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 #3

We would like to thank Reviewer #3 for her/his time devoted and the constructive and helpful comments.

General comment:

This is an interesting and well-presented paper. My main concern is that like many of the past papers that discuss the influence of climate change (either past or future) on tropospheric ozone, it is difficult to separate out the individual effects of different processes. You appear to be assuming that all (or the vast majority) of your signal is just the combination of changes in STT, together with changes of anthropogenic emissions under RCP6.0. But what about changes in water vapour, and natural emissions from lightning NOx and BVOCs (etc.)? Most of these are barely discussed in the paper, but I think they must be simultaneously changing, and having potentially large effects. Some authors have attempted to separate out some of these processes in the past (e.g., Wild, 2007; Doherty et al.,2013), but this is not easy. This wider context needs to be discussed to place some perspective on where changes in the STT rank compared to other climate change effects on ozone. If this can be included, and the points below, then I am happy to recommend publication in ACP.

We thank the Reviewer for the comments, to which we will respond point by point. We agree with the main comment of the Reviewer, regarding the future projected role of water vapour, lightning NOx and BVOCs on tropospheric ozone changes, and thus, we have included the appropriate discussion in the RM (Introduction, Methodology and Results). In the examined simulation, lighting NOx, soil NOx and BVOC emissions are online calculated by the MESSy submodels LNOX (Tost et al., 2007) and ONEMIS (Kerkweg et al., 2006), and therefore they consider the climate change and the induced effects on ozone chemical production/loss.

Specific comments

P1

L4: Clarify the temporal and spatial context of the 3% increase (i.e. from (1970-99) to (2070-99); is it a global average number, or related to the model grid size?)

The exceedance of 3% increase in fold frequency is seen over some regions. We have modified the respective phrase in the Revised Manuscript (RM) as follows: P1, L4-5 *"Statistically significant changes in tropopause fold frequencies from 1970-99 to 2070-99 are identified in both Hemispheres, regionally exceeding 3%,..".*

L8: maxima -> largest

Done

L9: Highest background fold frequencies, or changes?

It is the "highest fold frequencies changes". This has been modified in the RM.

Abstract: How is the (likely) shortened lifetime of tropospheric ozone in future, due to higher levels of water vapour, and hence bigger flux through O(1D)+H2O, taken into account? Also what about changes in lightning NOx emissions (and BVOC emissions, and other climate dependent processes…) that may affect tropospheric O3? Introduction: This should also mention other climate-driven influences on tropospheric O3 – ie water vapour, lightning NOx, biogenic VOC emissions, etc.

We agree with the Reviewer and thus we have included the following discussion in the RM:

Introduction, P2, L6-9 *"Moreover, climate-related changes in lightning NOx emissions, Biogenic Volatile Organic Compounds (BVOCs) emissions and water vapour content, are also key drivers of future tropospheric ozone changes, affecting its chemical production and loss processes (Wild, 2007; Fiore et al., 2012, 2015; Doherty et al., 2013)".*

Introduction, P3, L9-19: *"There is a high confidence that the increasing temperature will lead in a decline of lower tropospheric ozone through the enhanced water vapour abundances and the associated acceleration of ozone chemical loss (Fiore et al., 2012, 2015; Fu and Tian, 2019). Several studies indicate that the emissions of BVOCs are subject to increase in a warming climate, as they are temperature-sensitive, leading to a positive feedback on future ozone chemical production (Zeng et al., 2008; Weaver et al., 2009; Doherty et al., 2013). Yet, other studies considering the CO2 inhibition effect, report that this positive feedback on ozone may be offseted or even reverse negative (Tai et al., 2013; Hantson et al., 2017). Climate-related changes in lightning activity and the associated NOx emissions are thought to have complex implications for tropospheric ozone. While the enhancement in lightning NOx emissions in a warmer climate will increase baseline ozone, the induced enhancement in OH will result in CH4 reduction and thus, in a decline of ozone chemical production on greater timescales (Wild, 2007; Banerjee et al., 2014; Murray, 2016). Moreover, climate-induced changes in NOx emissions from soils and ozone precursors emissions from wildfires are also expected to modulate future ozone changes (Voulgarakis and Field, 2015; Romer et al., 2018)."*

Methodology, P4, L15-17: *"Lightning NOx emissions and emissions of BVOCs are online calculated by the MESSy submodels LNOX (Tost et al., 2007) and ONEMIS (Kerkweg et al., 2006), respectively, considering the effects of climate change."*

P4

L1: Presumably the stratospheric ozone tracer ignores rapid cycling processes involving O3, ie.: O3 + NO -> NO2 + O2 NO2 + hv -> NO + O O2 + O -> O3 Which form a null cycle. But presumably it does include Ox (O3+NO2) loss processes that interact with this cycle, such as O(1D)+H2O and NO2 dry deposition?

Yes, this is correct.

P5
L7 in the Northern Hemisphere, not at the Northern Hemisphere (and several other similar instances). 'At' is appropriate for a specific site, whereas 'in' is more appropriate for a larger region. I don't think this is just my dubious grammar.

Done

L22 Do you mean the hotspots in the REF distribution, or the changes?

We mean for the REF period. This has been modified in the RM to make it clearer (P6, L10).

L26 delete 'a'

Done

L27 It is a bit confusing that Figure 3 has colours for REF winds and contours for FUTREF changes, whilst Figure 4 has contours for REF fold frequencies and colours for changes. I suggest all the figures follow a consistent format?

We agree with the comment. Figure 3 is in the same format as Figure 4 in the RM. Figure 3 caption has been modified accordingly also.

P6
L12 lower tropospheric ozone?

We thank the Reviewer for the comment. We mean that "Clearly, temperature and humidity under a warmer climate play an important role in decreasing tropospheric ozone in the tropical Pacific, due to the increased rate of the ozone destruction reactions (Revell et al., 2015)", which has been updated in the RM (P6L33-P7L1). Moreover, we have updated the reference of Revell et al. (2015) with the appropriate one (P14, L28-30), as initially we have inadvertently included another one.

Section 3.2 What about lightning NOx? Does it change? And BVOCs?

Please, also see our response to Reviewer's #1 Specific Comment #6, where we present the future changes in ozone chemical production and loss. The future projections of soil NOx, total BVOCs, and lightning NOx emissions for the examined simulation (RC2-base-04) are provided in Figures 2, 3 and 4, respectively, in Jöckel et al., (2016), depicting an increase up to 2100. As the examined simulation is not sensitivity, we cannot separate the respective effects on ozone. Nevertheless, we have included the following discussion regarding the potential effects of both in future ozone changes.

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."*

Section 3.3 From your experiments it is not possible to separate the effects of stratospheric O3 recovery (due to ODS declines) and enhanced STE. Is that correct?

Yes, this is correct.

P7

L25 and l30 positively

Done

Section 4: Should EM be EMME?

Yes it should. Every instance of EM is replaced by EMME.

P18, Figure 4 caption: green not black. What are the units of fold frequency? "hatched with black circles" -> "indicated by black dots".

Done. The units of fold frequency are percentage (%) of fold occurrence during the respective period.

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