Response to referee comments

We would like to thank the referees and editor for the interest in our work and the helpful comments and suggestions to improve our manuscript. We have carefully considered all comments and the replies are listed below. The changes have been marked in the text using blue color.

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

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 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.

Response: Thanks for the comments. In this revised version, we added more analysis including Multi Linear Regression which was calculated using hourly data as you suggested, Meridional cross-sections over Nam Co Station derived from ERA-Interim data and a tracer for stratospheric ozone incursions which was obtained using the CAMx.

As a result of the reviews, we have refined the analysis of potential vorticity as a tracer for stratospheric air and we have also expanded the regression analysis to include tracers for stratospheric ozone transport using an air quality model. In the ACPD manuscript, we had used Potential Vorticity near the surface (500 hPa) to test for stratospheric incursions. However, this did not lead to a clear signal in the regression analysis. Based on new research, we have now found that if we use PVU at the 350 hPa level we detect an influence on the ozone time series. If we use PVU at 350 hPa above the Himalayas then this signal is even clearer. The description of the regression analysis has been expanded and the results updated accordingly.

An even better match for stratospheric incursions was obtained when we used ERA-Interim ozone concentrations aloft as boundary and initial conditions for the CAMx air quality model. Chemistry was turned off to obtain a passive tracer of stratospheric air at the measurement site. This gave a signal in the regression analysis that is even stronger than the new PVU analysis. The text was expanded and the results updated in the manuscript as follows (lines 124 – 131):

“A tracer for stratospheric ozone incursions at the measurement site was obtained using the CAMx (Comprehensive Air-quality Model with eXtensions) v6.30 model (Ramboll Environ, 2016). The model initial and boundary conditions were obtained from ERA-Interim ozone fields, retaining only concentrations above 80 ppb and higher than 400 hPa. CAMx simulations were performed using the WRF medium and fine domains (domains 2 and 3) in nested mode for the full 4 year time series. In order to serve as a tracer for direct transport, there was no chemistry in the model and ozone was treated as a passive tracer. The resulting time series of the tracer concentration at the measurement site was used as input in the multi-linear regression model. This is similar to the procedure described in de Foy et al. (2014) to estimate the impact of the free troposphere on surface reactive mercury concentrations.".

Fig. 4 and table 2 were added to explain the new analysis. The model suggested that up to 20% of the ozone variability was due to stratospheric incursions. Meridional cross-sections over Nam Co Station (Fig. 5) illustrated the position of downward transport of stratospheric ozone in different seasons.

SPECIFIC COMMENTS

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

Response: Thank you for your comment. Now we removed this sentence.

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).

Response: Thank you for pointing out. We rewrote this sentence as follows (lines 56 - 57):

"At NCO-P and Xianggelila, surface ozone maximum was observed in spring (Cristofanelli et al., 2010; Ma et al., 2014)".

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.

Response: The sentence has been rephrased for specific meaning as follows (lines 68 - 71):

"This study expands the understanding of baseline and variations in the surface ozone concentration and the transport processes that influence tropospheric ozone in the inland Tibetan Plateau. The long-term measurements of surface ozone; together with other reported surface ozone time series over the Tibetan Plateau represent valuable datasets for evaluating long-term regional-scale ozone trends".

Line 84: remove the capital letter from "The"

Response: Thanks. Changed as suggested in line 85.

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 reference instrument?

Response: Now we modified the description in the main text as (lines 97 - 98):

"Yearly instrument calibrations are performed against the Standard Reference Photometer (SRP) maintained by the WMO World Calibration Centre in Switzerland (EMPA)".

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.

Response: Thank you for your suggestion. We agree the hourly average concentration is a better proxy for assessing enhancements induced by STE events and now present results of the mupltiple regression analysis using hourly ozone concentrations. The MLR analysis estimates the impact of the WRF-FLEXPART clusters on the ozone levels at the measurement site. For the WRF-FLEXPART clusters, a separate time series was constructed for each cluster, with 1 for the hours experiencing that particular cluster and 0 otherwise. The model estimated a coefficient corresponding to enhanced or decreased ozone concentrations for each cluster.

Least-Squares methods are sensitive to outliers. This is why we remove them from the analysis in order to have a more robust analysis. There are many approaches to this problem described in the literature under the heading of "robust estimation" for example. The specific method used here is described in (de Foy et al., 2016a).

