Response to the Referees

"On the impact of future climate change on tropopause folds and tropospheric ozone"

by Dimitris Akritidis et al.

Dear Dr. Jimenez-Guerrero

We would like to thank the three Referees for their constructive comments and their time devoted on reviewing our manuscript, contributing to its improvement.

On the following pages, we present our point by point response (blue colour) to the comments raised by the Referees (black colour), along with the corresponding changes made in the manuscript. A version of the revised manuscript with our changes highlighted (blue and red) is also included.

Sincerely,

Dimitris Akritidis (on behalf of all the co-authors)

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.

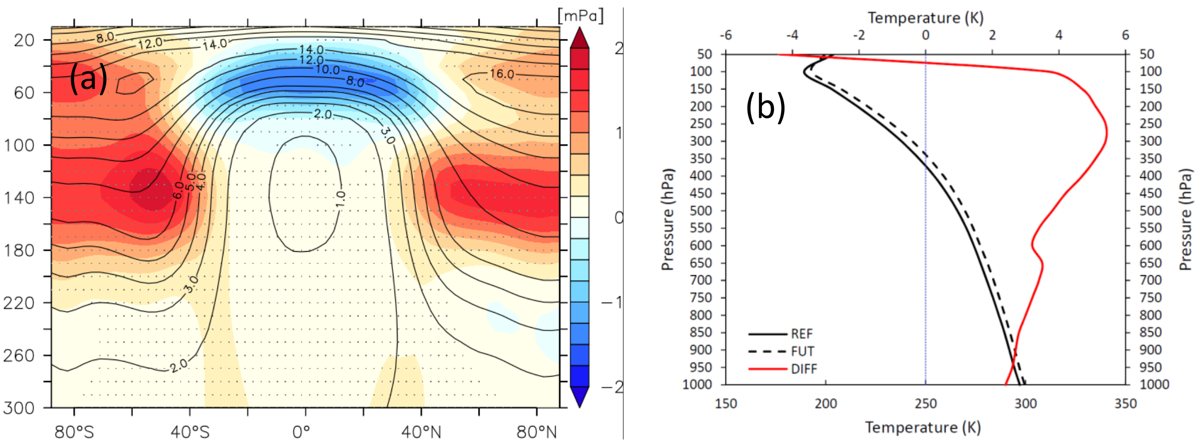

Figure R1.1. a) Zonal-mean O3 partial pressure differences between the FUT and REF periods (colour shaded). Contours depict the zonal-mean O3 partial pressure during the REF period b) Temperature (REF, FUT and differences) profiles over the tropics (20S-20N). The vertical axis stands for pressure (hPa).

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).

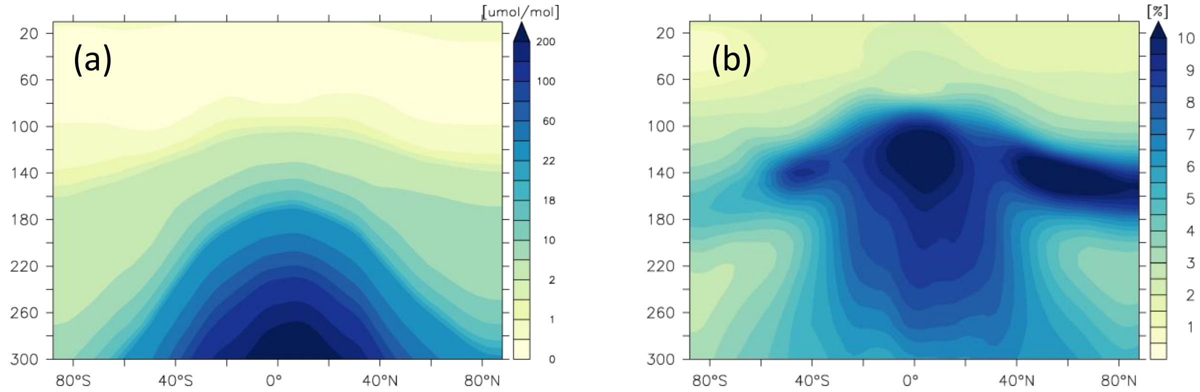

Figure R1.2. a) Zonal-mean water vapour mixing ratio a) differences and b) percentage differences between the FUT and REF periods. The vertical axis stands for pressure (hPa).

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)).

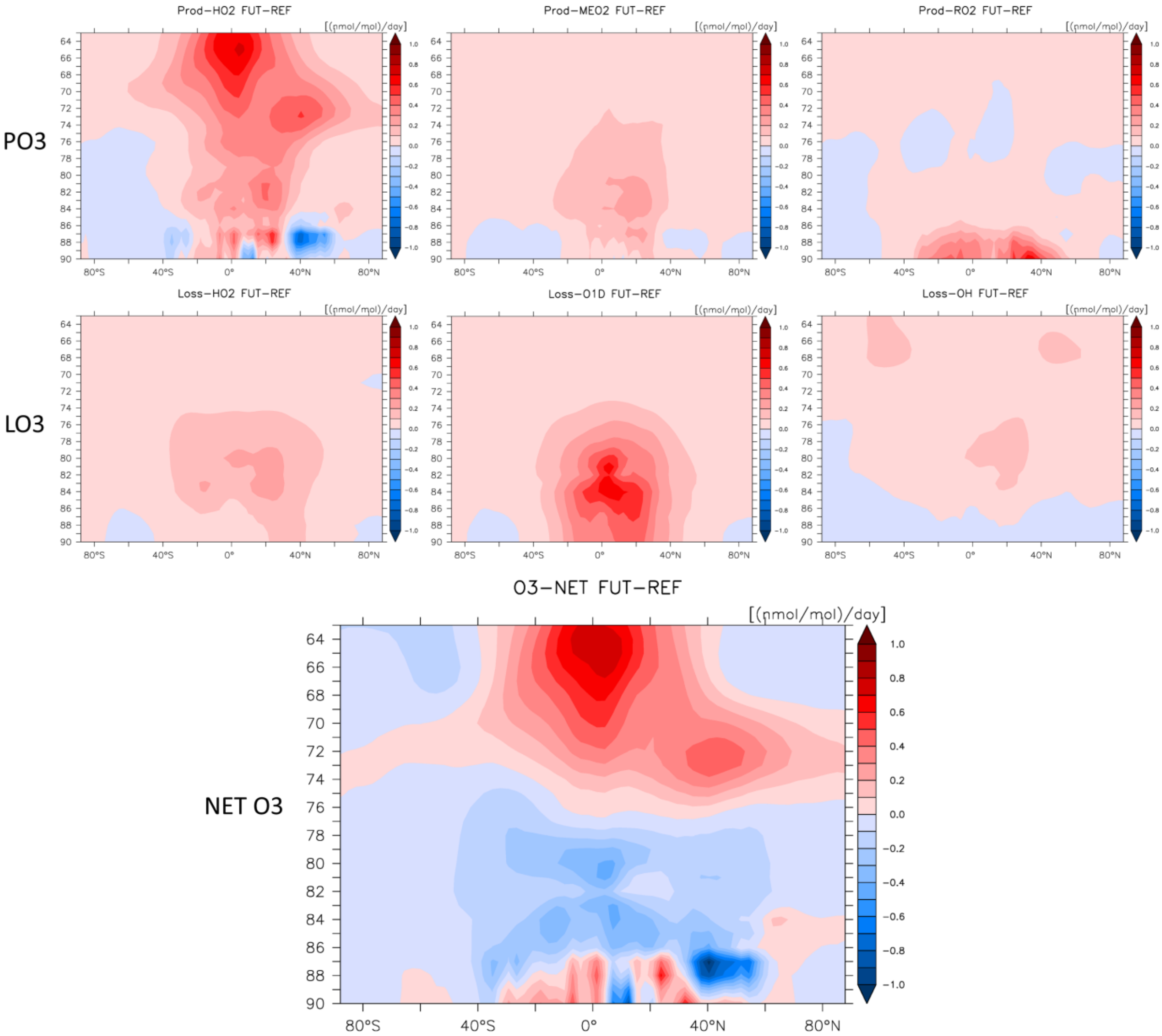

Figure R1.3. Differences in zonal-mean O3 production rates (from HO2, methyl peroxy radical and RO2) (top), in zonal-mean O3 loss rates (from HO2, O1D and OH) (middle) and in zonal-mean net O3 production rates (bottom) between the FUT and REF periods. The vertical axis stands for the model levels.

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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Anonymous Referee #2

We would like to thank Reviewer #2 for her/his time devoted and the constructive and helpful comments.

General comment:

The authors explore the roles of future climate change in tropospheric ozone changes using a global chemistry-climate model with artificial stratospheric ozone tracer. The results of this study emphasize the importance of downward transport of stratospheric ozone associated with tropopause folds. You've convinced me that changes in tropopause folds are regulated by upper-level jet. Also, I agree that projected increase of tropospheric ozone is associated with changes in BDC and STT. However, I find the linkage between the presence of folds and changes in ozone is relatively weak. I would expect shallow tropopause folds, which are located above 200hPa, account for the most changes in folding frequency. How do these shallow folding activities affect the ozone near 400-500hPa or even below? We know that summertime large-scale subsidence at 500hPa over Mediterranean is projected to change [Cherchi et al., Clim Dyn (2016)]. Perhaps the large changes in ozone near 400hPa are primarily associated with changes in descent, while the presence of tropopause folds is secondary. Except the one concern I've pointed out, this paper is very well structured and is certainly within the scope of ACP, although improvements can be applied to make it clearer. Therefore, I only have some minor comments.

