

Response to referee comments

We would like to thank the referees and editor again for 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.

Anonymous Referee #2

This is my second review of the paper. I would like to congrats to the authors because the manuscript is really improved since its first submission. Most of my suggestions have been caught by the authors and most of my concerns cleared. A few minor points must be considered at this point, thus I support publication on ACP after there are considered/fixed.

Response: Thanks for your valuable advices and comments.

Abstract, line 19: please, substitute “long-term” with “continuous”

Response: Thanks for your suggestion. Changed as suggested in line 19.

Abstract, line 23: here the low anthropogenic contribution from China in summer and from South Asia in spring must be cited.

Response: The sentence was added as follows (lines 24 - 25):

“and the anthropogenic contribution from South Asia in spring and China in summer may affect Nam Co Station occasionally”.

Pag 7, line 182: 47.6 +/- 11.6 ppb. Is it +/- 1-sigma? Please, specify.

Response: 47.6±11.6 is the mean ± standard deviation. Now we specified as follows (lines 220- 221): “(mean ± standard deviation)”.

It is noteworthy that July 2015 appeared much more higher that other July months. Maybe an interesting anomaly to investigate in a future paper!

Response: Thanks for your suggestion. We will do further study considering your advice.

Pag 9, line 233: “...because stratospheric intrusion contribution is characterised by a maximum in spring...”

Response: Thanks for your suggestion. Now we added the sentence as follows (lines 260 - 261): “because stratospheric intrusion contributions are seasonal,”.

Figure S4 is rather obscure to me. I cannot be able to understand which is the message that the authors aim to provide by this figure and what information is actually provided. What these histograms and scatterplots represent? Please, better explain in the text and in the figure caption. Moreover, even in the SM, I would like to see some explanation about this block-bootstrapped method. I'm not used with this method (and I guess other potential readers) and a simple explanation can help in better evaluating the obtained results without searching in other papers!

Response: Thanks for your suggestion. Now we added the sentences as follows (lines 275 - 284):

“The uncertainties in the model regression results are shown graphically in Fig. S4. The histograms show variation in the contribution to ozone variance for each group based on the 100 realizations of the model. Taking the CAMx tracer as an example, the model suggests that this term contributes 17.7% of the O₃ variance on average, but the results range from 12 to 24% and have a standard deviation of 2.6%. The scatterplots show the covariance between the model estimates for different groups. Most groups do not covary and hence there is no correlation between the x and y axis and the correlation coefficients are low. For example, the contribution from the WRF-FLEXPART clusters does not covary with the CAMx tracer, and $r^2 = 0.03$. In contrast, the seasonal signal covaries with the CAMx tracer ($r^2 = 0.5$) which suggests that there is an increased uncertainty in the estimates for these terms and that changes in estimates for the seasonal signal will lead to changes in the estimates for the CAMx tracer. In this case, this is because stratospheric intrusions occur in spring and hence there is an inescapable correlation between the two groups”.

Line 256: to consider the PV at 350 hPa has much more sense that considering PV at the surface, actually!

Response: Thanks for your comment. PV at 350 hPa was in positive correlation with surface ozone concentration at Nam Co Station.

Line 273: actually these structures over Himalayas are similar to those reported by Bracci et al. (JAMC, 2012). However, looking at figure 5, the 1-2 PV isolines do not appeared much more different in spring than in winter, please check it!

Response: PV isolines is one of the parameters in these monthly mean meridional cross-section. Zonal winds and ozone concentration appeared different in the spring and the winter, especially the longitude and the altitude of the hot spots of ozone which indicating that the surface ozone concentration at Nam Co Station is more easily affected by the air mass from stratosphere in the spring than the winter.

Line 283: in my opinion, by this analysis hardly you can affirm that STE “occurred right above Nam Co”. Please remove.

Response: Thanks for your comment. Now we removed this sentence.

Line 285-290: It is not clear which is the “take home message” of the SWD analysis. It is the local photochemistry which is supposed to play the most important role or the hemispheric-scale "photochemistry"? Please, clarify.

Response: Thanks for your comment. Local photochemistry is an important role to the monthly surface ozone variation at Nam Co Station and the intense solar radiation at Nam Co Station in the spring may lead to more photochemical production of surface ozone.

Line 294: please correct “Der”.

Response: Thanks for your comment. Changed as suggested in line 332.

Line 295: “the height of the folding is higher than those in the spring and summer”. I do not think that this info can be retrieved by Fig. 5. Maybe some works can be cite to support (e.g. Ojha, N., Pozzer, A., Akritidis, D., and Lelieveld, J.: Secondary ozone peaks in the troposphere over the Himalayas, Atmos. Chem. Phys., 17, 6743-6757, <https://doi.org/10.5194/acp-17-6743-2017>, 2017.)

Response: Thanks for your comment. Changed as suggested as (lines 330 – 333):

“Ojha et al. (2017) found that the potential vorticity layer in the summer was weaker than during the late winter and in the spring in the central Himalayan region which is to the south of Nam Co Station. In the autumn (Sep, Oct and Nov) and the winter (Dec, Jan and Feb), the mixing heights at Nam Co Station were much lower than those in the spring and the summer”.

Line 312: and high PBLH, thus suggesting an important role of thermal transport regimes of PBL air-masses to diurnal ozone variability”. Once again: it is not completely clear which is (for the authors) the driving processes for diurnal variability: (local?) photochemistry or transport?

Response: Thank you for pointing this out. Both vertical mixing and photochemical production are important to the diurnal variation of surface ozone at Nam Co Station.

Line 346: “...on the urban scale, due to NO titration under ambient conditions not favourable to photochemical production.”

Response: Thank you for pointing this out. Now we rewrote this sentence as (lines 378 - 380):

“It is possible that titration of ozone by NO_x at the urban sites as well as the differences in altitude and meteorology may lead to differences between the surface ozone concentrations at Nam Co Station and those at Lhasa and Dangxiong”.

Section 5.3: I must say that I’m still convinced that your clustering is not so effective. Looks for instance to the winter season: the single trajectories spanned over a wide latitudinal range, but the cluster centroids are not able to catch this variability. This must be stressed in the discussion...

Response: Thanks for your comment. We calculated the backward trajectories every 3 hours during the measurement period. Comparing the limited trajectories in the periphery, there were much more trajectories in the main body of trajectories which were grouped into clusters.

Line 378: please modify as following: “...may also reflect this possibility.”

Response: Thank you for pointing this out. Now we modified this sentence as (lines 411 - 412):

“The higher level of surface ozone at NCO-P (Cristofanelli et al., 2010) than at Nam Co Station in the spring may also reflect this possibility”.

Figure 11: it is almost impossible to read the inserts. I would suggest to rearrange figure 11 (see my attachment)

Response: Thanks for your comment. Now we rearranged figure 11 as you suggested.

Line 390: probably PSCF also picked up the contribution of the transport of pollution from the Indo-gangetic plains and Himalaya foothills...

Response: Thank you for pointing this out. Now we modified this sentence as (lines 424 - 425):

“and the transport of pollution from the Indo-Gangetic Plain and Himalayan foothills was also probably picked up by PSCF”.

Line 395 – 397: Actually, you showed that Mt. Waliguan, NCOP and other stations have very different seasonal and diurnal cycles. So I do not agree that they are representative of the entire TP! Please, check!

Response: Thanks for your comment. Now we removed “entire” from this sentence.

