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

Authors: Xiufeng Yin, Shichang Kang, Benjamin de Foy, Zhiyuan Cong, Jiali Luo, Lang Zhang, Yaoming Ma, Guoshuai Zhang, Dipesh Rupakheti, Qianggong Zhang

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 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 (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".

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.

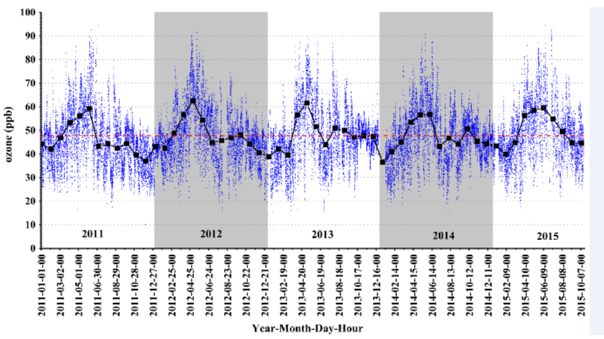

Fig. S1. Variation of surface ozone at Nam Co Station from January 2011 to October 2015. Hourly mean mixing ratios of surface ozone are in blue dots; monthly mean mixing ratios of surface ozone are in black dots; average mixing ratio of surface ozone during whole measurement period in red dash line.

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 r2 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 r2 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 (r2) 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 r2 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:

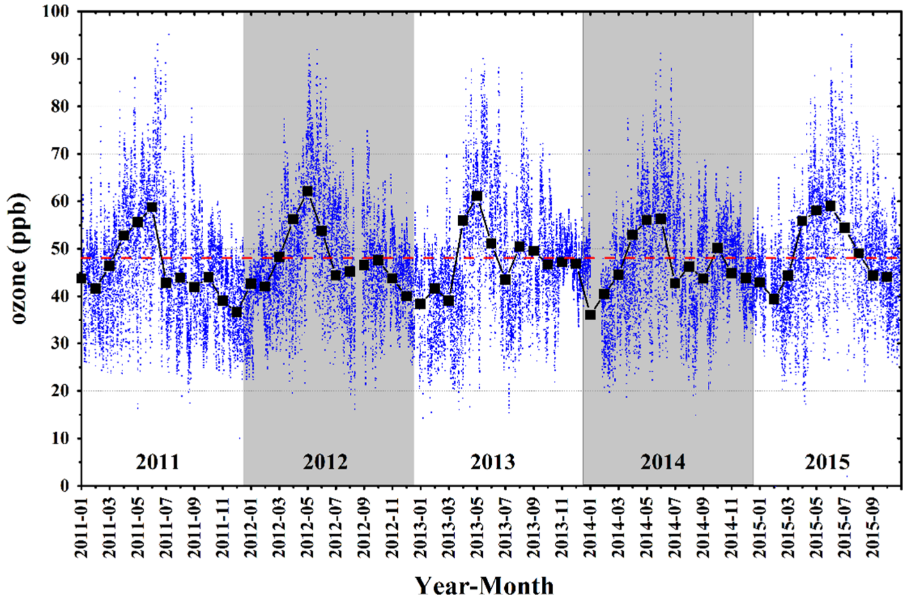

Fig. S1. Variation of surface ozone at Nam Co Station from January 2011 to October 2015. Hourly mean mixing ratios of surface ozone are in blue dots; monthly mean mixing ratios of surface ozone are in black squares; average mixing ratio of surface ozone during whole measurement period in red dash line.

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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).

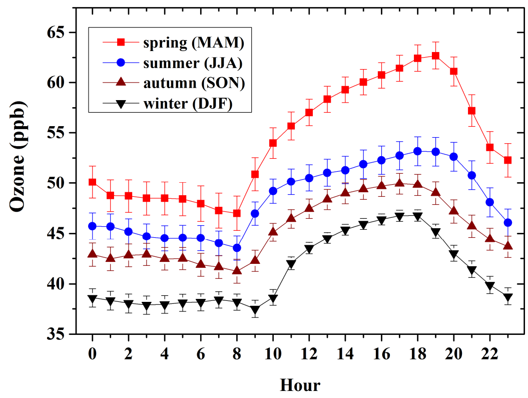

Fig. 3. Diurnal profiles of average hourly surface ozone at Nam Co Station by seasons. Error bars are 95% confidence levels.

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)".

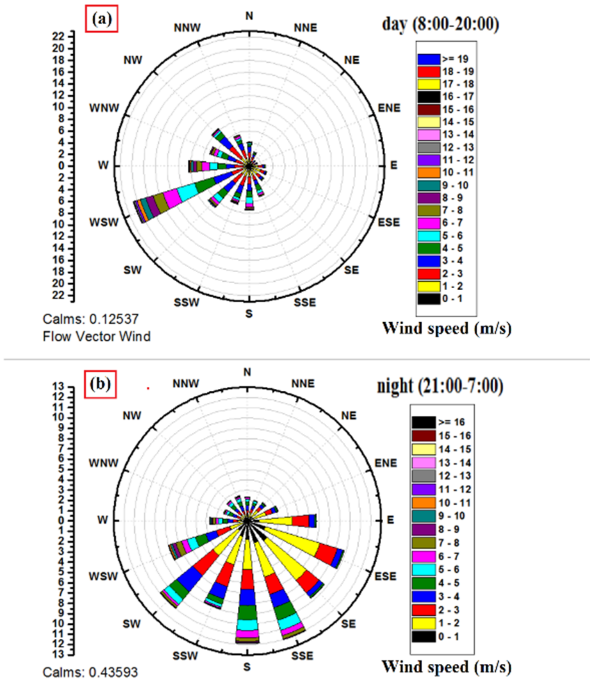

Fig. S7 Wind ross at Nam Co Station during the day (a) and at night (b).

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.

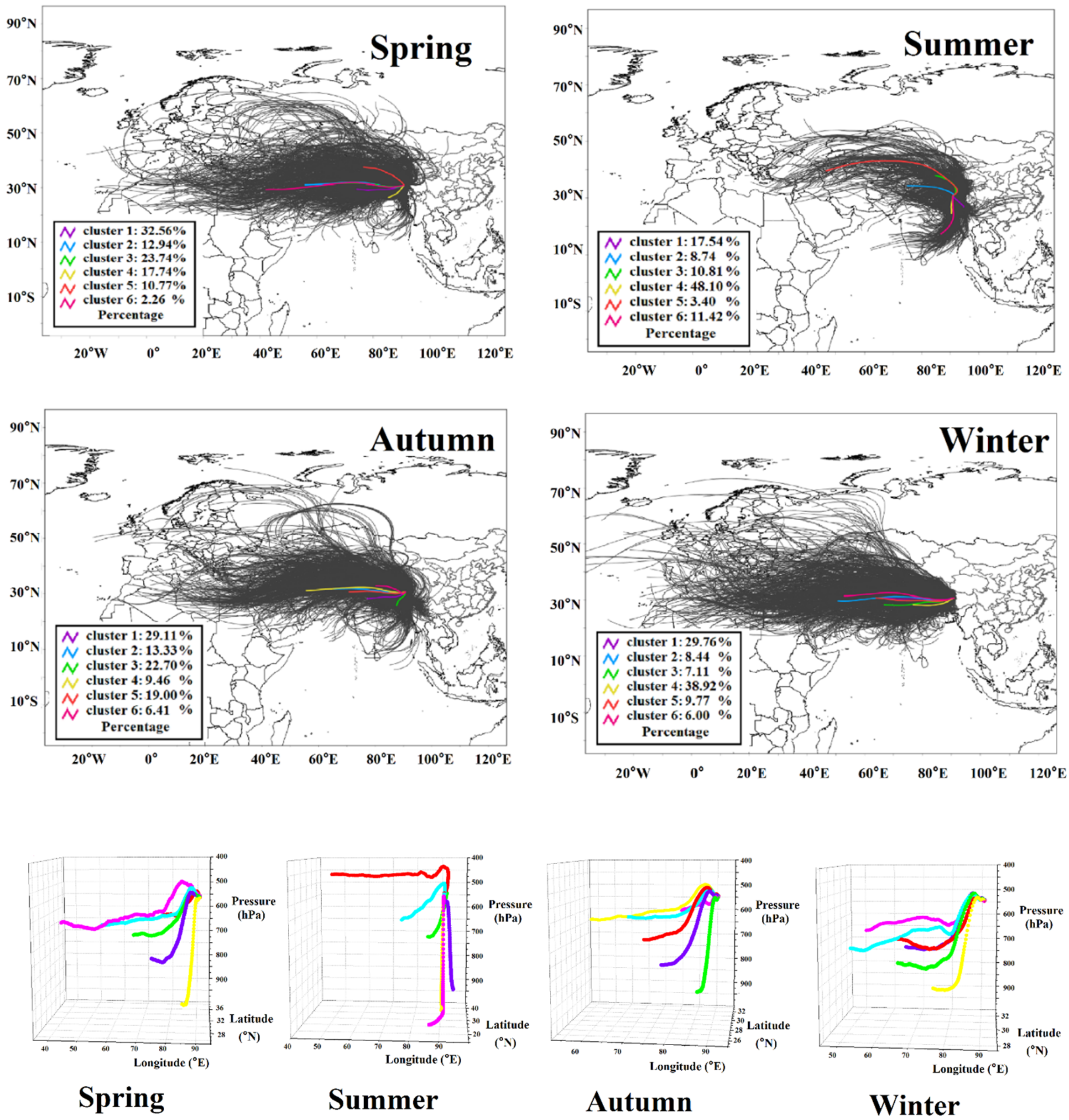

Fig. 10. Backward HYSPLIT trajectories for each measurement day (black lines in the maps), and mean back-trajectory for 6 HYSPLIT clusters (colored lines in the maps, 3D view shown on the right of the maps) arriving at Nam Co Station by season.

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

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Abstract:

Ozone is an important pollutant and greenhouse gas, and tropospheric ozone variations are generally associated with both natural and anthropogenic processes. As one of the most pristine and inaccessible regions in the world, the Tibetan Plateau has been considered as an ideal region for studying processes of the background atmosphere. Due to the vast area of the Tibetan Plateau, sites in the southern, northern and central regions exhibit different patterns of variation in surface ozone. Here, we present long-term measurements for ~5 years (January 2011 to October 2015) of surface ozone mixing ratios at Nam Co Station, which is a background site in the inland Tibetan Plateau. An average surface ozone mixing ratio of 47.6±11.6 ppb was recorded, and a large annual cycle was observed with maximum ozone mixing ratios in the spring and minimum ratios during the winter. The diurnal cycle is characterized by a minimum in the early morning and a maximum in the late afternoon. Nam Co Station represents a background region where surface ozone receives negligible local anthropogenic emissions. Surface ozone at Nam Co Station is mainly dominated by natural processes involving photochemical reactions, vertical mixing and stratospheric air mass downward transport. 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. By comparing measurements at Nam Co Station with those from other sites in the Tibetan Plateau, we aim to expand the understanding of ozone cycles and transport processes over the Tibetan Plateau. This work may provide a reference for model simulations in the future.

1 Introduction

The concentration of ozone in the troposphere showed sustained growth during the 20th century due to the increased emissions of anthropogenic precursors (Cooper et al., 2014). 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). In addition, ozone is a major precursor of hydroxyl (OH) and hydroperoxy (HO_2) radicals and it controls the oxidation capacity of the atmosphere (Brasseur et al., 1999). Furthermore, as the third most important greenhouse gas (after carbon dioxide (CO_2) and methane (CH_4)), tropospheric ozone contributes to global warming and has an estimated globally average radiative forcing of 0.40 ± 0.20 W m^{-2} with high confidence level (Myhre et al., 2013). 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).

The origin of tropospheric ozone and its temporal variation varies from site to site. Historically, the stratosphere was initially thought to be the main source of surface (tropospheric) ozone and a network of surface ozone monitoring sites was proposed (Junge, 1962). In the 1970s and 1980s, the effect of photochemical reactions in the troposphere on surface ozone became well recognized (Chameides and Walker, 1973; Crutzen, 1974) and photochemistry was identified as the dominant source of tropospheric ozone at some sites, as supported by models (Wu et al., 2007). 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. The study of surface ozone at background sites may enrich the understanding of surface ozone variation patterns.