We thank the Reviewer for the comments, to which we will respond point by point. Indeed the vast majority of tropopause folds are shallow. Nevertheless, considering that the average pressure of the tropopause in the extratropics is about 250 hPa and that the vertical extend (Δp) of shallow folds range from 50 to 200 hPa below the tropopause, the shallow foldings extend down to approximately 300-450 hPa. Of course, the large scale subsidence over specific regions, such as the summertime EMME, can further transport high ozone concentrations towards lower tropospheric levels in greater timescales. Thus, the folding mechanism enriches the upper and middle troposphere with high ozone concentrations, which might be further vertically transported under favorable meteorological conditions.

1. What's your rationale for choosing RCP6?

Please refer to our response in Reviewers' #1 Specific Comment #1.

2. Ozone is difficult to simulate in models due to biases in photochemistry processes and precursor emissions. Have you evaluated model performance in ozone? Discussion regarding how biases in EMAC would affect the estimated changes is necessary.

All ESCiMo simulations, including the RC2-base-04 simulation, are evaluated in the study by Jöckel et al. (2016) using the BSTCO (Bodeker Scientific combined total column ozone database; Bodeker et al., 2005) for total column ozone, the AURA Microwave Limb Sounder/Ozone Monitoring Instrument (MLS/OMI; Ziemke et al., 2011) for tropospheric and stratospheric partial column ozone and the ozonesonde dataset by Tilmes et al. (2012) for ozone profiles. In general the seasonal cycle and the spatial distribution of total column ozone are well reproduced in the simulation, with an overestimation of up to 9%. The following sentence has been extended in the RM: P4, L17-18 *"A detailed description of the simulation along with a comprehensive evaluation of ozone with satellite and ozonesonde measurements can be found in Jöckel et al. (2016).".*

3. In Fig.1, wintertime medium and deep fold frequency are much higher than those shown in Škerlak et al. (2015). Will it affect your results? Also, it'd be good to address that the climatological distribution of tropopause folds in your model is consistent with what shown in previous studies.

Given the fact that medium and deep folds are very rare to occur, as the order of magnitude of their frequencies in a global scale are -1 and -2 respectively (please mind the x10 and x100 notations in Figure 1) the impact on our results is expected to be very small. As concerns the climatological distribution of tropopause folds in RC2 simulation compared to previous studies, Figure 4 depicts the spatial distribution of tropopause folds frequency (green contours) for the REF period, which is also discussed compared to the climatology of Škerlak et al. (2015) in the manuscript as follows (P6, L8-13): *"The spatial distribution of fold frequencies during the REF period (green contours in Fig. 4), indicates that in principal folds occur in the regions with high zonal wind speed (colour shadings in Fig. 3). Noteworthy are the hotspots over Asia and Middle East during DJF and JJA, and over the southern Indian Ocean during JJA, whereas during the transition seasons the maxima are located over Asia in MAM, and over Asia and southern Indian Ocean in SON, being consistent with the ERA-Interim derived tropopause fold climatology of Škerlak et al. (2015)."*

4. I'm worried whether the future changes of tropopause folds are robust. Have you compared with other models?

To our knowledge, this is the first projection of tropopause fold frequencies under a future scenario, so we are not able to compare with other models and studies.

Detailed comments:

1. P18, Fig.4 caption, "black" -> "green"; "circles" -> "dots"

Done

2. P6, line 13, "contrary" -> "on the contrary"

Done

3. P7, "positevely" -> "positively"

Done

References

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Škerlak, B., Sprenger, M., Pfahl, 5 S., Tyrlis, E., and Wernli, H.: Tropopause Folds in ERA-Interim: Global Climatology and Relation to Extreme Weather Events, Journal of Geophysical Research: Atmospheres, 2015.

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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 $O(1D)+H_2O$, taken into account? Also what about changes in lightning NOx emissions (and BVOC emissions, and other climate dependent processes…) that may affect tropospheric O_3? Introduction: This should also mention other climate-driven influences on tropospheric O_3 – 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 CO_2 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 CH_4 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 O_3, ie.: $O_3 + NO \rightarrow NO_2 + O_2$ $NO_2 + hv \rightarrow NO + O$ $O_2 + O \rightarrow O_3$ 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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On the impact of future climate change on tropopause folds and tropospheric ozone

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Abstract. Using a transient simulation for the period 1960-2100 with the state-of-the-art ECHAM5/MESSy Atmospheric Chemistry (EMAC) global model and a tropopause fold identification algorithm, we explore the future projected changes in tropopause folds, Stratosphere-to-Troposphere Transport (STT) of ozone and tropospheric ozone under the RCP6.0 scenario. Statistically significant changes in tropopause fold frequencies from 1970-99 to 2070-99 are identified in both Hemispheres, occasionally regionally exceeding 3%, which are associated with the projected changes in the position and intensity of the subtropical jet streams. A strengthening of ozone STT is projected for future at in both Hemispheres, with an induced increase of transported stratospheric ozone tracer throughout the whole troposphere, reaching up to 10 $\mathrm{nmol/mol}$ in the upper troposphere, 8 $\mathrm{nmol/mol}$ in the middle troposphere and 3 nmol/mol near the surface. Notably, the regions exhibiting the maxima largest changes of ozone STT at 400 hPa, coincide with that of the highest fold frequencies changes, highlighting the role of tropopause folding mechanism in STT process under a changing climate. For both the eastern Mediterranean and Middle East (EMME), and the Afghanistan (AFG) regions, which are known as hotspots of fold activity and ozone STT during the summer period, the year-to-year variability of middle tropospheric ozone with stratospheric origin is largely explained by the short-term variations of ozone at 150 hPa and tropopause folds frequency. Finally, ozone in the lower troposphere is projected to decrease under the RCP6.0 scenario during MAM (March, April and May) and JJA (June, July and August) at in the Northern Hemisphere, and during DJF (December, January and February) at in the Southern Hemisphere, 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.

1 Introduction

Tropospheric ozone plays a key role in the oxidizing capacity of the atmosphere (Lelieveld et al., 2016), it is also a short-lived climate forcer being an important greenhouse gas, while near the surface it is a pollutant detrimental to human health, crops and ecosystems (Monks et al., 2015). The future tropospheric ozone changes in global scale depend on changes of the processes that control tropospheric ozone budget, namely, chemical ozone production and loss, Stratosphere-Troposphere Exchange (STE) and deposition (Young et al., 2013). The net stratospheric influx results from STE processes, comprised of Troposphere-to-Stratosphere Transport (TST) and Stratosphere-to-Troposphere Transport (STT) with tropopause folds considered as the main mechanism for stratospheric intrusions in STT events (Stohl et al., 2003). In the 21^{th} century, emissions of ozone precursor species, ozone depleting substances (ODSs) and long-lived greenhouse gases (GHGs) are expected to be the major factors governing ozone amounts and its distribution in the troposphere and the stratosphere ~~(Fiore et al., 2015; ?)~~ (Fiore et al., 2015; Revell et al., 2015). More specifically, future changes of the net stratospheric influx in STE are linked to changes of the stratospheric Brewer-Dobson Circulation (BDC) and the amount of ozone in the lowermost stratosphere, which are strongly influenced in a changing climate by the emissions of ODSs and GHGs (Oberländer-Hayn et al., 2016; Morgenstern et al., 2018). 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).

Predominantly, the foldings of the tropopause are of limited vertical extend and their global spatiotemporal distribution is mainly controlled by the location and intensity of the jet stream, as in principle, they are developed through ageostrophic flow in the proximity of the jet stream (Stohl et al., 2003). Deep folds extending down to the lower troposphere and occasionally to the ground surface may lead to irreversible mixing of stratospheric air into the troposphere, and thus, to chemical composition changes (Cristofanelli et al., 2006; Akritidis et al., 2010; Lin et al., 2015; Knowland et al., 2017). During the recent years, several modeling studies indicated that the stratospheric contribution to tropospheric and near-surface background ozone may be of greater importance than previously anticipated (Zhang et al., 2011; Lin et al., 2012; Zanis et al., 2014; Lefohn et al., 2014; Akritidis et al., 2016; Williams et al., 2019).