Line 425: I think that the term “aged” is confusing in this context. As shown by other works (Putero et al., 2016; Ojha et al., 2017...), STE affecting Himalayas can occur very far from the region (e.g. Mediterranean basin). So I wouldn't call STE occurring over TP as “aged”...

Response: Thanks for your comment. Now we removed “aged” from this sentence.

Line 428: ...”is similar” (please remove “most”)

Response: Thanks for your comment. Now we removed “most” from this sentence.

Line 429: please quantify “infrequent”: which is the percentage of occurrence?

Response: Thanks for your comment. Cluster 5 in the summer represents the backward trajectories from or passed through the Northwest China and it occurs only 3.4% during whole the summer. Cluster 3 in the summer also represents the trajectories from parts of the Northwest China and it occurs 10.81% during whole the summer.

Co-Editor Decision: Reconsider after minor revisions (Editor review)

Comments to the Author:

Dear authors,

Thank you for submitting your revised manuscript, which has been reviewed by two referees. After the revision the manuscript reads much better. However, further improvement is necessary to meet the ACP quality requirements. Please take into account referee #2's comments in your next revision. In addition I also have many points for your consideration. Please submit your revised manuscript with your point-by-point responses to referee and editor comments.

Sincerely,

Xiaobin Xu

Response: Thank you so much for your comment.

Co-editor comments:

(1) the English language needs to be polished further. There are many grammatical errors (e.g., incorrect use of "the", plural and singular, etc.). I strongly suggest that you either have the manuscript edited a native English-speaking expert or select the English language copy-editing.

Response: Thanks for your suggestion. Now we polished the manuscript by native English-speaking expert.

(2) Line 25, I think you mean "Model results indicate that the study site is affected ‘differently’ by the surrounding areas in different seasons."

Response: Thank you for pointing this out. Now we rewrote this sentence as (lines 26 - 27):

“Model results indicate that the study site is affected differently by the surrounding areas in different seasons”.

(3) Line 38, change "globally" to "global".

Response: Changed as you suggested in line 40.

(4) Lines 44-45, "... photochemistry was identified as the dominant source of tropospheric ozone at some sites,". I think photochemistry is a dominant source of tropospheric ozone over all sites of the world, not only "some sites".

Response: Thank you for pointing this out. Now we removed "at some sites" from this sentence.

(5) Line 46, what do you mean by "largely uniform synoptic systems"? A synoptic system or a large scale weather system has its structure characterized by many parameters, pressure, winds, temperature, etc. The spatial distributions of these parameters are not necessarily uniform. I think it is better to change "largely uniform synoptic systems" to "synoptic systems".

Response: Changed as you suggested. Now we removed "largely uniform" from this sentence.

(6) Lines 61-63, "The paucity of long-term surface ozone observations in the Tibetan Plateau, ... and can potentially lead to inaccurate simulation of surface ozone variation over the Tibetan Plateau". The accuracy of surface ozone simulation is determined by the capability of the model, the accuracy and fineness of emission inventory, the quality of meteorological data, etc., and should not be directionally influenced by the paucity of long-term surface ozone observations.

Response: Thank you for pointing this out. Now we modified this sentence as (lines 64 - 65):

"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 in the Tibetan Plateau".

(7) Lines 70-71, I do not think this sentence should be placed here.

Response: Thanks for your comment. Now we removed this sentence.

(8) Lines 74-75, it is better to move this sentence to line 49 (before "Due to...").

Response: Thanks for your comment. Changed as you suggested (lines 50 - 51).

(9) Lines 78-80, check the language and logic of this sentence.

Response: Thank you for pointing this out. Now we modified this sentence as (lines 78 - 79):

"Nam Co Station was established in September 2005 to monitor atmospheric conditions and enabled research of the atmospheric environment in the inland Tibetan Plateau (Kang et al., 2011)".

(10) Line 90, change "tourist" to "tourism".

Response: Changed as you suggested in line 90.

(11) Line 104, was the radiation measurement system calibrated? Was it stable?

Response: The radiation measurement system was calibrated and it was relatively stable.

(12) Line 113, are you sure there was no obvious difference among the backward trajectories for 500 m, 1000 m and 1500 m? This is usually not the case. Please check again.

Response: We calculated the backward trajectories for these three heights and it seems that the trajectories for different arrival heights had similar features and the pathways were similar. The trajectories calculated at 1500 m height traveled longer distances than those in 500 and 1000 m, but in the same direction.

(13) Lines 139-140, what do you mean by "offset by 10 ppb"? Add 10 ppb to all hourly ozone value? Why can a normal distribution be obtained by doing so?

Response: Thanks for your comment. We have added an explanation as follows (lines 163 - 164):

“We used the natural logarithm of O_3 concentrations offset by 10 ppb to approximate a normal distribution while reducing the long tail in the transformed variable that would be caused by taking the logarithm of low O_3 concentrations”.

(14) Line 194, some of the data gaps in Figs. 4 and S5 seem to be over a month. Are you sure you have >56% of data for EACH month?

Response: Thanks for your comment. Each month reported in this study had valid data more than 56%.

(15) What do you want to say with "the overall trends" here?

Response: Thank you for pointing this out. Now we modified this sentence as (lines 233 - 235):

“The surface ozone concentrations at Nam Co Station experienced similar annual cycles during each of the 5 years of measurements with slight variations (Fig. S1)”.

(16) Lines 212-214, Fig. 3 shows the daily minima in spring, summer and autumn are all at 8:00, contradicting to what you state here.

Response: Thank you for pointing this out. Now we modified this sentence as (lines 251 - 253):

“Mixing ratios went from low levels at night to high levels during the daytime. The diurnal profile was generally characterized by a later shift from low to high concentrations in the winter than in the rest of the year, most likely as a result of the later time of sunrise”.

(17) Section 4, please mathematically show your regression model. Without this it is very difficult to understand your method and results. (18) Lines 223-224, what do you actually mean with "For the boundary layer height quintiles were used"? (19) Lines 239-240, the WRF-FLEXPART produced six transport clusters. Why did you only obtain one contribution?

Response: Thanks for your comments. For these three comments, now we added the description of Multiple Linear Regression as (lines 136 - 192):

“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 and humidity), inter-annual variation factors, seasonal factors, diurnal factors, WRF boundary layer heights, WRF-FLEXPART trajectory clusters and the CAMx stratospheric ozone tracer.

Tests were performed with different variables and averaging times for the meteorological parameters, including hourly data, running averages of 3, 8 and 24 hours, and smoothed variables using Kolmogorov-Zurbenko filters (Rao et al., 1997). 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 Pearsons correlation coefficient was added to the inputs as long as the increase was greater than 0.005.

