Note: Reviewer's comments are presented in black font; authors' responses are presented in blue plain font; manuscript text quotations are presented in blue italics font.

Anonymous Referee #1

We would like to thank Reviewer #1 for her/his time devoted and the constructive and helpful comments.

General comment:

This manuscript by Akritidis et al. analyzes the impact of future changes in the tropopause fold frequency on concentrations of tropospheric ozone. The authors use an atmospheric chemistry global model and a well-known tropopause fold identification algorithm, to analyze variations in the stratosphere-to-troposphere transport (STT) of ozone, under the RCP6.0 scenario. The study is certainly of interest, since the topic of stratosphere-to-troposphere exchange (STE) is of great importance, especially for what concerns the future ozone variations, which would naturally undergo a decrease in the lower troposphere, as projected by precursors emissions reduction. This is an interesting study and a well written paper, and I recommend publication in ACP after addressing the comments listed below. In particular, the study could be more complete if also the role of troposphere-to-stratosphere transport (TST) is taken into account, especially to quantify whether the ozone reduction in the middle and upper troposphere (due to precursors emissions reduction) is "overcome" by the increase in ozone due to STT, which seems to occur globally.

We thank the Reviewer for the comments, to which we will respond point by point.

Specific comments:

1. Page 3, Line 7. The authors should motivate the choice of the RCP6.0 scenario. Apart from the RCP8.5, which was already assessed in the past, why not choosing, e.g., RCP2.6 or RCP4.5?

The examined simulation RC2-base-04 is part of the Earth System Chemistry integrated Modelling (ESCiMo) initiative chemistry–climate simulations, which have been conducted by the MESSy Consortium with the EMAC model following the recommendations by the Chemistry-Climate Model Initiative (CCMI). According to Eyring et al. (2013), the objective of REF-C2 (RC2) is to produce best estimates of the future ozone and climate changes up to 2100, under specific assumptions about GHG as well as tropospheric ozone and aerosol precursors that follow RCP 6.0 and a specific ODS scenario that follows the halogen scenario A1 from WMO (2011). The respective description of RC2 simulation has been modified in the Revised Manuscript (RM) as follows: P4, L3-7 *"More specifically, data from the simulation RC2-base-04 are used, which is part of the set of simulations performed within the ESCiMo project (Jöckel et al., 2016) following the recommendations by the CCMI. According to Eyring et al. (2013), the objective of REF-C2 (RC2) simulations is to* *produce best estimates of the future ozone and climate changes up to 2100, under specific assumptions about GHG, as well as tropospheric ozone and aerosol precursors that follow RCP 6.0, and a specific ODS scenario that follows the halogen scenario A1 from WMO (2011b)."*

2. Page 4, Lines 23–28. Do the authors take into account any limitations of the work by Škerlak et al. (2015)? How would these affect the comparison between the two methodologies?

For the fold detection we implement the same 3-D labelling algorithm as the one used in the study of Škerlak et al. (2015), thus the methodologies are the same. The differences found compared to Škerlak et al. (2015) are subject to the different meteorological input and the different vertical and horizontal resolution in each case. As Škerlak et al. (2015) use the ERA-Interim dataset, we consider this study as a reference to assess the performance of the RC2 simulation. Given the fact that RC2 is a free-running (without nudging) simulation, the spatiotemporal features of fold frequencies in RC2 are reproduced satisfactorily. Yet, there is an overestimation of fold frequencies compared to Škerlak et al. (2015). The respective discussion has been modified in the RM as follows: P5, L10-16 *"The results are similar, implying a good representation of present-time monthly folding frequency. Yet, a small systematic overestimation of EMAC fold frequencies is seen. Additionally, not only the hemispheric monthly fold frequencies are similar between data from simulation RC2-base-04 and data from ERA-Interim, but also the geographical distribution presents the same patterns (see Fig.4). Any discrepancies might be attributed to the fact that RC2-base-04 is a free-running simulation with different horizontal and vertical resolution. We can therefore consider that the data used in this work are comparable for present-time with state-of-the-art calculations based on the ERA-Interim dataset."*

3. Page 6, Lines 14–23. The strengthening of the BDC would imply more rising air in the tropics, which would then be reflected in a decrease of ozone in the tropical lower stratosphere. Is there any evidence on this, also based on TST (troposphere-to-stratosphere transport) studies? In particular, is Line 19 ("increased upwelling of tropospheric ozone-poor air into the lower stratosphere"), supported by any result? At line 20, the authors indicate a "global STE increase" as the main cause of tropospheric ozone increase, but would this include an increase in both of the two components, i.e., STT and TST, or does it refer to STT only?