It is common practice to normalize the input parameters for a regression analysis. This improves the stability and the robustness of the estimates. Please refer to the statistics literature for more detail.

The description of MLR in this study was adjusted in the manuscript as follows (section 2.4, lines

135 - 146)：

“A Multiple Linear Regression (MLR) model was used in this study to quantify the main factors affecting hourly surface ozone concentrations. The method follows the description provided in de Foy et al. (2016b and 2016c). The inputs to the MLR model include meteorological parameters (wind speed, temperature, solar radiation and humidity), inter-annual variation factors, seasonal factors, diurnal factors, WRF boundary layer heights, WRF-FLEXPART trajectory clusters and the CAMx stratospheric ozone tracer. To obtain a normal distribution, the MLR model was applied to the logarithm of the ozone concentration offset by 10 ppb. For the WRF-FLEXPART clusters, a separate time series was constructed for each cluster, with 1 for the hours experiencing that particular cluster and 0 otherwise. The model estimated a coefficient corresponding to enhanced or decreased ozone concentrations for each cluster. The inputs to the model were normalized linearly except for the ozone tracer which was transformed log-normally with 0 offset. Because the results of Least-Squares methods are sensitive to outliers, an Iteratively Reweighted Least Squares (IRLS) procedure was used to screen them out. Measurement times when the model residual was greater than two standard deviations of all the residuals were excluded from the analysis. This was repeated iteratively until the method converged on a stable set of outliers (de Foy et al., 2016a).

Tests were performed with different variables and averaging times for each, including hourly data, running averages of 3, 8 and 24 hours, and smoothed variables using Kolmogorov-Zurbenko filters (Rao et al., 1997). For the boundary layer height as well as for the wind speed and direction, the variables were decomposed into quintiles with separate regression factors for each quintile in order to enable non-linear influences of these variables in the model. The variables to be included in the regression were obtained iteratively. At each iteration, the variable leading to the greatest increase in the square of Pearson’s correlation coefficient was added to the inputs as long as the increase was greater than 0.005. Block-bootstrapping was used with a 24 hour block length to estimate the uncertainty in the results. 100 realizations of the final model were performed to obtained the standard deviation of the model uncertainties (de Foy et al., 2015).”

The revised results of MLR were added in the manuscript in section 4.1 (lines 219 - 271):

"An iterative procedure was used to include the variables that contributed the most to the correlation coefficient of the multi-linear regression model for hourly surface ozone. This model included scaling factors for each year in the time series as well as sine and cosine terms with periods of 12 and 6 months to account for seasonality. Individual factors were further included for each hour of the day except for 12:00 and 13:00 BJT which were taken as the baseline. Factors for the local wind speed and direction were included by quintile for wind speeds and quartile for wind directions leading to 20 factors. For the boundary layer height quintiles were used. The best fit was obtained by using the 3-hour running average of wind variables and the minimum 3-hour boundary layer height. 6 factors were included for each of the particle trajectory clusters. Temperature and specific humidity (q, g/kg) were both found to improve the model. Because these vary on both the diurnal and seasonal time scale, the Kolmogorov-Zurbenko filter was used to separate each into 2 time series. The seasonal component used 5 passes of a 13-point moving average (Rao et al., 1997). The diurnal component was the difference between the hourly and the seasonal time series. For the ozone tracer, the best fit was obtained by using the Kolmogorov-Zurbenko seasonal average of the hourly CAMx tracer.

Table 2 showed that this model had 27,310 hourly data points of which 26,005 were retained by the IRLS procedure. The correlation coefficient (r) was 0.77 for the entire time series and 0.81 without the outliers. The time series of the model was shown in Fig. 4 and the scatter plot between the measurements and the model were shown in Fig. S2. Note that because stratospheric incursions are seasonal, with a maximum in the spring, there was covariance between the stratospheric tracer and the seasonal signal. Uncertainties in the estimate of the contribution from one of these therefore impact the estimate from the other.