The regression model is described by the following equations (Eq. 1, 2, 3):

$$\begin{aligned} \log(O_3 + 10) = & \sum_{yr=2011}^{2014} \alpha_{yr} t_{yr} + f(seasons) + f(diurnal) + \sum_{i=1}^6 \alpha_{cl_i} t_{cl_i} + \sum_{i=1}^5 \alpha_{PBLH_i} t_{PBLH_i} \\ & + \sum_{j=1}^4 \sum_{i=5}^5 \alpha_{WS_{ij}} t_{WS_{ij}} + \alpha_{st} \log(Strat.Tracer) + \alpha_{bkg} \\ & + \epsilon \end{aligned} \quad (1)$$

$$f(seasons) = \alpha_{T_{SS}} T'_{SS} + \alpha_{q'_{SS}} q'_{SS} + \sum_{j=1}^2 \alpha_{s_j} \sin\left(\frac{2\pi jt}{365.25}\right) + \alpha_{c_j} \cos\left(\frac{2\pi jt}{365.25}\right) \quad (2)$$

$$f(diurnal) = \alpha_{T_{DU}} T'_{DU} + \alpha_{q'_{DU}} q'_{DU} + \sum_{i=1-11,14-24} \alpha_{hr_i} t_{hr_i} \quad (3)$$

α represents the regression coefficients that are determined by the model. O_3 is the hourly surface ozone concentration. Time vectors t are used to represent different temporal terms and vary between 0 and 1. For example, t_{yr} represents the variation from year to year. This means that t_{2011} is 1 during 2011 and 0 otherwise. The air mass clusters derived from WRF-FLEXPART are represented using t_{cl} : t_{cl_i} is 1 during cluster 1 and 0 otherwise.

The impact of winds and mixing heights on O_3 is expected to be non-linear. We therefore separated the data into quartiles and included a regression factor for each of the 5 points separating the quartiles (0%, 25%, 50%, 75% and 100%). For the boundary layer height, we have 5 time series represented by t_{PBLH_i} . Piecewise linear interpolation is used to give them a value of 1 at the edge of the quartile which then decreases to 0 by the next quartile. As an example, t_{PBLH_1} is 1 for the times when the mixing height is at the data minimum and it is 0 when the mixing height is above the first quartile. At times when the

mixing height is between these two levels, t_{PBLH_i} varies linearly.

For wind speed and direction, we performed the piecewise interpolation in 2 dimensions, with 5 factors for the wind speed and 4 factors for the wind direction (because the two extremes are the same: 0° and 360°). $t_{WS_{i,1}}$ represents times when the wind speeds are in the bottom quartile ($i = 1$) and when the wind direction is from the north ($j = 1$).

We used the natural logarithm of O_3 concentrations offset by 10 ppb to approximate a normal distribution while reducing the long tail in the transformed variable that would be caused by taking the logarithm of low O_3 concentrations. The background term is included as α_{bkg} and the residual between the regression model and the measurement time series is ϵ .

T is the temperature and q is the specific humidity. Both are normalized linearly as follows: $T' = (T - \mu(T))/\sigma(T)$. Because we were interested in identifying separately the seasonal and diurnal variation, we used a Kolmogorov-Zurbenko filter to separate T and q into a seasonal component (T_{SS} and q_{SS}) and a diurnal component (T_{DU} and q_{DU}) (Rao et al., 1997). The seasonal component used 5 passes of a 13-point moving average, and the diurnal component was the difference between the hourly and the seasonal time series. The seasonal terms also include harmonic terms with a 12 month and 6 month period as described in de Foy et al. (2016c).

The diurnal term includes diurnal temperature and specific humidity. In addition, we included a time series term for each hour of the day (t_{hr}) except for 12:00 and 13:00 which are taken to be reference hours.

The ‘‘Strat. Tracer’’ was simulated using the CAMx model as described in section 2.3. During testing, the best fit in the regression model was found using the seasonal component of the stratospheric tracer obtained from the Kolmogorov-Zurbenko filter in the same way as T_{SS} and q_{SS} .

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).

To estimate the uncertainty in the results, we used block-bootstrapping with a 24 hour block length, as described in de Foy et al. (2015). 100 realizations of the final model were performed. For each model

realization, days were selected randomly with replacement from the full set of days until a dataset containing the same total number of days was obtained. This accounts for both measurement errors and model errors as the instrument errors are assumed to be uncorrelated and model errors are unlikely to be correlated beyond a couple of days. The uncertainty in the results can then be estimated by calculating the standard deviation of the parameters obtained from the 100 model realizations.

When presenting results of the regression analysis, we obtained a single time series for each group by summing the individual parts. For example to estimate the impact of the WRF-FLEXPART clusters we make a sum of the 6 t_{chl} time series multiplied by the corresponding regression factors α_{chl} . For a log-transformed regression, the time series of each group can be interpreted as a scaling factor on the background term (α_{bkg}) and expressed as a percentage change. For a linear regression model, the time series can be expressed as a linear departure from the mean in the units of the measurements (ppb).

In all, there were 67 free parameters (α) in the regression model that were used to fit 27,310 data points”.

(20) Lines 248-249, "and that these can lead to enhancements of surface of ozone of 150% of the hourly standard deviation". What result support this statement? I can see it nowhere.

Response: Thanks for your comment. The sentence was based on an out-of-date figure, we apologize for this error. Now we rewrote this sentence (lines 266 - 267) and we have also better described the scaling factor in section 2.4 thanks to your comments:

“The regression model suggests that both stratospheric transport and seasonal variation can lead to enhancements in hourly concentrations of up to 20% of the baseline level of ozone”.

(21) Lines 301-305, “This is probably due to the time delay between the maximum ozone concentration and the maximum solar radiation” (lines 302-303). What do you mean with this sentence? The fact is that there is a delay between both. “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)”. It is too speculative and contradicting your points that are following. Local ozone formation may result in a good SWD-ozone correlation. However, a good SWD-ozone correlation may not necessarily be caused by local ozone formation. Wang et al. (2006) reported in situ photochemical production of ozone under the Waliguan spring and summer conditions that were very different from the Nam Co conditions. So, this citation does not support your speculation. I am not saying that there is no local formation of ozone at Nam Co. Theoretically local formation exists always where solar radiation is received. And ozone is also removed by local chemical and physical processes. If there is not net ozone production, you cannot attribute your SWD-ozone correlation to local ozone formation. Even if there is a net ozone production, you cannot simply do so with detailed analysis because the SWD-ozone

correlation may be caused by some other mechanisms. In fact, you are saying that the wind speed and PBLH are the main factors (lines 305-306). I strongly suggest that you delete this speculation and keep consistency.

Response: Thank you for pointing this out. Changed as you suggested. Now we removed the discussion of SWD-ozone correlation and modified the sentence as (lines 344 - 345):

“In addition, local photochemical production may also contribute to the higher concentration of surface ozone at Nam Co Station in the daytime”.

(22) Lines 345-346, “It is possible that the local NO_x emissions in these two urban regions reduce the average ozone on the urban scale”. Again, this is groundless. NO_x is the key precursor of ozone and promotes the photochemical production of ozone under favorable meteorological conditions. The meteorological conditions in Lhasa favor the ozone production as reported in Ran et al. (2014). Ozone at Dangxiong is controlled by large-scale physic-chemical processes, particularly the vertical mixing (Lin et al., 2015). Therefore, the differences between ozone values at Nam Co and those at Lhasa and Dangxiong should not be due to differences in NO_x emissions and may be caused some other factors, such differences in altitude, meteorology, etc.

Response: Thanks for your comment. Now we rewrote this sentence as (lines 378 - 380):

“It is possible that titration of ozone by NO_x at the urban sites as well as the differences in altitude and meteorology may lead to differences between the surface ozone concentrations at Nam Co Station and those at Lhasa and Dangxiong”.

(23) Lines 388-389, “In the winter, no obvious region was identified, which was likely due to low surface ozone mixing ratios in all these areas.” Not clear.

Response: Thank you for pointing this out. Now we modified this sentence as (lines 422 – 423):

“In the winter, no obvious region was identified as a potential source region”.

(24) Line 393, “universal concern”?! Please do not use inflated language.

Response: Thank you for pointing this out. Now we changed “universal” to “great” in line 427.

