

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.

Review (Anonymous Referee #1)

Surface ozone at Nam Co (4730 m a.s.l.) in the inland Tibetan Plateau: variation, synthesis
comparison and regional representativeness

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Summary of paper

The Tibetan Plateau is considered as an ideal region for studying processes of the background atmosphere. Sites in the southern, northern, and central regions of the Tibetan Plateau exhibit different patterns of variation in surface ozone. Measurements for the period January 2011 to October 2015 of surface ozone concentrations at Nam Co Station are summarized using mostly monthly averaged values. A large annual cycle was observed with maximum ozone mixing ratios occurring in the spring with minimum ratios occurring during the winter. The authors indicate that Nam Co Station represents a background region, where surface ozone receives negligible local anthropogenic emissions. The authors state that surface ozone at Nam Co Station is mainly dominated by natural processes involving photochemical reactions and potential local vertical mixing. Model results indicate that the study site is affected by the surrounding areas in different seasons and that air masses from the northern Tibetan Plateau lead to increased ozone levels in the summer. The authors believe that in contrast to the surface ozone levels measured at the edges of the Tibetan Plateau, those at Nam Co Station appear to be less affected by stratospheric intrusions and human activities, which makes Nam Co Station representative of vast background areas in the central Tibetan Plateau. By comparing measurements at Nam Co Station with those from other sites in the Tibetan Plateau and beyond, the authors' goal is to expand the understanding of ozone cycles and transport processes over the Tibetan Plateau.

General Comments

I would like to see another version of this manuscript after the authors have made their modifications.

A key question I have is to what extent do the authors believe that stratospheric intrusions (not necessarily originating directly above the site) influence the Nam Co station? The reason I am asking this question is that the authors state "In contrast to the surface ozone levels at the edges of the Tibetan Plateau, those at Nam Co Station are less affected by stratospheric intrusions and human activities which makes Nam Co Station representative of vast background areas in the central Tibetan Plateau." I am not sure what the authors are intending to say in this sentence. Does the sentence mean that stratospheric intrusions play an unimportant role at the site in influencing the surface ozone concentrations or do the authors mean that the Nam Co site is influenced by "aged" stratospheric intrusions but to a lesser extent than those intrusions that occur at the southern and northern portions of the Tibetan Plateau? Based on the detailed focus on stratospheric intrusions in the manuscript, I suspect that the authors believe that STE plays an important role at the Nam Co Station in enhancing surface concentrations during specific seasons but that STE plays *less* of a role when compared to stations located at the southern and northern portions of the Tibetan Plateau. I would appreciate it if the authors would clarify this.

Response: Thank you for pointing out this critical issue. We believe that stratosphere-troposphere exchange (STE) plays an important role on surface ozone at Nam Co Station, but one that is different from the STE that happens in the southern Tibetan Plateau (in the winter and the spring) and the northern Tibetan Plateau (in the summer), Nam Co Station was affected by STE indirectly most of the time. The air masses in high ozone level can be transported to Nam Co Station horizontally after the STE in the southern Tibetan Plateau and the northern Tibetan Plateau in different seasons.

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.

The authors devote a considerable amount of the manuscript to discussing the contribution from stratospheric intrusions during specific periods of the year. Using mostly monthly and annual average surface ozone mixing ratios, the authors report a large annual cycle with maximum ozone mixing ratios occurring in the spring, with minimum ratios occurring during the winter. As noted by the authors, during the spring, Nam Co was affected by aged stratospheric originating over the Himalayas rather than being influenced by transport from fresh stratospheric air masses directly above the station. In spring, the air masses that arrived at Nam Co Station were predominantly from the west and from the south, and the 3-D clusters indicated that the air masses traveled through the Himalayas before reaching Nam Co Station. The authors note that Cristofanelli et al. (2010), Putero et al. (2016) and Chen et al. (2011) found that the frequency of stratospheric intrusions in the Himalayas was high in spring, and slightly lower than during

the winter. Škerlak et al. (2014) showed that the seasonal average ozone flux from the stratosphere to the troposphere in the Himalayas was the highest in spring. The authors noted that air masses transported in the spring from the Himalayas led to higher concentrations of surface ozone at Nam Co Station.