Due to its small human population and low level of industrialization, the Tibetan Plateau is an ideal natural laboratory for studying surface ozone across remote regions of the Eurasian continent. Long term surface ozone measurements over the Tibetan Plateau have been conducted at Mt. Waliguan (northeast edge of the Tibetan Plateau) since 1994 (Xu et al., 2016), the Nepal Climate Observatory at Pyramid (NCO-P) which operates on the southern slope of the Himalayan region since 2006 (Cristofanelli et al., 2010) and the Xianggelila Regional Atmosphere Background Station at the southeastern rim of the Tibetan Plateau since 2007 (Ma et al., 2014). Analysis of long-term ozone mixing ratios at Waliguan Station has revealed steadily increasing concentrations over the past two decades (Xu et al., 2016) and has shown that maximum surface ozone occurs during the summer (Zhu et al., 2004). At NCO-P and Xianggelila, surface ozone maximum was observed in the spring (Cristofanelli et al., 2010; Ma et al., 2014). It is noteworthy that these three monitoring sites are on the boundaries of the Tibetan Plateau. In the vast inland area of the Tibetan Plateau, surface ozone measurements were only reported from Lhasa and Dangxiong for one year and two years, respectively. These measurements might be less representative of regional surface ozone variation due to their proximity to human settlements and relatively short duration of the measurements (Ran et al., 2014; Lin et al. 2015). The paucity of long-term surface ozone observations in the Tibetan Plateau, especially in the inland region, limits our understanding of the regional background ozone level and the factors that influence it and can potentially lead to inaccurate simulation of surface ozone variation over the Tibetan Plateau.

Surface ozone mixing ratios were monitored for ~5 years (January 2011 to October 2015) at Nam Co Station on the shore of Nam Co Lake (30°30′-30°56′N, 90°16′-91°03′E). In this study, we investigated the seasonal and diurnal variations of surface ozone and its influencing factors. We then evaluated surface ozone variability using combined observations over the Tibetan Plateau and beyond. Finally, we discussed the potential representativeness of surface ozone at Nam Co Station as the regional background of surface ozone in the inland Tibetan Plateau. 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.

2 Measurements and Methods

2.1 Measurement site

The Tibetan Plateau (27°N–45°N, 70°E–105°E, average elevation ~ 4 km) is the highest and most extensive highland in the world and has been called the 'Third Pole' (Yao et al., 2012). The Nam Co Comprehensive Observation and Research Station (hereafter referred to as the Nam Co Station, 30°46.44′N, 90°59.31′E, 4730 m a.s.l.) is a high altitude scientific research center located between the southeast shore of Nam Co Lake (1 km from the station) and the foothills of the northern Nyainqêntanglha Mountains (15 km from the station) in the southern-central region of the Tibetan Plateau (Fig. 1). Nam Co Station was established to monitor atmospheric conditions in September 2005 and provided a long-term record of the atmospheric environment in the Tibetan Plateau (Kang et al., 2011). Nam Co Station is in a natural flat field (220 × 100 m) and records meteorological, ecological, and atmospheric data, including surface ozone mixing ratios (Cong et al., 2007; Li et al., 2007; Huang et al., 2012; Liu et al., 2015; de Foy et al., 2016a). The climate at Nam Co Station is dry and cold, representing a typical climate regime in the high mountain region. The solar radiation at Nam Co Station is stronger than that at other sites at the same latitude due to the high altitude and thin air. Three synoptic systems influence the atmosphere at Nam Co Station: the South Asian anticyclone (which controls the 100-hPa upper layer), a subtropical high-pressure system, and southeast warm and wet airflow (during the monsoon season) (Qiao and Zhang, 1994). No major anthropogenic sources of atmospheric emissions exist near Nam Co Station. The urban area closest to the station is Dangxiong County, which is located on the southern slopes of the Nyainqêntanglha Mountain Range approximately 60 km south of Nam Co. Dangxiong is lower in elevation than Nam Co Station by more than 500 m. No large industries are located within 100 km of Nam Co Station. Local traffic is limited to a small number of vehicles traveling through the area during the tourist season.

2.2 Measurements: surface ozone and meteorology

The surface ozone mixing ratios were measured using a UV photometric instrument (Thermo Environmental Instruments, USA, Model 49i), which uses absorption of radiation at 254 nm and has a dual cell design. The ambient air inlet (Teflon tube) was 1.5 m above the roof and 4 m above the ground. The instrument has zero noise, 0.25 parts per billion (ppb) RMS (root mean square error) (60 s average time), a low detection limit of 0.5 ppb, a precision of 1 ppb and a response time of 20 s (10 s lag time). The instrument was calibrated using a 49i-PS calibrator (Thermo Environmental Instruments, USA) before measurements and during the monitoring periods and yearly instrument calibrations are performed against the Standard Reference Photometer (SRP) maintained by the WMO World Calibration Centre in Switzerland (EMPA). Field operators checked the instruments and created a monitoring log file every day. Due to the extreme winter weather that occurs at Nam Co Station, measurements were intermittently interrupted because of unstable power supply (due to damage from strong winds to the electrical wires) and equipment maintenance. All data displayed in this study are in UTC+8 (Beijing Time, BJT), and solar noon at Nam Co Station is at 13:56 in UTC+8.

Measurements of temperature, relative humidity, wind speed, wind direction and downward shortwave radiation (SWD) were conducted at Nam Co Station using an automatic weather station system (Milos520, Vaisala) and a radiation measurement system (CNR-1) (Ma et al., 2008).

2.3 Meteorological simulations

Backward trajectories and clusters were calculated by NOAA-HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Rolph, 2003, http://ready.arl.noaa.gov/HYSPLIT.php) using TrajStat, which is a free software plugin of MeteoInfo (Wang, 2014). Gridded meteorological data for backward trajectories in HYSPLIT and Planetary boundary layer height (PBLH) 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 in HYSPLIT 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 the 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.

To identify the impact of different air masses in a multiple linear regression model, WRF-FLEXPART (Stohl et al., 2005; Brioude et al., 2013) was used to obtain the clusters of particle trajectories reaching the Nam Co Station. 1000 particles were released per hour in the bottom 100 m surface layer above Nam Co Station and were tracked in backward mode for 4 days (de Foy et al., 2016a). Residence Time Analysis (RTA) (Ashbaugh et al., 1985) was used to create gridded fields representing the dominant transport paths of air masses impacting the measurement site (Wang et al., 2016; Wang et al., 2017). A k-means algorithm was used to classify the transport patterns into clusters (Wang et al., 2016). Six clusters were found to represent the dominant flow patterns to the Nam Co Station simulated using WRF-FLEXPART.

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.

The ECMWF ERA-Interim data (Dee et al., 2011) was used to analyze the upper troposphere and lower stratosphere structures of the meridional cross-section over Nam Co Station.

2.4 Multiple Linear Regression Model

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).

2.5 Potential Source Contribution Function

The Potential Source Contribution Function (PSCF) assumes that back-trajectories arriving at times of higher mixing ratios likely point to the more significant pollution directions (Ashbaugh et al., 1985). PSCF has been applied in previous studies to locate air masses associated with high levels of surface ozone for different sites (Kaiser et al., 2007; Dimitriou and Kassomenos, 2015). In this study, PSCF was calculated by using trajectories which were calculated by HYSPLIT. The top of the model was set to 10000 m. The PSCF values for the grid cells in the study domain are based on a count of the trajectory segment (hourly trajectory positions) that terminate within each cell (Ashbaugh et al., 1985). Let n_{ij} be the total number of endpoints that fall in the *ij*th cell during whole simulation period. Let m_{ij} represents the number of points in the same cell that have arrival times at the sampling site corresponding to surface ozone mixing ratios higher than a set criterion. In this study, we calculate the PSCF based on trajectories corresponding to concentrations that exceed the mean level of surface ozone. The PSCF value for the *ij*th cell is then defined as:

$$\mathrm{PSCF}_{ij} = \frac{m_{ij}}{n_{ij}}$$

The PSCF value can be interpreted as the conditional probability that the ozone mixing ratios at measurement site is greater than the mean mixing ratios if the air parcel passes though the ijth cell before arriving at the measurement site. 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. These cells are indicative of areas of 'high potential' contributions for the chemical constituent.

Identical $PSCF_{ij}$ values can be obtained from cells with very different counts of back-trajectory points (e.g. grid cell A with m_{ij}=5000 and n_{ij}=10000 and grid cell B with m_{ij} = 5 and n_{ij} = 10). In this extreme situation grid cell A has 1000 times more air parcels passing through than grid cell B. Because of the sparse particle count in grid cell B, the PSCF values are more uncertain. To account for the uncertainty due to low values of n_{ij}, the PSCF values were scaled by a weighting function W_{ij} (Polissar et al., 1999). The weighting function reduced the PSCF values when the total number of the endpoints in a cell was less than about three times the average value of the end points per each cell. In this case, W_{ij} 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 N_{ave} represents the mean n_{ij} of all grid cells. The weighted PSCF values were obtained by multiplying the original PSCF values by the weighting factor.

3 Surface ozone behavior at Nam Co Station

3.1 Mean mixing ratio

The mean surface ozone mixing ratio at Nam Co Station during the entire observational period was 47.6 ± 11.6 ppb, and the yearly average surface ozone mixing ratio was between 46.0 and 48.9 ppb (Table 1). During the whole monitoring period, the lowest hourly mixing ratio at Nam Co Station was 10.1 ppb, which was observed on December 3rd, 2011; and the highest hourly mixing ratio was 94.7 ppb, which was recorded on June 11th, 2011, resulting in a range of ~85 ppb.

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). 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).

3.2 Seasonal pattern

Every month considered in this study had more than 400 hours of available data (valid data for each month >56%). The overall trends of surface ozone at Nam Co Station showed similar annual cycles with slight variations (Fig. S1). The monthly average mixing ratios of ozone from 2011 to 2015 at Nam Co Station showed clear seasonal features (Fig. 2): 1) remarkably high values in the late spring-early summer; 2) low values in the winter; 3) little fluctuation during the remainder of the year except for the late spring-early summer and 4) a small peak around October in the second half of the year. Three winter months (December, January and February) had the lowest monthly mean surface ozone mixing ratios (41.0±7.6 ppb – 41.5±7.0 ppb) of the year, with variations smaller than 0.5 ppb. Monthly mean surface ozone mixing ratios increased from February to March by ~3.5 ppb, and a sharp increase from 44.5±10.4 ppb to 54.7±11.6 ppb occurred in March-April. The monthly mean mixing ratios remained above 54 ppb for the next 3 months (April, May and June), with the highest monthly mean mixing ratios occurring in May (58.6±12.2 ppb). After a large decrease in June-July (from 55.5±12.7 ppb to 44.9±11.9 ppb), the monthly mean mixing ratios of surface ozone during the second half of the year remained at low levels (ranging from 41.5±7.0 ppb to 48.0±8.6 ppb), with a small increase in October.

3.3 Diurnal variation

The diurnal cycles at Nam Co Station showed low ozone mixing ratios at night and high ozone mixing ratios during the day, with a unimodal pattern. After a rapid increase during the morning (8:00-11:00) of 6 ppb, the surface ozone mixing ratio at Nam Co continued to increase until reaching a maximum at 18:00 (53.2±10.9 ppb); it then decreased continuously to its lowest level at 8:00 the next day. Field observations revealed that the ozone mixing ratios reached an average of 50.6±10.9 ppb during the day (9:00-20:00) and an average of 44.6±11.2 ppb during the night and early morning (21:00-8:00).

All seasons displayed similar diurnal ozone mixing ratio cycles at Nam Co Station (Fig. 3). The diurnal cycle shift from low level at night to high level during the daytime was generally characterized by early shifts in the spring and the summer and late shifts in the winter, which was most likely related to seasonal differences in sunrise times. Relatively large diurnal amplitudes were observed in the spring, with much smaller diurnal amplitudes observed during the summer, the autumn and the winter.

