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

Anonymous Referee #2

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

The authors have used the ECHAM5/MESSy Atmospheric Chemistry model (EMAC) to quantify the influence of ozone transported from the stratosphere (through tropopause folds) on tropospheric ozone abundances over the eastern Mediterranean and the Middle East. This is a region that is a crossroads for transport of pollution from Europe, Asia, and North America and where there is persistent high summertime abundances of tropospheric ozone. In recent years there has been an increasing number of studies focusing on ozone in this region. This manuscript is a valuable addition to this growing body of literature. It helps establish the importance of tropopause folding events as a key mechanism driving the summertime buildup of ozone over the Mediterranean and the Middle East. I would therefore recommend the manuscript for publication after the authors have addressed my comments below.

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

Main Comments

1) The model resolution is 2.8 x 2.8 degrees, which is coarse compared to studies such as Lin et al. (JGR, doi:10.1029/2012JD018151, 2012), who used a model with a resolution of 0.5 x 0.5 degrees to study stratospheric intrusions. Although the EMAC model is nudged toward ECMWF ERA-Interim data, it would be helpful to see how the model compares to the ERA-Interim fields. On page 5, lines 2-4, it states that "a more extensive comparison … suggest that both spatial and temporal characteristics of tropopause fold frequencies are well captured by the EMAC modeling system (not shown)." However, given the coarse model resolution used here, I believe that it is important to show the results of these comparisons to establish the fidelity of the model in capturing tropopause folds.

We agree with the Reviewer. As the model horizontal resolution was also mentioned by Reviewer #1, we have included a new paragraph in the revised manuscript (Page 3, Lines 20-28), where we further discuss the benefits of a finer horizontal resolution. Following the Reviewer's suggestion, we have included as Supplementary Material (Figure S1) the EMAC monthly mean climatology of shallow ($50 \le \Delta p < 200$ hPa), medium ($200 \le \Delta p < 350$ hPa) and deep ($\Delta p \ge 350$

