Response to Reviewers comments on "Secondary ozone peaks in the troposphere over the Himalayas"

Anonymous Referee #2

GENERAL COMMENTS: The authors use soundings from an Indian station (Nainital) sampled over the course of one year to identify 'secondary ozone peaks' (SOP). According to the authors 3-4 profiles are available per month. Six profiles are presented showing an SOP. A comparison with the EMAC model at T42L90 is used to extend the limited data set over a time period of 15 years (2000-2014) to assess the impact of such events on the ozone column over the Himalayan region. During the monsoon season they find virtually no SOPs over the region of interest. According to the authors such SOPs contribute 7-9 DU of ozone to the tropospheric ozone columns during SOP occurrence. They also show, that the SOPs are only a minor effect and do not significantly enhance the ozone column over the whole year. The quantification of ozone transport across the tropopause is important and as such this study could in principle add to this. Overall the paper is well written and the graphics are clear and appropriate. However, the paper needs some major clarifications: I missed clear descriptions of terms and definitions given the central topic SOP: How do the authors define an SOP? They only provide a definition for the model analysis later in the manuscript, but does this also work for the soundings, which have a much higher resolution? How do they distinguish an SOP from a tropopause fold or do they imply folds as SOPs? This is not clearly stated at all also in the introduction. Directly linked to this they don't discuss the transience or irreversibility of the phenomena, which are however crucial for the irreversibility of ozone flux and the persistence of the effect. I also missed a careful analysis of the transport and mixing process, as stated in the abstract. The authors should and could provide this, but currently they show coincident fields, but not a process. Given these points there I recommend the paper for publication after the following points have been addressed.

Response: We thank the reviewer for the careful evaluation of the manuscript and his/her constructive comments and suggestions. The paper has been revised as discussed in responses to the individual comments.

Major points:

Comment 1: In section 2 the authors should provide a clear definition for SOPs, which have been applied to the soundings. Further: What is the vertical resolution of the soundings and which role does the resolution of the sounding play for definition and the final column ozone estimate? The authors also do not discuss the effect of the limited vertical and horizontal resolution of the model. How many layers do they miss compared to high resolution sonde profile and how would this affect the number of peaks and the ozone column?

Response: SOPs are basically a significant enhancement in ozone mixing ratios as compared to the lower troposphere (by at least 50%). Additionally these are not a direct downward transport and ozone levels above SOPs are again lower (here we considered at least 20%). The defining conditions for SOPs are added in the Section 2 (Page: 5, Lines:148-150).

Sounding data was reported originally at 100 m vertical resolution. As mentioned by the reviewer, the identification of SOP (and their effects) could be affected by the model vertical resolution, if this would be coarser than the vertical extent of SOPs (10-12 km: about 2km). As the EMAC simulations were conducted at a vertical resolution of 0.5 km (i.e. four times finer than the typical SOP extend) by using 90 vertical levels, we are able to reproduce all the six events. It must be stressed that also in the observations SOP were observed both in the high-resolution 100 m sounding data (Ojha et al., 2014) and in the 500-m resolution data (equivalent to model vertical resolution at the tropopause) used for the model evaluation (see Fig. 2).

Moreover, the EMAC modeling system (with the exact same horizontal and vertical resolution) was found capable to reproduce the observed (ERA-Interim) spatiotemporal features of tropopause fold occurrences (Akritidis et al., 2016), indicating that the current model resolution is sufficient for resolving similar processes near the tropopause region.

Comment 2: The authors should pay more attention to the reversibility of the SOPs. As long as the SOPs keep their high PV values as indicated in Figures 2-4, the ozone peaks will not permanently contribute to the tropospheric ozone budget, since they do not mix as shown in Fig.4 by the O3s. Figures 7-9 show O3s structures in the troposphere which are collocated to the tropopause (i.e. PV structure). The authors could e.g. diagnose the evolution of O3S on an isentropic surface relative to the evolution of PV to diagnose a persistent effect of the SOPs on tropospheric ozone. Maybe an additional plot of wind gradients or Richardson number would give some further indication for the process.
Response: The calculations of tropospheric ozone budget are revised by implementing the PV criteria suggested by the reviewer as described in the response to reviewer 2's major comment 4.
Moreover, to investigate the mixing of the transported stratospheric air with tropospheric air in the vicinity of SOPs, we present a turbulence-index (TI) (new Fig. 10), as described in Ellrod and Knapp (1992), to detect Clear Air Turbulence (CAT) areas and potential mixing, similar to the approach followed by Traub and Lelieveld (2003).
The enhanced TI values during the SOPs above Nainital indicate higher probability of mixing between stratospheric and tropospheric air, supporting the irreversible nature of the associated STT (Page: 9,10; Lines: 307-314).

