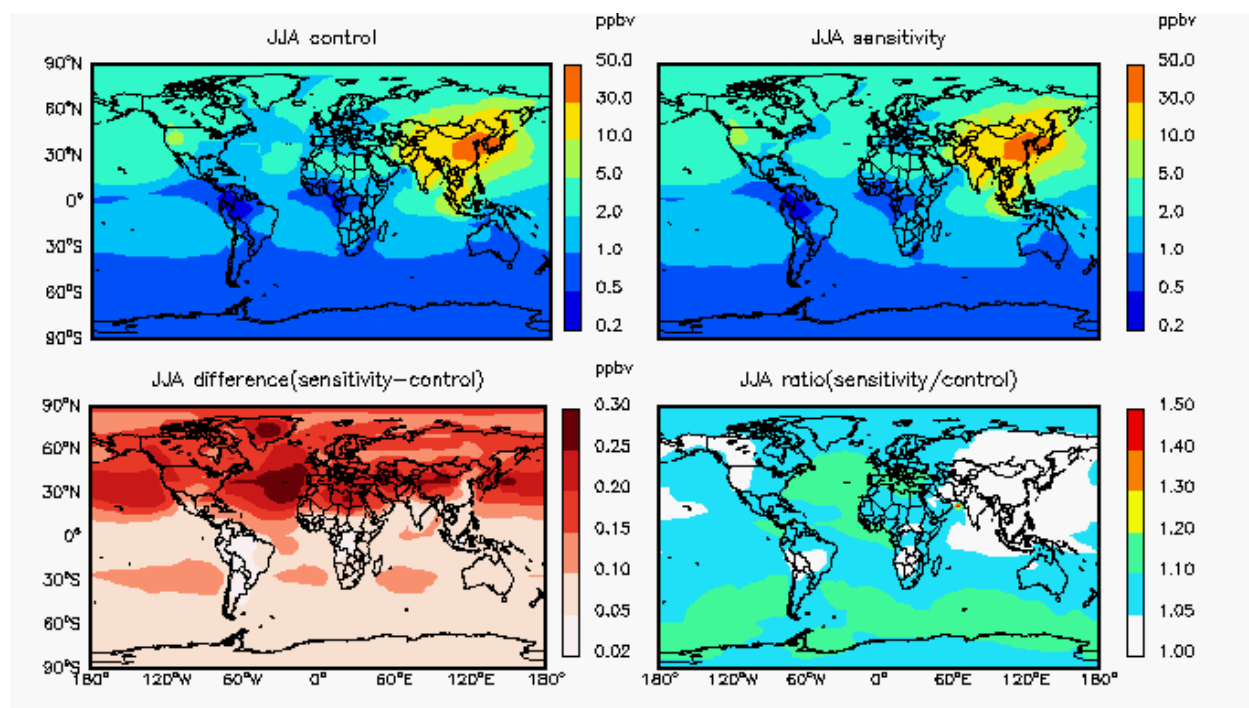


Figure A2. JJA intercontinental transport of ozone

Referee #2

1. Climate change would be an important factor for stratospheric ozone recovery. If the stratospheric ozone recovered, there are definitely corresponding changes in environmental (climate and emission) conditions. Therefore, without considering it, the results would be less persuasive and contain more uncertainties. For example, (1) Climate change would sensitively affect the cloud coverage, which further affect the radiation transfer. (2) Climate change resulted temperature profile would affect the stratosphere and troposphere ozone chemistry. etc. A conclusion based on certain climate change scenarios or estimated uncertainty in climate change would show more meaningful results.

Response:

Point well taken. Indeed, climate change is expected to affect atmospheric composition in both the stratosphere and troposphere. However, in this study, we only focus on the impacts of stratospheric ozone recovery on troposphere chemistry through changes in radiation and photochemical reactions due to two primary reasons/factors:

- (a) Limitation in model capability – the CTM we used is not capable of simulating the impacts of climate change on strat. ozone recovery. From other sources, like the WMO report, we were only able to obtain data as compiled in Table 1 reflecting an assumed “full” recovery of the strat. ozone.
- (b) To our knowledge, there hasn’t been any study on the potential impacts of strat. ozone recovery on photolysis and long range transport of troposphere ozone with a global 3D model. In contrast, there have been numerous literature studies on the other factors affecting tropospheric chemistry in the context of global change, like the impacts of climate change on tropospheric chemistry. Therefore, we focus our study on the sensitivity of tropospheric photochemistry to strat. ozone recovery.