4 Factors affecting surface ozone variation at Nam Co Station

4.1 Impact of factors on seasonal variation

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 $18.2 \pm 2.6\%$ of the ozone variance at the site and the WRF-FLEXPART wind transport clusters (Fig. S3) contributed $6.5 \pm 1.7\%$. Local winds accounted for $31.0 \pm 1.8\%$, seasonal variations (including the 12 and 6-month sine and cosine terms, and the seasonal temperature and humidity terms) accounted for $35.3 \pm 3.0\%$, diurnal signals (including the hourly terms and the diurnal temperature and humidity signals) accounted for $7.4 \pm 0.8\%$, the annual signal for $1.5 \pm 0.5\%$ and the WRF boundary layer height for $0.1 \pm 0.1\%$ 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.

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.

4.2 Impacts of photochemical production and vertical mixing on diurnal variation

In the regression model, solar radiation was not selected as an input by the automatic procedure based on the improvement in the correlation coefficient of the model due to individual time series. The solar radiation time series does match the surface ozone concentration, but it was not as good as a match as local winds and the diurnal profile. This is probably due to the time delay between the maximum ozone concentration and the maximum solar radiation. Hourly average SWD showed a positive correlation with hourly average surface ozone (correlation coefficient=0.77) which indicated that the potential of local ozone formation by photochemical production during the daytime contributed to the peak in the afternoon (Wang et al., 2006). Wind speed and PBLH are also generally regarded as the main factors influencing the diurnal cycle of surface ozone. High wind speed was found to covary with turbulent downward mixing in previous studies in the Tibetan Plateau (Tang et al., 2002; Ma et al., 2014; Lin et al., 2015). 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). Hourly average wind speed and PBLH at Nam Co Station showed positive correlation with hourly average surface ozone (Fig. 7). The correlation coefficient between hourly average surface ozone and hourly average wind speed was 0.95 and the correlation coefficient between hourly average surface ozone and hourly average PBLH was 0.92. These results indicated that high level of surface ozone was associated with high wind speed and high PBLH.

5 Synthesis comparison of surface ozone variation across the Tibetan Plateau and beyond

5.1 Diurnal variation

Diurnal surface ozone patterns varied among sites across the Tibetan Plateau (Fig. 8). Nam Co Station, Xianggelila, Lhasa and Dangxiong showed similar diurnal surface ozone patterns as discussed in section 4.2.

Diurnal surface ozone at NCO-P showed different patterns in different seasons (Fig. 8), and thermal circulation was the main influential factor (Cristofanelli et al., 2010). Surface ozone mixing ratio at Waliguan experienced a minimum around noon and a maximum at night (Fig. 8), which is indicative of a mountain-valley breeze (local anabatic and catabatic winds) (Xue et al., 2011). Specifically, more boundary layer air affected Waliguan and resulted in lower surface ozone at noon; whereas at night, more air masses from free tropospheric increased the surface ozone level (Xu et al., 2011). It should be noted that the amplitudes in the diurnal variations at Waliguan were much smaller than those at other sites.

In all, diurnal surface ozone variations across the Tibetan Plateau were generally controlled by site-specific meteorological conditions and photochemical production. Sites located in plains or valleys exhibited daytime maxima of ozone associated with vertical mixing and photochemical production whereas mountain top sites experienced daytime ozone minima associated with up-slope flow of low-ozone air.

5.2 Seasonal variation

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). The seasonal variation of ozone at sites across the Tibetan Plateau and at the ridge of Himalayas can be divided into the Summer-maximum and Spring-maximum type based on the location of the sites:

A) The northern Tibetan Plateau: Summer-maximum type.

In the northern Tibetan Plateau (Waliguan site), surface ozone showed a maximum in the summer and a minimum in the winter (Fig. 9A). The summer maximum of surface ozone at Waliguan was linked to the impact of a high ozone band between 35°N-45°N over 70°E-125°E (Zhu et al., 2004). Similarly, Qinghai Lake site also showed a maximum in the summer (Shen et al., 2014). Horizontal and vertical transports have been regarded as major contributor to surface ozone at these two sites (Zhu et al., 2004; Shen et al., 2014).

B) The central Tibetan Plateau: Spring-maximum type.

Sites in the central Tibetan Plateau including Nam Co Station showed maximum ozone during the 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. It is possible that the local NOx emissions in these two urban regions reduce the average ozone on the urban scale. A study at Dangxiong revealed that the greater rainfall in the summer caused the surface ozone levels to remain relatively low during the warm period (July-September) (Lin et al., 2015). At Lhasa, photochemistry was the main factor affecting surface ozone in the spring and the summer, whereas transport largely contributed to the observed ozone mixing ratios in the autumn and the winter (Ran et al., 2014). Large-scale background of surface ozone in the spring considered an important influence on Dangxiong and Lhasa in the spring (Lin et al., 2015; Ran et al., 2014).

C) The southern Tibetan Plateau and the southern ridge of the Himalayas: Spring-maximum type.

In the southern Tibetan Plateau and the southern ridge of the Himalayas, Xianggelila and NCO-P each had a single surface ozone peak in the spring (pre-monsoon) and a minimum in the summer (monsoon) with a difference between the two exceeding 30 ppb. This pattern is different from those of the northern and central Tibetan Plateau (Fig. 9C). At NCO-P, frequent stratospheric intrusions were recorded in all seasons except during the monsoon season (Cristofanelli et al., 2010). A similar frequency of downward transport was identified at Xianggelila, including less frequent intrusions in the summer (Ma et al., 2014).

5.3 Backward trajectories and PSCF results of surface ozone at Nam Co Station

Backward trajectories and PSCF were utilized to identify the air masses associated with high levels of surface ozone at Nam Co Station and to assess the regional representativity of surface ozone at Nam Co. In the 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). 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 the spring, and slightly lower than during the winter. This was confirmed by analysis of the ERA-Interim data set showed that the seasonal average ozone flux from the stratosphere to the troposphere in the Himalayas was high in the spring (Škerlak et al., 2014). 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. In the summer, there are more backward trajectories coming from the northern Tibetan Plateau than in the other seasons (Fig. 10). During the summer, the northern Tibetan Plateau is the hot spot of stratosphere-to-troposphere ozone flux; and during autumn this flux remains higher than the one in the southern Tibetan Plateau (Škerlak et al., 2014). The summer peak of surface ozone at Waliguan also suggests that the northern Tibetan Plateau and northwestern China (a band between 35°N-45°N over 70°E-125°E) have their highest level of surface ozone in the summer (Zhu et al., 2004).

HYSPLIT backward trajectories arriving at Nam Co Station in the spring and the summer were classified in 6 clusters respectively (Fig. 11). In the spring, clusters which came from the southern Tibetan Plateau had higher mean surface ozone levels than clusters which came from the northern Tibetan Plateau. Air masses transported from the Himalayas therefore led to higher concentrations of surface ozone at Nam Co Station. The higher level of surface ozone at NCO-P (Cristofanelli et al., 2010) than at Nam Co Station in the spring may also be the result of this. 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.

Using PSCF, we have identified air masses associated with higher surface ozone at Nam Co Station in different seasons (Fig. 12) and throughout the measurement periods (Fig. S8). The Himalayas region to the south of Nam Co Station and South

Asian countries including Nepal, India Pakistan, Bangladesh and Bhutan had high PSCF weight values in both spring and summer. The large areas of northwestern China, including the northern Tibetan Plateau, were the regions in additional potential high PSCF weight values in the summer. The PSCF values at both the southern Tibetan Plateau and the northern Tibetan Plateau in the autumn were smaller than those in the spring and the summer. In the autumn, the inland Tibetan Plateau seems to have a larger impact on the study site than regions more on the edge of the Tibetan Plateau. In the winter, no obvious region was identified, which was likely due to low surface ozone mixing ratios in all these areas. Considering the results in section 4.3, PSCF probably picked up the contribution from STE as a signal from the south in the spring and from the north in the summer.

5.4 Implication for measurement and study of surface ozone in the inland Tibetan Plateau and beyond

The changes of the 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.

6 Summary

Surface ozone mixing ratios and meteorological parameters were continuously measured from January 2011 to October 2015 at Nam Co Station in the inland Tibetan Plateau. The inter-annual mixing ratios of surface ozone were stable with an average of 47.6±11.6 ppb throughout the monitoring period. The surface ozone mixing ratios at Nam Co Station were high in the spring and low in the winter. The diurnal cycle indicated that the ozone mixing ratio continued to increase after sunrise until sunset and was higher in the daytime than at night.

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.

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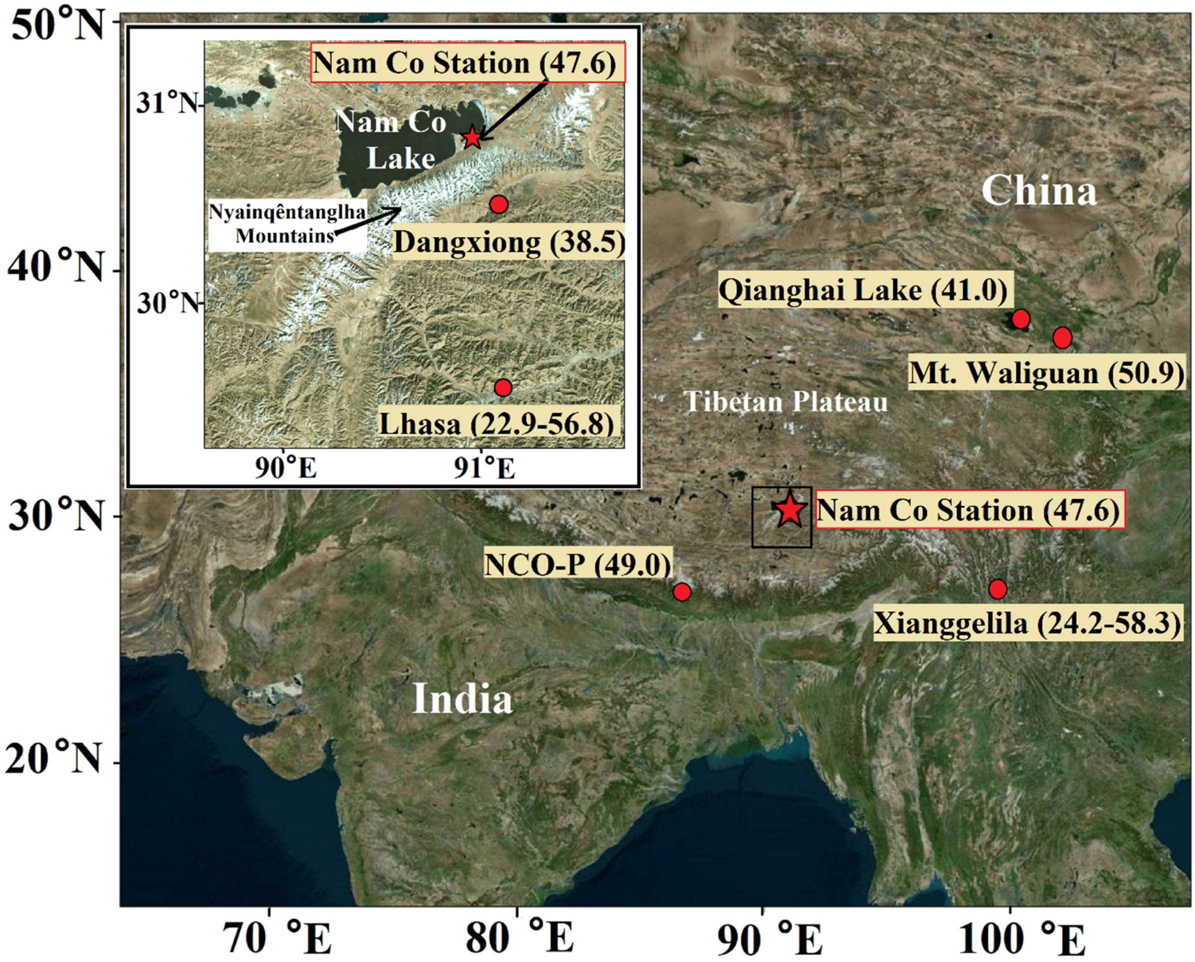


Fig. 1. Geographical location of Nam Co Station and other sites in the Tibetan Plateau. Values in the parenthesis refers to the average or range of surface ozone in ppb as obtained from Cristofanelli et al., 2010; Lin et al., 2015; Shen et al., 2014; Xu et al., 2011; Ma et al., 2014; Ran et al., 2014.

