Response to Reviewer #2

First of all, we would like to thank the reviewer for their comments.

General

This paper analyses several scenarios of greenhouse gas development in the 21st century, in particular increases in nitrous oxide and methane, and its impact on ozone. This is an interesting and important topic which is well appropriate for ACP. The paper is well presented and Figures are of good quality.

I have some points of critique as well. I suggest more discussion of changes in the Brewer Dobson circulation, of chemistry driven by stratospheric temperature change (e.g., $N_2O + O(^1D)$), and of the stratospheric chlorine loading and its impact on the predicted behaviour of ozone. The decreasing stratospheric chlorine loading is certainly the dominant effect on stratospheric ozone over the time period considered.

While it is true that changes in chlorine loading, the Brewer-Dobson circulation and stratospheric temperature have a large impact on ozone through the 21st century, this is not the focus of the paper that we have written. The paper we have written focusses exclusively on the effects of changes in CH_4 and N_2O on ozone. That specific focus is ensured by varying only the CH_4 or N_2O concentration boundary conditions between the simulations. Because the CO_2 concentration boundary condition is identical in every simulation, and because CO_2 provides the main radiative forcing of the climate system, including the Brewer-Dobson circulation, we do not expect to see differences in the strength the Brewer-Dobson circulation between our simulations. Our simulations therefore cannot address any aspects of the effects of changes in the Brewer-Dobson circulation on ozone. That's not what they were designed for. The same holds true for temperature and chlorine loading on the simulated ozone differences. However, we have incorporated these factors into our discussion.

Furthermore, I suggest more discussion about the observed latitudinal structure of the response of stratospheric ozone on N_2O and methane changes.

We have followed this suggestion and have discussed in greater detail the latitudinal structure of the response of stratospheric ozone to changes in N_2O and CH_4.

And I am not convinced that CO oxidation is the reason for enhanced ozone production and an enhanced N_2O scenario (Fig. 4).

We have provided references that support the fact that CO oxidation leads to enhanced ozone in the troposphere and lower stratosphere and have provided the detailed chemistry that shows how this happens. We have also included other possible reasons for enhanced ozone production.

In summary, I suggest several revisions to the paper (see also details below) but expect to find a revised version acceptable for publication in ACP.

Specific Comments

The NO_x levels in the stratosphere are determined by the reaction N_2O with $O(^1D)$, which is temperature dependent. Thus, an increase in N_2O in the future does not necessarily lead to an increase in NO_x. Further, changes in greenhouse gases lead to changes in the Brewer-Dobson circulation which likely have an impact the relation between N_2O (and methane) increases in the troposphere and stratospheric ozone loss cycles. I suggest more discussion of these issues in the paper. In part, the issues mentioned above have been discussed in a recent publication by the authors (in Geophys. Res. Lett.) so that it might be helpful to refer to this paper in the discussion. This could also help clarifying in what respect the results in the present paper reach beyond those in the earlier paper (I do not dispute that they do reach beyond).

We have extended our discussion on the role of stratospheric NO_x changes in driving changes in ozone as suggested.

Furthermore, increase in stratospheric water vapour has an impact also on polar ozone (e.g., Kirk-Davidoff et al., 1999; Feck et al., 2008; Vogel et al., 2011) and possibly even on mid-latitude ozone (e.g., Ravishankara, 2012) via a coupling with chlorine chemistry. This aspect is not treated in the paper. Possibly, such effects are less an issue for the present study as it aims at a situation when chlorine levels will have significantly decreased compared to the maximum values. Nonetheless, this issue should be discussed in the paper.
Contrary to the results of these papers, we find that ozone in the polar regions is enhanced where water vapour is enhanced, so it would seem that chlorine chemistry does not play a significant role in ozone chemistry by the 2090s decade (which is the focus of our analysis). We have noted this in the manuscript.

In particular as Figs. 4 and 5 demonstrate that (apart from the tropical lower stratosphere) the greatest effect of N_2O and methane increases occur in the tropics in the SOCOL simulations discussed here. What is the reason for this model behaviour? Can any impact of chlorine chemistry on this model behaviour be excluded?
Figures 4 and 5 show that the greatest effect of N_2O and CH_4 increase is in polar regions; we have discussed potential reasons for this in the text.

Moreover, chemical ozone production via R2-R5 (and driven by NOx increase) is put forward as a reason for the positive ozone signal in the tropics visible e.g. in Fig. 4. Note that the positive ozone signal reaches up to altitudes of about 30 hPa. Are there references which could support this idea? Could not tropical ozone might also change through a change in tropical up-welling (e.g., Randel et al., 2007; Ploeger et al., 2011), which might well occur between the scenarios compared in Fig. 4? The paper could be clearer here to preset the ozone production via R2-R5 either a possible explanation or to back up the conclusion with information (e.g., ozone production rates) from the model simulations.
Tropical upwelling is the same between all simulations because the CO_2 and SST scenarios are the same. Therefore, tropical upwelling cannot be responsible for the positive ozone signal. We have provided references that support the fact that CO oxidation leads to enhanced ozone in the troposphere and lower stratosphere and have provided the detailed chemistry that shows how this happens. We have also discussed the idea that increased oxygen photolysis due to reduced overhead ozone abundances could drive the increase in ozone production.

