Anonymous Referee #1
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This is a useful paper that presents the changes in tropospheric ozone and its precursors through the 21st century for one widely-used emissions/concentration scenario. It clearly demonstrates in a quantitative manner that changes in ozone precursor emissions dominate these changes, and that increased methane concentrations make a substantial contribution to this. It also highlights that climate changes under this scenario have little net impact on mid-tropospheric ozone, but that this masks increased ozone influx from the stratosphere and from lightning along with increased ozone destruction from humidity/temperature changes. These competing factors are not quantified, which is disappointing, and this constitutes one of my main concerns the manuscript, as outlined below. However, the paper is worthy of publication once the points below have been addressed suitably.

General Comments:
The model used has a number of weaknesses, and the authors appear to avoid discussion of surface ozone or its changes wherever they can. On the positive side, the authors have justified their choices and have demonstrated that although there are some large biases in the mid troposphere compared with current observations, these are generally self-consistent and can be rationalized if not fully explained. However, the biases may be heavily influenced by what is happening at the surface, and so it would be very helpful for the reader if a few sentences were included in the evaluation section (3.1) on model performance at the surface.
First of all, we would like to thank Reviewer 1 for their helpful comments on both the initially submitted and ACPD versions of this manuscript. As suggested here, we have added some discussion on surface ozone to the evaluation section in Section 3.1:

Surface ozone in SOCOL is biased on a similar order of magnitude in the Northern Hemisphere compared with the mid-troposphere, with ozone over Europe, the US and Asia up to 20 ppb higher in 2000 compared with the ACCMIP ensemble mean (Young et al., 2013).

The least satisfying part of the paper is the final paragraph before the conclusions, which highlights the potential impact on ozone of changes in lightning, but is unable to quantify this. While the paper demonstrates that this term together with the change in stratospheric influx (which is quantified) balance the increased destruction from humidity and temperature (which is not quantified), it is not clear if the two unknown terms are negligible (sub-ppb level) or substantial (4-5 ppb or more, but counteracting each other). Some attempt to establish this, or at least to speculate in a semi-quantitative manner, would add substantial value to the paper. A short model run with scaled lightning NOx emissions would be sufficient to estimate this, and hence quantify both terms. What have other studies found?
Indeed, since increased ozone destruction from humidity and temperature, and increased ozone production from lightning NO_x emissions are both linked to climate change, we cannot separate the two terms in our simulations. However previous studies with scaled lightning NO_x emissions provide an estimate of the impact of lightning NO_x on ozone, and so we have incorporated these results into this section of the paper:

As discussed in Section 3.2, methane leads to ozone production in the presence of NO_x. Along with humidity and STE, lightning NO_x emissions may increase in a warmer climate, either due to increased frequency of thunderclouds (and therefore lightning), or more intense thunderstorms (Schumann and Huntrieser 2007; Price 2013). Figure 9a shows lightning NO_x emissions from SOCOL averaged over the 1960s, and shows that most lightning is produced over Africa and South America. Lightning NO_x emissions increase over the continents by 61% between 1960-2100 (Fig. 9b), and by 48% between 2000-2100. Smyshlyaev et al. (2010) found that ozone increased between 10-20% when

lightning NO_x emissions increased by 2 Tg(N) $year^{-1}$ (depending on latitude and season), and up to 90% with a 20 Tg(N) $year^{-1}$ increase in lightning NO_x. Banerjee et al. (2014) calculated increases in lightning NO_x emissions of 33% (2 Tg(N) $year^{-1}$) and 78% (4.7 Tg(N) $year^{-1}$) between 2000-2100 in simulations using RCP 4.5 and RCP 8.5, respectively. In our fEmis simulation (~~Recalling that we~~which used RCP 6.0, a scenario of intermediate severity compared to RCP 4.5 and RCP 8.5), we calculate a 48% increase in lightning NO_x emissions over the same period, which is broadly consistent with their findings. Banerjee et al. (2014)~~.~~ also showed that under RCP 8.5, the increase in lightning NO_x emissions of 78% caused ozone increases of up to 30% in the troposphere (maximizing between the equator and 30 °S). Although we cannot quantify ozone increases induced by lightning NO_x emissions in our simulations, the studies referred to here indicate the likely magnitude of increase (20-30%). Together with STE, ozone increases induced by lightning NOx emissions are largely offset by the temperature-induced increased rates of ozone destruction in the troposphere. ~~However,~~ Finally, we note that the results also depend on the chosen lightning parameterization, which is coupled to the cloud top heights~~.~~ Grewe (2009) showed that lightning NO_x emissions might also slightly decrease, when stronger but fewer convective events occur in a future climate.