A log-transformed model provided estimates of the contribution to the variance in the hourly ozone by different factors. Because some of these co-vary, we grouped them together in order to calculate the fraction of variation as shown in Table 2. The stratospheric ozone tracer from the CAMx model contributed 18.2±2.6% of the ozone variance at the site and the WRF-FLEXPART wind transport clusters (Fig. S3) contributed 6.5±1.7%. Local winds accounted for 31.0±1.8%, seasonal variations (including the 12 and 6-month sine and cosine terms, and the seasonal temperature and humidity terms) accounted for 35.3±3.0%, diurnal signals (including the hourly terms and the diurnal temperature and humidity signals) accounted for 7.4±0.8%, the annual signal for 1.5±0.5% and the WRF boundary layer height for 0.1±0.1% of the variance. Fig. S4 showed the histograms of the contribution terms as well as the covariance of the results by group, as determined by the block-bootstrapped method.

Figure 4 showed the contribution of the stratospheric ozone tracer and the seasonal signal. Because the model was log-transformed, these were expressed as percentage enhancements or reductions relative to the model determined baseline. The model suggested that up to 20% of the ozone variability was due to stratospheric incursions, and that these can lead to enhancements of surface of ozone of 150% of the hourly standard deviation.

As a separate test, the regression model was performed with linear transformations instead of log-transformations. The results were shown in Table 2. Although the fit was not as good, the results were remarkably similar. The contribution of the stratospheric tracer was lower, mainly because there were individual peaks which had a larger influence in the linearly transformed model than in the log-transformed model. Fig. S5 (corresponding to Fig. 4) showed the linear results. Although the mean contribution of the stratospheric tracer to surface ozone concentrations was only 1 ppb over the entire time series, it can reach above 20 ppb during specific events in the spring.

Potential vorticity from the ERA-Interim model at 500 hPa, which was near the surface at Nam Co, was not found to contribute to the simulated ozone time series. However, at 350 hPa a positive correlation was found. The correlation was even larger if we took the potential vorticity at 350 hPa above the Himalayas. Total column ozone correlated more weakly with surface ozone than potential vorticity and was not found to improve the regression model. As for potential vorticity, the correlation coefficient for total column ozone was higher above the Himalayas than at the measurement site. Fig. S6 showed the 24-hour running average of the surface ozone and the stratospheric tracer at the measurement site, and the total column ozone and the potential vorticity from ERA-Interim above the Himalayas.

We performed a separate model run where we replaced the stratospheric tracer with the potential vorticity time series at 350 hPa above the Himalayas. The model found the best fit using the Kolmogorov-Zurbenko seasonally filtered time series of potential vorticity. The model had a slightly lower correlation coefficient, and lower contribution of the potential vorticity tracer (5.8%) than the model using the CAMx stratospheric tracer. This suggests that the CAMx stratospheric tracer was a better indicator of stratospheric ozone incursions than the time series of potential vorticity.

The regression model was also performed by season, as shown in Table S1. This shows that the largest stratospheric incursions occurred in the spring (Mar, Apr, May) with 20% contribution to ozone variation, and did not impact surface ozone in the fall (Sep, Oct, Nov). The air mass transport clusters accounted for nearly 10% of the ozone variation in the summer (Jun, Jul, Aug) but very little otherwise."

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

Response: Thanks for your suggestion. We hope to keep the data displayed in UTC+8 (Beijing Time) in this study because all the measurements in this study were recorded in UTC+8 and all the models in this study were also calculated in UTC+8.

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?

Response: We used TrajStat for clusters calculation and reference was listed in the main text; now we added a reference in the main text (line 116) (Sirois and Bottenheim, 1995), and the descriptions of backward trajectory clusters methodology and the algorithm in this article were very detailed. We revised the description of HYSPLIT (lines 109 - 116):

"Gridded meteorological data for backward trajectories in HYSPLIT were obtained from Global Data Assimilation System (GDAS-1) by the U.S. National Oceanic and Atmospheric Administration (NOAA) with 1°×1° latitude and longitude horizontal resolution and vertical levels of 23 from 1000 hPa to 20 hPa (http://www.arl.noaa.gov/gdas1.php). The backward trajectories arrival height was set at 500 m (500 m, 1000 m and 1500 m were tested as arrival height and there was no obvious difference in results) above the surface and the total run times was 120 hours for each backward trajectory and in time interval of 3 hours during whole measurement period. The vertical motion was calculated using the default model selection, which used the meteorological model's vertical velocity fields. Angle distance (Sirois and Bottenheim, 1995) was selected to calculate clusters in this study".

The set-up of WRF-FLEXPART can refer to de Foy et al. (2016a) which was mentioned in the main text (lines 119 - 120).

