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

Anonymous Reviewer #1

GENERAL COMMENTS: This paper by Ojha et al. presents an interesting study about the occurrence of secondary ozone peaks (SOPs) in the troposphere over the Himalaya/Indian region. The work is mainly based on the combined use of a limited set of vertical ozone sounding available at a single measurement site (Nainital in the Himalayan region) and on the outputs from the EMAC model, with the purposes of elucidating the processes leading to the occurrence of SOPs, characterizing their temporal variability and assessing their contribution to tropospheric ozone. The measurement data at Nainital were also used to evaluate the capacity of EMAC in reproducing SOPs. The paper is well within the scopes of ACP and, potentially, it can seriously help in better clarifying this specific (but rather frequent) tropospheric phenomenon and its implication on tropospheric O3 budget over the region. However, I think that the authors should better take advantage of the long-term (2000 – 2014) EMAC data-set to provide a more robust characterization of this class of events both in terms of their origin, dynamical features and impact on long-term O3 variability. Thus, I recommend publication, but after that some major efforts will be implemented towards this direction. I'm rather confident that the authors can implement the requested changes in a reasonable small amount of time.

Response: We thank the reviewer for the careful evaluation of the manuscript and his/her constructive comments and suggestions. The paper has been revised and now includes more analysis of long-term model simulations, as discussed in responses to the individual comments.

SPECIFIC COMMENTS

Comment 1: In the introduction you should better describe the paramount importance of clarifying processes affecting tropospheric ozone variability in the Southern Asia and Himalayas, two global hot spot for climate, atmospheric composition and anthropogenic pressures (see e.g. http://www.unep.org/pdf/ABCSummaryFinal.pdf).

Response: The suggested information is mentioned in the revised version (Page:3, Lines:86-95) as "Additionally, model simulations are required to both trace the source regions and quantify the effect of SOPs on the tropospheric ozone budget. Such investigations are of key importance as the Indo-Gangetic Plain (IGP) and Himalaya region are global hotspot regions in terms of anthropogenic pressures that could impose threats to Asia's water and food security (Ramanathan et al., ABC report, 2008). Satellite-based studies corroborate the high pollution loading over northern India and the nearby IGP including the Tropospheric Column Ozone (TCO) over South Asia (Fishman et al., 2003).The IGP is a regional hotspot of the so called "Atmospheric Brown Clouds (ABC)", consisting of brown haze formed by sub-micron size aerosol particles, emitted from a wide range of anthropogenic and natural sources. It has been shown that ABC reduce the amount of sunlight reaching the Earth's surface by as much as 10 to 15 %, and enhance atmospheric solar heating by as much as 50% (Ramanathan et al., 2007)."

Comment 2: The verification of EMAC model capacity in reproducing SOP is based on a limited number of vertical soundings (only 6). The comparison provided by Figure 2 is encouraging about the ability of EMAC (despite the relatively coarse horizontal resolution 2.8 x 2.8 deg) in reproducing the SOPs. However, you should mention that this very limited amount of data prevent a systematic assessment. Did you try to inspect soundings at other locations in the same region (e.g. New Delhi, see http://woudc.org/data/explore.php) to make the data-set for verification larger? Can you provide some references of earlier works showing comparison of EMAC vertical ozone profiles with measurements (maybe Jöckel et al, 2016 can be profitably cited)?

Response: The reference to Jöckel et al, 2016 is added for comparison of EMAC ozone fields with aircraft-based measurements from the IAGOS-CARIBIC program (Page: 7, Lines:206). It is shown that the simulation used in this work (RC1SD-base-10a) overestimates ozone concentrations, although mostly in the lower troposphere, while in the tropopause regions a reasonable agreement is obtained, compared to satellite and aircraft observations: "The seasonal cycle is reproduced, but the lower values in the troposphere are generally overestimated by up to 40% by the model. In the stratosphere, differences are smaller, as the model underestimates measurements by 5%, reaching 30% only in summer (Jöckel et al, 2016). Comparison with ozonesondes launched in Delhi has been added in the supplement (Fig. S2), which also shows an overestimation in the lower troposphere. Ozone mixing ratios in the middle troposphere show good agreement with the observations (Page: 7, Lines:204-205).

