

Interactive comment on "Surface ozone at Nam Co (4730 m a.s.l.) in the inland Tibetan Plateau: variation, synthesis comparison and regional representativeness" *by* Xiufeng Yin et al.

Anonymous Referee #2

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GENERAL COMMENTS This work by Yin et al. presents an overview of about 5 years of continuous near-surface ozone observations at the Nam Co station which is located in the central Tibetan Plateau. The scope of the paper is rather ambitious: to characterize the typical variability of near-surface O3 at this measurement site, to compare it with other sites in the Tibetan Plateau (and beyond) and to demonstrate that this site is representative for the whole Tibetan Plateau. The presented data-set is of great interest (and I suggest to share it in the framework of international initiatives like WMO/GAW or TOAR/JOIN). However, the paper is a little bit confusing and for a great part relies too much in other studies, resembling more a "review" than a research paper. Moreover, some important conclusions were based too much on qualitative assertions. As

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an instance, in my opinion, the authors failed in demonstrating that: "The unique geographical characteristics make Nam Co Station more representative of the baseline of surface ozone in the extensive inland of Tibetan Plateau than other existing monitoring sites", as they report in the Summary. More analyses/comparisons are needed to assess this point! My impression is that the authors mixed together several different analyses without a well-defined scientific track. For instance, at least two different model (FLEXPART- WRF and HYSPLIT) were used with the same aim (characterize O3 variability as function of air-mass transport) but without any critical comparison or integration. The fact that O3 is positively correlated with some meteorological parameters is not of great scientific novelty and (the most important point) I suspect that the linear model results were significantly affected/biased by the use of daily average values (at least for ozone). The discussion about the role of STE is simply based on a subjective (mainly visual) analysis of O3 variability with stratospheric "tracers" (not specific analyses or tool have been used). For these reasons, I suggest to resubmit the paper after than some essential modifications have been made. In the following I provide some suggestion to help authors towards this aim.

SPECIFIC COMMENTS Line 43-45: I think that this sentence is meaningless.

Line 55: this is wrong. At NCO-P the highest contributions from STE is in WINTER. This is clearly stated by Cristofanelli et al. ACP (2010) and Putero et al. ACP (2016). The pre-monsoon (spring) O3 peaks was strongly affected by the transport of pollution from the lower troposphere (Himalayas foothills and Indo-Gangetic Plains). See e.g. Putero et al. Atmospheric Pollution (2013); Bonasoni et al., ACP (2010).

Line 69-70: this sentence is too generic. Specify what kind of ozone-related climatic and environmental effect can be assessed and by which methodology.

Line 84: remove the capital letter from "The"

SECTION 2 Line 95: how did you evaluate change in sensitivity? By which frequency the analyser was calibrated? The calibrator 49iPS was calibrated against which refer-

ence instrument?

Section 2.4: Which is the time resolution of the inputs to the MLR Model (hourly, daily)? How did you consider the FLEXPART trajectory cluster in the regression analysis? Why did you normalize the input parameters? Why did you exclude outliers? The last three sentence are rather obscure to me (from line 126). Please, provide a clear step-by-step description of the methodology. By only considering the maximum 8-hour average ozone concentration, you discharge all the information about variability at hourly scale (which is rather important)...and this is the reason why you find out a great role of radiation! At least, this must be clearly stated in the revised manuscript.

Line 100: I think that I would be better and more useful to refer the measurements to the "local time" instead of "Beijing time".

Line 110: please provide more info about the HYSPLIT simulation set-up. Which meteorological gridded data-set has been used to calculate back-trajectories (GFS)? By which time resolution did you calculate back-trajectories (Once a day? Every hour?)? How did you take into account uncertainties due to the complex topography surrounding the Plateau? Also provide more info about the cluster methodology and provide a description of the algorithm. Provide web access indication to the TRAJplot software. I think that both NOAA (for providing GDAS and HYSPLIT) and TRAJPlot developers must be acknowledged in this paper. I guess WRF-FLEXPART is much more accurate in reproducing air-mass origin and transport to Nam CO. However, please provide more technical details about the model set-up. It is not clear to me which is the reason to use HYSPLIT when WRF-FLEXPART is available. Please, explain. Did you compare the results obtained with FLEXPART and HYSPLIT? Line 117: "Six clusters were found...". Does this sentence refer to HYSPLIT or FLEXPART? Not clear

Section 2.5: What model did you use for this analysis (HYSPLIT or WRF-FLEXPART)? Did you consider some altitude/pressure level thresholds of back-trajectory points to allow the PSCF calculation? If not, hardly you can relate the obtained results with

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surface emissions....The W values are a key parameter for the interpretation of the obtained results. How did you define them? Did you perform a sensitivity study by changing the weighting factor?