For the summer months, the authors note that there were more backward trajectories coming from the northern Tibetan Plateau than in other seasons. HYSPLIT backward trajectories arriving at Nam Co Station in the summer were classified into 6 clusters. Clusters which came from the northern Tibetan Plateau had higher mean surface ozone levels than clusters which came from the southern Tibetan Plateau. The authors indicate that the air masses that arrived at Nam Co Station from the northern Tibetan Plateau and northwestern China by horizontal wind transport likely resulted in the higher ozone concentrations at Nam Co Station during the summer. However, Trajectories 2 and 3 during the summertime also contain high ozone concentrations (Fig. 11).

During the summer, according to Škerlak et al. (2014), the northern Tibetan Plateau is the hot spot of stratosphere-to-troposphere ozone flux. Do other trajectories (e.g., 2 and 3) during the summertime also exhibit possible contributions from STE? A further reading of Škerlak et al. (2014) indicates that the hotspot region of the Tibetan Plateau is most likely affected by stratospheric intrusions during the months of DJF, MAM, and JJA (page 926 of Škerlak et al., 2014). Škerlak et al. (2014) indicate that there are **intense** deep STT ozone fluxes over the Tibetan Plateau during MAM and JJA. Škerlak et al. (2014) indicate that the global hotspots, where surface ozone concentrations are most likely influenced by STE, is the Tibetan Plateau in all seasons except for SON (page 934).

Response: Thank you for your comments.

As noted by Škerlak et al. (2014), surface ozone in Tibetan Plateau (considered as a whole) was most likely influenced by STE in all seasons except for autumn (SON) (page 934 in Škerlak et al., 2014). Nevertheless, when we look into different parts of Tibetan Plateau and even northwestern China, STE was not occurred synchronously. The peak of stratosphere to the troposphere ozone flux was found over the Himalayas and the southern side of the Tibetan Plateau in spring (MAM) (page 926 in Škerlak et al., 2014); while the stratosphere to the troposphere ozone flux occurred in the northern Tibetan Plateau and northwestern China is much higher than those in the southern Tibetan Plateau in summer (JJA) (Fig. 16 and page 926 in Škerlak et al., 2014).

To facilitate the understanding of STE over the Tibetan Plateau, we have added meridional cross-sections over Nam Co Station (Fig. 5) to indicate the position (altitude and longitude) of the strongest STE in the meridional cross-section (over Nam Co Station) in different months. We also added related discussion on the meridional cross-sections (lines 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 (Dec, 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”.

As indicated above, the authors mostly used monthly and annual average surface ozone mixing ratios to characterize the ozone concentrations at the Nam Co Station. The use of monthly or annual average concentrations “smoothes” the variability associated with hourly average concentrations. Thus, if one were interested in assessing the magnitude of the ozone concentration enhancements that may be associated with STT events, he or she might wish to focus on the frequency and time of year when high hourly average concentrations occur. Although I am not suggesting that the authors have to perform an additional assessment, I think the authors, using *hourly* average concentrations, have an opportunity to include in their current manuscript an expanded discussion on the potential importance of aged stratospheric air originating at other locations that is transported to the site.

Response: Thank you for your comments. We agree the hourly average concentration is a better proxy for assessing enhancements induced by STE events and now present results of the multiple regression analysis using hourly ozone concentrations. The description of MLR in this study was adjusted in the manuscript as follows (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).”