Changes in ozone precursor emissions have the largest effect on future tropospheric ozone concentrations. Future reductions in most ozone precursor emissions, which is a common feature across the Representative Concentration Pathways (RCPs), drive tropospheric ozone decreases, except for RCP8.5 that show an increase due to much larger methane concentrations compared to the other RCPs ~~(Stevenson et al., 2006; Naik et al., 2013; Young et al., 2013; Sekiya and Sudo, 2014; ?; Banerjee et al., 2016; Meul et al.,~~ (Stevenson et al., 2006; Naik et al., 2013; Young et al., 2013; Sekiya and Sudo, 2014; Revell et al., 2015; Banerjee et al., 2016; Meul et al. . Future decreases in ODS may lead to an ozone increase essentially everywhere in the atmosphere, with the largest percentage changes in the upper stratosphere and the lower stratosphere at high latitudes due to the anticipated ozone recovery, while changes in GHGs may lead to a decrease in the tropical lower stratosphere and an increase of STE due to strengthening of the BDC (Morgenstern et al., 2018). The 2014 Ozone Assessment (Carpenter et al., 2014) highlighted that Chemistry Climate Models (CCMs) robustly predict a long term acceleration of the BDC in response to anthropogenic climate change (Hardiman et al., 2014; Palmeiro et al., 2014) which also stands for the new CCMI (Chemistry-Climate Model Initiative) simulations (Morgenstern et al., 2018).

Several recent studies with CCMs provide evidence that both the acceleration of the BDC and stratospheric ozone recovery will tend to increase the future global tropospheric ozone burden through enhanced STE with the magnitude of the change depending on the RCP scenario, partially offsetting tropospheric ozone decreases associated with reductions in ozone precursor emissions (Sekiya and Sudo, 2014; Banerjee et al., 2016; Meul et al., 2018). Banerjee et al. (2016) showed that BDC strengthening under the RCP8.5 has the largest impact on tropospheric ozone over the tropics and subtropics, while stratospheric ozone recovery from declining ODSs becomes more important in the mid-latitudes and extratropics. Meul et al. (2018)

simulated that the global mean annual STT is projected to increase by 53% between the years 2000 and 2100 under RCP8.5 and it will be smaller for RCP6.0, but the resulting relative change in the contribution of ozone with stratospheric origin to ozone in the troposphere is of comparable magnitude in both scenarios. The covariability between STE and tropospheric ozone from observations was used to deduce that the projected future strengthening of the BDC alone (without accounting for ozone recovery), could lead to an increase in zonal-mean tropospheric ozone of 2% by the end of the 21st century (Neu et al., 2014). Hess et al. (2015) extrapolating their model results from present time to future, concluded that a 30% increase in the ozone flux by 2100 due to BDC strengthening would result in a 3% increase in surface ozone and a 6% increase in mid-tropospheric ozone. However, Morgenstern et al. (2018) using simulations from multiple CCMs showed that the surface ozone response to anthropogenic forcings from well-mixed GHGS and ODS remains uncertain, reflecting uncertainties related to STE.

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).

It is therefore crucial to conduct more studies towards this direction, in order to increase confidence in the future projected changes of tropospheric ozone and its associated drivers.

This study aims to assess the impacts of future climate change under the RCP6.0 scenario on tropopause folds and tropospheric ozone, using a free-running hindcast and projection ECHAM5/MESSy (EMAC) simulation for the period 1960-2100. To this end, a 3-D labeling algorithm is implemented to detect tropopause folds in EMAC simulation. Besides ozone, a tracer for stratospheric ozone is also employed to investigate the projected changes in STE of ozone. Section 2 presents the main characteristics of the EMAC model and describes the 3-D labeling algorithm used to detect the folding events. Section 3 and 4 show the key results of the current study, and finally, Section 5 summarizes the main conclusions.

2 Methodology

2.1 EMAC model

The ECHAM5/MESSy Atmospheric Chemistry (EMAC) global model is a numerical chemistry and climate simulation system that includes sub-models describing tropospheric and middle atmosphere processes and their interactions with ocean, land and human activities (Jöckel et al., 2010). It uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes. The core atmospheric model is the 5^{th} generation circulation model (ECHAM5, Roeckner et al., 2006). The EMAC model has been extensively evaluated for gas tracers ~~(e.g. Pozzer et al., 2007)~~ (e.g. Pozzer et al., 2007; Jöckel et al and for aerosols ~~(e.g. Pringle et al., 2010; Pozzer et al., 2012; Astitha et al., 2012)~~(e.g. Pringle et al., 2010; Pozzer et al., 2012; Astitha et a . For the present study we use ECHAM5 version 5.3.02 and MESSy version 2.51. 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). The model horizontal resolution is T42L90MA, i.e. with a spherical truncation of T42 (corresponding to a quadratic Gaussian grid of approx. 2.8 by 2.8 degrees in latitude and longitude) with 90 vertical hybrid pressure levels up to 0.01 hPa.

The simulation covers the time frame 1960-2100 (10 years spin-up from 1950 to 1959) driven by prescribed Sea Surface Temperature (SST) and Sea Ice Coverage (SIC) taken from simulations with the global climate model HadGEM2-ES (Collins et al., 2011; Martin et al., 2011) for the Coupled Model Intercomparison Project phase 5 (CMIP5). Anthropogenic emissions are incorporated as prescribed emission fluxes following the CCMI recommendations (Eyring et al., 2013). In more detail, the emissions data set consists of a combination of ACCMIP (Lamarque et al., 2010, for the 1950-2000 period) and RCP6.0 data (Fujino et al., 2006, for the 2000 and on). 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. A detailed description of the simulation along with a comprehensive evaluation of ozone with satellite and ozonesonde measurements can be found in ~~(Jöckel et al., 2016, and references therein)~~Jöckel et al. (2016, and references therein).

Along with ozone chemistry, EMAC also includes a tracer for ozone of stratospheric origin, denoted by O3s, which provides an indicator of the stratospheric contribution to tropospheric ozone. In the stratosphere, O3s is equal to ozone values, while in the troposphere it follows the transport and destruction processes of ozone. When O3s returns to the stratosphere it is reset to stratospheric values; however, since it is initialized above 100 hPa, only a very small fraction is recirculated by multiple crossings of the tropopause (Roelofs and Lelieveld, 1997).

2.2 Tropopause fold identification

In this work the algorithm developed by Sprenger et al. (2003) and improved by Škerlak et al. (2015) has been adopted and applied in order to detect tropopause folds in EMAC simulation (as in Akritidis et al. (2016)), using the 3-D fields of potential vorticity, potential temperature and specific humidity. As in several previous studies (Hoskins et al., 1985; Holton et al., 1995; Stohl et al., 2003; Sprenger et al., 2003), the tropopause is defined as the combination of the isosurfaces of potential vorticity at ±2 PVU and potential temperature at 380 K, whichever is lower (refered as dynamical tropopause). For each grid point a tropopause fold is designated where multiple crossings of the dynamical tropopause are detected in instantaneous vertical profiles. Subsequently, the upper (p_U), middle (p_M), and lower (p_L) pressure levels of tropopause crossings are determined and the pressure difference $\Delta p = p_M - p_U$ between the upper and middle tropopause crossings is calculated (for more details see

Fig.1 in Tyrlis et al. (2014)). The above pressure difference reveals the vertical extent of the tropopause fold, and is used to classify the identified folds into three categories (for more details see Škerlak et al., 2015):

- shallow folds, $50 \leq \Delta p < 200$ hPa
- medium folds, $200 \leq \Delta p < 350$ hPa
- deep folds, $\Delta p \geq 350$ hPa

Before the results from simulation RC2-base-04 can be used to estimate the future projected changes of fold frequencies, the capability to reproduce present-time folding frequencies must be first ckecked. Therefore the model results have been compared with the monthly fold frequencies climatology compiled by Škerlak et al. (2015). The climatology has been calculated using the same identification algorithm used in this work from the ERA-interim dataset (Dee et al., 2011). Figure 1 shows the mean hemispheric (0-65°N and 0-65°S) monthly frequencies of different folding categories calculated from the results of simulation RC2-base-04 for the period 1979-2012, the exact same one covered by the work of Škerlak et al. (2015). This figure can be compared with Figure 7 of the Škerlak et al. (2015) manuscript. 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 (~~not shown).~~ 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 Future projected changes

To explore the future projected changes in EMAC meteorological and chemical parameters under the RCP6.0 emissions scenario, we consider two 30-years time periods: a) present-day climate used as reference (REF) spanning from 1970 to 1999 and b) future climate (FUT) spanning from 2070 to 2099. The selection of a 30-years period for the climate representation is complying with World Meteorological Organization's (WMO) suggestion (WMO, 2011a). All seasons in the manuscript refer to boreal seasons (winter: DJF, spring: MAM, summer: JJA, autumn: SON).

3.1 Jet streams and tropopause folds

At first, the impact on atmospheric circulation under the RCP6.0 scenario is explored. As it is depicted from Figure 2 there is a distinct upward and poleward shift in the Southern Hemisphere (SH) midlatitude jet during all seasons, which is also identified during DJF and SON ~~at~~ in the Northern Hemisphere (NH), yet less pronounced. A poleward-upward shift of the westerly jet in response to greenhouse warming, was reported by several previous studies using individual models (Butler et al., 2010; Orbe et al., 2015; Doherty et al., 2017) or ensembles of models participating in the Intergovernmental Panel on Climate Change

(IPCC) 4^{th} assessment report (Lorenz and DeWeaver, 2007), and the Coupled Model Intercomparison Project phase 3 (CMIP3) and phase 5 (CMIP5) (Swart and Fyfe, 2012; Delcambre et al., 2013; Yim et al., 2016). Moreover, a rise of the tropopause is seen during all seasons ~~at~~ in both the NH and SH extratropics, which in an annual basis is estimated about 8.8 hPa and 5.8 hPa respectively. A more comprehensive view of the present circulation patterns and their future changes is presented in Figure 3. ~~At~~ In the NH, a poleward shift of the zonal wind at 250 hPa is found during DJF over the Atlantic and the eastern Asia, and an equatorward shift over the NH Central and Eastern Pacific, while ~~at~~ in the SH a poleward shift is seen over the Indian Ocean. During JJA, an equatorward shift of the NH subtropical jet stream is depicted over central and eastern Asia, while ~~at~~ in the SH a poleward shift is seen over Australia.