Specific Comments:

Abstract l.12: add "compared to 1960" somewhere in this sentence.

Done – changed to:

Changes in ozone precursor emissions have the largest effect, leading to a global-mean increase in tropospheric ozone which maximises in the early 21st century at 23% compared to 1960.

p.483, l.12: visibility doesn't just affect traffic.

True, but disruption of traffic is one of the more disruptive effects on human activities. We have modified by adding "for example":

tropospheric ozone has harmful effects because it is an air pollutant, with adverse effects on crop yields (and therefore food security), visibility (affecting, for example, all forms of traffic) and human health (West et al., 2007).

p.484, l.2: the distribution and abundance of ozone may also be affected by changes in transport and convection.

We have noted this and added to the manuscript:

Depending on the sensitivity of ozone budget reactions to humidity and temperature, the distribution and abundance of tropospheric ozone may also be affected by climate change and changes in transport and convection through the 21st century.

p.486, l.18: What are the implications of adding NMVOC as CO? This method captures some of the increase in O3 and decrease in OH that might be expected from NMVOC, but over much longer timescales and hence in different locations.

Considering NMVOCs as an additional source of CO by directly adding a certain fraction of emitted NMVOCs to CO led to a substantial improvement of the simulated CO and OH levels. Before that modification SOCOL showed a severe low bias in near-surface CO mixing ratios and a pronounced high bias in OH. We agree that this rather simple approach neglects the different timescales of the

involved chemical conversions, but nevertheless it leads to an improved model performance all over the globe.

p.489, l.10: "constant removal value": a clear explanation is needed here. Is HNO3 given a simple first-order loss globally throughout the troposphere, or only where there are clouds or rainfall? What lifetime is assumed?

2.5% of HNO3 in the gas phase is removed everywhere up to 160 hPa, in every time step, independent of clouds or rainfall. The lifetime of HNO_3 is not taken into account. We have added some details here to the manuscript:

In the model setup for the present study, a constant removal value was applied to the HNO_3 gas phase at each time step (2.5% of gas-phase HNO_3 was removed everywhere up to 160 hPa, independent of clouds or rainfall).

p.489, l.16: how might inclusion of anthropogenic NMVOC affect these biases? Nitrate formation and RO2 could increase NOx removal and ozone formation may be localized where lifetimes are much shorter, reducing O3?

As mentioned by Young et al. (2013) a high bias in NH tropospheric ozone and a low bias in the SH are widespread features in current chemistry-climate models, irrespective of the complexity of the applied chemical mechanism. That means that the inclusion of additional NMVOC species into SOCOL does not necessarily lead to an improved representation of tropospheric ozone in both hemispheres. Recently, the HNO_3 washout routine in SOCOL has been modified from a constant removal rate to a cloud cover- and precipitation-based approach. While this change indeed led to an enhanced NO_x removal throughout the NH, the signal in tropospheric ozone was less homogeneous.

p.490, l.22: emissions are a source of uncertainty, but are the same as those used in other model studies, e.g., ACCMIP. How do the NO2 biases compare with those from other models?