(25) Line 394, “varied” or “varying”?

Response: Thank you for pointing this out. Now we changed “varied” to “varying” in line 428.

(26) Line 404-406, “Its mountainous landform facilitates mountain-valley breezes and may sometimes pump up anthropogenic emissions especially during the winter (Xue et al., 2011)”. This is wrong! The impacts of anthropogenic emissions on the Waliguan station occur mainly in summer. And the Xue et al. (2011) paper is about a summer study.

Response: Thanks for your comment. Now we rewrote this sentence as (lines 438 - 439):

“and the impacts of anthropogenic emissions on Waliguan occur mainly in the summer”.

(27) Lines 423-424, “The analysis suggests that stratospheric intrusions account for around 20% of the variability in surface ozone concentrations at Nam Co Station” is a misleading statement. Fig. 4 shows that stratospheric ozone makes at maximum a contribution of 20% to the variation of surface ozone at

Nam Co. Most of the time, the stratospheric contribution is either zero or very low. I think you should state "...that the maximum contribution of stratospheric intrusions to surface ozone at Nam Co Station is about 20%" or give an average contribution.

Response: Thanks for your comment. Changed as you suggested. Now we rewrote this sentence as (lines 457 - 458):

"The analysis suggests that the maximum contribution of stratospheric intrusions to variability in surface ozone at Nam Co Station is approximately 20%".

(28) Please rewrite the third paragraph of Section 6 (Summary). This paragraph should be based on the major results in Section 5. I do not think this is the case in the current version.

Response: Thanks for your comment. Now we rewrote this paragraph as (lines 461- 464):

"Surface ozone at Nam Co Station showed distinct seasonal and diurnal variation patterns as compared with other sites in the Himalayas and the northern Tibetan Plateau. The monthly maximum of surface ozone at Nam Co Station, which is in the inland Tibetan Plateau was later in the year than at the sites in the southern Tibetan Plateau and the southern ridge of the Himalayas, but earlier in the year than at the sites in the northern Tibetan Plateau".

(29) Line 466, "Our measurements provide a baseline of tropospheric ozone at a remote site in the Tibetan Plateau". Be careful, your measurements of surface ozone at any site cannot provide a baseline (or whatever) of tropospheric ozone, which should be described by tropospheric column content and vertical distribution of ozone globally or over a region.

Response: Thanks for your comment. Changed as you suggested. Now we rewrote this sentence as (lines 465 - 466):

"Our measurements contribute to the understanding of ozone cycles and related physico-chemical and transport processes over the Tibetan Plateau".

(30) Fig. 4, the "scaling factor" is not explained and not clear.

Response: Thanks for your comment. Now we added sentences as (lines 264 - 267):

"In the log-transformed model, the time series for each group corresponds to a scaling factor that is applied to the baseline ozone concentration. This is shown in Fig. 4 for the stratospheric tracer and for the seasonal scaling. The regression model suggests that both stratospheric transport and seasonal variation can lead to enhancements in hourly concentrations of up to 20% of the baseline level of ozone".

(31) Fig. 5, what are the red dots on the sub-plots? The location of Nam Co? If so, there are relative positions should not be varying.

Response: The red dots show the position of the top of boundary layer height at Nam Co Station. We also clarify it in the caption of Fig. 5.

(32) Fig. 10, the color selection of this figure is poor. Some of the trajectory clusters are hardly differentiated. I suggest that you use light grey for individual trajectories and well contrasted colors for the clusters.

Response: Thanks for your comment. Changed as you suggested.

(33) Fig. 12, the PSFC data look very strange. Why are the differences among season so large? See for example the difference between spring and winter? How did you obtain the weighted values? Were there calculated on a yearly or seasonal basis?

Response: We used the mean concentration of surface ozone as the threshold in PSCF calculation. Due to low surface ozone concentration in winter, less data were used in PSCF calculation and grid cells in the winter had less air parcels passed through. Then the PSCF values multiplied smaller weighting value and resulted in the difference of weighted PSCF among different seasons. The weighted PSCF values were obtained by multiplying the original PSCF values by the weighting factor. There were calculated seasonally but the weighting function was unified.

(34) Tables 2 and S1, it is not so straightforward what you are presenting in the tables. Please study the ACP instructions for the requirements on tables.

Response: Thanks for your comment. Changed as you suggested.

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

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Abstract:

15 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 **continuous** measurements of surface ozone mixing ratios at Nam Co Station over a period of ~5 years (January 2011 to October 2015), which is a background site in the inland Tibetan Plateau. An average surface ozone mixing ratio of 47.6 ± 11.6 ppb (**mean \pm standard deviation**) 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 inputs, **and the anthropogenic contribution from South Asia in spring and China in summer may**

25 **affect Nam Co Station occasionally**. Surface ozone at Nam Co Station is mainly dominated by natural processes involving photochemical reactions, vertical mixing and downward transport of stratospheric air mass. **Model results indicate that the study site is affected differently 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 are 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,

30 we aim to expand the understanding of ozone cycles and transport processes over the Tibetan Plateau. This work may provide a reference for future model simulations.

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 (HO₂) 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 (CO₂) and methane (CH₄)), tropospheric ozone contributes to global warming and has an estimated [global](#) average radiative forcing of $0.40 \pm 0.20 \text{ W m}^{-2}$ at a high confidence level (Myhre et al., 2013). Average monthly and annual concentrations of tropospheric ozone are used to assess and improve global modeling results (Wild and Prather, 2006; Roelofs et al., 2003).

The origin of tropospheric ozone and its temporal variation vary 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, as supported by models (Wu et al., 2007). Background sites can represent areas with surface ozone concentrations that are under the control of 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.

[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’ \(Yao et al., 2012\).](#) 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 (northeastern edge of the Tibetan Plateau) since 1994 (Xu et al., 2016), the Nepal Climate Observatory at Pyramid (NCO-P) which has operated 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, which has been in operation 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, maximum surface ozone was observed in the spring (Cristofanelli et

60 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 the 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
65 understanding of the regional background ozone level and the factors that influence it in 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 influential factors. We then evaluated surface ozone variability using combined observations over the Tibetan
Plateau and beyond. Finally, we discuss the potential representativeness of surface ozone at Nam Co Station as the regional
70 background of surface ozone in the inland Tibetan Plateau. This study expands the understanding of the concentration and
variations in the surface ozone concentration and the transport processes that influence tropospheric ozone in the inland Tibetan
Plateau.

2 Measurements and Methods

2.1 Measurement site

75 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 southeastern 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 in September 2005 to monitor atmospheric
conditions and enabled research of the atmospheric environment in the inland Tibetan Plateau (Kang et al., 2011). Nam Co
80 Station is located 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 a 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),
85 a subtropical high-pressure system, and southeasterly 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 [tourism](#) 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 the 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 were 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 to the electrical wires caused by strong winds) and equipment maintenance. All data displayed in this study are in UTC+8 format (Beijing Time, BJT), and solar noon at Nam Co Station occurs at 13:56 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 using 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 the planetary boundary layer height (PBLH) were obtained from the Global Data Assimilation System (GDAS-1) operated by the U.S. National Oceanic and Atmospheric Administration (NOAA) with $1^{\circ} \times 1^{\circ}$ latitude and longitude horizontal resolution and 23 vertical levels from 1000 hPa to 20 hPa (<http://www.arl.noaa.gov/gdas1.php>). The backward trajectory arrival height in HYSPLIT was set at 500 m (500, 1000 and 1500 m were tested as arrival heights and there was no obvious difference in the results) above the surface and the total run times was 120 hours for each backward trajectory with a time intervals of 3

115 hours throughout 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.