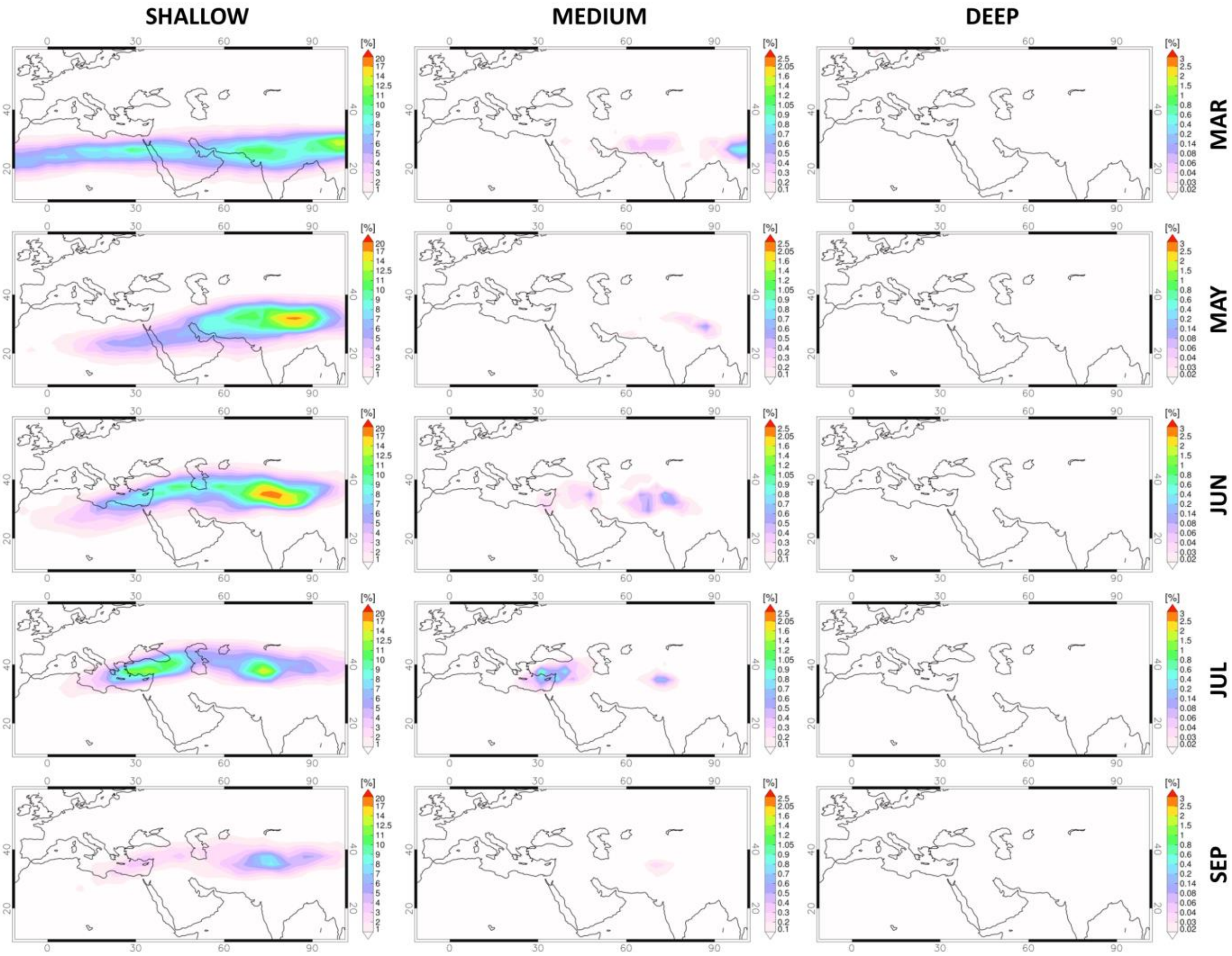

Figure S1. Monthly mean climatology (1979-2012) of shallow (50 ≤ Δp < 200 hPa), medium (200 ≤ Δp < 350 hPa) and deep (Δp ≥ 350 hPa) fold frequency during March, May, June, July and September, for intercomparison with Fig. 2 of Tyrlis et al. (2014).

hPa) fold frequency during March, May, June, July and September, for a straightforward intercomparison with the results of the study by Tyrlis et al. (2014). The selection of the months by Tyrlis et al. (2014) is based on the seasonal evolution of both tropopause folds and south Asian Monsoon activities. The results of this comparison are further discussed in the revised manuscript and the respective paragraph has been modified as follows (Page 5, Lines 15-26): *"To evaluate the ability of the EMAC model to capture tropopause fold activity, we compare results with the findings of Tyrlis et al. (2014), based on the ERA-Interim reanalysis data. The monthly mean climatology (1979-2012) of shallow (50 ≤ Δp < 200 hPa), medium (200 ≤ Δp < 350 hPa) and deep (Δp ≥ 350 hPa) fold frequency during several months is depicted in Figure S1 (Supplementary Material), for intercomparison with Figure 2 of Tyrlis et al. (2014). Both temporal and spatial patterns of EMAC-simulated shallow (more frequent) and medium fold frequencies are found to be in good agreement with the ERA-Interim reanalysis data. The very* *rare occurence of deep folds in ERA-Interim data (with a peak frequency of about 0.1%) is not reproduced by EMAC, probably due to its coarser horizontal resolution. Figure 2 presents the summer (JJA) climatology of total folding activity calculated from EMAC simulations. A distinct hot spot of tropopause fold activity is found over the EMME region, as a result of the dynamical interaction between the subtropical jet and the Asian monsoon anticyclone (Tyrlis et al., 2014), with maximum values of total fold frequency up to 15% over southern Turkey. The above pattern of summertime fold frequency is in line with the results of recent studies (Tyrlis et al., 2014; Škerlak et al., 2015) based on the ERA-Interim reanalysis data."*.

2) It is unclear how the anomalies that are shown in Figures 5-7 are calculated. More information than what is given in line 3 on page 6 would be helpful and would make it easier to interpret the results that are presented.

We agree with the Reviewer's suggestion to provide more information on how the anomalies are calculated. The following sentence was included in the revised manuscript (Page 6, Lines 21-23): *"The anomalies of O3 and O3s presented hereafter, are calculated as the differences between the average concentrations during fold events (average over 1866 timesteps) and the average concentrations during the remainder summer timesteps (average over 5864 timesteps)."*

Minor comments

1) Page 1, lines 13-16: This is a long sentence. Please try breaking it into two sentences, separately addressing the long-range transport and radiative effects.

Done. We have split the respective sentence into two as follows (Page 1, Lines 13-16): *"Compared to ozone near the surface, ozone in the free troposphere can be transported over greater distances due to its relatively longer lifetime and the higher wind velocities. Moreover, owing to its high radiative forcing efficiency in the upper troposphere ozone concentration changes have proportionally greater impact on climate compared to the lower troposphere (Lacis et al., 1990)."*

2) Page 1, lines 16-18: The sentence starting with "Tropospheric ozone originates…" is long and difficult to read. Also, What about methane? It is not a volatile organic compound, but it is an important precursor of tropospheric ozone.