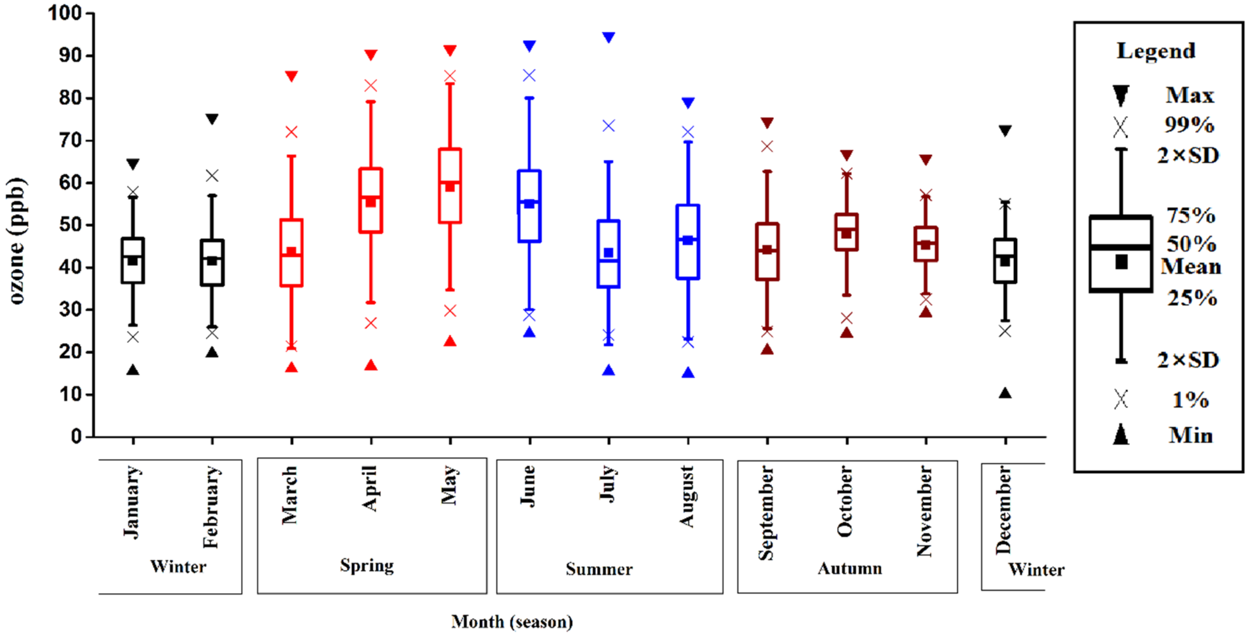


Fig. 2. Monthly average and statistical parameters of surface ozone at Nam Co Station during the whole measurement period (spring (MAM) in red; summer (JJA) in blue; autumn (SON) in dark red; winter (DJF) in black).

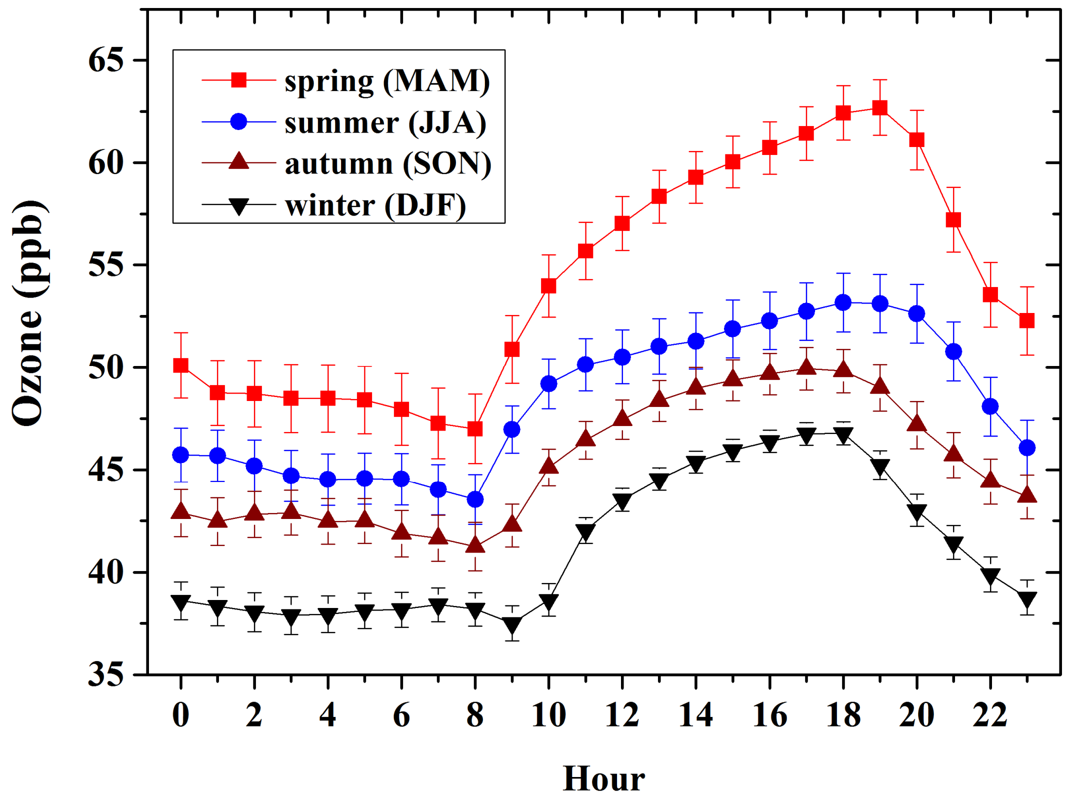


Fig. 3. Diurnal profiles of average hourly surface ozone at Nam Co Station by seasons. Error bars are 95% confidence levels.

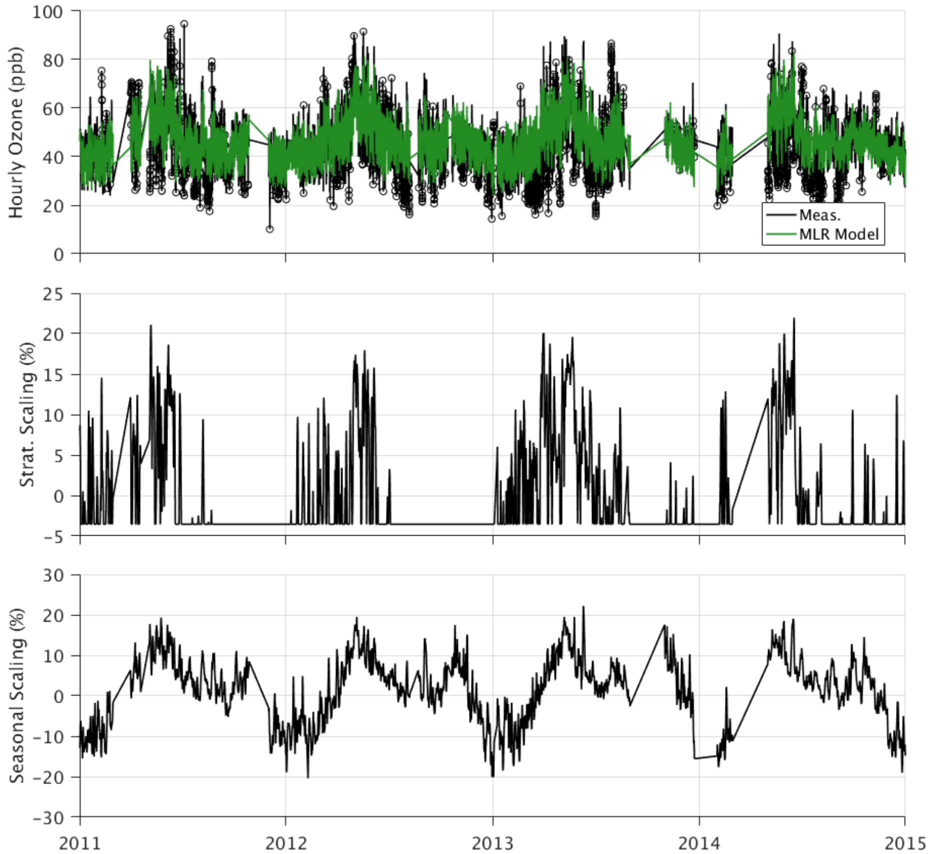


Fig. 4. Top: Surface hourly measurements of ozone at Nam Co (black) and multi-linear regression (MLR) model fit (green). Outliers rejected by the Iteratively Reweighted Least Squares Procedure are shown as circles. Middle: Scaling factor of the stratospheric ozone tracer simulated using CAMx. Bottom: Scaling factor due to the seasonal factors including the 12 and 6-month sine and cosines, and the seasonal temperature and specific humidity time series.

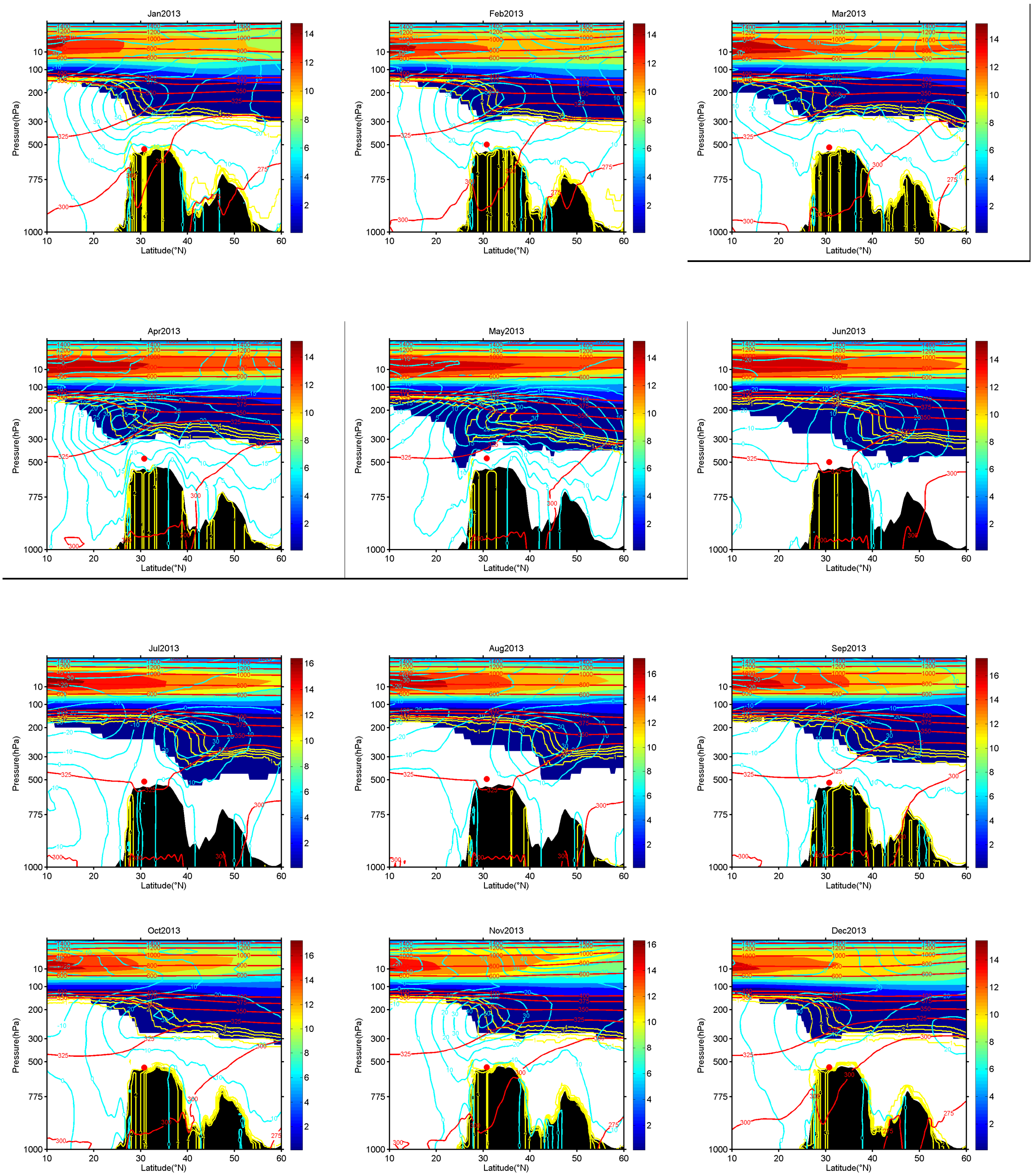


Fig. 5. Monthly mean meridional cross-section at 91° E (over Nam Co Station) at 20:00 UTC+8 in 2013, derived from ERA-Interim data, including zonal winds (cyan contours, m/s), potential vorticity (yellow lines, contours of 1, 2, 3, 4 potential vorticity unit), ozone (solid color, ×10^{6} kg/kg) and potential temperature (red contours, K). The color bar is the scale of ozone concentration. The area in black shows the cross section of the Tibetan Plateau terrain. The red dots show the position of the top of PBL.