SECTION 3

Line 158: please attribute the origin of these anomalous events

Line 161: for the period 2006 – 2011 Putero et al (2013) found an average O3 of 48.7 ppb at NCOP, while Cristofanelli et al. (2010) over two year investigation pointed out an average value of 49 ppb. Thus, I would say that average value at Nam Co and NCO-P are comparable. Please correct.

Line 162: different factors influence background O3 levels, i.e. altitudes, latitude, site classification (mountain, coastal, marine). The authors must better address this comparison taking into account all these factors.

Line 166: So, did you consider months with at least a 60% data coverage. Please specify this point rather than indicating the number of hours.

Section 3.3: would remove Fig 3 and leave only Fig 4 (where diurnal variability are also more evident). However, for each hourly average you must add an error bar denoting the 95% confidence level of the mean average value. At this point, a description of typical local wind variability (wind speed and direction) must be added to evaluate possible influence of diurnal wind breeze on O3 variability.

Section 4.1: This analysis of stratospheric intrusion is too raw. I would like to see a more specific investigation (see e.g. Cristofanelli et al., 2010; Putero et al., 2016; Trickl et al., ACP, 2010). The authors only described in a very qualitative and oversimplified way (basically by "visual" inspection) the time series of stratospheric air markers (any statistical analysis or selection methodology is applied). Moreover, the assumption that stratospheric intrusion can be directly related to the daily maximum of ozone is wrong. Due to mixing and dilution processes, stratospheric air-masses are often characterized

by O3 values which are even lower than those due to photochemistry. Moreover, these events are often characterized by short time duration (even lower than 1 day), thus simply comparing time series of stratospheric tracers with a daily time resolution can mask the real influence of STE. The final sentence: "Nam Co was affected by aged stratospheric air originating over the Himalayas rather than being affected by transport from fresh stratospheric air masses directly above Nam Co Station ", it's not clear to me. Quantify "aged".

Section 4.2: I suggest to perform this analysis also on a seasonal basis. Since most of the used predictors are characterized by significant seasonal cycles, this would provide more hints about the role of single factors in driving O3 variability. Figure S4 it's not clear at all. What is the scale reported on the right bar?

Line 210: "impacts from air masses aloft". Be more specific!

Line 213: "why these air-masses are depleted in O3". I suspect simply because they were related to southern air-mass advection during the monsoon. Please provide a description of the seasonal frequency of occurrence of air-mass transport patterns reported by Fig. S4. You stated that: "For the whole measurements period, it seems that transport of surface ozone is not the main influencing factor to the daily surface ozone variations in the multiple linear regression model". I'm not convinced. As showed by other works (see Di Carlo, JGR, 2007). The role of dynamic is important at hourly timescale. By analysing data as daily averages you ruled out by default these contributions! By comparing the time series of O3 observations with the regression model (Fig. 5), it is rather clear than the model was not able to reproduce the spring peak. To my opinion, this is a clear hint toward an important contribution of transport and dynamics.

Section 4.3: If data analyzed are daily averages, the correlation coefficient here provided (R: 0.77) does not describe the "local" (in-situ) role of photochemistry. This must be described by analysing the hourly data-set as you did for wind speed and PBLH. Which is the correlation coefficient between hourly ozone and hourly SWD? As sug-

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gested by Fig.7, the higher correlation with wind speed and PNLH suggest that dynamics is the most important factor explaining diurnal O3 variability. I suggest to apply the linear correlation model both for daily and hourly values and to comment differences in the results.

Line 245: "the background ozone at the site": this is contradictory, the background cannot be local!

SECTION 5. It is not clear why in Figure 8 you reported "normalized O3" for NCOP. Please explain what kind of normalization was applied. At Xianggelila, Ma et a. (2014) reported that at diurnal scale O3 was strongly correlated with wind speed (as occurred also at Nam CO) and that " the transport and deposition will be the key factors influencing the diurnal variations of surface O3 at Xianggelila, a remote and clean site, rather than local photochemical processes". Also at Dangxiong, Lin et al. (2015), suggested that the correlation with high wind speed and O3 during the afternoon pointed out the important role of transport in affecting O3 more than photochemistry. I would bet that the same is true for Nam CO.