125 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) were used to analyze the upper troposphere and lower stratosphere structures of the meridional cross-section over Nam Co Station.

135 **2.4 Multiple Linear Regression Model**

A multiple linear regression (MLR) model was used in this study to quantify the main factors affecting the 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 and humidity), inter-annual variation factors, seasonal factors, diurnal factors, WRF boundary layer heights, WRF-FLEXPART trajectory clusters and the CAMx stratospheric ozone tracer.

Tests were performed with different variables and averaging times for the meteorological parameters, including hourly

data, running averages of 3, 8 and 24 hours, and smoothed variables using Kolmogorov-Zurbenko filters (Rao et al., 1997). 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 Pearsons correlation coefficient was added to the inputs as long as the increase was greater than 0.005.

145 The regression model is described by the following equations (Eq. 1, 2, 3):

$$\log(O_3 + 10) = \sum_{yr=2011}^{2014} \alpha_{yr} t_{yr} + f(seasons) + f(diurnal) + \sum_{i=1}^6 \alpha_{cl_i} t_{cl_i} + \sum_{i=1}^5 \alpha_{PBLH_i} t_{PBLH_i} + \sum_{j=1}^4 \sum_{i=5}^5 \alpha_{WS_{i,j}} t_{WS_{i,j}} + \alpha_{st} \log(Strat.Tracer) + \alpha_{bkg} + \epsilon \quad (1)$$

$$f(seasons) = \alpha_{T_{SS}} T'_{SS} + \alpha_{T_{SS}} q'_{SS} + \sum_{j=1}^2 \alpha_{s_j} \sin\left(\frac{2\pi jt}{365.25}\right) + \alpha_{c_j} \cos\left(\frac{2\pi jt}{365.25}\right) \quad (2)$$

$$f(diurnal) = \alpha_{T_{DU}} T'_{DU} + \alpha_{q_{DU}} q'_{DU} + \sum_{i=1-11,14-24} \alpha_{hr_i} t_{hr_i} \quad (3)$$

150 α represents the regression coefficients that are determined by the model. O_3 is the hourly surface ozone concentration. Time vectors t are used to represent different temporal terms and vary between 0 and 1. For example, t_{yr} represents the variation from year to year. This means that t_{2011} is 1 during 2011 and 0 otherwise. The air mass clusters derived from WRF-FLEXPART are represented using t_{cl} : t_{cl_i} is 1 during cluster 1 and 0 otherwise.

155 The impact of winds and mixing heights on O_3 is expected to be non-linear. We therefore separated the data into quartiles and included a regression factor for each of the 5 points separating the quartiles (0%, 25%, 50%, 75% and 100%). For the boundary layer height, we have 5 time series represented by t_{PBLH_i} . Piecewise linear interpolation is used to give them a value of 1 at the edge of the quartile which then decreases to 0 by the next quartile. As an example, t_{PBLH_1} is 1 for the times when the mixing height is at the data minimum and it is 0 when the mixing height is above the first quartile. At times when the mixing height is between these two levels, t_{PBLH_i} varies linearly.

160 For wind speed and direction, we performed the piecewise interpolation in 2 dimensions, with 5 factors for the wind speed and 4 factors for the wind direction (because the two extremes are the same: 0° and 360°). $t_{WS_{i,j}}$ represents times when the wind speeds are in the bottom quartile ($i = 1$) and when the wind direction is from the north ($j = 1$).

165 We used the natural logarithm of O_3 concentrations offset by 10 ppb to approximate a normal distribution while reducing the long tail in the transformed variable that would be caused by taking the logarithm of low O_3 concentrations. The background term is included as α_{bkg} and the residual between the regression model and the measurement time series is ϵ .

T is the temperature and q is the specific humidity. Both are normalized linearly as follows: $T' = (T - \mu(T))/\sigma(T)$. Because we were interested in identifying separately the seasonal and diurnal variation, we used a Kolmogorov-Zurbenko filter to separate T and q into a seasonal component (T_{SS} and q_{SS}) and a diurnal component (T_{DU} and q_{DU}) (Rao et al., 1997). The seasonal component used 5 passes of a 13-point moving average, and the diurnal component was the difference between the hourly and the seasonal time series. The seasonal terms also include harmonic terms with a 12 month and 6 month period as described in de Foy et al. (2016c).

The diurnal term includes diurnal temperature and specific humidity. In addition, we included a time series term for each hour of the day (t_{hr}) except for 12:00 and 13:00 which are taken to be reference hours.

The “Strat. Tracer” was simulated using the CAMx model as described in section 2.3. During testing, the best fit in the regression model was found using the seasonal component of the stratospheric tracer obtained from the Kolmogorov-Zurbenko filter in the same way as T_{SS} and q_{SS} .

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).

To estimate the uncertainty in the results, we used block-bootstrapping with a 24 hour block length, as described in de Foy et al. (2015). 100 realizations of the final model were performed. For each model realization, days were selected randomly with replacement from the full set of days until a dataset containing the same total number of days was obtained. This accounts for both measurement errors and model errors as the instrument errors are assumed to be uncorrelated and model errors are unlikely to be correlated beyond a couple of days. The uncertainty in the results can then be estimated by calculating the standard deviation of the parameters obtained from the 100 model realizations.

When presenting results of the regression analysis, we obtained a single time series for each group by summing the individual parts. For example to estimate the impact of the WRF-FLEXPART clusters we make a sum of the 6 t_{cli} time series multiplied by the corresponding regression factors α_{cli} . For a log-transformed regression, the time series of each group can be interpreted as a scaling factor on the background term (α_{bkg}) and expressed as a percentage change. For a linear regression model, the time series can be expressed as a linear departure from the mean in the units of the measurements (ppb).

In all, there were 67 free parameters (α) in the regression model that were used to fit 27,310 data points.

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 using trajectories that 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 were based on a count of the trajectory segment (hourly trajectory positions) that terminated 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 the simulation period. Let m_{ij} represents the number of points in the same cell with arrival times at the sampling site that correspond to surface ozone mixing ratios higher than a set criterion. In this study, we calculated the PSCF based on trajectories corresponding to concentrations that exceeded the mean level of surface ozone during measurement. The PSCF value for the ij th cell was then defined as:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}}$$

The PSCF value can be interpreted as the conditional probability that the ozone mixing ratios at the measurement site are greater than the mean mixing ratios if the air parcel passes through the ij th 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 endpoints in a cell was less than approximately three times the average value of the end points per cell. In this case, W_{ij} was set as follows:

$$W_{ij} \begin{cases} 1.00 & n_{ij} > 3N_{ave} \\ 0.70 & 3N_{ave} > n_{ij} > 1.5N_{ave} \\ 0.42 & 1.5N_{ave} > n_{ij} > N_{ave} \\ 0.05 & N_{ave} > n_{ij} \end{cases}$$

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

220 The mean surface ozone mixing ratio at Nam Co Station during the entire observational period was 47.6 ± 11.6 ppb (mean \pm standard deviation), 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.