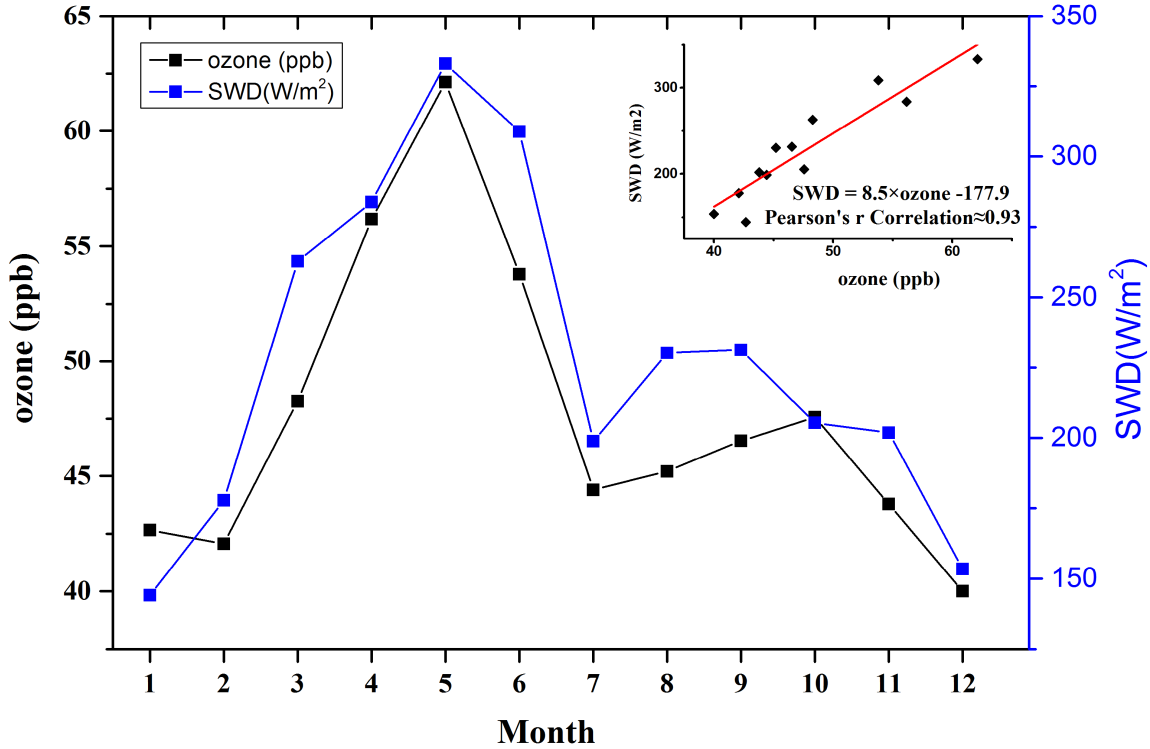


Fig. 6. Comparison between monthly average surface ozone (black) and monthly average SWD (downward shortwave radiation, blue) at Nam Co Station in 2012.

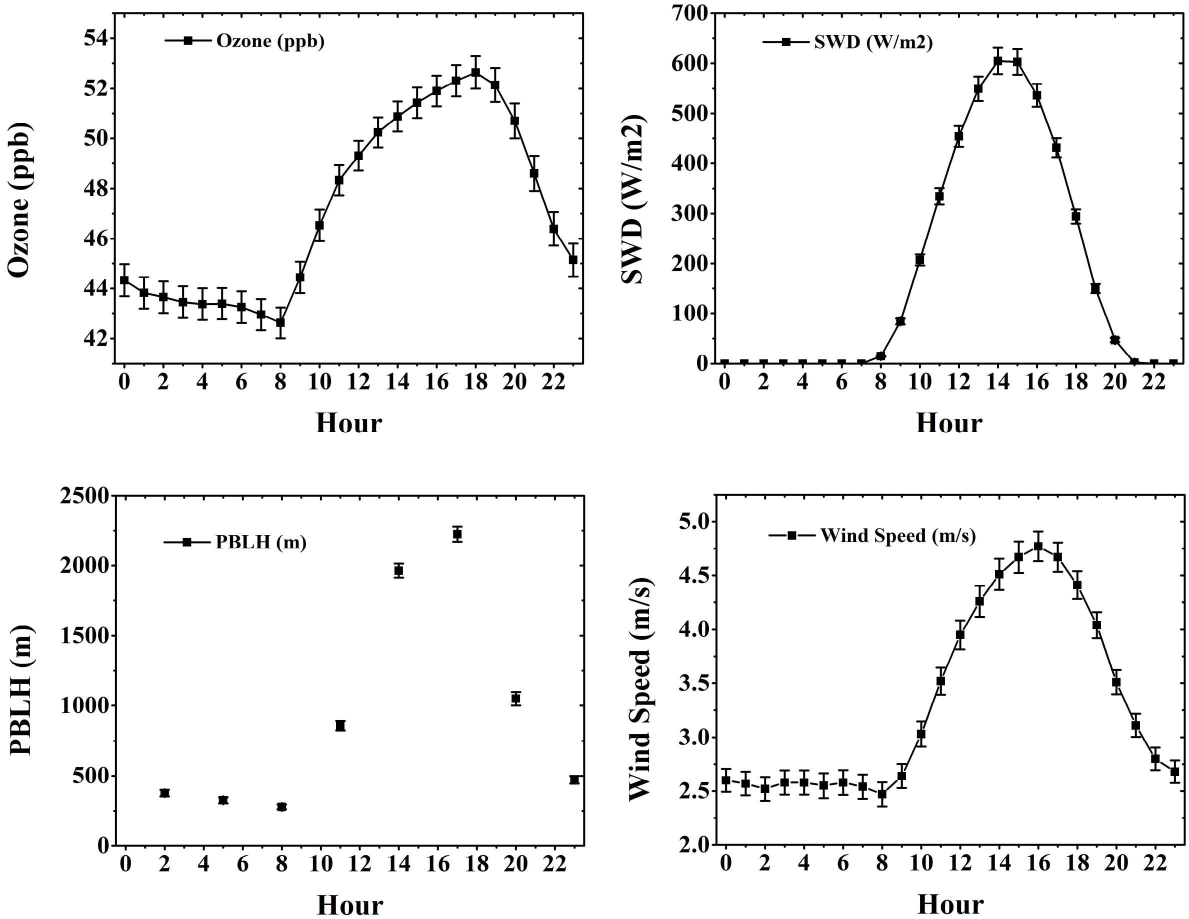


Fig. 7 Diurnal variations of hourly average of surface ozone, SWD (downward shortwave radiation), wind speed and PBLH (planetary boundary layer height) during the whole measurement period at Nam Co Station. Error bars are 95% confidence levels.

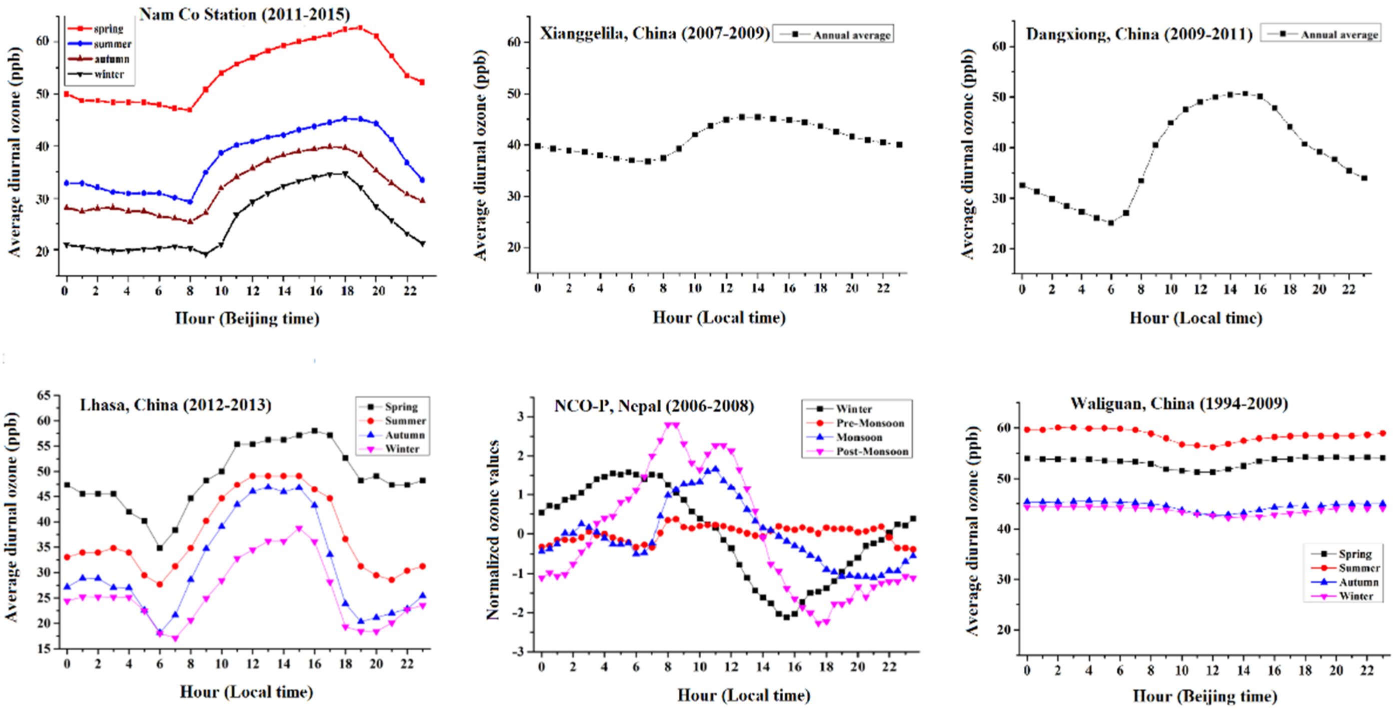


Fig. 8. Comparison of diurnal profiles of surface ozone concentration at different sites in the Tibetan Plateau (referred to Ma et al., 2014; Lin et al., 2015; Ran et al., 2014; Cristofanelli et al., 2010; Xu et al., 2011.) Measurement years at different sites are displayed in brackets.