Section 5.2: In my opinion the classification of the seasonal ozone regimes I-III is oversimplified (see the nice work by Tarasova e al., 2007, ACP). I suggest the authors to skip this first part (line 243-263) and discuss the O3 variability at the Tibetan sites as a function of the characterization provided by Tarasova et al. 2007. Line 256: please provide adequate references. Line 260: I think that this sentence only refers to summer season. Please, specify. Line 275: The possible impact of NO titration to the appearance of lower ozone levels at the the Tibetan sites should be better assessed/showed. For instance, you can report diurnal variability as a function of different seasons for these sites. NCO-P is not located over the Tibetan Plateau but at the southern ridge of Himalayas. Please correct.

Line 290: Figure 10 is hard to read and clusters look very similar each other's (except than for those related to southerly circulation). What kind of cluster algorithm was

used? It looks that a large part of the information carried by the back-trajectories was missed by this clustering. Nevertheless, in agreement with this analysis, during Spring only a fraction (about 18%) of back-trajectories crossed the Himalayas. This must be clearly stated.

Line 292: Actually, Skerlak et al. (2014) reports a maximum of deep STT over the Tibetan Plateau and not only over Himalayas! In my opinion, your conclusion that O3 is higher at NCO-P due to a larger contribution from stratosphere is wrong. Looking at your Fig. 9, it looks that O3 values at NCO-P and Nam Co were well comparable on March and May. O3 was higher at NCO-P in April, but (as I reported below) the contribution of polluted air-masses in driving O3 variability at NCO-P during this season cannot be neglected!

Line 294: I think that at this point the transport of polluted air-masses from Himalaya foothills and IGP to high Himalayas must be considered (see Bonasoni et al., 2010; Putero et al., 2013; Luthi et al., 2015)! This contributed to the appearance of the premonsoon maximum at NCOP and possibly the cross-Himalaya transport can also affect Tibetan Plateau.

Line 296: which cluster was associated to the northern TP? It is not possible to recognize it from Figure 10 (please increase the fonts used for legend!)

Line 297: I read carefully Skerlak et al (2014) but I was not able to found any reference to the higher stratospheric flux over the northern Plateau in respect to the southern Plateau in autumn. Indeed, looking at their Fig. 6, this not looks to be the case.

Line 301-304: Is this confirmed also by WRF-FLEXPART clustering?

Line 305-312: were these results confirmed by the HYSPLIT clustering? I expect that WRF-FLEXPART could have much more skill than HYSPLIT (based on global meteorological fields with coarse spatial resolution)in analysing spatial "contributions" for elevated O3 values at Nam CO. However, you must attribute the seasonal variability

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of the "contributions" you found by WRF-FLEXPART (by what kind of emissions, precursors are emitted over each identified regions?). Moreover, you should discuss and quantify the uncertainties related with this analysis. Also some details were missed: as an instance, for the seasonal analysis you used as O3 threshold values ,the seasonal averages or the whole period average? What happens if different threshold were applied (e.g. 75th or 90th percentiles of ozone distribution)? Probabilities higher than 1.0 were reported in the legends: I think this is inconsistent...please check!

Section 5.4: This section about representativeness of Nam CO is mostly based on an intuitive/subjective approach and from review of previous works. Even if I'm personally convinced that Nam Co is an interesting background site, the authors must perform much work if their want to unambiguously assess the spatial representativeness of the station. See for instance Henne et al., ACP, 10, 3561–3581, 2010. I do not think that a " consistent diurnal variability of ozone regardless of season" can be used as proof to claim the large spatial representativeness of the station. Moreover, it seems that the authors do not consider STE as part of the "global" background ozone: from my point of view, this is completely wrong. If not specific analyses are accrued out, I strongly recommend to eliminate this section and limit some lines of comment in the summary Section.

Line 332: please quantify the spatial scale of this "long-range" contribution

SUMMARY Line 343: "Nam Co represents a wide background region in the Tibetan Plateau". In my opinion this need more quantification efforts, since this sentence is too generic/qualitative.

Line 349: "Synthesis comparison...". The authors did not convince me about the small impact of STE.

ACKWNOLEDGMENTS You must acknowledge NOAA for providing HYSPLIT model and GFS meteorological files. I suppose that also the TrajPlot developers must be acknowledged! Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-175, 2017.

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