The decrease of tropical lower stratospheric ozone under an increase of GHGs due to a BDC strengthening and the induced upwelling enhancement has been reported from several studies, such as Zeng et al. (2010), Young et al. (2013), Banerjee et al. (2016) and Abalos et al. (2017). Specifically, Abalos et al. (2017) suggested an increase in the tropical upwelling, and thus a stronger vertical TST in the future. The decrease of tropical lower stratospheric ozone in EMAC RC2 simulation is presented

in Figure R1.1a, depicting the differences of zonal-mean ozone partial pressure between the FUT and REF periods. Moreover, in Figure R1.1b we present the temperature profiles over the tropics (20S-20N) for the REF and FUT period, as well the difference between them. It seems that the projected warming in the upper troposphere combined with the projected cooling in lower stratosphere results in enhanced upwelling through the tropopause and towards the lower stratosphere, which also agrees with the findings of Lin et al. (2017). The following discussion has been included in the RM: P7, L15-19 *"This tropical lower stratospheric ozone decrease under an increase of GHGs, due to a BDC strengthening and the induced upwelling enhancement, has been reported in other studies as well (e.g. Zeng et al., 2010; Young et al., 2013; Banerjee et al., 2016; Abalos et al., 2017). Specifically, Abalos et al. (2017) using the artificial tracer e90, suggested an increase in the tropical upwelling, and thus a stronger vertical TST in the future."*
Regarding the "global STE increase" we agree with the reviewer, as we indeed refer to "global STT increase". This has been changed accordingly in the RM.

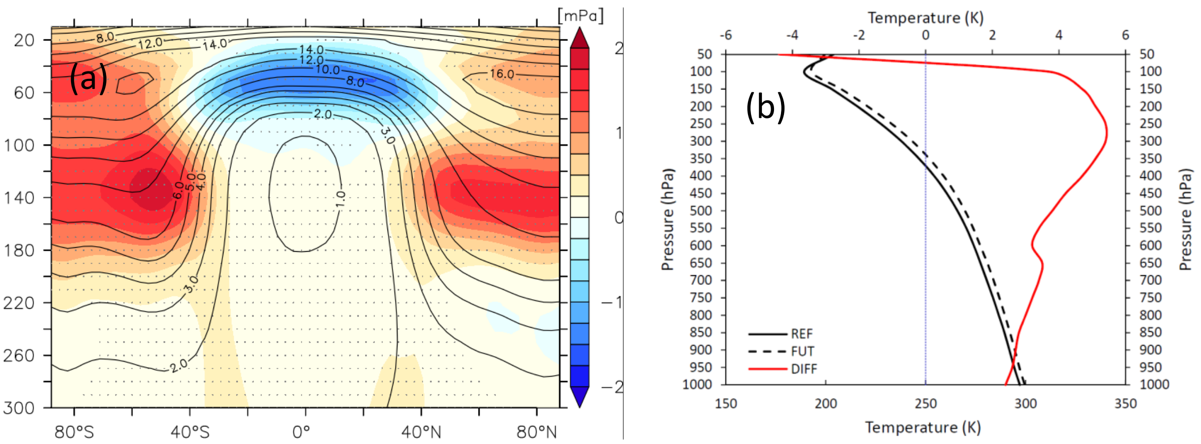

Figure R1.1. a) Zonal-mean O3 partial pressure differences between the FUT and REF periods (colour shaded). Contours depict the zonal-mean O3 partial pressure during the REF period b) Temperature (REF, FUT and differences) profiles over the tropics (20S-20N). The vertical axis stands for pressure (hPa).

4. Page 7, Lines 2–3. In which way is the increase in GHGs concentrations related to the increase in STE of ozone?

Meul et al. (2018) in their sensitivity simulations with EMAC model accounted for GHG increase (RCP8.5) only, ODS decrease only and both, finding that the GHG increase is the main driver of the increased ozone mass flux into the troposphere through the strengthening of the BDC and the increase of the net ozone production in the stratosphere. The respective sentence has been modified in the RM as follows: P7, L30-32 *"Meul et al. (2018) in their future projected simulations under the RCP8.5 GHGs scenario with the EMAC model noted a similar increase in ozone STT through the strengthening of the BDC and the increase of the net ozone production in the stratosphere, which was attributed to the rising GHGs concentrations".*

5. Page 7, Lines 12–14. Again, the role and quantification of TST in not taken into account here. What role would it play in modulating the increase of ozone STE reported in the paper?

An explicit quantification of TST is beyond the scope of this paper, as the EMAC model doesn't include the appropriate tracer (like e90 tracer). However, the effect of TST is shown over the tropics with enhanced upwelling leading to higher water vapour mixing ratios (see Figure R1.2 below) and lower ozone in the lower stratosphere (Figure R1.1a).