We agree with the Reviewer that methane is also an important ozone precursor, which reaction with hydroxyl radical initiates a sequence of reactions that result in ozone production. To this end, methane was also included as an ozone precursor in the revised manuscript. Following the Reviewer's suggestion we have modified the respective sentence as follows (Page 1, Lines 16-19): *"The main sources of ozone in the troposphere are (i) photochemical production through a sequence of reactions from its precursors (nitrogen oxide, volatile organic compounds, carbon* *monoxide and methane) (Crutzen, 1974) and, (ii) downward transport from the stratosphere (Danielsen, 1968)."*

3) Page 2, line 2: Li et al. (GRL. Vol. 28, 3235-3238, 2001), which first highlighted the summertime ozone buildup over the Middle East, should be referenced here.
Done. We thank the Reviewer for noticing this out.

4) Page 4, line 6: "Optimal" in what sense? How was it determined that the vertical resolution should be for optimal tropopause fold representation?
What the authors meant is that the high vertical resolution of the model near the tropopause contributes to a more realistic representation of the tropopause fold process. Therefore, we have replaced the word *"optimal"* with the word *"realistic"* (Page 4, Line 19).

5) Page 6, line 5: At what level is this 7 ppb increase found? What is the maximum increase in ozone at 700 hPa? The values at 700 hPa seem to be much less than 7 ppb.
We thank the Reviewer for this comment. Indeed the values at 700 hPa are not up to 7 ppb as inadvertently is stated in the manuscript. The increase of ozone due to fold activity is up to 7 ppb at 400, 500 and 600 hPa, and up to 4 ppb at 700 hPa. The respective discussion has been modified accordingly in the revised manuscript as follows (Page 6, Lines 25-27): *"A distinct positive pattern is found in the middle troposphere (Fig. 5a, b and c) mainly over the EMME region, revealing an increase of ozone up to 7 ppb due to fold activity. An increase of ozone is also clear at 700 hPa (Fig. 5d) with mixing ratios of up to 4 ppb."*

6) Figure 6: What do the negative values in the stratosphere mean in panels c) and d)?
The positive O3s anomalies in the troposphere and the negative O3s anomalies aloft are simply due to mass redistribution in the vertical, taking into consideration the mass balance.

7) Figure 6: The anomalies seem to be descending across the potential temperature surfaces, indicating that the downward transport is not purely isentropic. In this region descent is also associated with radiative cooling. How consistent is the rate of cross isentropic transport with the cooling rates in this region and the timescale for downward transport in these folding events?
Despite the isentropic framework (in fact near isentropic) of stratospheric intrusions that result on the formation of filamentary structures within the troposphere, the lifetime and evolution of these filaments are influenced by non-conservative processes such as radiative cooling and heating, turbulence (e.g. generated by

wind shear, convection, breaking gravity waves and radiation) and molecular diffusion, which cascade the filaments gradually to smaller scales down to the molecular level (Forster and Wirth, 2000; Stohl et al., 2003).

The point raised by the Reviewer for the role of radiative cooling in the evolution of the intruded filamentary structures is an interesting point, but at the same time a complicated issue, which cannot be readily unraveled in our case from Figure 6 which shows a mean situation of a number of different selected cases characterized as fold events. This issue could be more readily addressed through idealized model experiments or specific case studies, where we could isolate the effect of radiative cooling from other important diabatic processes, such as turbulent mixing. For example, Forster and Wirth (2000) addressed the issue of radiative decay of stratospheric filaments in the troposphere in idealized model experiments for different filaments (thick or thin) and assuming solely a PV anomaly or a PV anomaly associated with an anomaly in ozone and water vapour, without taking into account turbulent mixing.

A quantitative answer on Reviewer's question is beyond the scope of this manuscript, but we address this point and the relevant limitations within the manuscript as follows (Page 3, Lines 8-12): *"Subsequent to transport into the troposphere, air with stratospheric origin is quasi-adiabatically stirred by large-scale cyclonic and anticyclonic disturbances, which may lead to the formation of elongated streamers or isolated coherent structures. These can further dissipate and cascade down to smaller scales by non-conservative processes (such as radiative cooling/heating and turbulence), thus leading to irreversible mixing with the surrounding air (Shapiro, 1980; Appenzeller and Davies,1992; Forster and Wirth, 2000)."*

All the extra references used are added in the revised manuscript.

Please also note a few minor changes in the manuscript:

- Page 2, Line 22 *"temperature increase may raise"* is replaced with *"climate warming may intensify"*.
- Page 3, Line 2 *"Specifically,"* is removed.
- Page 3, Line 31 *"which is explored"* is replaced with *"explored here"*.
- Page 4, Line 20 *"enough"* is replaced with *"sufficient"*.
- Page 4 Lines 24-25 the phrase *", however, since it is initialized above 100 hPa, only a very small fraction is recirculated by multiple crossings of the tropopause"* is added.
- Page 10, Line 5 *"modeling system"* is replaced with *"model"*.

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