225 The mean surface ozone mixing ratio at Nam Co Station was within the reference range reported for the Himalayas and Tibetan Plateau; it was higher than the ratios for the two nearest urban sites: Lhasa (Ran et al., 2014) and Dangxiong (Lin et al., 2015) and was 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 any other sites over the Tibetan Plateau were generally higher than the range of 20-45 ppb measured at background sites in the
230 mid-latitudes of the Northern Hemispheres. This was in agreement with the higher concentrations typically observed 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 surface ozone concentrations at Nam Co Station experienced similar annual cycles during each of the 5 years of measurements
235 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. The mean monthly surface ozone
240 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), albeit with a small increase in October.

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). [Mixing ratios went from low levels at night to high levels during the daytime. The diurnal profile was generally characterized by a later shift from low to high concentrations in the winter than in the rest of the year, most likely as a result of the later time of sunrise.](#) 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 factors on seasonal variation

The regression model had 27,310 hourly data points of which 26,005 were retained by the IRLS procedure, see Table 2. The correlation coefficient (r) was 0.77 for the entire time series and 0.81 without the outliers. The time series of the model is shown in Fig. 4 and scatter plots between the measurements and the model [are shown in Fig. S2.](#) Note that [because stratospheric intrusion contributions are seasonal,](#) there was covariance between the stratospheric tracer and the seasonal signal. Uncertainties in the estimate of the contribution from one of these therefore [impacted](#) the estimate from the other.

[As described in section 2.4, we combined the time series in the regression model into 7 distinct groups and present the results for these merged factors. In the log-transformed model, the time series for each group corresponds to a scaling factor that is applied to the baseline ozone concentration. This is shown in Fig. 4 for the stratospheric tracer and for the seasonal scaling. The regression model suggests that both stratospheric transport and seasonal variation can lead to enhancements in hourly concentrations of up to 20% of the baseline level of ozone.](#)

The contribution of each group to the O₃ variation can be calculated as the variance of the time series for that group divided by the sum of the variances for all the groups shown in Table 2 and described in Eqs. 1-3. The uncertainty of the results was calculated using the standard deviation of the 100 realizations of the model using block-bootstrapping. 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. The uncertainties in the model regression results are shown graphically in Fig. S4. The histograms show variation in the contribution to ozone variance for each group based on the 100 realizations of the model. Taking the CAMx tracer as an example, the model suggests that this term contributes 17.7% of the O₃ variance on average, but the results range from 12 to 24% and have a standard deviation of 2.6%. The scatterplots show the covariance between the model estimates for different groups. Most groups do not covary and hence there is no correlation between the x and y axis and the correlation coefficients are low. For example, the contribution from the WRF-FLEXPART clusters does not covary with the CAMx tracer, and $r^2 = 0.03$. In contrast, the seasonal signal covaries with the CAMx tracer ($r^2 = 0.5$) which suggests that there is an increased uncertainty in the estimates for these terms and that changes in estimates for the seasonal signal will lead to changes in the estimates for the CAMx tracer. In this case, this is because stratospheric intrusions occur in spring and hence there is an inescapable correlation between the two groups.

As a separate test, the regression model was performed with linear transformations instead of log-transformations. The model is similar to Eq. 1 but with the logarithm terms replaced by a linear normalization for the ozone concentration and for the stratospheric tracer. The advantage of this model is that the results can be interpreted as ozone enhancements from each group in ppb. The results are 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) shows 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 shows 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.

300 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 is a better indicator of stratospheric ozone incursions than the time series of potential vorticity.

305 The regression model was also performed by season, as shown in Table S1. This results show 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.

To visualize the transport of ozone from the stratosphere to the troposphere, we analyzed the upper troposphere and lower
310 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 occurred in the southern Tibetan Plateau and closer to Nam Co Station in the spring. Cosmogenic ³⁵S results (Lin et al., 2016) also indicated that in the spring, Nam Co was affected by aged stratospheric
315 air originating over the Himalayas rather than being affected by transport from fresh stratospheric air masses above Nam Co Station. The larger diurnal amplitude of surface ozone in the spring than in other seasons (Fig. 3, mentioned in section 3.3) may be related to four factors: (1) position of the STE (stratosphere–troposphere exchange) 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 occurs in the southern Tibetan Plateau which is close to Nam Co Station. Furthermore, PBLH at Nam Co Station
320 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. 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
325 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. Ojha et al. (2017) found that the potential vorticity layer in the summer was weaker than during the late winter and in the spring in the central Himalayan region which is to the south of Nam Co Station. In the autumn (Sep, Oct and Nov) and the winter (Dec, Jan and Feb), the mixing heights at Nam Co Station were much lower than those in the spring and the summer. Furthermore, SWD in the autumn and the winter was 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 vertical mixing and photochemical production on diurnal variation

Wind speed and PBLH are 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). A lake-land breeze influenced Nam Co Station, and the wind speed in the daytime was higher than that at night (Fig. S7). The hourly average wind speed and PBLH at Nam Co Station showed a positive correlation with the 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 the hourly average surface ozone and the hourly average PBLH was 0.92. These results indicate that high levels of surface ozone are associated with high wind speeds and high mixing heights. In addition, local photochemical production may also contribute to the higher concentration of surface ozone at Nam Co Station in the daytime.

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 most influential factor (Cristofanelli et al., 2010). The 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 the free troposphere 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 general, diurnal surface ozone variations across the Tibetan Plateau were typically controlled by site-specific meteorological conditions and photochemical production. Sites located in plains or valleys exhibited daytime ozone maxima associated with vertical mixing and photochemical production, whereas mountain-top sites exhibited daytime ozone minima associated with up-slope flow of low-ozone air.

360 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; Ordóñez 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; 365 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 the 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.

370 In the northern Tibetan Plateau (Waliguan site), surface ozone experienced 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, the Qinghai Lake site also experienced a maximum in the summer (Shen et al., 2014). Horizontal and vertical wind transport have been regarded as major contributor to surface ozone at these two sites (Zhu et al., 2004; Shen et al., 2014).

375 B) The central Tibetan Plateau: spring-maximum type.

Sites in the central Tibetan Plateau including Nam Co Station experienced maximum ozone levels during the late spring-early summer and relatively low levels during the remainder of the 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 titration of ozone by NO_x at the urban sites as well as the differences in altitude and meteorology may lead to differences between the surface ozone concentrations at Nam Co Station and those at Lhasa and Dangxiong.](#) 380 A study at Dangxiong revealed that the higher 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 summer,

whereas transport largely contributed to the observed ozone mixing ratios in the autumn and the winter (Ran et al., 2014). The large-scale background of surface ozone in the spring is considered an important influence on Dangxiong and Lhasa during this season (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 the 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 Station. In the spring, the air masses that arrived at Nam Co Station were predominantly from the west and 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, which 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 had an effect on the level of surface ozone at Nam Co Station through transport. In the summer, there are more backward trajectories originating 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 summer were classified into 6 clusters respectively (Fig. 11). In the spring, clusters that originated from the southern Tibetan Plateau had higher mean surface ozone

410 levels than clusters that originated 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 reflect this possibility.](#) In the summer, clusters from the northern Tibetan Plateau had higher mean surface ozone levels than clusters that originated 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
415 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 the spring and summer. The large areas of northwestern China, including the northern Tibetan Plateau, were the additional regions with
420 potentially high PSCF weight values in the summer. The PSCF values for both the southern and northern Tibetan Plateau in the autumn were smaller than those in the spring and summer. In the autumn, the inland Tibetan Plateau appeared 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 as a potential source region,](#) which was likely due to low surface ozone mixing ratios in all these areas. PSCF probably picked up the contribution from STE as a signal from the south in the spring and from the north in the summer, [and
425 the transport of pollution from the Indo-Gangetic Plain and Himalayan foothills was also probably picked up by PSCF.](#)