To the best of our knowledge, none of the ACCMIP studies analyse NO_2 biases. We are unable to find a multi-model study (with the models using the same emissions we did) which compares model-simulated NO_2 with observations.

p.493, l.25: the mid-tropospheric signal is clearly not representative of the surface, but it is broadly consistent with it. How does the surface ozone compare with that seen in recent model studies, e.g., ACCMIP?

We have changed "representative" to "broadly consistent," and addressed how surface ozone compares with ACCMIP as above.

p.497, l.14: "on atmosphere": word missing here.

We have corrected this:

Although the fEmis simulation was designed to assess the impacts of climate change on the atmosphere (Eyring et al., 2013a),

p.497, l.26: "... up to 6 ppb almost everywhere" is unclear; 6 ppb is a maximum, and not everywhere. Either present this as a range (2-6 ppb?) or drop "almost everywhere".

We have deleted "almost everywhere":

Here, with NO_x, NMVOCs and CO fixed in the fEmis simulation, ozone increases up to 6 ppb ~~almost everywhere~~ (a global-mean increase of 6%).

Figure 3: Panels c and d showing the ratio would be clearer with a dichromatic color scale centered at 1 (perhaps red/blue like Fig 6). Is this figure (and Fig 4) a multi-annual average?
Yes, as noted in the caption, the figures are multi-annual averages from 1960-1969. The colour scales on panels c and d have been changed as suggested:

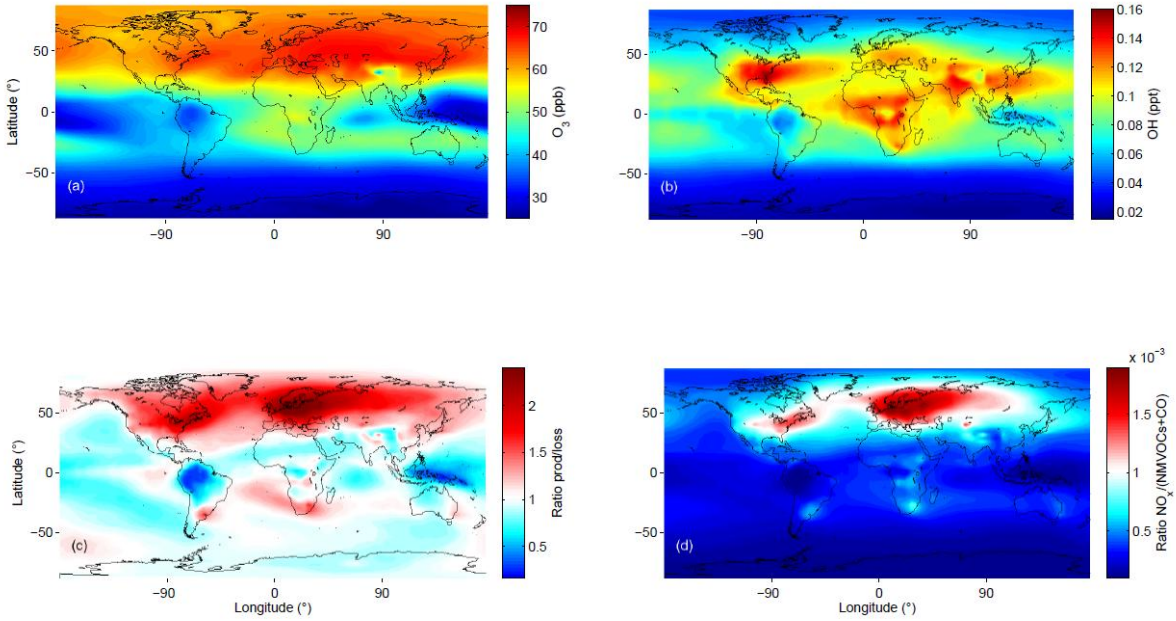

Figure 3. Results from the REF-C2 simulation, 1960-1969 average, 500 hPa. (a) Ozone; (b) OH; (c) Ratio of ozone production over loss; (d) Ratio of NO_x: NMVOCs+CO.