Finally, global averages of ozone loss rates are shown in Fig. 3. However, ozone loss rates have a different meaning in a photochemically controlled regime, where loss rates and chemical production are in close balance (upper stratosphere and tropics) and in a dynamically controlled regime (e.g., polar spring and lower stratosphere). So I am not sure how to interpret a global mean of this quantity. My suggestion is to discuss and show only regional averages of this quantity not global averages. Investigating the ozone loss rates in regions might also help explaining why ozone reacts differently in different latitudes on N_2O and methane increases in the simulations (Figs. 4 and 5).
As suggested by the reviewer, we have replaced Fig. 3 with pressure-latitude plots of the change in ozone loss rates, and have modified our discussion accordingly.

Comments in Detail
• page 17584, l. 9: quantify 'decrease'
We have quantified the ozone change for both the N_2O and CH_4 sets of simulations.

• l. 11: state why the overall effect is an increase in total ozone

We have done this.

• l. 24: The radiative impact of methane and N_2O should be discussed. Could they contribute to stratospheric cooling in the model results?
Fleming *et al.* (2011) (ACP, "A model study of the impact of source gas changes on the stratosphere for 1850-2100"), shows that N_2O has a negligible impact on stratospheric temperature. CH_4 has a much smaller impact in the stratosphere than CO_2, and leads to net cooling. This is via increased water vapour, which we do discuss in the paper.

• p. 17585, l. 17: add 'and adjustments'
We have done so.

• p. 17586, l. 1,2: this statement is not true for polar ozone depletion. Although R1 is important for Antarctica, the concentration of methane is not relevant.
We have added a sentence after this reaction (now Reaction (R3)) to address this concern raised by the reviewer.

• p. 17589, l. 17: What is the evidence from the model results for this conclusion? As it stands, this statement is too speculative.
We have included a reference to the appropriate figure.

• l. 28,29: again, what is the evidence from the model results for this attribution?
We have referenced the appropriate figures in our discussion.

• p. 17590, l. 4: why is the strongest effect in the Arctic?
We have extended our discussion to include this.

• l 14: 'significantly different' from zero . . .
We have made the correction.

• l. 19./20: why is the sensitivity enhanced in the polar regions?
We have extended our discussion here.

• What is the evidence from the model results that enhanced N_2O leads to ozone production?
We have no direct evidence from the model since the necessary model output fields to quantitatively make this connection were not saved. However, other studies have found similar effects and we refer explicitly to those studies.

• p. 17591, l. 10.,11.: This increase in ozone is driven mainly by the decrease of the stratospheric chlorine levels. This should be clarified.
We have done so.

• l. 21: state the result of the 'vertically-resolved relationship' not just the fact that it was 'shown'.
We have done so.

• p. 17592: Ref. Brasseur and Solomon: publisher is missing.
Thank you for bringing this error to our attention. We have made the necessary correction.

• Fig. 5: it is obvious from this figure that the strongest impact of methane increase is at about 20–50 hPa at the poles. This result seems important to me and I suggest that it should be discussed in more detail.

We agree and have included a discussion of this in the manuscript.

• Figs. 6, 7, 8, and 9: These fits and the estimated uncertainty are based on only four points. I think this needs to be taken into account in the interpretation of these results.
We have noted this point in the manuscript.

References

Feck, T., Grooß, J.-U., and Riese, M.: Sensitivity of Arctic ozone loss to stratospheric H_2O, Geophys. Res. Lett., 35, doi:10.1029/2007GL031334, 2008.
Kirk-Davidoff, D. B., Hintsa, E. J., Anderson, J. G., and Keith, D. W.: The effect of climate change on ozone depletion through changes in stratospheric water vapour, Nature, 402, 399–401, doi:10.1038/46521, 1999.
Ploeger, F., Fueglistaler, S., Grooß, J.-U., Günther, G., Konopka, P., Liu, Y. S., Müller, R., Ravegnani, F., Schiller, C., Ulanovski, A., and Riese, M.: Insight from ozone and water vapour on transport in the tropical tropopause layer (TTL), Atmos. Chem. Phys., pp. 407– 419, doi:10.5194/acp-11-407-2011, 2011.
Randel, W. J., Park, M., Wu, F., and Livesey, N.: A large annual cycle in ozone above the tropical tropopause linked to the Brewer-Dobson circulation, J. Atmos. Sci., 64, 4479–4488, 2007.
Ravishankara, A. R.: Water Vapor in the Lower Stratosphere, Science, 337, 809–810, doi: 10.1126/science.1227004, 2012.
Vogel, B., Feck, T., and Grooß, J.-U.: Impact of stratospheric water vapor enhancements caused by CH4 and H2 increase on polar ozone loss, J. Geophys. Res., 116, D05 301, doi: 10.1029/2010JD014234, 2011.