HYSPLIT and WRF-FLEXPART were both widely used. FLEXPART and HYSPLIT were used for different purposes in this study. We used WRF-FLEXPART to generate inputs to MLR model which is better than HYSPLIT and we used HYSPLIT to generate the trajectories be used in PSCF calculation.

Line 117: "Six clusters were found. . .". Does this sentence refer to HYSPLIT or FLEXPART? Not clear . . ..

Response: Sentence was rewritten as follows (lines 122 - 123):

"Six clusters were found to represent the dominant flow patterns to the Nam Co Station by using WRF-FLEXPART".

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 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?

Response: In this study, PSCF was calculated by using trajectories which were calculated by HYSPLIT. The top of the model was set to 10000 m.

W values was set as follows: $$Wij\begin{cases}1.00 & n_{ij} > 3N_{ave}\\ 0.70 & 3N_{ave} > n_{ij} > 1.5N_{ave}\\ 0.42 & 1.5N_{ave} > n_{ij} > N_{ave}\\ 0.05 & N_{ave} > n_{ij}\end{cases}$$

where Nave represents the mean nij of all grid cells. The weighted PSCF values were obtained by multiplying the original PSCF values by the weighting factor. We used several values of W and found that by using the values listed in our manuscript, the most information can be kept in PSCF result.

SECTION 3

Line 158: please attribute the origin of these anomalous events

Response: Thanks for your suggestion. Here we just want to show general characteristics of ozone levels. In this paper, we focus on ozone levels and its temporal changes over the long-term monitoring period, diagnoses of specific ozone elevation can be meaningful but is beyond the scope of the current study. We plan to investigate ozone anomalous events and using more data from sites around Nam Co in the near future.

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.

Response: Thank you for pointing this out. Now we rewrote this sentence as (lines 186 - 189):

"The mean surface ozone mixing ratio at Nam Co Station was within the reference range reported for the Himalayas and Tibetan Plateau, and it was higher than the ratios for the two nearest urban sites: Lhasa (Ran et al., 2014) and Dangxiong (Lin et al., 2015); and comparable to of two sites on the edge of the Tibetan Plateau: Waliguan Station (Xu et al., 2011) and NCO-P (5079 m) (Cristofanelli et al., 2010) (see Fig. 1 for station locations).".

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.

Response: We agree. As stated in Vingarzan (2004), "The ozone concentration in any given area results from a combination of formation, transport, destruction and deposition", here we would just like to make a simple comparison to show the baseline of surface ozone at Nam Co in a global context. A comprehensive comparison in terms of altitudes, latitudes and site classification is more informative but is beyond the scope of the current paper. In addition, the range of 20-45 ppb is actually surface ozone baseline at background sites over the mid-latitudes in the Northern Hemisphere as indicated in Vingarzan (2004). We rephrased the sentences to make it clear and concise (lines 189 - 192):

"Surface ozone mixing ratios at Nam Co as well as other sites over the Tibetan Plateau were generally higher than the range of 20-45 ppb measured at background sites in the mid-latitudes of the Northern Hemispheres. This was in agreement with the higher concentrations typically seen at sites located in the free troposphere (Vingarzan, 2004)".

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.

Response: Thanks for your suggestion. We specified this point as follows (line 194):

"Every month considered in this study had more than 400 hours of available data (valid data for each month >56%)".

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.

Response: Thanks for your suggestion.

Fig. 3 was removed now and we adjusted Fig. 4 as you suggested (now it is Fig. 3).

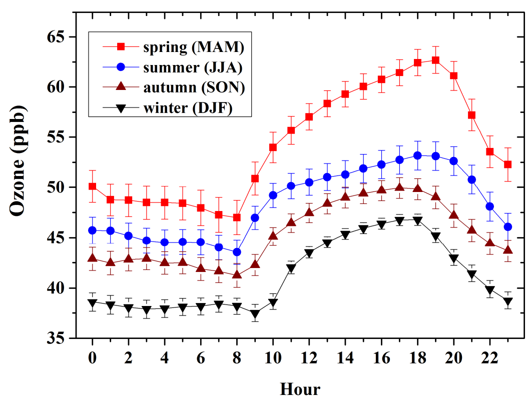

Fig. 3. Diurnal profiles of average hourly surface ozone at Nam Co Station by seasons. Error bars are 95% confidence levels.

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.