The impacts of RCP6.0 emissions scenario on tropopause folds frequency are hereafter investigated, considering as folds all folds with $\Delta p \geq 50$ hPa (shallow, medium and deep). Figure 4 presents the projected fold frequency changes between the FUT and REF periods along with the climatology of fold frequencies during the REF period for every season. The spatial distribution of fold frequencies during the REF period (green contours in Fig. 4), indicates that in principal folds occur in the regions with high zonal wind speed (~~colour shadings~~ green contours in Fig. 3). Noteworthy are the hotspots during the REF period over Asia and Middle East during DJF and JJA, and over the southern Indian Ocean during JJA, whereas during the transition seasons the maxima are located over Asia in MAM, and over Asia and southern Indian Ocean in SON, being consistent with the ERA-Interim derived tropopause fold climatology of Škerlak et al. (2015). The projected changes in fold occurrence for the FUT period with respect to the REF period during DJF reveal ~~a~~ a distinct pattern of decrease/increase of folds frequency over south Asia/NH Pacific Ocean, associated with the adjacent decrease/increase of zonal wind in the upper troposphere depicted from Figure 3a. During JJA, the equatorward shift of the subtropical jet stream over central Asia implies in an dipole pattern of decrease/increase in fold frequencies, while in the SH a decrease/increase in fold occurrence is found over Southern Africa/Indian Ocean as a response to the projected changes in the upper tropospheric zonal winds. During MAM a distinct increase in folds frequency prevails in a zone extending across the NH Pacific Ocean, and a decrease ~~at~~ in the north of India, while during SON more frequent folding events are projected over the NH Western Pacific Ocean and the Indian Ocean.

3.2 Tropospheric ozone

Here we explore the future changes in tropospheric ozone under the RCP6.0 GHGs scenario. Figure 5 presents the projected changes of zonal-mean ozone along with its climatological values during the REF period in a seasonal basis. The highest concentrations of zonal-mean ozone in the troposphere during the REF period are found ~~at~~ in the NH mid-latitudes during MAM and JJA and ~~at~~ in the SH mid-latitudes during SON. With respect to the REF period, a decrease of zonal-mean ozone in the lower troposphere up to 3 $\mathrm{nmol/mol}$ is projected for the FUT period during MAM and JJA in the NH, and similarly during DJF (austral summer) ~~at~~ in the SH, resulting from the RCP6.0 future ozone precursor emissions reduction, which as expected dominates during the seasons with more intense photochemistry. Additionally, even if we have no change in precursor emissions, as has been also outlined in the 5^{th} IPCC Assessment Report (Kirtman et al., 2013), there is high confidence that in unpolluted regions, higher water vapour abundances and temperatures in a warmer climate enhance ozone destruction, leading to lower baseline ozone levels, while there is medium confidence that in polluted regions it is expected to increase surface ozone. 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). Clearly, temperature and humidity under a warmer climate play an important role ~~for lower~~ in decreasing tropospheric ozone in the tropical Pacific, due to the increased rate of the ozone destruction reactions ~~(?). Contrary~~(Revell et al., 2015) . 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)). On the contrary, in the extratropical lower stratosphere and the upper and middle troposphere ozone is projected to increase during all seasons. The largest increases in the upper and middle troposphere, of up to 12 $\mathrm{nmol/mol}$, are seen in the subtropics and in the vicinity of the jet streams where tropopause folds formation and the induced STT are favoured. The more pronounced increases of ozone are found in the NH/SH during MAM/SON throughout the entire free troposphere. 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. As regards the lower stratosphere, an increase of ozone is projected outside the tropics reflecting the recovery of stratospheric ozone. In the tropical lower stratosphere, the projected decrease of ozone is presumably related to the BDC strengthening and the induced increased upwelling of tropospheric ozone-poor air into the lower stratosphere. ~~These patterns of tropospheric ozone increase are probably resulting from a global STE 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). In the free troposphere, it seems that the beneficial reduction of ozone precursor emissions is canceled out by the projected increase of stratospheric ozone influx~~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.

3.3 Stratospheric ozone tracer (O3s)

To estimate the impact of STE on tropospheric ozone, the projected changes of O3s are examined here. Same as in Figure 5, Figure 6 depicts the differences of zonal-mean O3s concentrations between the FUT and REF periods. An increase of O3s occurs almost throughout the troposphere during all seasons. In the NH, the peak of O3s enhancement is found in the subtropics and in the vicinity of the NH jet stream during DJF and MAM (Fig. 6a and ~~c~~b), while in the SH the respective positive maxima are seen during JJA and SON (Fig. 6~~b~~ c and d), similarly near the position of the SH jet stream. These increases of O3s in the NH/SH subtropical upper and middle troposphere, reveal an increase of isentropic cross-tropopause ozone transport, through tropopause folds that in principal occur near the NH/SH subtropical jet streams. In general, the positive O3s patterns resemble that of tropospheric ozone (Fig. 5), indicating that the projected increase of tropospheric ozone is ~~mainly~~ largely driven by the increase in STT and the induced vertical transport of stratospheric ozone in the underlying troposphere, as it is was also reported from previous modeling studies employing a tracer for stratospheric ozone in future projected sensitivity simulations (Banerjee et al., 2016; Meul et al., 2018). Meul et al. (2018) in their future projected simulations under the RCP8.5 GHGs scenario with the EMAC model noted a similar increase in ~~STE of ozone~~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. A small decrease of O3s occurring mainly in the SH/NH lower troposphere during DJF/JJA, is associated with an increased chemical O3s loss due to a slight increase in OH and HO_2 and their reaction rate with ozone (due to increased temperature).

The spatial distribution of O3s projected changes at 400 hPa is presented in Figure 7, to identify the global hot spots of climate change impact on ozone STT. Overall, an increase of ozone with stratospheric origin is projected in the middle troposphere (400 hPa) during all seasons, reflecting the recovery of stratospheric ozone and the associated increase of ozone STE. Notably, the maxima of O3s increase coincides mainly with the respective maxima of tropopause folds frequency increase (see Fig. 4). In more detail, during DJF the peaks of future O3s increases (up to 12 nmol/mol) are found over the NH Pacific Ocean (Fig. 7a), while during JJA the respective peaks (exceeding 12 nmol/mol) are mainly occured over the central Asia and the Indian Ocean (Fig. 7c). All in all, the emerging increase in ozone STE under the RCP6.0 GHGs scenario is ~~mainly~~ mostly driven by the strengthening of BDC and the recovery of stratospheric ozone, still for regions where tropopause folds are projected to occur more often, the downward transport of ozone from the stratosphere seems to be more pronounced.

4 Hot spots of ozone STT

STT is of great importance for ozone levels and variability in the upper/middle troposphere over regions where the meteorological conditions favor the formation of tropopause folds and the downward transport (Roelofs and Lelieveld, 1997; Sprenger and Wernli, 2003), such as the eastern Mediterranean and the Middle East (EMME) (Li et al., 2001; Zanis et al., 2014; Akritidis et al., 2016), and the broader Afghanistan area (AFG) (Tyrlis et al., 2014; Ojha et al., 2017) during summer, and especially the July-August period. To explore the links of the tropopause folds frequency and stratospheric ozone, with the interannual variability of middle tropospheric ozone with stratospheric origin over the ~~EM~~ EMME (20-45°E,30-40°N) and AFG (60-80°E, 30-40°N) regions, the mean July-August timeseries of tropopause folds frequency, ozone at 150 hPa, and O3s at 400 and 500 hPa for the period 1960-2099 were constructed. Figure 8a presents the mean July-August fields of tropopause folds frequency during the REF period revealing a pronounced fold activity over the depicted ~~EM~~ EMME and AFG regions. For the ~~EM~~ EMME region (Fig. 8b), the interannual variability of mean July-August O3s at 400 hPa (500 hPa) is found to be ~~positevely~~ positively correlated at the 99% significance level with the mean July-August tropopause folds frequency and ozone at 150 hPa, with values of r=0.53 (r=0.43) and r=0.56 (r=0.49), respectively. Employing a multiple linear regression analysis, folds frequency and ozone at 150 hPa are found to explain the 58% (42%) of the variance of O3s at 400 hPa (500 hPa). As regards the AFG region, the variance of the projected mean July-August O3s concentrations at 400 hPa (500 hPa) explained by folds frequency and ozone at 150 hPa is 73% (68%). The year-to-year variability of July-August O3s at 400 hPa (500 hPa) is found positevely positively correlated at the 99% significance level with both fold frequency r=0.64 (r=0.58) and ozone at 150 hPa r=0.64 (r=0.64).