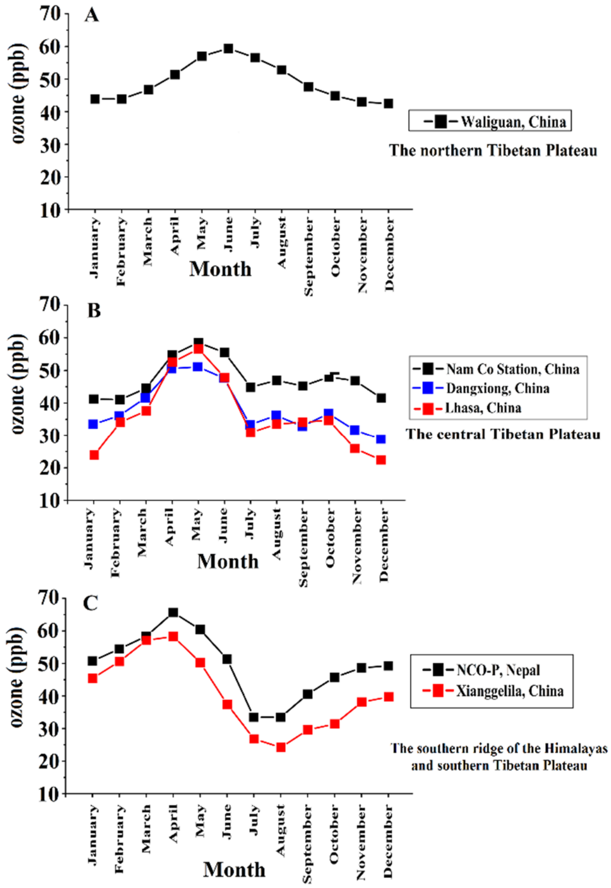


Fig. 9. Monthly variation of surface ozone at different sites in the Tibetan Plateau (right, A: The northern Tibetan Plateau: Summer-maximum type; B: The central Tibetan Plateau: Spring-maximum type and C: The southern Tibetan Plateau and the southern ridge of the Himalayas: Spring-maximum type) (referred to Ma et al., 2014; Lin et al., 2015; Ran et al., 2014; Cristofanelli et al., 2010; Zhu et al., 2004).

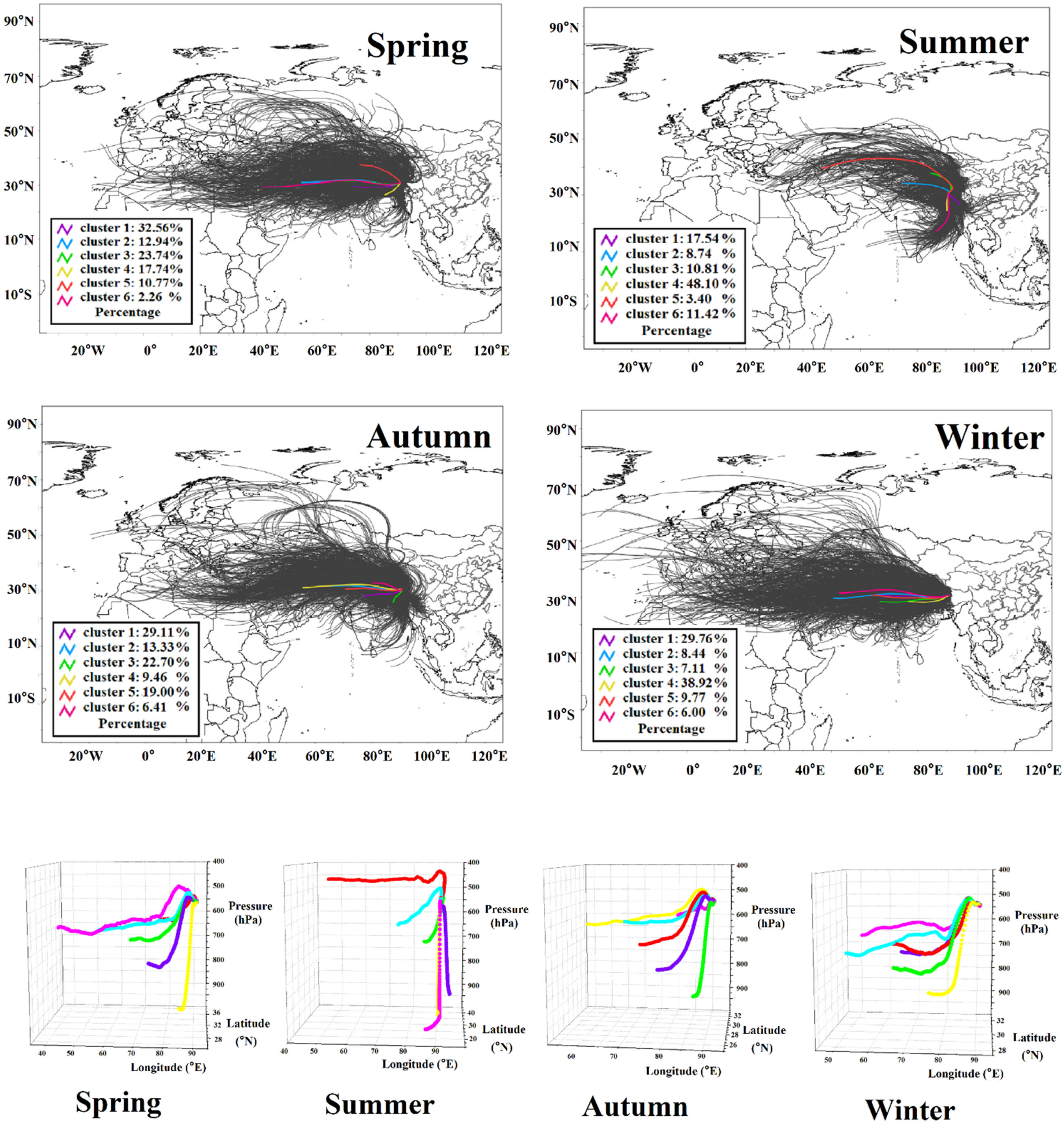


Fig. 10. Backward HYSPLIT trajectories for each measurement day (black lines in the maps), and mean back-trajectory for 6 HYSPLIT clusters (colored lines in the maps, 3D view shown on the right of the maps) arriving at Nam Co Station by season.

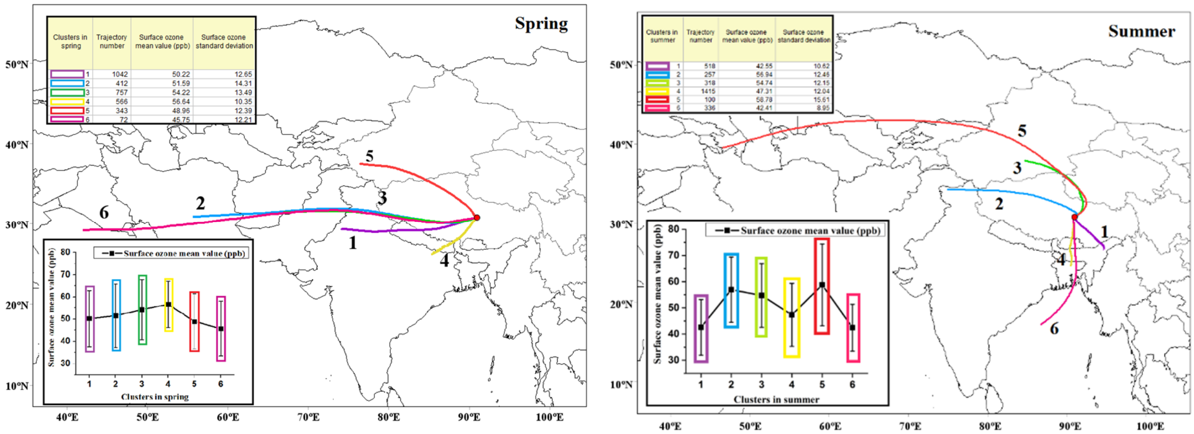


Fig. 11. Mean trajectory of 6 HYSPLIT clusters arriving at Nam Co Station in the spring and the summer. Subplot shows the range of surface ozone mixing ratios measured at Nam Co Station by cluster.

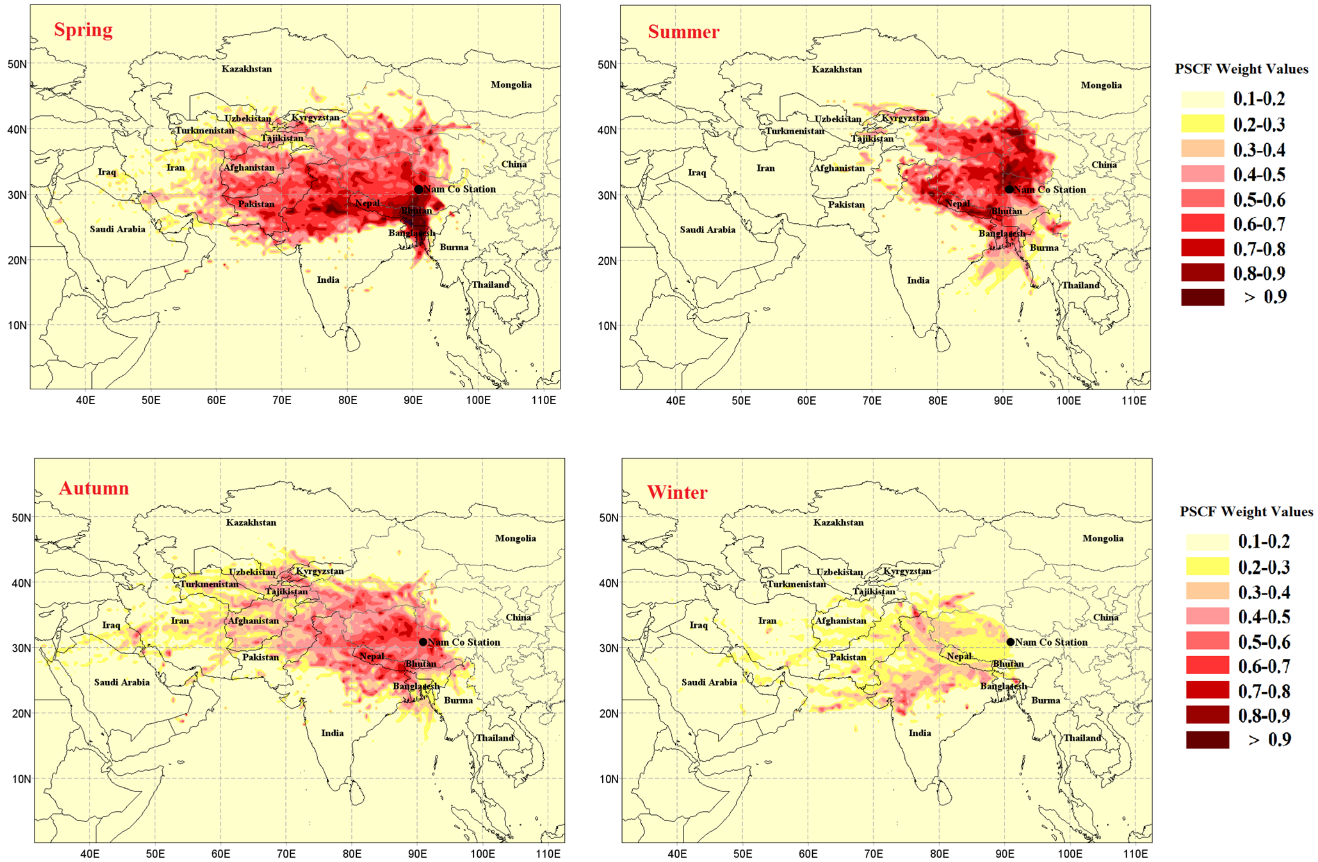


Fig. 12. Likely source areas of air mass associated with higher surface ozone concentrations at Nam Co Station by season identified using PSCF (Potential Source Contribution Function).

Table 1. Statistical summary of surface ozone at Nam Co from 2011 to 2015.

Year (valid time during whole year %)	Ozone (ppb)	Range (ppb)
2011 (75.25%)	46.0±12.1	10.1-94.7
2012 (90.30%)	48.1±11.4	14.3-91.5
2013 (75.90%)	47.5±12.3	15.5-89.7
2014 (70.05%)	47.5±10.6	14.9-90.8
2015 (66.21%)	48.9±12.0	17.3-94.7
Total	47.6±11.6	10.1-94.7

Table 2. Multi-Linear Regression Model for Hourly Ozone (2011-2014) for 3 different models.