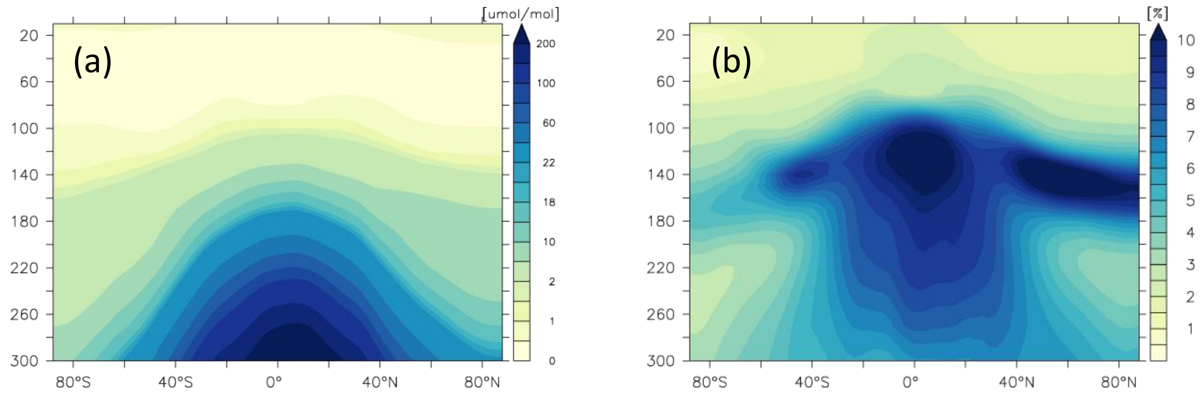

Figure R1.2. a) Zonal-mean water vapour mixing ratio a) differences and b) percentage differences between the FUT and REF periods. The vertical axis stands for pressure (hPa).

6. Page 8, Lines 28–31. Would it be possible to "quantify" the effect of these two contributions (i.e., reduction of ozone precursors emissions and increase of ozone STT), so that one could quantitatively see that the ozone decrease due to emissions reduction is effectively canceled out by the global ozone increase due to STT?

The increase of tropospheric ozone due to the STT increase is depicted in Figures 6 and 7. Quantification of the role of ozone precursor's emissions reduction on ozone is not possible since this is not a sensitivity simulation. Nevertheless, to investigate the mechanisms assisting/cancelling the STT-related tropospheric ozone increase, we have calculated the future projected changes of the main ozone chemical production and loss processes, presented in Figure R1.3. Overall, a reduction of net ozone production is projected in the lower and middle troposphere, as a result of a) the reduction of anthropogenic emissions of ozone precursors leading to decrease of ozone production (Prod-HO2) in the lower troposphere and b) the increase of water vapour leading to increase of ozone destruction (Loss-O1D) in the lower and middle troposphere. Moreover, the increase of ozone in the lower troposphere through RO2 probably indicates the impact of the BVOC emissions of ozone precursor's increase due to the global warming. In the upper troposphere, the dominant feature is the increase of ozone production (Prod-HO2) likely resulting from the enhanced lightning NOx emissions, again due to a warmer climate and the associated enhanced convection activity. Both BVOC and lightning NOx emissions in RC2-base-04 simulation are increasing in future (see Figures 3 and 4 in Jöckel et al. (2016)).