Fig. S1 provides potentially important information about the day-to-day variability of the hourly concentrations. I have reproduced Fig. S1 below. The figure illustrates the variability of the hourly average concentrations for the period from January 2011 until October 2015. As anticipated, the frequency of the highest hourly average concentrations (e.g., 70 ppb to > 90 ppb) occurs during the springtime and early summertime (Fig. S1). This agrees with the authors' observations based on the monthly average concentrations. However, unlike the pattern described based on the monthly averages, high hourly average concentrations are also occurring during the winter and summertime for some of the years. During the months of SON, the frequency of high hourly average concentrations is much lower than those values exhibited during the DJF, MAM, and JJA seasons. Thus, there appears to be different patterns observed when using the monthly average concentration results with those using the hourly average concentration results.

Investigating the pattern for when the highest hourly average concentrations occur, it appears that this pattern is similar to the one described by Škerlak et al. (2014), which indicated stratospheric intrusion hotspots in the Tibetan Plateau during the months of DJF, MAM, and JJA. If the authors wish to, they have the opportunity to expand their discussion in their manuscript to comment on the degree to which the observed enhanced hourly average ozone concentrations may be associated at the Nam Co Station with STE.

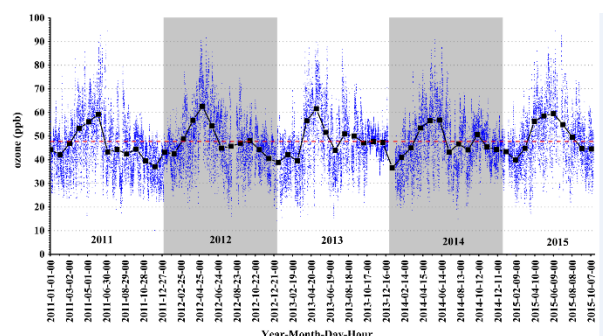


Fig. S1. Variation of surface ozone at Nam Co Station from January 2011 to October 2015. Hourly mean mixing ratios of surface ozone are in blue dots; monthly mean mixing ratios of surface ozone are in black dots; average mixing ratio of surface ozone during whole measurement period in red dash line.

Response: Thank you for your suggestion.

Now we also investigated the STE happened by the meridional cross-section at 91°E (over Nam Co Station) monthly (Fig. 5) and the enhanced hourly average surface ozone concentrations at Nam Co Station associated with STE were analyzed by using CAMx stratospheric tracers (Table 2). Downward transport of stratospheric ozone contributed to high level of surface ozone at Nam Co Station. Following your suggestion, we have performed the Multiple Linear Regression (MLR) model by seasons using log-transforms and CAMx stratospheric tracers (Table S1). The regression model suggests that CAMx tracers contributed much more to surface ozone at Nam Co Station in the spring than during the rest of the year. The minimum impact of the CAMx tracers was during the autumn, which might be a reason for the low incidence of high hourly average concentration of surface ozone during the month of SON. MLR results indicated that although the mean contribution of the stratospheric tracer to surface ozone concentrations is only 1 ppb over the entire time series, it can reach above 20 ppb during specific events in the spring.

Specific Line-by-Line Comments

1. Title: I would suggest that the title be slightly modified as follows: Surface ozone at Nam Co in the inland Tibetan Plateau: variation, synthesis comparison and regional representativeness.

Response: Thank you for your suggestion. Title was changed according to your suggestion as follows:

“Surface ozone at Nam Co in the inland Tibetan Plateau: variation, synthesis comparison and regional representativeness”.

2. Lines 24-25: The authors state "Model results indicate that the study site is affected by the surrounding areas in different seasons and that air masses from the northern Tibetan Plateau lead to increased ozone levels in the summer." I think the authors are not necessarily indicating that there is an increase during the summer at Nam Co due to air masses from the northern Tibetan Plateau but that the

air masses from the northern Tibetan Plateau *contribute* to the enhancement of ozone levels measured at the site. The word "increase" gives the impression that relative to the spring, the summer monthly averages are higher. The monthly average levels at the site are lower than those observed during the spring and therefore, I am suggesting a slight change in the wording.