Log, using CAMx Strat Tracer		Linear, using CAMx Strat Tracer		Log, using ERA-Interim PV	
No. All	27310	No. All	27310	No. All	27310
No. IRLS	26005	No. IRLS	25934	No. IRLS	25985
r (All)	0.77	r (All)	0.75	r (All)	0.75
r (IRLS)	0.81	r (IRLS)	0.79	r (IRLS)	0.80
Contribution to Variance (%) by Group					
CAMx Tracers	18.2	CAMx Tracers	12.5	PV	5.8
WRF-FLEXPART Clusters	6.5	WRF-FLEXPART Clusters	6.8	WRF-FLEXPART Clusters	6.4
Local Winds	31.0	Local Winds	28.6	Local Winds	29.4
Seasonal Signal	35.3	Seasonal Signal	44.2	Seasonal Signal	52.1
Diurnal Signal	7.4	Diurnal Signal	6.7	Diurnal Signal	5.7
Annual Signal	1.5	Annual Signal	0.7	Annual Signal	0.5
WRF PBLH	0.1	WRF PBLH	0.4	WRF PBLH	0.2

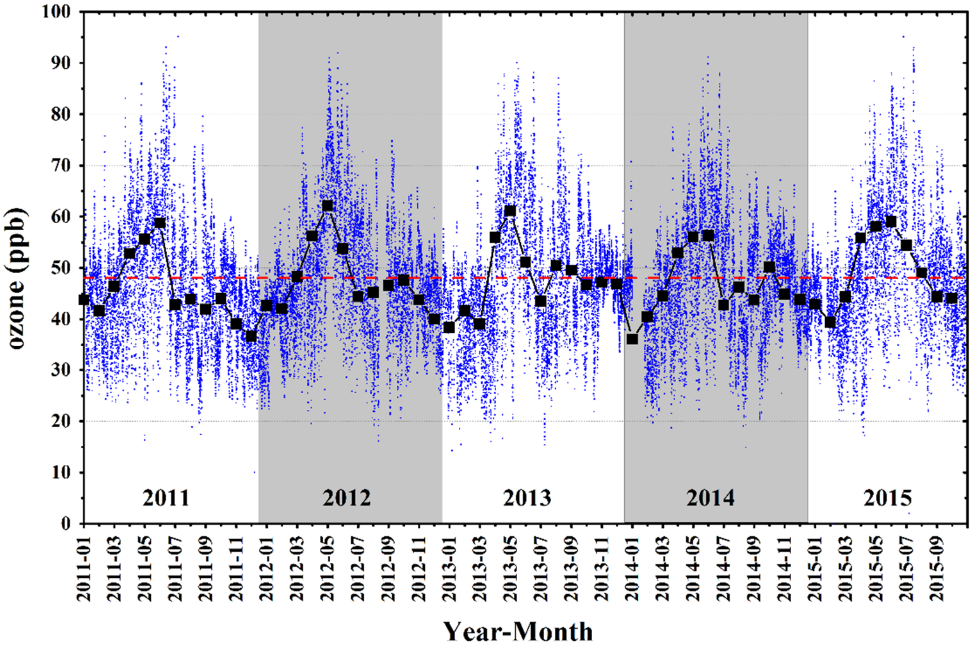
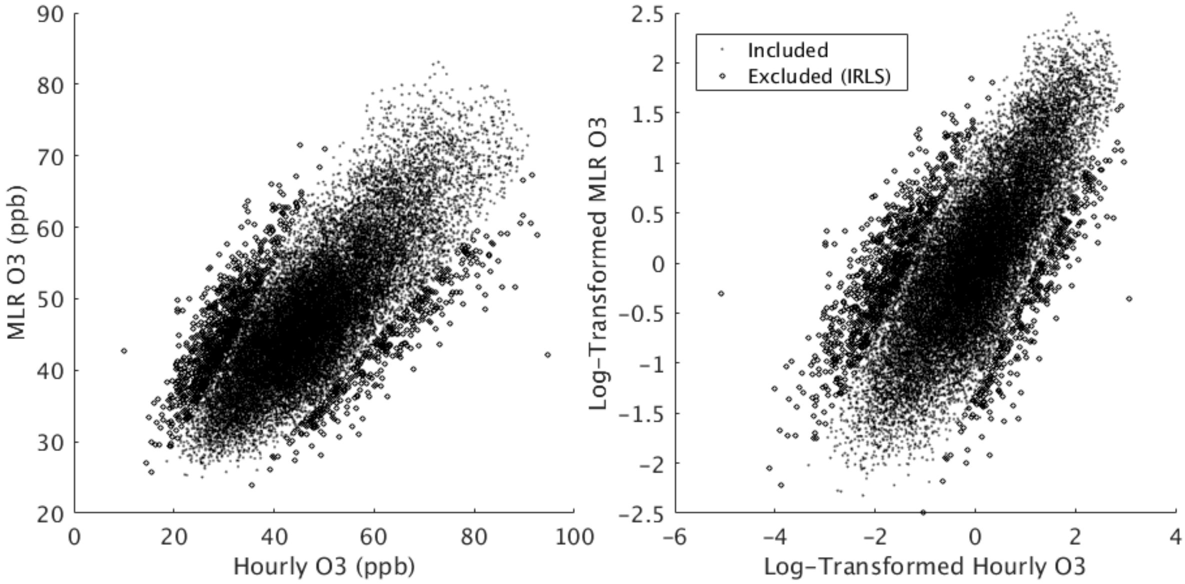


Fig. S1. Variation of surface ozone at Nam Co Station from January 2011 to October 2015. Hourly mean mixing ratios of surface ozone are in blue dots; monthly mean mixing ratios of surface ozone are in black squares; average mixing ratio of surface ozone during whole measurement period in red dash line.



965 **Fig. S2: Scatter plot of model surface ozone mixing ratio against observed surface ozone mixing ratio at Nam Co Station for the Multiple Linear Regression (MLR) model. The dots are points that are included in the regression; the circles are points that were excluded as outliers by the Iteratively Reweighted Least Squares (IRLS) method.**

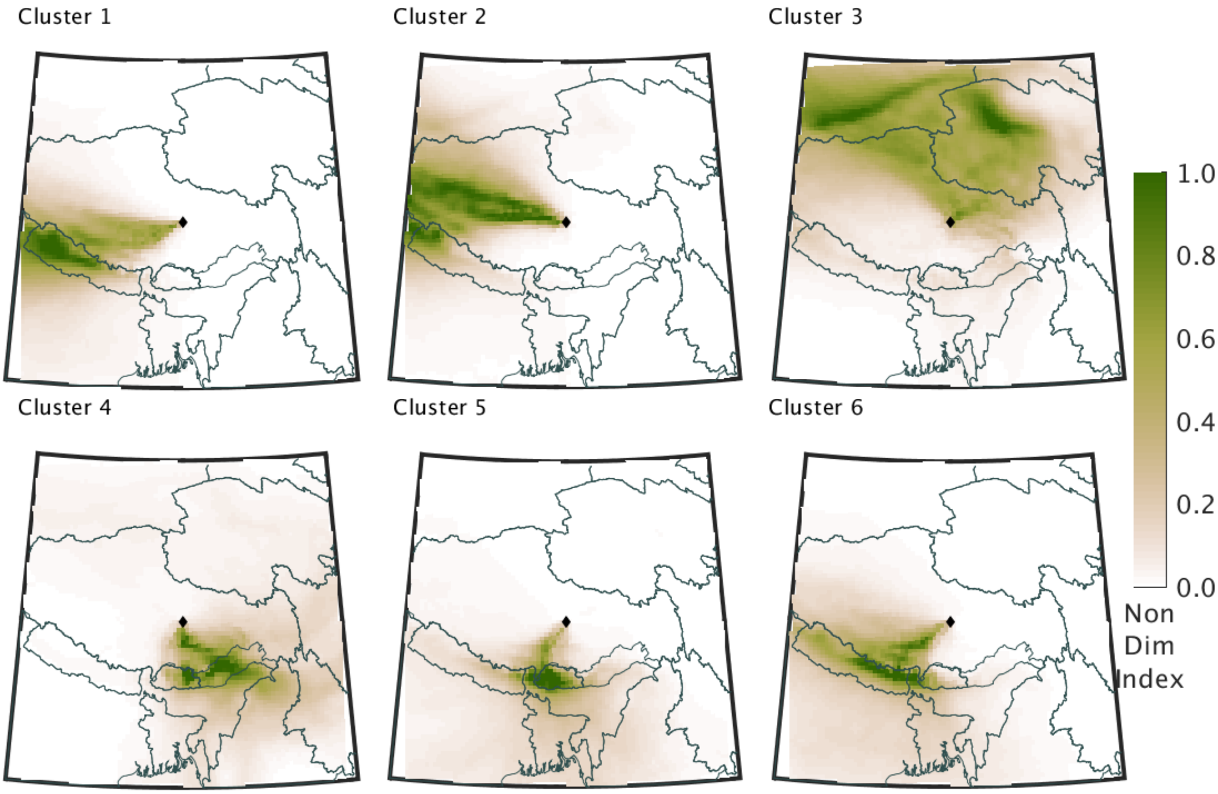


Fig. S3. Average Residence Time Analysis grids for each WRF-FLEXPART trajectory clusters at Nam Co Station. The black diamond represents the sampling site.

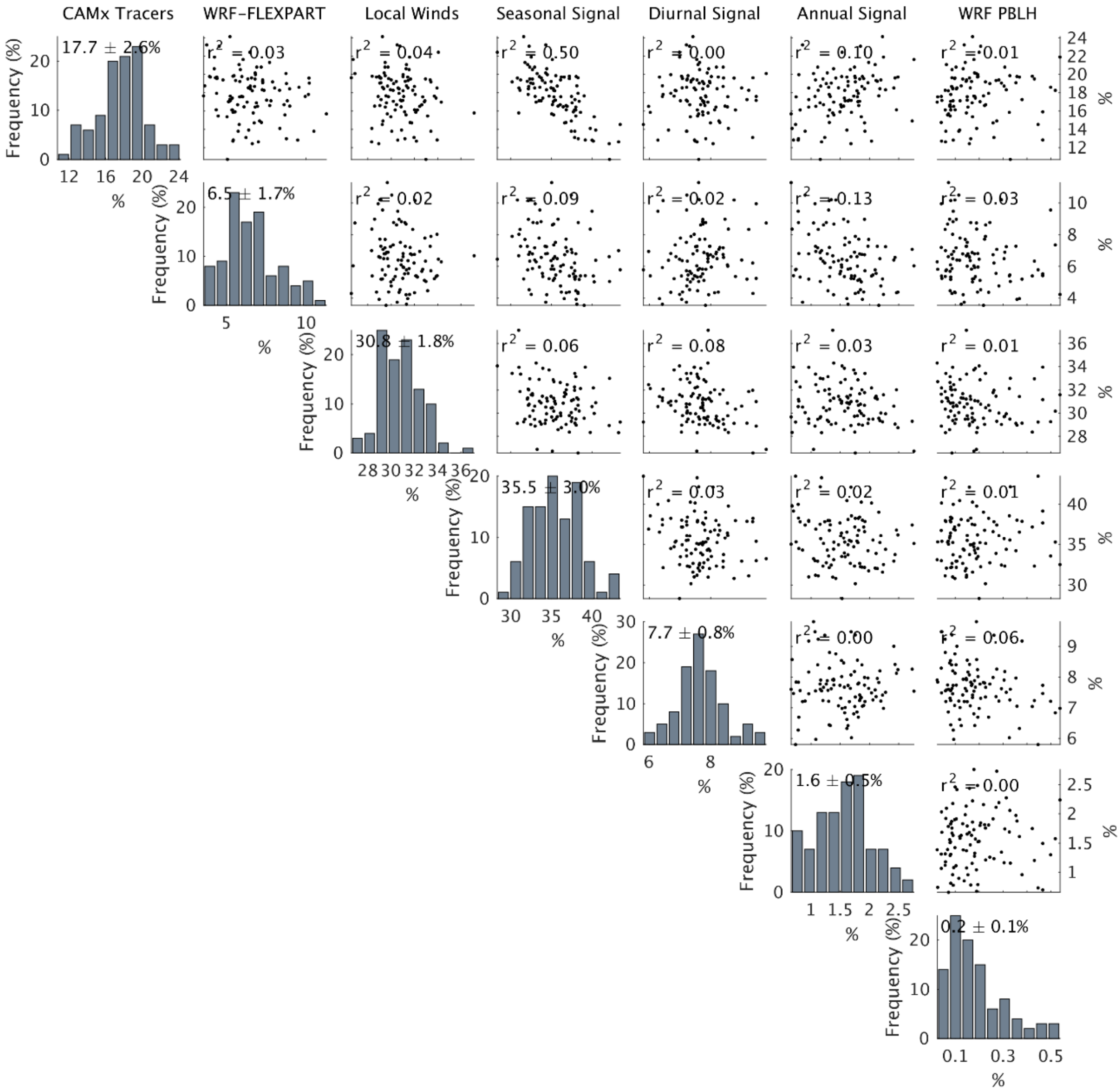


Fig. S4. Uncertainty and covariation of the Multi-Linear Regression estimates of the contribution to ozone variance by group, based on 100 realizations of the model using block-bootstrapping. Histograms show the distribution of the contribution estimates and the scatter plots show the cross-correlation of the estimates. Mean and standard deviation of the estimates are shown in the histogram, squared Pearson correlation coefficients are shown in the scatter plots.