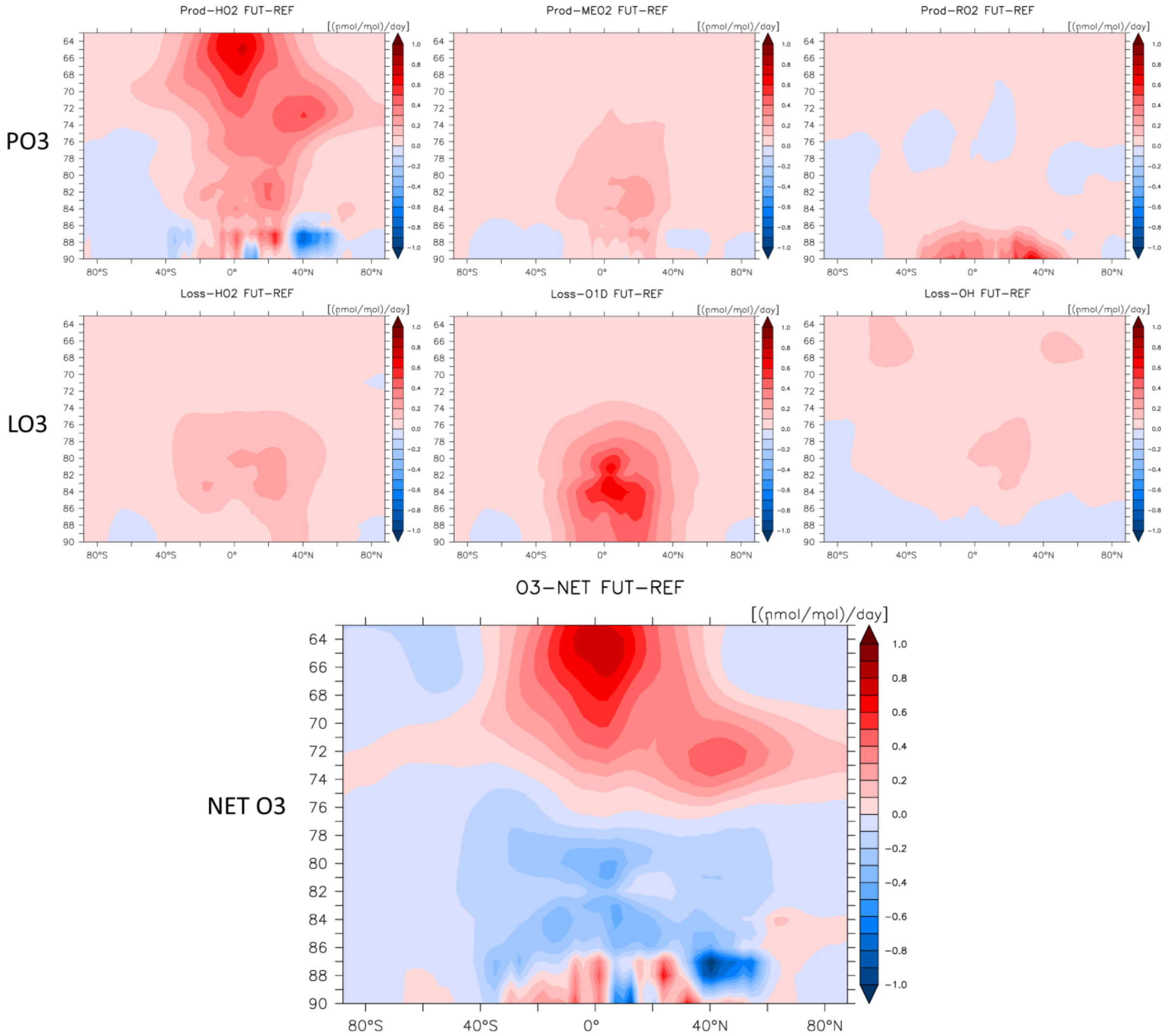

Figure R1.3. Differences in zonal-mean O3 production rates (from HO2, methyl peroxy radical and RO2) (top), in zonal-mean O3 loss rates (from HO2, O1D and OH) (middle) and in zonal-mean net O3 production rates (bottom) between the FUT and REF periods. The vertical axis stands for the model levels.

According to the previous, we have modified several parts of the manuscript:

P1, L15-17 *"..due to the decline of ozone precursors emissions and the enhanced ozone loss from higher water vapour abundances, while in the rest of the troposphere ozone shows a remarkable increase owing mainly to the STT strengthening and the stratospheric ozone recovery."*

P6, L31-33 *"This is also the case in the examined simulation, as the projected increase of water vapour mixing ratios is contributing to the decrease of lower tropospheric ozone through its enhanced chemical loss (not shown)."*

P7, L1-3 *"The aforementioned decreases in lower tropospheric ozone, are overcoming the appearing increases in ozone chemical production (not shown), which are likely associated with the enhanced emissions of BVOCs and lightning NOx (see figures 2, 3 and 4 in Jöckel et al. (2016))."*

P7, L7-12 *"These patterns of tropospheric ozone increase are due largely to a global STT increase, linked to stratospheric ozone recovery and a strengthening of BDC, as suggested by previous studies based on simulations with CCMs (Banerjee et al., 2016; Morgenstern et al., 2018). The enhanced lightning NOx, are also likely to act auxiliary in the direction of increasing tropospheric ozone. In the free troposphere, it seems that the beneficial reduction of ozone precursor emissions and the ozone decline due to higher water vapour content, is cancelled out by the projected increase of stratospheric ozone influx and ozone chemical production from BVOC and lighting NOx."*

P8, L7-8 *"..is mostly driven by the strengthening of BDC and the recovery of stratospheric ozone,.."*

P9, L23-25 *"Ozone in the lower troposphere and near the surface decreases under the projected decline in ozone precursor's emissions and the effect of increased water vapour content. In the middle and upper troposphere the projected strengthening of ozone STT contributes to the increase of ozone globally."*

Technical corrections:

1. Page 5, Line 21. "Green contours", please revise Fig. 4 caption, i.e., "black"→"green".

Done.

2. Pag. 6, Lines 28–29. Please check correspondence between Figure numbering and seasons.

Done.

3. Figure 7. "concnentrations"→"concentrations".

Done.

4. Page 7, Line 21. "EM" or "EMME"? Please be consistent.

We thank the Reviewer for the comment. It is EMME. This has been modifies accordingly in the RM.

5. Page 7, Lines 25 and 30. "positevely"!"positively".

Done.

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