Response: This sentence was changed according to your suggestion as follows (lines 25 -27):

“Model results indicate that the study site is affected by the surrounding areas in different seasons: air masses from the southern Tibetan Plateau contribute to the high ozone levels in the spring and enhanced ozone levels in the summer were associated with air masses from the northern Tibetan Plateau”.

3. Lines 34-35: I would suggest references that represent comprehensive summaries of human health and vegetation effects, such as LRTAP Convention (2015), REVIHAAP (2013), and US EPA (2013).

Response: We added these references and sentence was rewritten as follows (lines 33 -35):

“High levels of surface ozone are currently a major environmental concern because of the harm ozone poses to health and vegetation at the surface (LRTAP, 2015; REVIHAAP, 2013; US EPA, 2013; Mauzerall and Wang, 2001; Desqueyroux et al., 2002)”.

4. Line 45-46: The sentence: "In this situation, background sites can represent areas with surface ozone concentrations that are under the control of largely uniform synoptic systems and are minimally affected by local anthropogenic sources." What does "in this situation" refer to?

Response: We used “in this situation” to refer that “the surface ozone over the entire world can’t be represented by one site or few sites. But background site can represent extended area in a relatively similar environment”. It seems “in this situation” was misleading and redundant. Now we removed “in this situation” from the main text.

5. Lines 141-143: The sentence "In cells with high PSCF values are associated with the arrival of air parcels at the receptor site that have pollutant mixing ratios that exceed the criterion value" does not appear to be complete. Should the sentence start with "Cells with high PSCF...”?

Response: Thank you so much. We changed “In cells” to “Cells” in the manuscript and sentence was rewritten as follows (line 168 - 170) “Cells with high PSCF values are associated with the arrival of air parcels at the receptor site that have pollutant mixing ratios that exceed the criterion value”.

6. Line 155: The sentence states "The mean surface ozone mixing ratio at Nam Co Station during the entire observational period was 47.6 ± 11.6 ppb..." I am not suggesting any change in this sentence but I do want to point out that the authors on Lines 33 and 34 state that "High levels of surface ozone are currently a major environmental concern because of the harm ozone poses to health and vegetation." This is a correct statement. However, researchers who assess human health and vegetation effects focus on the occurrence of high, as well as mid-level hourly average concentrations, and normally do not focus on high annual average concentrations. Annual, seasonal, or monthly average ozone concentrations are not necessarily the best metrics to use when assessing either human health or vegetation effects. While monthly and annual average concentrations are used for assessing the performance of global modeling results, these metrics are not necessarily relevant for assessing human health and vegetation effects.

Response: Thank you for pointing this out. We added the sentence as follows (lines 39 - 40):

"For global modeling, monthly and annual average concentrations of tropospheric ozone are used for assessing and improving the modeling results (Wild and Prather, 2006; Roelofs et al., 2003)".

7. Page 156: Table 1 indicates that the data capture was as follows: 2011 (75.25%), 2012 (90.30%), 2013 (75.90%), 2014 (70.05%), and 2015 (66.21%). Was the 66.21% data capture observed in 2015 related to the entire 12 months or was this value the data capture for the period January – October 2015?

Response: 66.21% in 2015 was the valid data during whole 2015 from January to December, and these valid data in 2015 was started from January 2015 to October 2015.

8. Lines 182-183: The authors state "The transition between high levels during the daytime and low levels during the nighttime was fast." I would appreciate it if the authors could please explain why the transition was fast.

Response: Thank you for your comment. The transition was probably caused by the vertical mixing and photochemical production which was induced by sunrise. We removed this sentence as we were not going to further expand this point.

9. Lines 186-187: The authors state "Relatively large diurnal amplitudes were observed in spring, with much smaller diurnal amplitudes observed during summer, autumn and winter." Can the authors offer an explanation for this observation? Could this observation be associated with STE making it to the

ground during the spring more frequently than during the other seasons?

Response: We added the explanation for the relatively large diurnal amplitudes in the spring in the manuscript as follows (lines 279 - 290):

“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”.