Response: Wind rose at Nam Co Station during the day (a) and at night (b) was now added as Fig. S7. Description of local wind variability was added as follows (lined 308 - 309):

"There was a lake-land breeze influencing Nam Co Station and the wind speed in the daytime was higher than those at night (Fig. S7)".

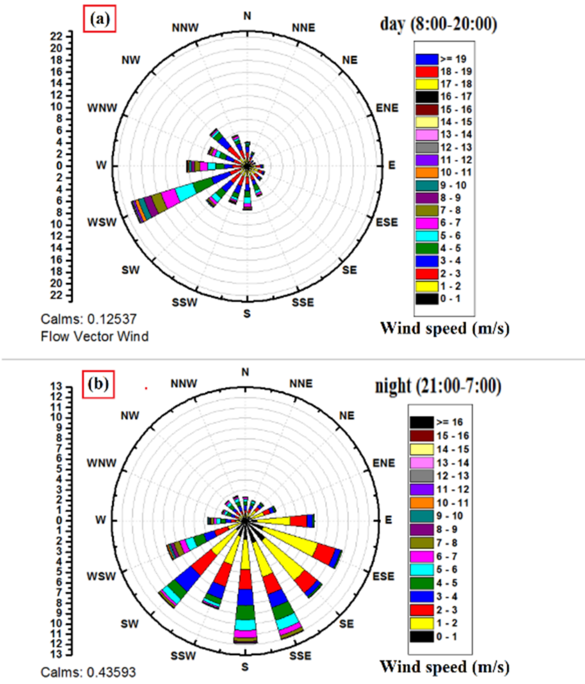

Fig. S7 Wind ross at Nam Co Station during the day (a) and at night (b).

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 suggested 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.

Response: Thanks for your suggestion. This was also a concern of Referee #1. We now present results using hourly data from the regression model. In the ACPD paper, we had found that potential vorticity near the surface did not correspond to higher ozone concentrations in the regression model. However, new research has found that potential vorticity aloft (350 hPa) did correspond to higher ozone. This suggests that PVU aloft could be used as a tracer of STE's. We also performed extensive new simulations of the impact of ERA-Interim stratospheric ozone at the surface using an air quality model. This provided time series of stratospheric tracers at the surface which were found to contribute up to 20% of the ozone variability at the site. Thanks to these new results, we have rewritten section 4, please see our response to your comments above.

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

Response: Thanks for your suggestion. This sentence was changed as follows (lines 328 -333):

"The seasonal variation of surface ozone mixing ratios at different sites around the world is influenced by many factors including: stratospheric intrusion, photochemical production, long-range transport of ozone or its precursors, local vertical mixing and even deposition (Vingarzan, 2004; Ordónez et al., 2005; Tang et al., 2009; Reidmiller et al., 2009; Cristofanelli et al., 2010; Langner et al., 2012; Ma et al., 2014; Lin et al., 2015; Ran et al., 2014; Xu et al., 2011; Macdonald et al., 2011; Pochanart et al., 2003; Derwent et al., 2016; Lin et al., 2014; Tarasova et al., 2009; Gilge et al., 2010; Wang et al., 2011; Wang et al., 2009; Zhu et al., 2004; Zhang et al, 2015; Nagashima et al., 2010)".

SECTION 5. It is not clear why in Figure 8 you reported "normalized O3" for NCOP. Please explain what kind of normalization was applied.

Response: We made Fig. 8 based on Cristofanelli et al. (2010) who reported the diurnal cycle of normalized ozone values. Cristofanelli et al. (2010) investigated the average diurnal variation of normalized O3 values obtained by subtracting daily means from the actual 30-min ozone concentrations.

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.

Response: We investigated the relationship between surface ozone and wind speed. The correlation coefficient between surface ozone and wind speed was 0.95 which indicating that high level of surface ozone was associated with high wind speed. It is important to note that local wind speed also correlates with time of day and with the evolution of the boundary layer height. In our regression model, we include all these factors and estimate the uncertainty due to the covariance by carrying out a block-bootstrapping analysis. The regression analysis suggests that local winds account for 31% of the ozone variability at the site (line 240, table 2).

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.

Response: Thanks for your suggestion. Now we removed "Type of seasonal variation of surface ozone in the Northern Hemisphere" from the manuscript.

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.