Comment 3: I suggest to calculate a statistical amount of trajectories in the model and to evaluate the evolution of O3s, O3 and PV along the trajectories? I can't see, how the current Lagrangian analysis provides a robust view on any exchange on the basis of one trajectory per case and I suggest to remove Fig.5 and 6. At least the authors could show plots of ozone timeseries along the trajectories in Fig.6. Instead of the current Fig.6 the authors could plot the ratio of O3S/O3 to illustrate the stratospheric entry (with PV as contour to differentiate between transience versus irreversibility). This would much more strengthen the paper. Alternatively the authors could use the ERA Interim data, which drive the EMAC to perform trajectory calculations with a statistical amount of data. This would also much better help to identify the process of ozone transport and mixing into the troposphere by diagnosing PV change.
Response: Statistical amount of trajectories are computed and evolution of O_3s, O_3 and PV is presented (New Figures 6, S7, and S8). Air masses are enriched the ozone of stratospheric origin during transport to Nainital causing SOPs. A significant fraction of trajectories during non SOP timesteps originates over the south west having lower O_3s (< 90 nmol mol^{-1}). The trajectories which do get higher contributions of stratospheric ozone are found to be diluted during the transport making the enhancements above Nainital too small to be an SOP (Page:8, Lines 264-272).

Further, as suggested in place of previous Fig. 6, we show the ratio O_3s/O_3 to illustrate the transport from stratosphere and its advection towards Nainital (Fig. 7 in revised version). This is discussed in the manuscript as "The O_3s/O_3 ratio is mostly found to be close to unity (≥0.9) near the altitude (pressure) of air mass trajectory during transport, except on 7th Jun and 25th Oct (0.5–0.8). The intrusions enriching tropospheric air masses with stratospheric O_3 are clearly visible. More specifically, a significant stratospheric contribution to tropospheric ozone is found in the upper/middle troposphere during the 5-day period before the event, with the associated PV values (< 2 PVU) indicating mixing of stratospheric air into the troposphere" (Page: 9; Lines: 277-282).

Comment 4: For the estimate of the effect of the SOPs on the tropospheric ozone column the authors should extend their analysis. As long as they don't account for the PV change, their results are not related to the tropospheric ozone budget. I suggest to compare in addition to Fig. 10 O3 and O3s for PV < 2 only for periods with and without SOPs. This would give the ozone which stays in the troposphere and leads to an enhancement during periods of SOPs, which would strengthen the importance of the results.
Response: Budget calculation is revised by accounting for PV change (Fig. 12 in the revised manuscript). The effect on tropospheric column ozone is found to be slightly lower (3.3-7.5DU; up to 21%) when PV criteria is applied, as compared to when PV criteria is relaxed and timesteps with PV higher than 2 are also included (4-9 DU; up to 26%) (Page: 12, Lines: 383-389).

Minor comments: l.53: If SOPs occur in the lower stratosphere, how are these defined? They can't be the result of the same mechanism as tropospheric SOPs, are they comparable?
Response: Here we focused on the SOPs in the troposphere. We find that stratosphere troposphere exchange is the main source of SOPs in the troposphere. Differential advection of ozone poor and ozone rich air could lead to secondary ozone peaks in the stratosphere (Lemoine, 2004).

l.100: Whats the output frequency of the model?
Response: 10 hours. Mentioned in the revised version (Page:4, Line: 112). Detailed description of the simulation can be found in Joeckel et al. (2016).