10. Lines 194-196: The authors state "35S results (Lin et al., 2016) also support this result by showing 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." Should the ";" be placed with a "," to make a complete sentence?

Response: Changed as suggested. This sentence was rewritten as follows (lines 277 - 279):

“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”.

11. Lines 188-200: The authors state "A multiple linear regression model was used to quantify the contributions of various factors (including temperature, clear sky solar radiation, potential vorticity, wind speed, humidity, annual cycle, interannual variation and WRF-FLEXPART trajectory clusters) to the

measured maximum daily 8-hour average surface ozone." If in the authors' multiple linear regression model the variables (i.e., temperature, clear sky solar radiation, potential vorticity, wind speed, humidity, annual cycle, interannual variation and WRF-FLEXPART trajectory clusters) were not independent, what would be the effect on the outcome of the results using the model?

Response: We use block-bootstrapping to estimate the uncertainty in the results, including the impact of covariation in the inputs (de Foy et al., 2015). The results are presented for groups of variables arranged by the time scale of the variability and the type of inputs. These are mostly orthogonal to each other, although some of them have an inherent correlation. For example, the diurnal variation terms have an r^2 of 0.21 with the boundary layer height and 0.19 with the local winds. Because we nonetheless wish to estimate the different contributions of these terms, we keep them separate in the analysis. Likewise, the CAMx stratospheric tracer and the seasonal time series have an r^2 of 0.17. Because the stratospheric impacts are greater in the spring than during the rest of the year, the correlation between the two time series is inescapable. The block-bootstrapping method can be used to estimate the corresponding uncertainty in the results. Figure S4 shows the covariation of the results of the MLR analysis. The correlation coefficient squared (r^2) of the contribution from the diurnal terms with the local winds is 0.08 and for the boundary layer height it is 0.06. This suggests that the correlation between the time series does not have a large impact on the results. For stratospheric tracer and the seasonal time series, the r^2 is 0.5 which suggests that the correlation of the time series has a stronger impact on the estimation of the contribution of each term to the ozone variance in the measurements. A larger estimate of the contribution of the stratospheric tracer will lead to a lower estimate of the seasonal term and vice versa. This is reflected in the larger uncertainty in the estimates, as shown in Fig. S4.

12. Lines 209-211: The authors state "Specific humidity was the second largest contributor (20%; Table 2) with a negative coefficient indicating that higher surface ozone was associated with drier conditions possibly due to transport of continental air masses; or impacts from air masses aloft." If the Nam Co Station were influenced by "aged" stratospheric intrusions, would the lower humidity still be associated with the "aged" transported air from the stratosphere originating over the Himalayas after several days? Perhaps a short comment in the manuscript might be in order.

Response: Both continental air masses and air masses aloft can lead to low specific humidity, so we try to find another stratospheric incursion indicator. Now we used CAMx tracer instead of specific

humidity to identify the impact from the stratospheric incursion and CAMx tracer was a better indicator of stratospheric ozone incursion (lines 263 - 267):

“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”.

13. Lines 212-214: The authors state "The negative coefficient indicates that air masses transported from the south to Nam Co were associated with lower surface ozone. 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." However, in Lines 287-290, the authors indicate that "Backward trajectories and PSCF were utilized to identify the source of surface ozone at Nam Co Station and to assess the regional representativity of surface ozone at Nam Co. In spring, the air masses that arrived at Nam Co Station were predominantly from the west and from the south, and the 3-D clusters indicated that the air masses traveled through the Himalayas before reaching Nam Co Station (Fig. 10)." If the air masses traveled through the Himalayas during the spring before reaching the Nam Co Station, at times would not the air masses represent "aged" stratospheric intrusions and wouldn't these air masses influence the daily surface ozone variation? Is there a difference in the conclusions reached using the multiple linear regression model versus the back trajectory and the PSCF analyses? Perhaps I am missing something here.