Comment 3: Pag 6, line 169: I would like to see the bias expressed as %. This would better help in understanding the deviation of the model from the measurements.
Response: Suggestion is incorporated.

Comment 4: Pag 6 line 176: "However, these. . .for completeness". I cannot understand this sentence. Do you mean that the selection by Ojha et al. (2014) is not accurate? Please, rephrase!
Response: No we only meant that the events were identified visually in that paper, while with availability of long-term data in this paper we make a criteria to calculate their frequency. The sentence is rephrased.

Comment 5: It is a pity that the "core" Section "3.2 Origin of SOPs" is discussing only the results from the six selected profiles at Nainital! I strongly encourage the authors to use the 15-year EMAC outputs to investigate in a more systematic way and for a long-term perspective this point. Also the back-trajectories investigation can be carried out for the whole 2000-2014 period by using NCEP re-analysis. I would suggest to use the SOP events identified over the period 2000 – 2014 and aggregate them on a seasonal basis to provide indication about the amount of ozone transported from the stratosphere during SOP (by comparing average O3 , O3s and PV vertical profiles).
Response: Section 3.2 is revised to incorporate the reviewer's suggestions by analyzing 15-year EMAC outputs for a long-term perspective.
Average vertical profile of PV during SOPs, derived from a long-term model simulation (2000–2014), shows similar structure, as shown for the individual events. Average PV values during SOPs are found to be significantly higher (e. g. 3.0±1.3 PVU in winter, 1.8±0.5 during summer monsoon) as compared to timesteps without SOP (0.3±0.2 to 1.5±1.3) (Page: 7, Lines: 219-225 and Supplementary Fig. S3, Table S1).

Evolution of O_3s, O_3 and PV along statistical amount of trajectories is presented (New Fig. 6 in manuscript and Fig. S7, S8 in the supplement). 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).

SOP events identified over the period 2000 – 2014 are aggregated on a seasonal basis and average profiles of O3 , O3s and PV vertical profiles are presented (New Fig. 5, Fig. S3 and Table S1). The amount of ozone transported from the stratosphere during SOPs is also indicated. The average amount of ozone transported from the stratosphere to the SOPs is estimated to be the highest during spring (162.5±40 nmol mol^{-1}), followed by winter (149.4 ± 35 nmol mol-1). In contrast the contribution of tropospheric photochemical sources to the SOPs is highest during the summer monsoon (30 nmol mol^{-1}) (Page-8, Lines:236-239).

Comment 6: Figure 5. Basing on the Figure caption, the TF locations 5 days before the events are reported in the maps. However, all the back-trajectories showed very fast transport: 5 days before the arrival to Nainital the air-masses were (at least) off of the north Africa western coast-lines. Thus, which is the relationship with the identified TFs? I suppose the authors would say that the TF DURING the air-mass transport were reported. . . Moreover, how long the back-trajectories are? No information are provided along the manuscript. . . Also seasonal composites over the period 2000 – 2014 about the spatial locations of tropopause folding related to SOP events can be presented (see e.g. Figure 4 by Putero et al., 2016 but for tropopause crossing). What about days without SOPs? I guess that no (or fewer) tropopause foldings were crossed by back-trajectories for these cases. . .To provide a "climatological" long-term perspective, you should also consider the possibility to present a composite for Fig. 7 and Fig. 8 as a function of the seasons for the period 2000 – 2014.
Response: Yes, we meant TFs DURING the air-mass transport, now clarified in the caption. This figure has been now moved to the supplement (Fig. S4) (Reviewer 2, comment 3). Length of trajectories (5 days) is now mentioned in the section 2.4 as well as in the figure caption. Seasonal composites of the spatial locations of folds (Fig. S6) shows higher frequency of occurrence during SOPs. The days without SOPs have minimal effects of O_3s transport due to fewer folds along the transport path, and dilution of any effects before reaching the Himalayas.

Comment 7: Pag 7, line 228: "This variability in LRT. . ..in Fig.5". It is not clear to me. Please, explain better this kind of association. . .
Response: We meant to say that the dramatic changes in tropopause pressure along the trajectory (e.g. from 100 to 200 hPa on 11th Feb) could be associated with the tropopause folds. The sentence is suitably revised.

Comment 8: Pag 7, line 232: please define "medium".
Response: The folds having a vertical extent of 200 to 350 hPa are defined as medium folds. This is mentioned in the revised manuscript. Further details are available by Škerlak et al. (2015).