We have updated the title of our MS to “Effects of stratospheric ozone recovery on photochemistry in the troposphere” to better reflect the major point of this study.

We have also added in the MS discussion/comparison to other factors/drivers of atmospheric composition in the context of global change.

2. Radiation distribution change resulted change in water vapor profile (so is the climate change) directly affect the concentrations of OH in the atmosphere. How does it affect the result?

Response:

Yes, theoretically the changes in radiation associated with the strato. ozone recovery would affect the energy budget of the earth system and hence water vapor in the atmosphere. However, we think these changes (in the energy budget and water vapor) would be negligibly small (compared to the changes in ozone photolysis rate) since the ozone column change only

affects UV radiation (< 310 nm) but not the visible lights (~400-700 nm) which dominate the incoming solar radiation flux (energy input) to the earth system.

3. In page 21430, more clear expression on the method is needed. In the sensitivity run, the stratospheric ozone is assumed to recover to 1980 levels. Is it a modification to initial value ? or hold it as a constant field? If using the Linoz stratospheric ozone package, how the current emissions and environment (climate) adjust the 1980 stratospheric ozone field with time in simulations?

Response:

In our model, the stratosphere-troposphere-exchange (STE) and calculations of photolysis rates in the troposphere are treated in two separate processes. Since our focus in this study is to look at the perturbations in photochemistry, we keep the STE (as simulated by Linoz) in the sensitivity run the same as that in the control run but only adjusted the column ozone used for calculating photolysis rates following the scaling factors in Table 1.

We have added clarification in the MS –

“We carry out two groups of simulations to derive the sensitivity of troposphere photochemistry to stratospheric ozone recovery: one as the control run and the other as sensitivity run where the stratospheric ozone is assumed to fully recover to its pre-1980 levels. We follow the WMO report (Fioletov, 2006;Randel, 2003) for the stratospheric ozone depletion in the past decades, which varies significantly with season and latitudes. The expected increases in stratospheric ozone column associated with the ozone recovery in the coming decades are shown in Table 1.

To isolate the potential impact of stratospheric ozone recovery on tropospheric chemistry through changes in UV radiation and therefore photolysis rates from other effects such as the changes in stratospheric-tropospheric-exchange (STE) associated with transport (Zeng et al., 2010), we only apply the perturbations to stratospheric ozone column in calculating the actinic flux and photolysis rates of various species. The STE of ozone remains unchanged from the control run to the sensitivity run.”

4. Surface ozone is sensitive to changes in VOC concentrations too. How the radiation change affect the photosynthesis rate and further affect the biogenic VOC emissions? Does it will affect the conclusion that the largest changes in surface ozone appear to be over the oceans?

Response:

This is an interesting point. We did a little bit research and found that the photosynthetically active radiation (PAR) spectrum (~400-700 nm) corresponds more or less with the visible lights (http://en.wikipedia.org/wiki/Photosynthetically_active_radiation). As discussed above, the ozone column change would only affect UV radiation but not the PAR radiation. Therefore, we do not expect any significant changes in the photosynthesis rate and biogenic VOC emissions.

5. A comprehensive study on the ozone transport from stratosphere to troposphere is valuable to enhance this study. Previous studies [Barré et al., 2012; Hsu et al., 2005] have shown that this stratosphere-troposphere ozone exchange is frequent, which would be significant for the tropospheric ozone change after the stratosphere ozone recovery. A simple method [e.g. Hsu et al., 2005] would be enough to examine this issue.

Response:

Yes, the stratospheric ozone recovery can affect tropospheric composition and chemistry through two different channels: (a) changing STE; (b) changing UV radiation and therefore photolysis rates. As the reviewer pointed out, there have been multiple literature studies on the former effect but not on the latter, therefore we focus on the latter effect in this study. Another reason is that, the model used in our study was mainly designed/developed for tropospheric chemistry, so we feel it may not be the best tool to investigate the changes in STE. But we have added discussion and comparison to those literature studies on the former effect in the MS.