5 Conclusions

This study investigates the future projected changes in tropopause folds, ozone STT and tropospheric ozone under the RCP6.0 emissions scenario, using a transient simulation with EMAC CCM from 1960 to 2100 and a tropopause fold identification algorithm. In particular, we examined the long-term change in tropopause folds frequency and the potential links with atmospheric circulation changes. Moreover, the long-term changes in tropospheric ozone and ozone STT were also explored and associated with the respective variations in fold activity. The most noteworthy findings of the present study can be summarized as follows:

- Robust changes in atmospheric circulation are identified under the RCP6.0 GHGs emissions scenario. A poleward and upward shift of the NH subtropical jet is projected for DJF and SON, while a strengthening of zonal-mean wind in the upper troposphere is seen equatorward for JJA. The responses are more pronounced at in the SH showing a distinct poleward shift for DJF and MAM, with a strengthening of zonal-mean wind poleward during JJA and SON.

- The spatial patterns of the projected changes in NH and SH subtropical jets seem to drive the respective patterns of tropopause folds frequency future changes, with a negative/positive dipole structure found over south Asia/NH Pacific Ocean during DJF and MAM. The most prominent features during JJA are a distinct increase of fold activity over the Indian Ocean exceeding 3%, and a negative/positive dipole structure centered over the greater Afghanistan region.

- The regions exhibiting the highest increases in tropopause folds occurrence in future are those with the more pronounced projected increases of O3s in the middle troposphere (400 hPa). The projected changes of zonal-mean O3s concentrations reveal a strengthening of ozone STT at the middle latitudes of both hemispheres during all seasons, which is more distinct at in the NH during DJF and MAM (up to 6 $\mathrm{nmol/mol}$ down to 500 hPa), and at in the SH during JJA and SON (up to 8 nmol/mol down to 500 hPa). Although the future increase in ozone STT on a global scale seems to be forced from stratospheric ozone recovery and strengthening of BDC (Banerjee et al., 2016; Meul et al., 2018), regionally, the degree of increase in the downward transport of stratospheric ozone is partially driven from the long-term changes of fold activity.

- For specific regions considered as global STT hotspots, namely the summertime EMME and AFG, the projected year-to-year variability of middle tropospheric ozone with stratospheric origin seems to be largely governed from both the variabilities of ozone at 150 hPa and folds frequency, as they explain 60% and 68% of the variance of mean July-August O3s concentrations at 400 hPa for EMME and AFG respectively, over the period 1960-2100.

- Ozone in the lower troposphere and near the surface decreases under the projected decline in ozone precursors emissions ~~during MAM and JJA at the NH, and during DJF at the SH, as photochemical ozone production is more dominant~~

~~during these seasons~~and the effect of increased water vapour content. In the middle and upper troposphere the projected strengthening of ozone STT ~~results in a distinct~~ contributes to the increase of ozone globally~~, that seems to cancel out the aforementioned ozone decrease due to emissions reduction~~.

In summary, the findings of this study are in the same direction with other studies based in different CCMs (Zeng et al., 2010; Banerjee et al., 2016; Meul et al., 2018), increasing confidence in the direction of an increased ozone STT and induced increases in middle/upper tropospheric ozone in the future under the RCP6.0 emissions scenario. The role of tropopause folds activity in a changing climate seems to be a considerable factor for both the levels and variability of ozone STT.

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Competing interests. The authors declare that they have no conflict of interest.

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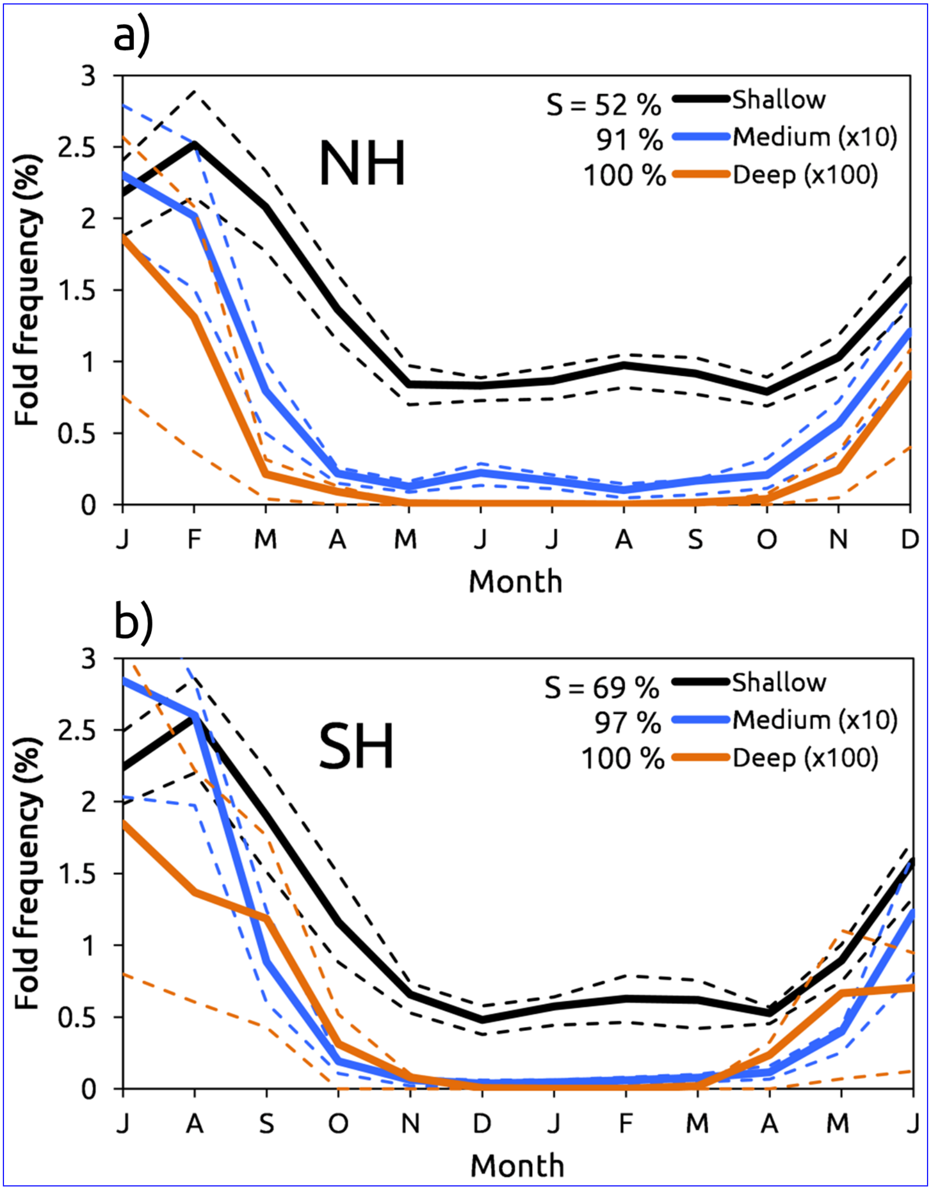

Figure 1. Seasonal cycle of tropopause fold frequencies (%) for (a) the NH (0-65°N) and (b) the SH (0-65°S) over the period 1979-2012 for intercomparison with Figure 7 from Škerlak et al. (2015). The solid lines stand for the mean values, while the dashed coloured lines stand for the 25% and 75% percentiles. The seasonality $S = \frac{max-min}{max+min}$ of each seasonal cycle is also shown.