Comment 9: Pag. 8, line 241: please provide in the text longitude boundaries for these regions.
Response: Suggestion is incorporated.

Comment 10: Pag 8, line 243: despite your statement at pag 8 line 236, basing on that plot, it looks that a STE is actually occurring also for the June event (a tongue of air-mass rich in O3 extended down to 500 hPa southward than 30N)!
Response: We agree. The statement that stratospheric effect is not found on 7th June is removed. As pointed out by the reviewer, text is also revised considering that some effects also reach southward than 30N (Abstract: Page 1, line 9; Results: Page 9, Lines:303-304; Conclusions: Page: 12, Lines 411-412).

Comment 11: Section 3.4 The authors must provide some information about the long-term SOP trend over the region of interest: this information is very valuable also taking into account the current debate about the occurrence and attribution of tropospheric ozone trends (see e.g. http://www.igacproject.org/TOAR). Trends in seasonal/yearly frequencies or physical features (e.g. altitude) of SOP and the related O3 contribution are detected? Also the information that no long-term trends were detected is nevertheless valuable.
Response: We evaluated trends in SOP frequencies on seasonal and yearly basis, however, as expected due to their origin from dynamical processes, frequency of SOPs discern strong

inter-annual variation as shown in the new Figure S9 in the Supplement. We discuss this in the revised manuscript (Page:13, Lines: 425-429) and provide relevant references.

Comment 12: Figure 10: I would add the percentage contributions of SOPs to monthly TCO values. What the error bars represent?
Response: The percentage contributions of SOPs to monthly TCO values is added. Error bars represent the standard deviation derived from the temporal variations over the period of 2000-2014. Now mentioned in the figure caption.

Comment 13: Conclusions In general this Section reports very important general statements about SOP but which are mostly based on the analysis of just 6 case studies (see lines 323- 220). I would recommend to try to increase the robustness of these interesting hints by adopting a long-term perspective basing on EMAC simulation.
Response: Conclusions are revised to add the additional results based on long-term model simulations (as mentioned in response to comment 5 also) (Page:12, Lines: 395, 399-401).

Comment 14: Line 335: "The minimum in the. . .mixing". I would also mention the northward displacement of subtropical jet stream during summer monsoon.
Response: Suggestion is incorporated.

Comment 15: Line 339: are you able to provide any indication about the impact of this increase in terms of radiative forcing over the region?
Response: Radiative forcing is not explicitly investigated in this paper. To provide an indication, a 4-9 DU higher tropospheric column ozone (due to enhancement at the SOP altitude) would correspond to an increase in surface temperature by 0.07 to 0.16 degree, as we take into account the vertical profile of ozone forcing (Lacis et al., 1990). This is discussed in the revised manuscript (Page: 13, Lines: 422-424).

TECHNICALS
Comment 1: Figure 1. I would skip the typical event plot since it is also reported in Figure 2.
Response: Suggestion is incorporated.

Comment 2: Figure 6: x-axis and y-axis. I suppose the black line is the back-trajectory pressure level: it should be reported in the caption.
Response: Suggestion is incorporated.

Comment 3: Figure 7 (Figure 8): please indicate in the caption the latitude (longitude) value for which the cross section is produced.
Response: Suggestion is incorporated.

Comment 4: Figure 9-10: what the error bars represent?
Response: The error bar in Fig. 9 represents the standard deviation of SOP frequency in each month among different years from 2000-2014 (Page: 10, Lines: 336-337). In Fig. 10 , it shows the standard deviation in the temporal variation of tropospheric ozone column among all the time steps during 2000-2014. This is now added in the figure caption.

References:
Fishman, J., Wozniak, A. E., and Creilson, J. K.: Global distribution of tropospheric ozone from satellite measurements using the empirically corrected tropospheric ozone residual

technique: Identification of the regional aspects of air pollution, Atmos. Chem. Phys., 3, 893-907, doi:10.5194/acp-3-893-2003, 2003.

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Lacis, A. A., D. J. Wuebbles, and J. A. Logan (1990), Radiative forcing of climate by changes in the vertical distribution of ozone, J. Geophys. Res., 95(D7), 9971–9981, doi:10.1029/JD095iD07p09971.

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