Response: Thanks for your suggestion. We will try our best to look into the diurnal variability as a function of different seasons for these sites in future. The description of the location of NCO-P was changed as suggested (lines 353 - 355):

"In the southern Tibetan Plateau and the southern ridge of the Himalayas, Xianggelila and NCO-P each had a single surface ozone peak in spring (pre-monsoon) and a minimum in summer (monsoon) with a difference between the two exceeding 30 ppb".

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

Response: Thank you for pointing out. Now we revised Figure 10 to make it easier to read.

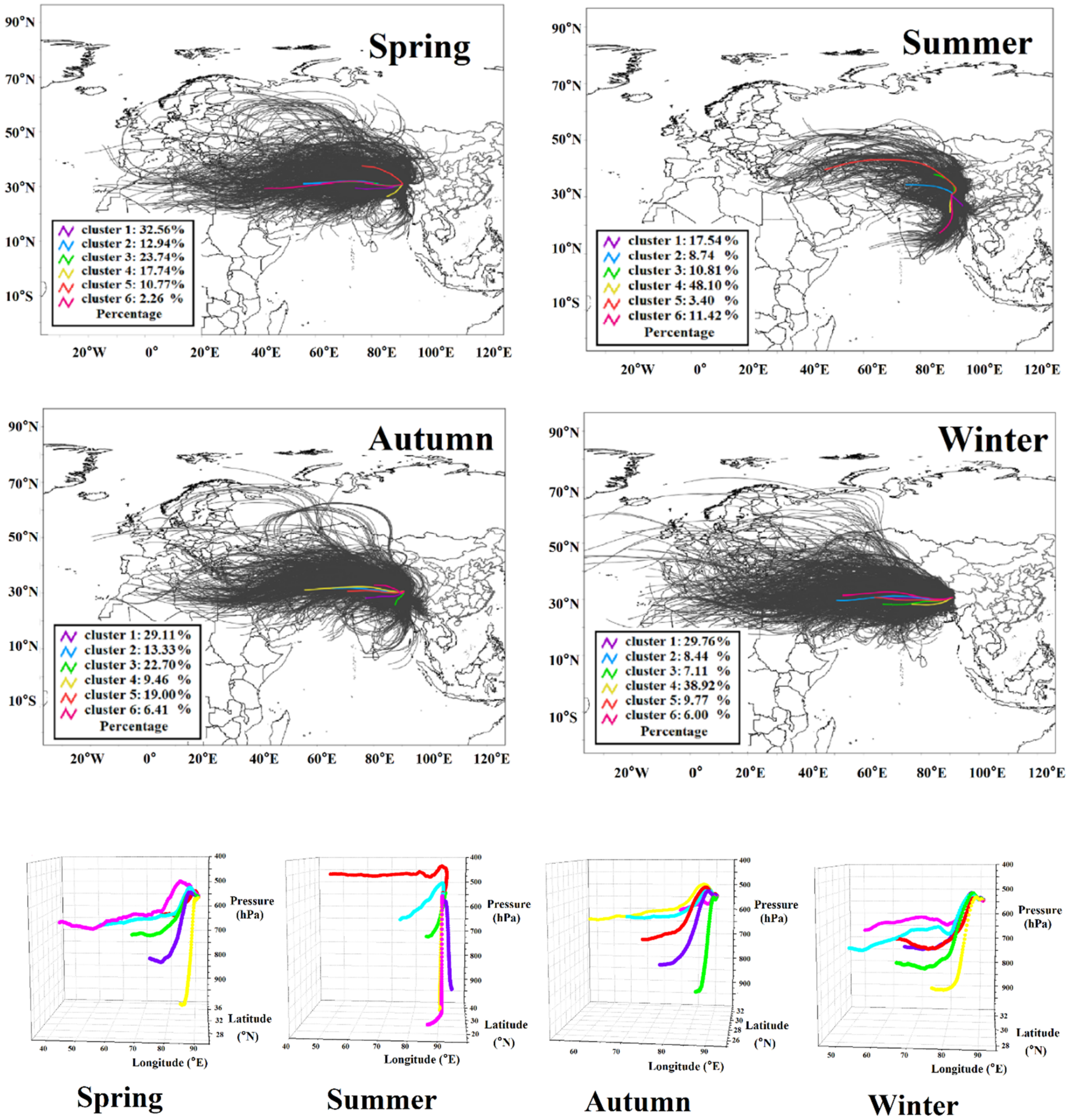

Fig. 10. Backward HYSPLIT trajectories for each measurement day (black lines in the maps), and mean back-trajectory for 6 HYSPLIT clusters (colored lines in the maps, 3D view shown on the right of the maps) arriving at Nam Co Station by season.