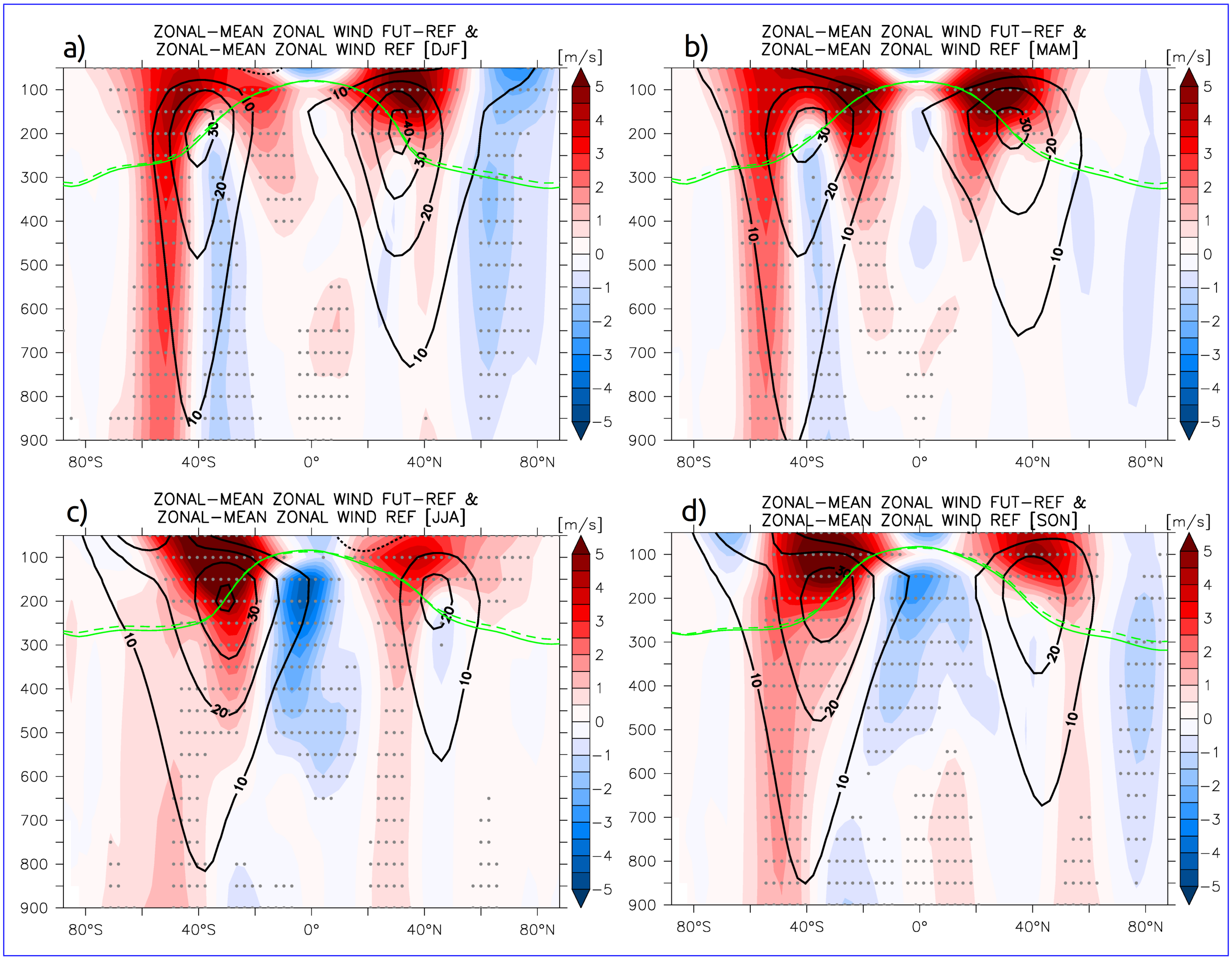

Figure 2. Zonal-mean zonal wind differences (m/s) between the FUT and REF periods for DJF (a), MAM (b), JJA (c) and SON (d). The black contours indicate the zonal-mean zonal wind climatology (m/s) for the REF period. The green solid/dashed line denotes the height of the tropopause during the REF/FUT period. Grey dots denote statistically significant changes at the 99% confidence level.

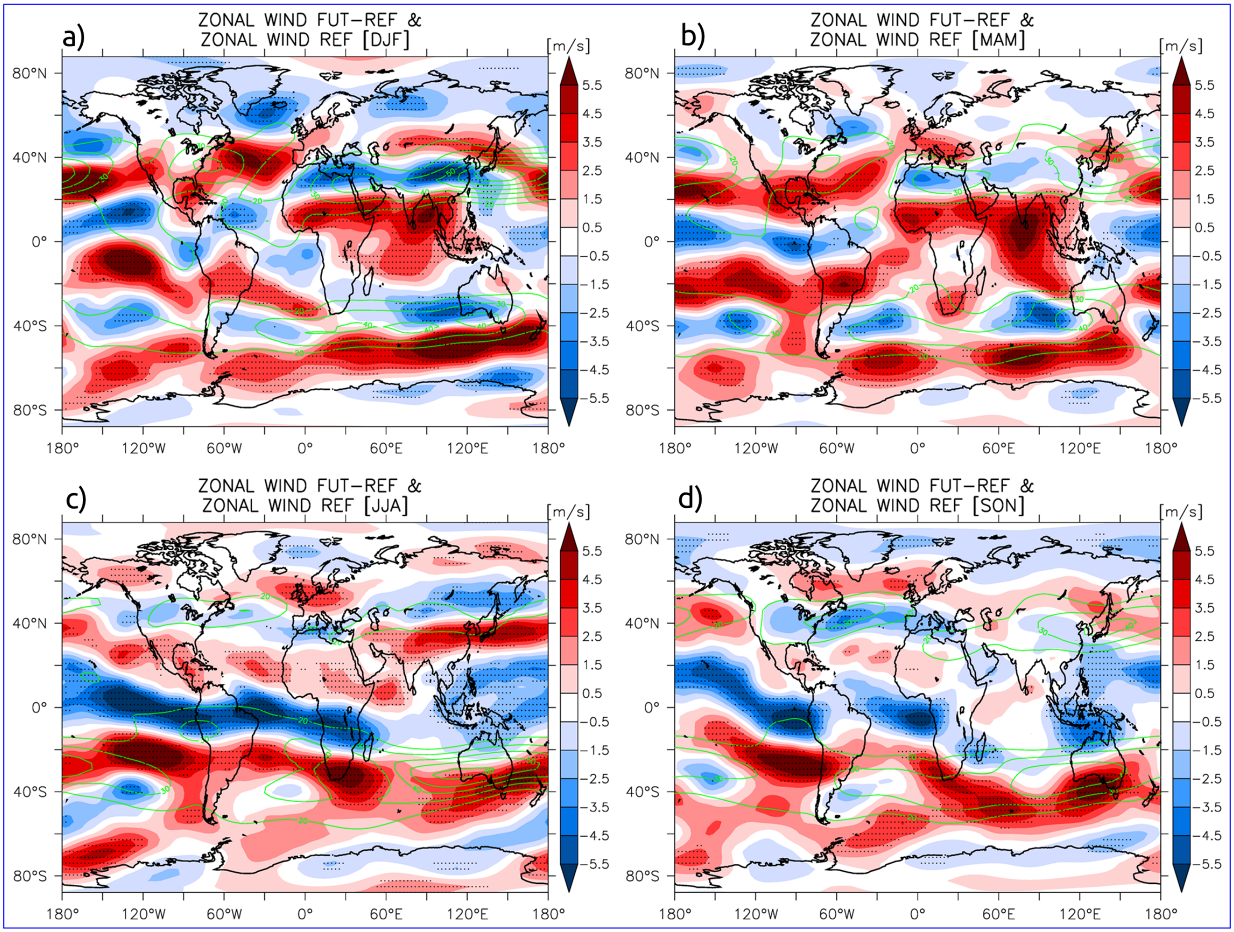

Figure 3. Mean zonal wind differences at 250 hPa (shaded; ~~m/s~~m/s) ~~during~~ between the FUT and REF ~~period~~ periods for DJF (a), MAM (b), JJA (c) and SON (d). The ~~red~~ green contours represent the mean zonal wind ~~differences between~~ at 250 hPa (m/s) during the ~~FUT and~~ REF ~~periods~~period. The regions where the changes are statistically significant at the 99% confidence level are hatched with black dots.