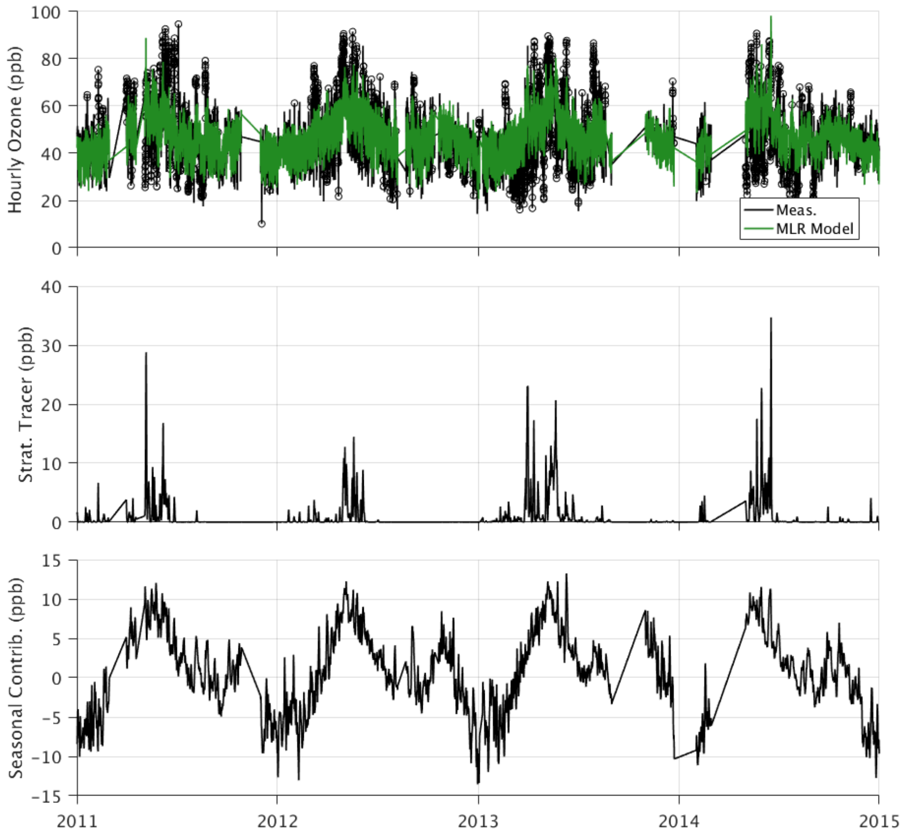


Fig. S5. Same as Fig. 4 but for the linear model. Because the model is linear, the contributions of the stratospheric tracer and the seasonal signal are in concentrations (ppb) rather than scaling factors.

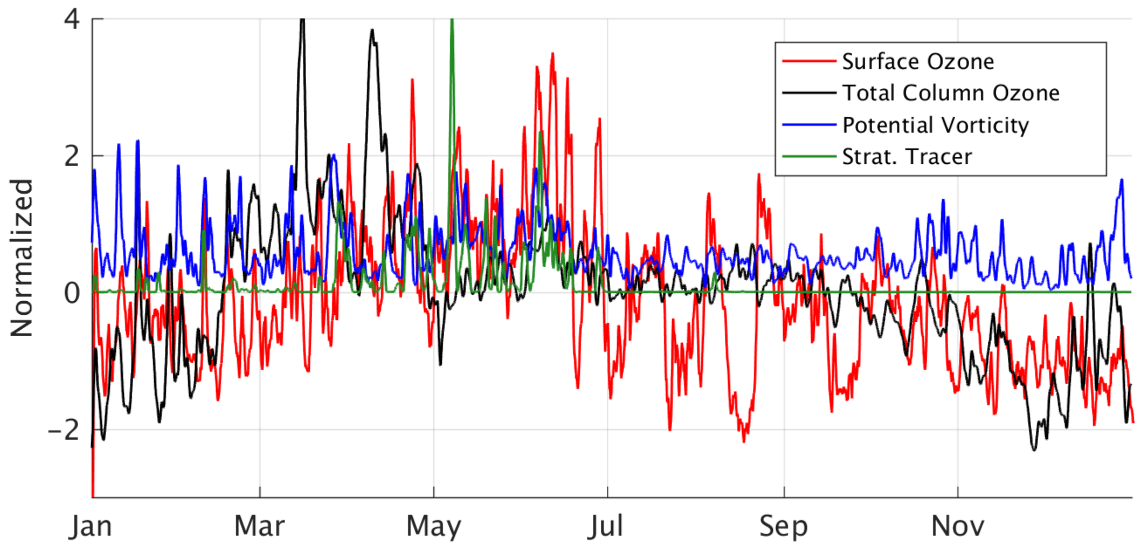


Fig. S6. Time series of 24-hour running average of surface ozone measured at Nam Co for 2011; ERA-Interim Total Column Ozone above the Himalayas, ERA-Interim Potential Vorticity at 350 hPa above the Himalayas (units of PVU); and stratospheric ozone tracer simulated by CAMx (units of 0.1 * ppb).

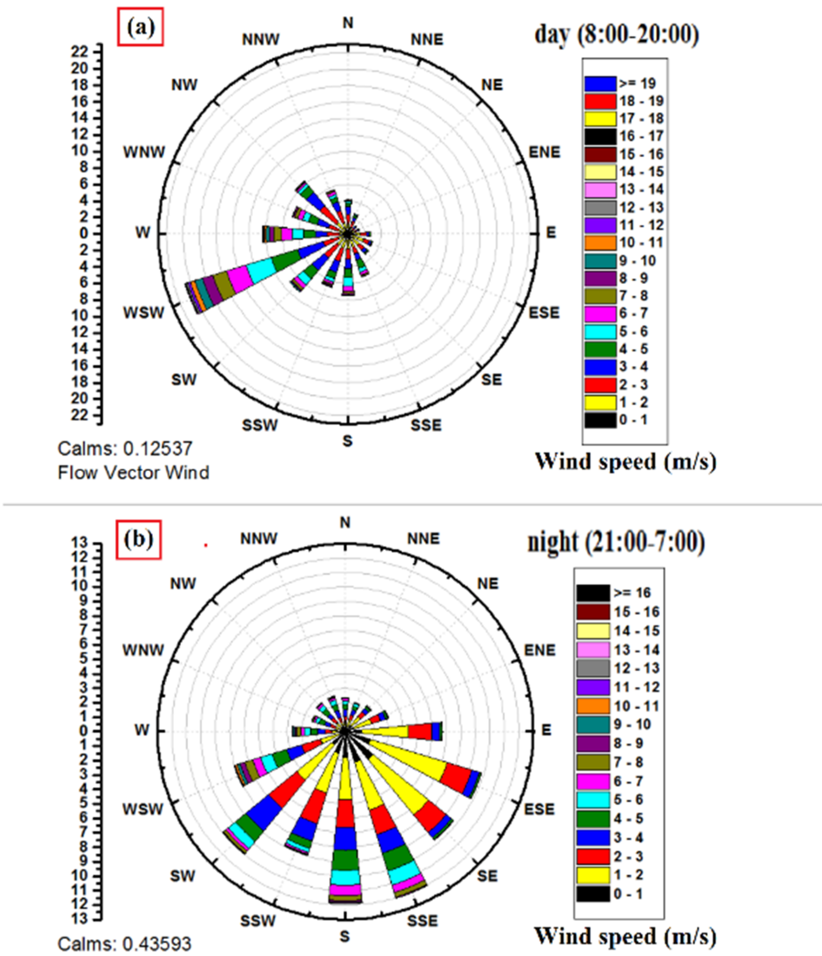


Fig. S7 Wind rose at Nam Co Station during the day (a) and at night (b).

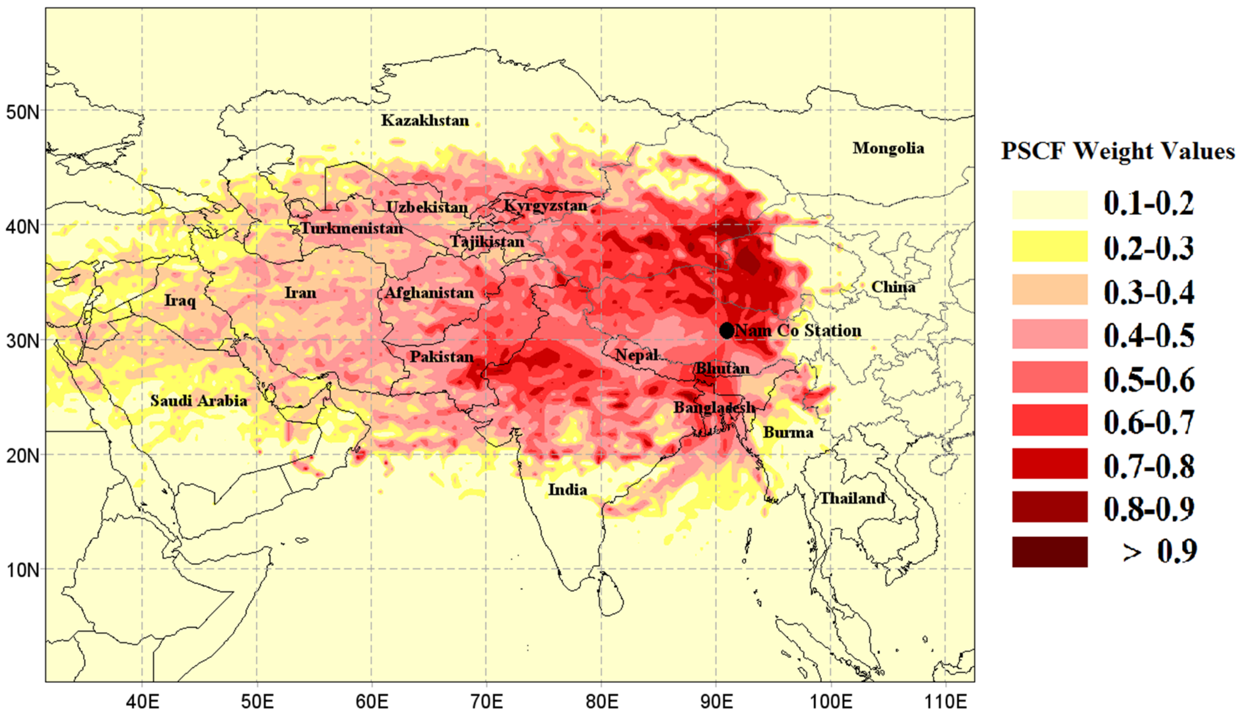


Fig. S8. Likely source areas of air masses associated with higher surface ozone concentrations at Nam Co Station during the whole measurement period identified by PSCF (Potential Source Contribution Function).

Table S1. Multi-Linear Regression Model by season using log-transforms and CAMx stratospheric tracers (corresponding to Model 1 in Table 2).

Statistical Metrics	Spring	Summer	Autumn	Winter
No. All	6043	7992	6157	7118
No. IRLS	5750	7615	5878	6759
r (All)	0.83	0.77	0.68	0.74
r (IRLS)	0.86	0.81	0.73	0.79
Contribution to Variance (%) by Group				
CAMx Tracers	21.10	3.85	0.41	4.41
WRF-FLEXPART Clusters	0.91	8.99	0.70	0.46
Local Winds	28.30	29.30	38.40	54.20
Seasonal Signal	33.60	45.60	44.30	20.60
Diurnal Signal	6.32	8.58	10.30	15.40
Annual Signal	7.87	2.80	3.94	3.43
WRF PBLH	1.85	0.84	2.00	1.55