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

The changes in the atmospheric environment of the Tibetan Plateau are of [great](#) concern due to its rapid responses and feedbacks to regional and global climate changes. The Tibetan Plateau covers vast areas with [varying](#) topography; however, comprehensive monitoring sites are limited and sporadically distributed. Analysis of the atmospheric composition at Waliguan
430 in the north and Everest in the south of the Tibetan Plateau has shown that they are representative of high-altitude background sites for the 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
435 (Decesari et al., 2010; Maione et al., 2011; Putero et al., 2014). In addition, Mount Everest has been identified as a hot spot 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) and the impacts of anthropogenic emissions on Waliguan occur mainly in the summer (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 directly influenced by stratospheric intrusions than NCO-P and is minimally influenced by local anthropogenic emission. Nam Co Station showed distinct seasonal and diurnal variation patterns compared with those sites in the Himalayas and the northern Tibetan Plateau presented earlier. Our measurements of surface ozone at Nam Co Station are essential datasets 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 influential 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 mixing ratio of surface ozone at Nam Co Station 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 the maximum contribution of stratospheric intrusions to variability in surface ozone at Nam Co Station is approximately 20%. Further analysis of tropopause folding suggests that Nam Co Station is affected by air masses associated with stratospheric intrusions transported from the southern and northern Tibetan Plateau, mainly during the spring and the summer, respectively.

Surface ozone at Nam Co Station showed distinct seasonal and diurnal variation patterns as compared with other sites in the Himalayas and the northern Tibetan Plateau. The monthly maximum of surface ozone at Nam Co Station, which is in the inland Tibetan Plateau occurred later in the year compared with the sites in the southern Tibetan Plateau and the southern ridge of the Himalayas, but earlier in the year than at the sites in the northern Tibetan Plateau.

465 Our measurements 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 influential mechanisms.

Data availability

All the data presented in this paper can be made available for scientific purposes upon request to the corresponding
470 authors (Qianggong Zhang (qianggong.zhang@itpcas.ac.cn) or Shichang Kang (shichang.kang@lzb.ac.cn)).

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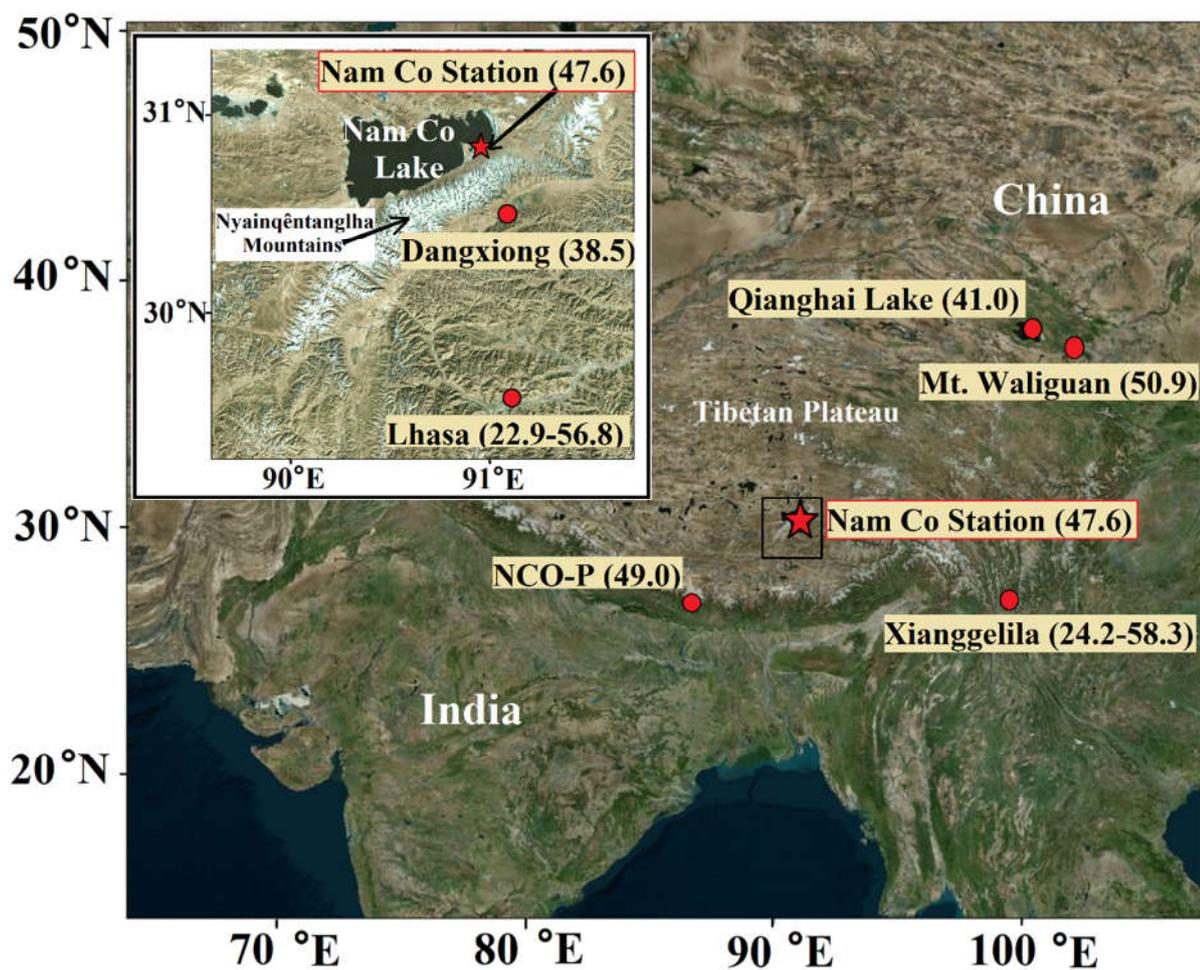
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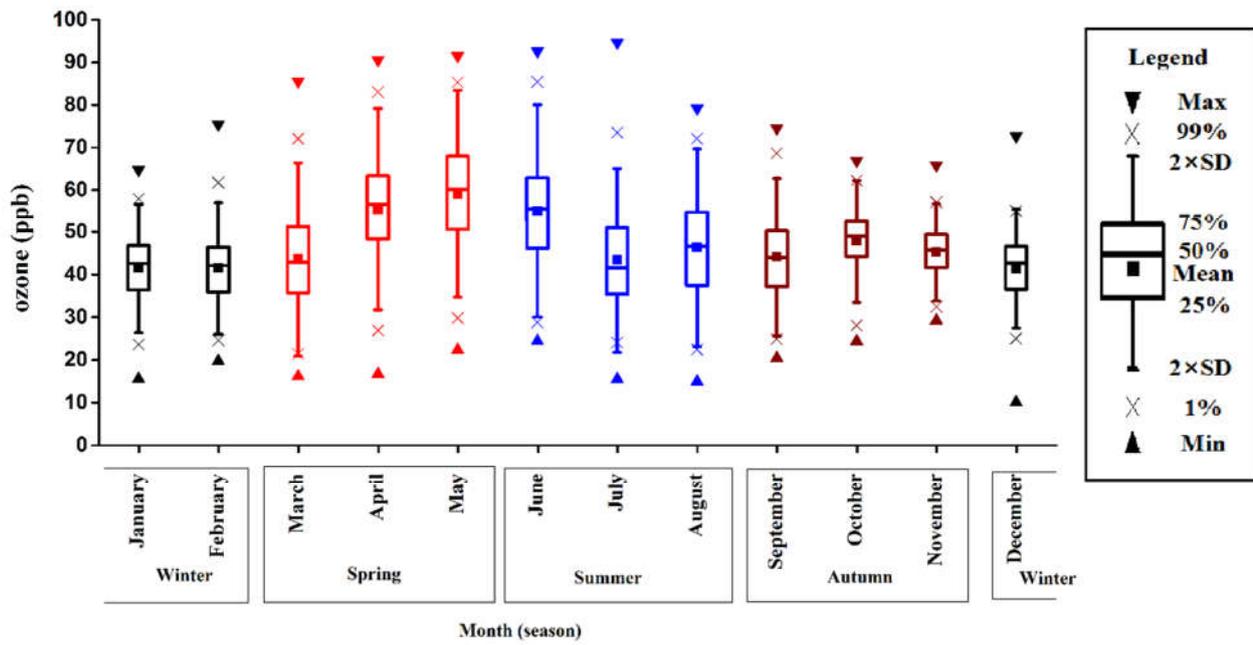
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Fig. 1. Geographical location of Nam Co Station and other sites in the Tibetan Plateau. Values in the parenthesis refers to the average or range of surface ozone in ppb as obtained from Cristofanelli et al., 2010; Lin et al., 2015; Shen et al., 2014;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