Response: We considered the transport by cluster in MLR and it was the secondary factor. The MLR results suggested that lower levels of surface ozone were associated with air masses 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 was not separate from the stratospheric tracer and it is 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. PSCF results are different from MLR but not inconsistent.

14. Lines 256-258: The authors state "This type has a plateau of high surface ozone in spring and summer and a minimum in winter. Sites of this type occur in regions with strong ozone precursor emissions in the summer (such as the central European continent) or in regions where stratospheric intrusion occurs frequently in summer." Could the authors please provide examples for specific regions of the world where stratospheric intrusions frequently occur during the summer. Perhaps the results from Škerlak et al. (2014) might be a good source.

Response: Thank you for pointing this out. Regions including the Pamirs, Tian Shan, north-central US, Anatolia, northern side of the Tibetan Plateau, the east and west coasts of Australia, the northern Tasman Sea and Wilkes Land in East Antarctica were the places had stratospheric intrusions frequently during the summer (JJA) (Škerlak et al., 2014). But now we removed this part as suggestion from referee #2.

15. Lines 271-273: The authors state "Sites in the central Tibetan Plateau including Nam Co Station showed maximum ozone during late spring-early summer and relatively low levels in the remainder of year (Fig. 9B), corresponding to the Spring-maximum type. Compared with the surface ozone levels at Nam Co Station, those at Lhasa and Dangxiong were much lower." This conclusion is based upon the use of monthly average concentrations. Is there any indication that the use of the frequency of high hourly average concentrations might provide a different pattern?

Response: It is a good suggestion. We are looking forward to having collaborations with the researchers who work on the surface ozone measurement at Lhasa and Dangxiong. But now, we can only get the monthly average concentrations of surface ozone at Lhasa and Dangxiong from their publications and we were unable to investigate the pattern by using the frequency of high hourly average concentrations of surface ozone at these three sites now. We will try our best to investigate this in future.

16. Lines 313-314: The authors state "The atmospheric environment of the Tibetan Plateau and its relationship to regional and global change are of universal concern due to the rapid responses and feedbacks specific to the "Third Pole". I would appreciate it if the authors would please expand on this sentence to explain what they mean.

Response: The sentence has been rewritten as follows to make it clear and concise (lines 393 - 394):

"The changes of the atmospheric environment of the Tibetan Plateau are of universal concern due

to its rapid responses and feedback to regional and global climate changes”.

17. Line 324-327: The authors state "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 local anthropogenic emissions especially during the winter (Xue et al., 2011)." I was under the impression that local anthropogenic sources are small near Mt. Waliguan. Mt. Waliguan is far from major cities, such as Xining (90 km) and Lanzhou (260 km) in the eastern sector. I would appreciate it if the authors would further elaborate concerning the enhancement at Mt. Waliguan from local anthropogenic emissions.

Response: Xue et al. (2011) stated “further analysis of backward trajectories for the recent 10 years indicated that WLG was frequently (~50% of air masses) influenced by the air from the east, suggesting an important role of anthropogenic emissions in central and eastern China in shaping the summertime surface ozone and other atmospheric trace constituents at WLG and over the Tibetan Plateau.” Zhang et al. (2011) stated “pollution episodes at WLG were characterized by significantly enhanced mixing ratios and large and erratic variations. This apparently reflects influence of regional emission sources on WLG”; “in summer, the most elevated CO mixing ratios are associated with cluster 3 which passed through the urbanized area southeast of WLG (e.g. Lanzhou city, the central region and southeast of Gansu province)” and “compared to the JFJ, air masses identified at WLG as polluted contained more CO relative to the background values and displayed large and irregular fluctuations suggesting greater influence from regional emission sources”. Xue et al. (2013) stated at Waliguan, “the daytime upslope flow of boundary-layer air and nighttime downslope flow of free tropospheric air resulted in a reversed diurnal variation of trace gases at WLG. This unusual phenomenon could be explained by transport of anthropogenic pollution during the night. Transport of anthropogenic pollution from the northeast/east, where Xining and Lanzhou are located, is likely responsible for the enhanced levels of CO and VOCs during the nighttime at WLG.” Refer to the description in these publications, Waliguan can be affected by the polluted air masses from regional emission sources and the enhanced levels of CO and VOCs during the nighttime at WLG was associated with upslope flow (night wind in mountain–valley breezes).