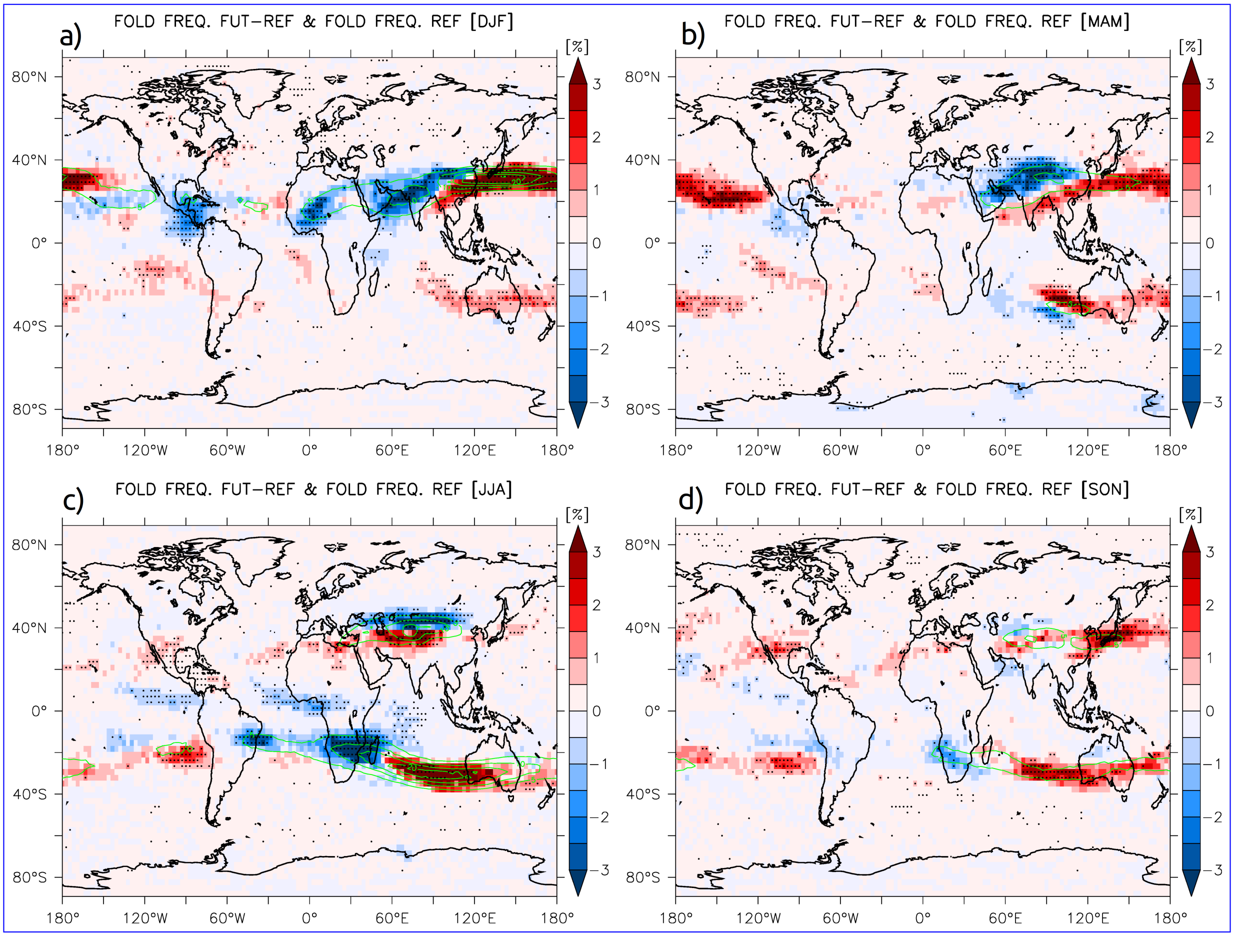

Figure 4. Mean tropopause folds frequency differences (shaded; %) between the FUT and REF ~~period~~ periods for DJF (a), MAM (b), JJA (c) and SON (d). The ~~black~~ green contours denote the tropopause folds frequency (%) during the REF period. The regions where the changes are statistically significant at the 99% confidence level are hatched with black ~~circles~~dots.

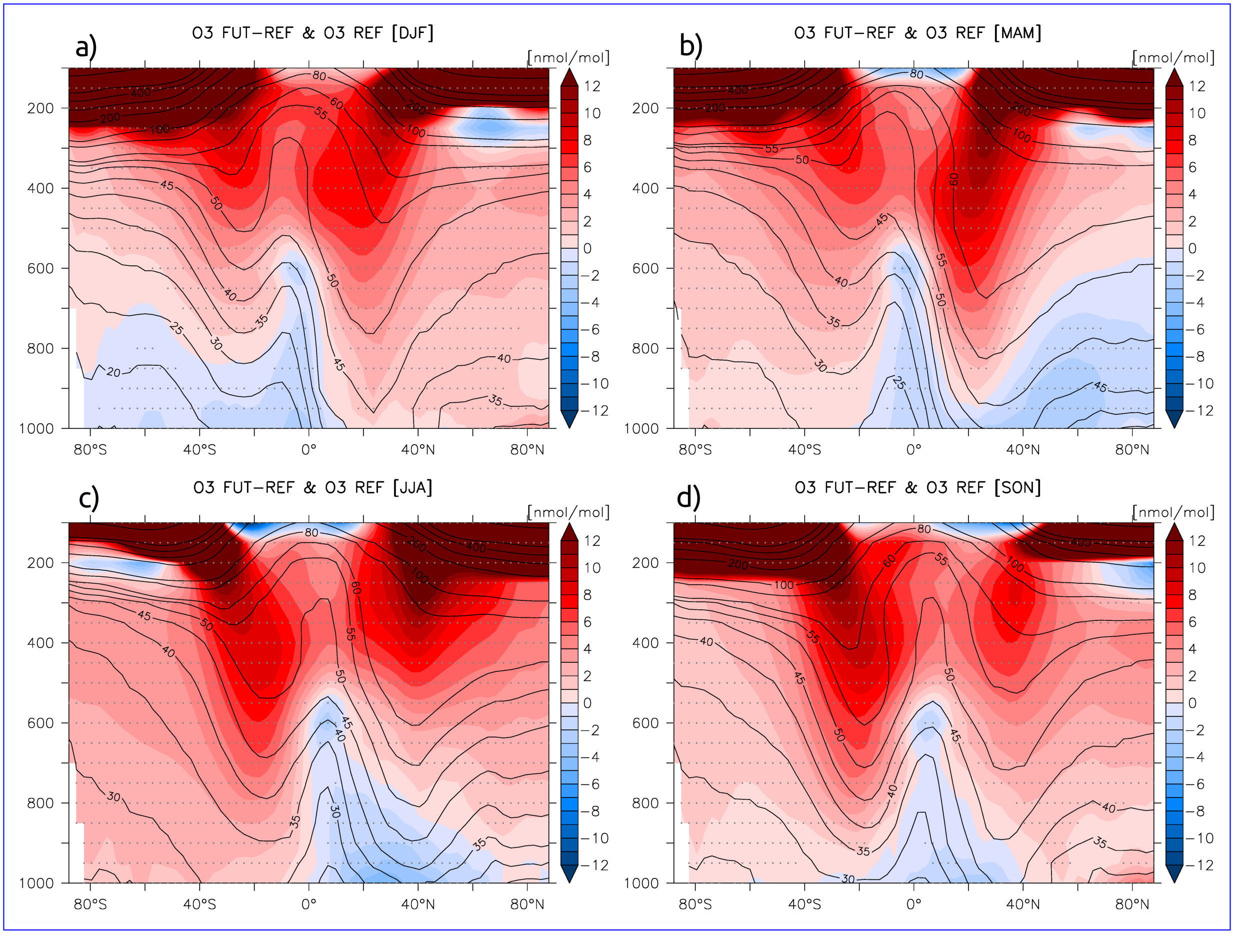

Figure 5. Differences of zonal-mean ozone concentrations between the FUT and REF periods (shaded; $\mathrm{nmol/mol}$) for DJF (a), MAM (b), JJA (c) and SON (d). The black contours denote the zonal-mean ozone concentrations ($\mathrm{nmol/mol}$) during the REF period. Grey dots denote statistically significant changes at the 99% confidence level.

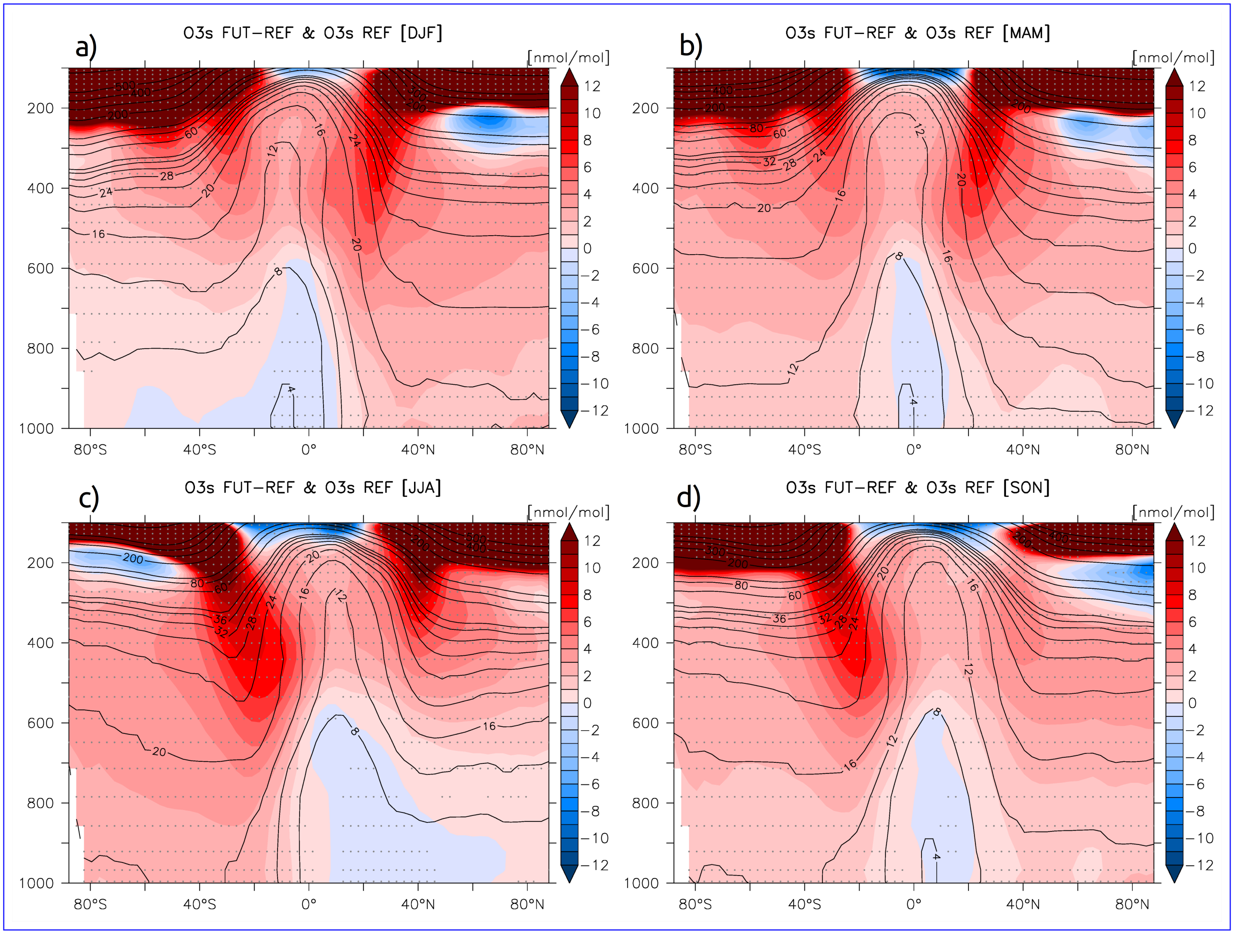

Figure 6. Differences of zonal-mean stratospheric ozone tracer (O3s) concentrations between the FUT and REF periods (shaded; nmol/mol) for DJF (a), MAM (b), JJA (c) and SON (d). The black contours denote the zonal-mean stratospheric ozone tracer concentrations (nmol/mol) during the REF period. Grey dots denote statistically significant changes at the 99% confidence level.