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.

Response: Angle distance (Sirois and Bottenheim, 1995) was selected to calculate clusters in this study. In spring, cluster 1 (32.56%) and cluster 4 (17.74%) were the clusters crossed the Himalayas in different pathways.

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.

Response: We thought the ozone values at NCO-P in March, April and May was much higher than those at Nam Co Station accordingly.

Polluted air-masses can contribute to the surface ozone variability at NCO-P and contribution can also contribute to the elevated level of surface ozone at Nam Co Station. Now we added the description for this as follows (lines 366 - 368):

"The contribution of polluted air masses in driving ozone variability at the southern ridge of the Himalayas was remarkable in the spring and it may also have an effect on the level of surface ozone at Nam Co Station through transport".

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!)

Response: Thank you for pointing out. Cluster 2, 3 and 5 were associated to the northern Tibetan Plateau. Now we made Figure 10 easier to read.

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.

Response: Thanks for your comment. Now meridional cross-sections over Nam Co Station (Fig. 5) were added to indicate the position (altitude and longitude) of the strongest STE in the meridional cross-section (over Nam Co Station) in different months (line 272 - 298):

"In order to visualize the transport of ozone from the stratosphere to the troposphere, we analyzed the upper troposphere and lower stratosphere structures of the meridional cross-section of monthly mean ERA-Interim data above Nam Co Station (Fig. 5). In the spring (Mar, Apr and May), the dynamical tropopause (identified by the isolines of 1 and 2 potential vorticity unit) exhibited a folded structure over the Tibetan Plateau. This tropopause folding can lead to a downward transport of ozone from the stratosphere to the troposphere. Tropopause folding happened in the southern Tibetan Plateau and close to Nam Co Station in the spring. Cosmogenic ^{35}S results (Lin et al., 2016) also indicated that in the spring, 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. The larger diurnal amplitude of surface ozone in the spring than other seasons (Fig. 3, mentioned in section 3.3) may be related to four factors: (1) position of STE hot spot; (2) frequency of STE; (3) PBLH at Nam Co Station and (4) solar radiation at Nam Co Station. In the spring, plots of tropopause folding suggest that STE mostly happens in the southern Tibetan Plateau which is close to Nam Co Station and that STE even happens right above Nam Co Station. Furthermore, PBLH at Nam Co Station was higher in the spring than during the rest of the year. The higher PBLH in the spring facilitated the impact of downward transport from the stratosphere to Nam Co Station. The spring also has more intense solar radiation than the summer because the Monsoon leads to increased cloudiness in the summer. The Pearson's correlation coefficient between monthly SWD and surface ozone was ~0.93 in 2012 (2012 was selected because it had a more complete dataset than the other years) (Fig. 6) indicating that monthly surface ozone variability at Nam Co Station was associated with solar radiation. This was expected as increased solar radiation promotes the photochemical production of surface ozone in the spring, which is similar to the mechanism at other background sites (Monks 2000). Consequently, more photochemical production of ozone is expected in the spring. In the summer (Jun, Jul and Aug), the jet core moved to the northern Tibetan Plateau and tropopause folding was relatively farther from Nam Co Station than those in the spring. Consequently, there was a smaller impact of stratospheric air at Nam Co Station. With tropopause folding further north in the summer, the air masses from the northern Tibetan Plateau may contribute more to the surface ozone levels at Nam Co Station than the air masses from the southern Tibetan Plateau. In the autumn (Sep, Oct and Nov) and the winter (Der, Jan and Feb), the heights of folding were higher than those in the spring and the summer; and the PBLHs in the autumn and the winter were much lower than those in the spring and the summer. Furthermore, SWD in the autumn and the winter were weaker than those in the spring and the summer. These factors contributed to the relatively low level of surface ozone at Nam Co Station in the autumn and the winter."

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

Response: While we used WRF-FLEXPART and HYSPLIT for different purpose, this is also confirmed by WRF-FLEXPART.

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 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!

Response: Thank you for pointing this out.

These results were calculated by PSCF which using backward trajectories calculated by HYSPLIT.