l.117: "Tropopause folds are identified..." : How do the results compare to Sprenger et al,2003 or Skerlak, 2014 (over the Himalayas)?
Response: The mean seasonal (DJF, MAM, JJA, SON) climatology of shallow (50≤Δp<200 hPa), medium (200≤Δp<350 hPa) and deep (Δp≥350 hPa) tropopause fold frequencies (%) over the period 2000-2014 are presented (Fig. S5), for intercomparison with the studies of Sprenger et al. (2003) and Škerlak et al. (2015).
The EMAC-simulated fold occurrences are generally in agreement with the findings of the aforementioned studies both spatially and temporally, especially for shallow folds which constitute the majority of folds. Moreover, in agreement with Škerlak et al. (2015), the fold maxima over the Himalayas are found during MAM and DJF, while the minimum fold frequencies are found during JJA.

l.146: Why don't you use a larger number of trajectories and perform a robust analysis?
Response: Trajectories at every time step are computed for May 2002 (the month having highest SOP frequency). Evolution of O_3s, O_3 and PV along the trajectories are analyzed for SOP and No SOP time steps (New Fig. 6, S7, S8). Also see response to your comment 3.

Comment: l.155,156: Why is the model interpolated and not simply evaluated at the model levels, which would avoid interpolation errors particularly in the vertical? Is the output interpolated in time?
Response: Interpolation errors are minimal as model's vertical resolution is also ~500 m on which we are taking the observational profiles for comparison. A time weighted mean of the model profiles have been obtained by weighing higher the profile which is closer in time of the observation (also see Ojha et al., 2016). We suggest (and verified) that this procedure would better include the temporal evolution, as compared to directly taking the profile closest in time.

l.167-172: How do the relative ozone enhancements of compare to the observations instead of the absolute values?

Response: The relative ozone enhancements are also reproduced, in general, with in the variabilities (see Table 1).

l.285-287: 285-287: Clarify: What is meant with" PV structures and subtropical jetstreams"? Do you mean tropopause folds below the jet?

Response: We have modified the phrase in the revised manuscript as follows:"PV structures, induced by fluctuations of the zonal flow and tropopause folds development along the subtropical jet-stream".

References

Akritidis, D., Pozzer, A., Zanis, P., Tyrlis, E., Škerlak, B., Sprenger, M., and Lelieveld, J.: On the role of tropopause folds in summertime tropospheric ozone over the eastern Mediterranean and the Middle East, Atmos. Chem. Phys., 16, 14025-14039, doi:10.5194/acp-16-14025-2016, 2016.

Ellrod, G., and D. Knapp, An objective clear-air turbulence forecasting technique: Verification and operational use, Weather Forecast., 7, 150–165, 1992.

Lemoine, R.: Secondary maxima in ozone profiles, Atmos. Chem. Phys., 4, 1085-1096, doi:10.5194/acp-4-1085-2004, 2004.

Ojha, N., M. Naja, T. Sarangi, R. Kumar, P. Bhardwaj, S. Lal, S. Venkataramani, R. Sagar, A. Kumar, H. C. Chandola: On the processes influencing the vertical distribution of ozone over the central Himalayas: Analysis of yearlong ozonesonde observations, *Atmos. Environ.*, 88, 201-211, ISSN 1352-2310, doi:10.1016/j.atmosenv.2014.01.031, 2014.

Ojha, N., Pozzer, A., Rauthe-Schöch, A., Baker, A. K., Yoon, J., Brenninkmeijer, C. A. M., and Lelieveld, J.: Ozone and carbon monoxide over India during the summer monsoon: regional emissions and transport, Atmos. Chem. Phys., 16, 3013-3032, doi:10.5194/acp-16-3013-2016, 2016.

Sprenger, M., M. Croci Maspoli, and H. Wernli, Tropopause folds and cross-tropopause exchange: A global investigation based upon ECMWF analyses for the time period March 2000 to February 2001, J. Geophys. Res., 108, 8518, doi:10.1029/2002JD002587, D12, 2003.

Škerlak, B., M. Sprenger, S. Pfahl, E. Tyrlis, and H. Wernli, Tropopause folds in ERA- Interim: Global climatology and relation to extreme weather events, J. Geophys. Res. Atmos., 120, 4860–4877, doi:10.1002/2014JD022787, 2015.

Traub, M., and J. Lelieveld, Cross-tropopause transport over the eastern Mediterranean, J. Geophys. Res., 108(D23), 4712, doi:10.1029/2003JD003754, 2003.