We adjusted this sentence as follows (lines 403 - 406):

“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)".

18. Lines 332-335: The authors state "During the summer, surface ozone concentrations at Nam Co Station are higher than the northern hemisphere average, which suggests that there are impacts of long-range transport. Nam Co is less influenced by stratospheric intrusions than NCOP on the slopes of Mount Everest, and it is minimally influenced by local anthropogenic emission as evidenced by the constant long-term variation of surface ozone and consistent diurnal variation regardless of season, as discussed above." What is the influence of stratospheric intrusions on Nam Co during the summer? Škerlak et al. (2014) appear to indicate that it is important during the summer. If the surface ozone concentrations during the summer at Nam Co Station are higher than the northern hemisphere average, could the suggested long-range transport be associated with "aged" air masses from the stratosphere that are being transported to the site? I think it would help the reader to clarify what the authors mean by "there are impacts of long-range transport."

Response: Thanks for your comment. We add meridional cross-sections over Nam Co Station (Fig. 5) to indicate the position (altitude and longitude) of the strongest STE in the meridional cross-section (over Nam Co Station). In summer, the hot spot of STE was in the northern Tibetan Plateau and air masses from this region elevated surface ozone concentration at Nam Co Station in summer which was also showed in Fig. 11. Air masses with high concentration of ozone in stratosphere were probably first transported to the northern Tibetan Plateau then transported horizontally to Nam Co Station.

We added the description for this point in the manuscript as follows (lines 290 -294):

"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".

(lines 378 - 381):

"In the summer, clusters from the northern Tibetan Plateau had higher mean surface ozone levels

than clusters which came from the southern Tibetan Plateau. The air masses that arrived at Nam Co Station from the northern Tibetan Plateau and northwestern China by horizontal wind transport likely resulted in the higher ozone concentrations at Nam Co Station in the summer”.

19. Line 340: The summary needs to be expanded. It is very minimal at this time.

Response: The summary has been expanded to including major results and conclusions. Parts of summary were rewritten as follows (lines 420 - 436):

“The baseline of surface ozone is mainly controlled by various natural factors. Downward transport of air masses, air masses from the southern Tibetan Plateau in the spring and from the northern Tibetan Plateau in the summer contributed to the elevated monthly concentrations of ozone at the surface. Diurnal peaks of surface ozone in the afternoon were associated with high SWD, high PBLH and high wind speed. The analysis suggests that stratospheric intrusions account for around 20% of the variability in surface ozone concentrations at Nam Co Station. Further analysis of tropopause folding suggest that Nam Co Station is affected by “aged” air masses associated with stratospheric intrusions transported from the southern and northern Tibetan Plateau, mainly during the spring and the summer, respectively.

Synthesis comparison of ozone variability at regional and hemispheric scales revealed that the seasonality of surface ozone at Nam Co Station is most similar to other background sites in the Northern Hemisphere, albeit with slightly higher fluctuations in the summer season due to infrequent occurrences of air mass transport from Northwest China. Surface ozone at Nam Co showed distinct seasonal and diurnal variation patterns as compared with those sites in the Himalayas and the northern Tibetan Plateau. The monthly maximum of surface ozone at Nam Co Station was later in the year than the sites in the southern Tibetan Plateau and the southern ridge of the Himalayas, but earlier than the sites in the northern Tibetan Plateau.