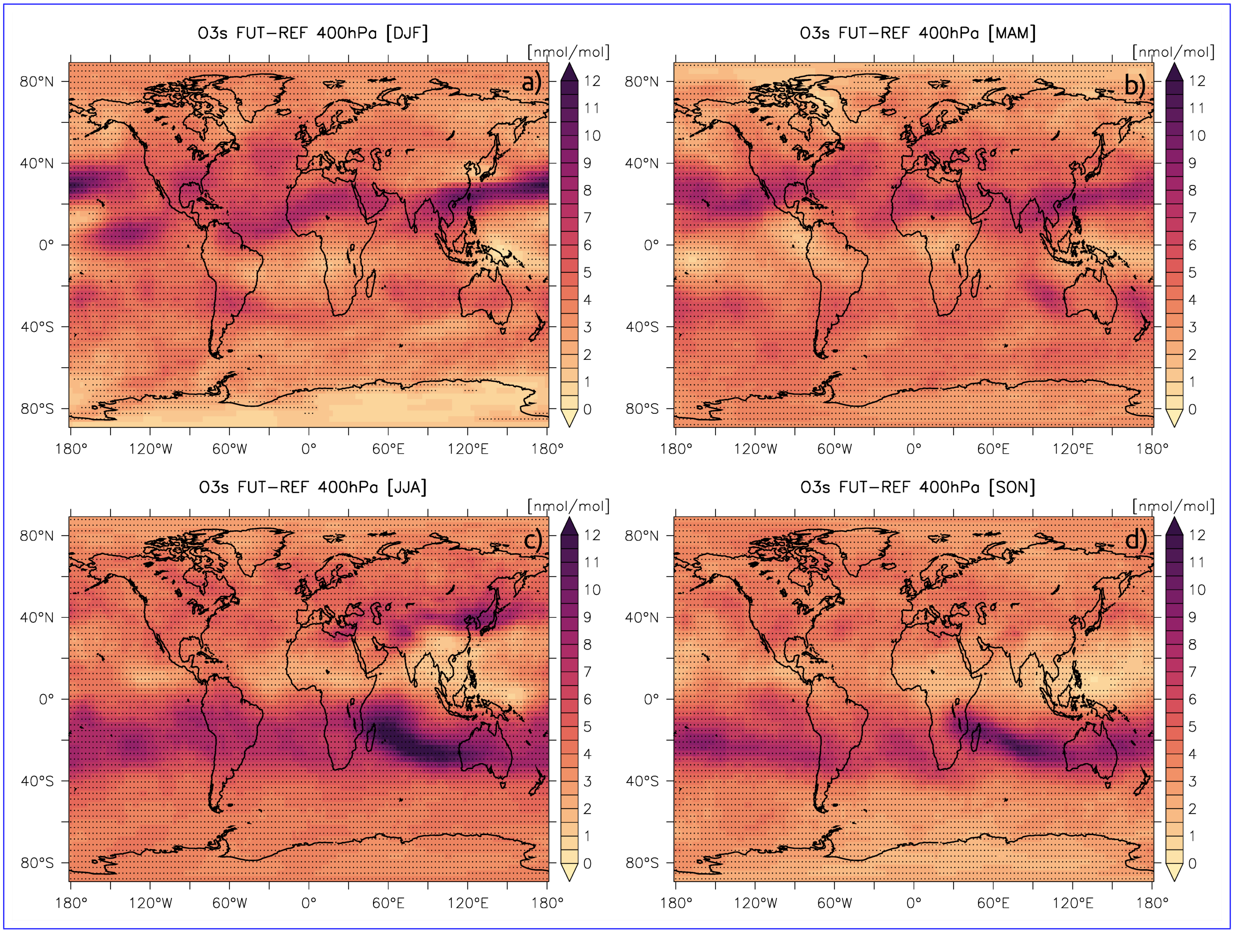

Figure 7. Mean stratospheric ozone tracer (O3s) ~~conenentrations~~ concentrations differences (shaded; nmol/mol) between the FUT and REF periods at 400 hPa for DJF (a), MAM (b), JJA (c) and SON (d). Black dots denote statistically significant differences at the 99% significance level.

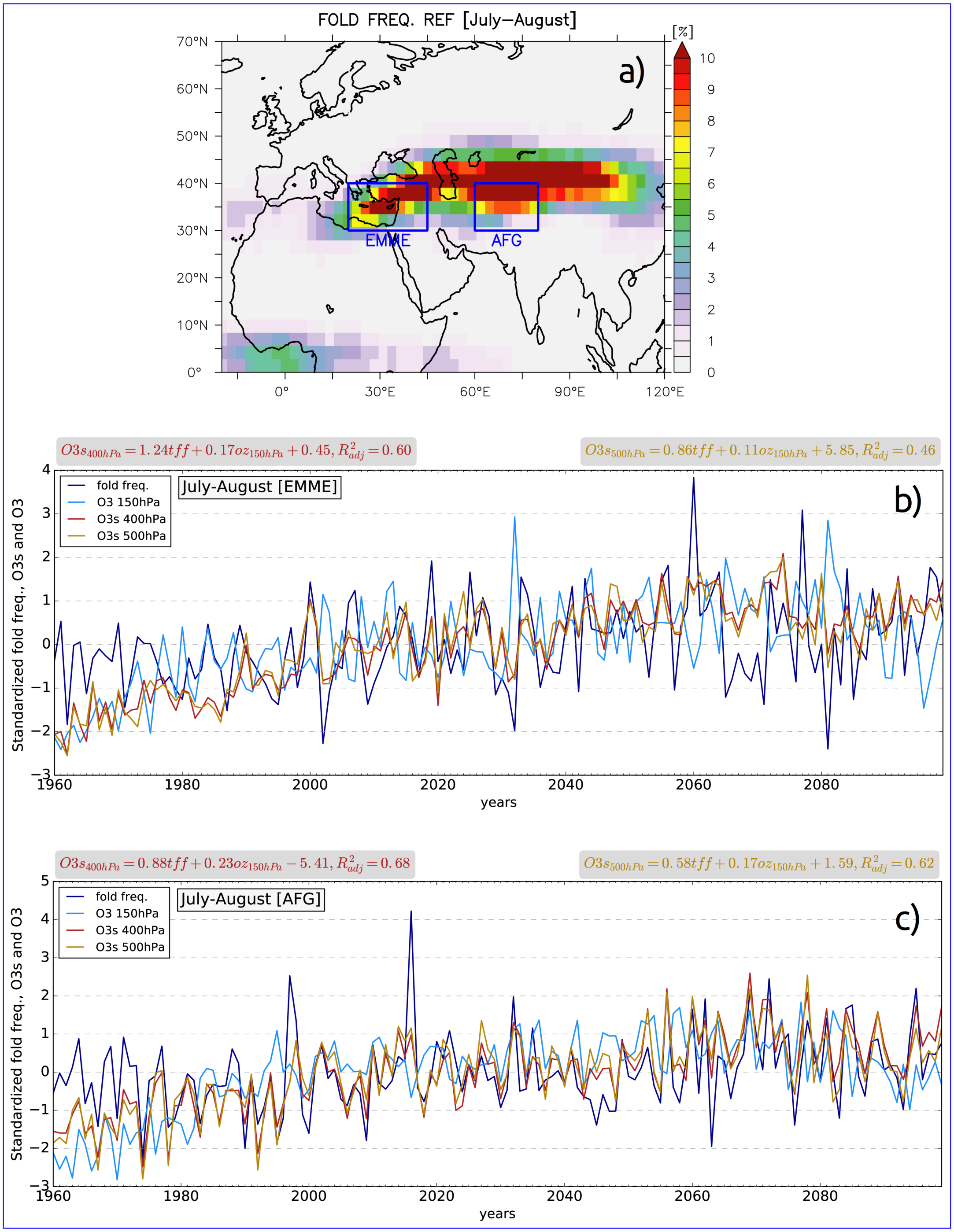

Figure 8. Mean July-August tropopause fold frequency (%) during the REF period and the examined EMME (20-45°E,30-40°N) and AFG (60-80°E, 30-40°N) regions (a). Timeseries of standardized mean July-August tropopause folds frequency (dark blue line), O3 at 150 hPa (light blue line), O3s at 400 hPa (dark red line) and O3s at 500 hPa (orange line) over ~~EM~~ EMME (b) and AFG (c) for the period 1960-2100. Regression equations for O3s at 400 and 500 hPa are also shown at the top of the charts with dark red and orange colours respectively.