We considered the transport by cluster in MLR and it was a secondary factor. The MLR result suggested that lower levels of surface ozone were associated with air masses that came from the south (it was possibly related to the pollution emitted from Dangxiong and Lhasa) and higher levels of surface ozone were identified when air masses were from the north. PSCF results do not identify the stratospheric tracer separately and it is therefore possible that PSCF picked up the contribution from STE as a signal from the south in the spring and from the north in the summer. The fact that the MLR results account for the stratospheric tracers separately explains why we obtained PSCF results that are different but not inconsistent from the MLR model.

With respect to the seasonality of the WRF-Flexpart results we have added a new table S1 that shows the MLR results by season. These also suggest that most of the STE's occur in the spring

As we mentioned in section 2.5, in this study, PSCF was calculated basing on trajectories corresponding to concentrations that exceed the mean level of surface ozone. When we used 75th and 90th percentiles of surface ozone distribution as the threshold in PSCF, there were a lot of information being missed.

We checked legend which was automatic generated by MeteoInfo and now the legend in figure was revised.

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.

Response: We rewrote this section and named it as "Implication for measurement and study of surface ozone in the inland Tibetan Plateau and beyond". This section was rewritten as follows (lines 393 - 413):

"The changes of atmospheric environment of the Tibetan Plateau are of universal concern due to its rapid responses and feedbacks to regional and global climate changes. The Tibetan Plateau covers vast areas with varied topography; however, comprehensive monitoring sites are few and sporadically distributed. Analysis of atmospheric composition at Waliguan in the north and Everest in the south of the Tibetan Plateau have shown that they are representative of high-altitude background sites for the entire Tibetan Plateau. It is noteworthy that the Tibetan Plateau, as a whole, is primarily regulated by the interplay of the Indian summer monsoon and the westerlies; and the atmospheric environment over the Tibetan Plateau is heterogeneous. Mount Everest is representative of the Himalayas on the southern edge of the Tibetan Plateau and is the sentinel of South Asia where anthropogenic atmospheric pollution has been increasingly recognized as disturbing the high mountain regions (Decesari et al., 2010; Maione et al., 2011; Putero et al., 2014). In addition, Mount Everest has been identified as a hotspot for stratospheric- tropospheric exchange (Cristofanelli et al., 2010; Škerlak et al., 2014) where the surface ozone is elevated from the baseline during the spring due to frequent stratospheric intrusions. Waliguan, in the northern Tibetan Plateau, is occasionally influenced by regional polluted air masses (Zhu et al., 2004; Xue et al., 2011; Zhang et al., 2011). Its mountainous landform facilitates mountain-valley breezes and may sometimes pump up anthropogenic emissions especially during the winter (Xue et al., 2011). Nam Co Station, in the inland Tibetan Plateau, is distant from both South Asia and northwestern China, it has been found to be influenced by episodic long-range transport of air pollution from South Asia (Xia et al, 2011; Lüthi et al., 2015), evidenced by the study of aerosol and precipitation chemistry at Nam Co Station (Cong et al., 2007; Cong et al., 2010). As for surface ozone, Nam Co Station is less influenced by stratospheric intrusions directly than NCO-P, and is minimally influenced by local anthropogenic emission. It showed distinct seasonal and diurnal variation patterns as compared with those sites in the Himalayas and the northern Tibetan Plateau as presented earlier. Our measurements of surface ozone at Nam Co are essential baseline data of the inland Tibetan Plateau, more long-term measurements are needed to enable a better spatial coverage and a comprehensive understanding of regional surface ozone variations and underlying influence mechanisms".

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

Response: Long-range transport of air pollutants referred to the atmospheric transport of air pollutants within a moving air mass for a distance greater than 100 kilometers.

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.

Response: We removed this sentences from the revised manuscript.

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

Response: We have tried our best to give more quantized evidence and modify our description in manuscript now.

ACKWNOLEDGMENTS You must acknowledge NOAA for providing HYSPLIT model and GFS meteorological files. I suppose that also the TrajPlot developers must be acknowledged

Response: Thanks for your suggestion. Added as you suggested. Acknowledgements were rewritten as follows (lines 438 - 442):

"The authors are grateful to NOAA for providing HYSPLIT model and GFS meteorological files. The authors thank Yaqiang Wang who is the developer of MeteoInfo and give selfless help. Finally, the authors would like to thank the editor and referees of this manuscript for their helpful comments and suggestions. This study was supported by the National Natural Science Foundation of China (41371088, and 41630754) and the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (XDB03030504)".

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