Our measurements provide a baseline of tropospheric ozone at a remote site in the Tibetan Plateau, and contribute to the understanding of ozone cycles and related physico-chemical and transport processes over the Tibetan Plateau. More long-term measurements of surface ozone at field sites covering the spatially extensive Tibetan Plateau are needed to improve our understanding of surface ozone variations and the underlying influence mechanisms”.

20. Lines 348-349: The authors state " Synthesis comparison indicated that Nam Co is less

influenced by stratospheric intrusions and anthropogenic disturbances than sites along the rim of the Tibetan Plateau." I would appreciate it if the authors could please clarify this sentence. Should the sentence read "While the Nam Co Station is less influenced by stratospheric intrusions and anthropogenic disturbances than sites along the rim of the Tibetan Plateau, the site does exhibit during specific months large contributions associated with transported "aged" air masses associated with stratospheric intrusions." I do not wish to impose this interpretation on the authors, but rather elicit from them if this is what they are attempting to say. If not, could they please provide a concise sentence that clearly describes their conclusion on the importance of stratospheric intrusions associated with long-range transport in enhancing the surface ozone concentrations at Nam Co. I think this would help the reader.

Response: Thanks for your comment. We rewrote this sentence as follows (lines 423 - 426):

“The analysis suggests that stratospheric intrusions account for around 20% of the variability in surface ozone concentrations at Nam Co Station. Further analysis of tropopause folding suggest that Nam Co Station is affected by “aged” air masses associated with stratospheric intrusions transported from the southern and northern Tibetan Plateau, mainly during the spring and the summer, respectively”.

21. Supplement: Fig. S1. I would suggest improving the readability of the title of the x-axis (Year-Month-Day-Hour). It seems to not be clear on my copy. Does the first symbol in the time series identified as 2011-01-01 in Fig. S1 represent the January average or just the 2011-01-01 point? I am not sure what the first dot represents. The meaning of the first dot is confusing.

Response: Thanks for your suggestion. We added a new version of Fig. S1 in manuscript as follows:

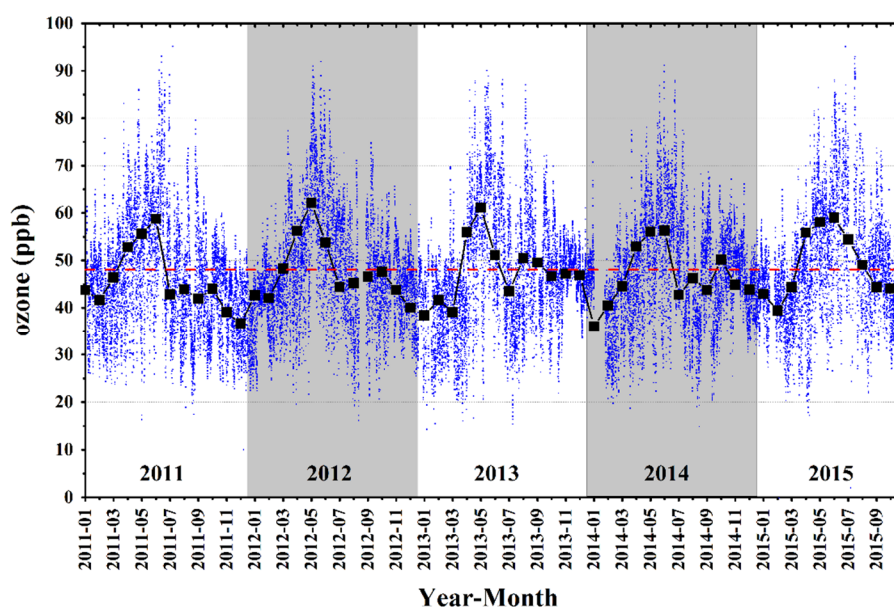


Fig. S1. Variation of surface ozone at Nam Co Station from January 2011 to October 2015. Hourly mean mixing ratios of surface ozone are in blue dots; monthly mean mixing ratios of surface ozone are in black squares; average mixing ratio of surface ozone during whole measurement period in red dash line.

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