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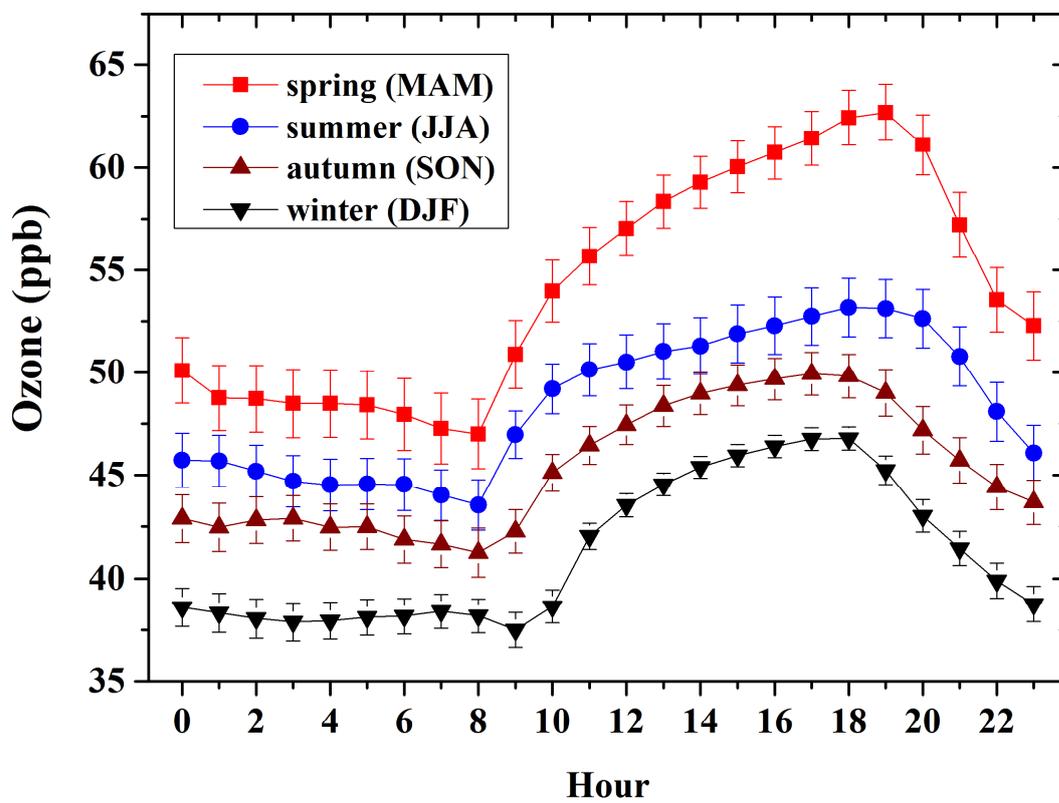
690 Fig. 2. Monthly average and statistical parameters of surface ozone at Nam Co Station during the whole
 measurement period (spring (MAM) in red; summer (JJA) in blue; autumn (SON) in dark red; winter (DJF) in
 black).

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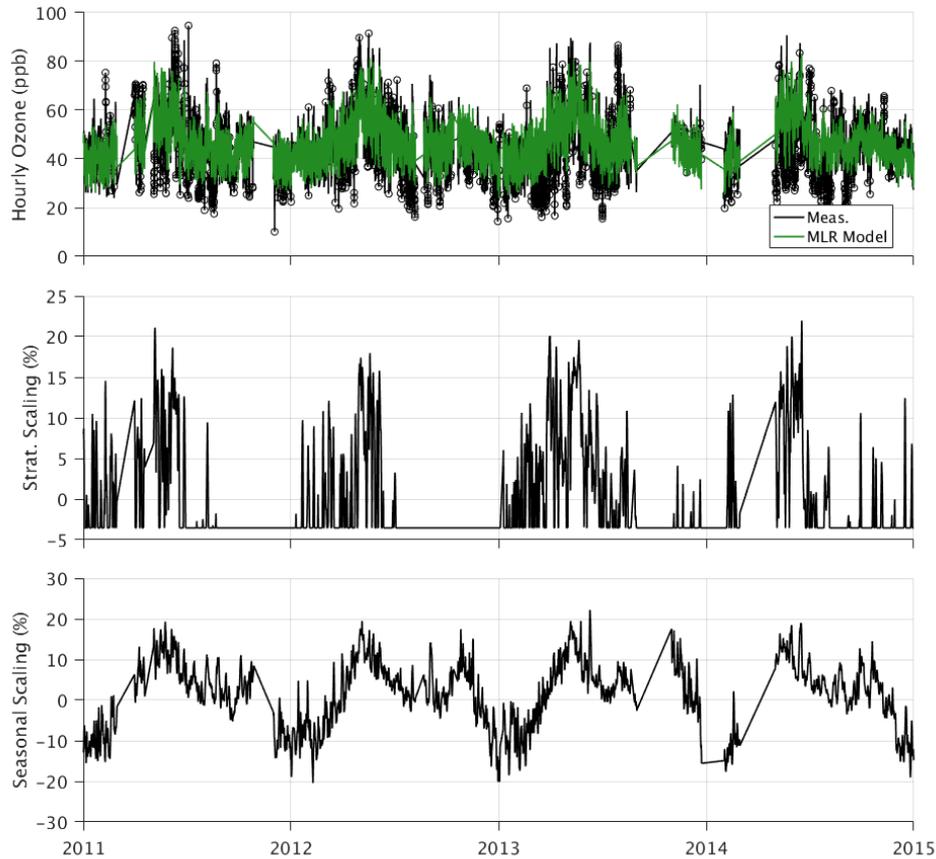


715 **Fig. 3. Diurnal profiles of average hourly surface ozone at Nam Co Station by seasons. Error bars show the 95% confidence intervals.**

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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: Impact of the CAMx stratospheric tracer on surface ozone concentration in the regression model expressed as a percentage change relative to the model average. Bottom: Impact of the seasonal factor (Eq. 2) on surface ozone concentration.

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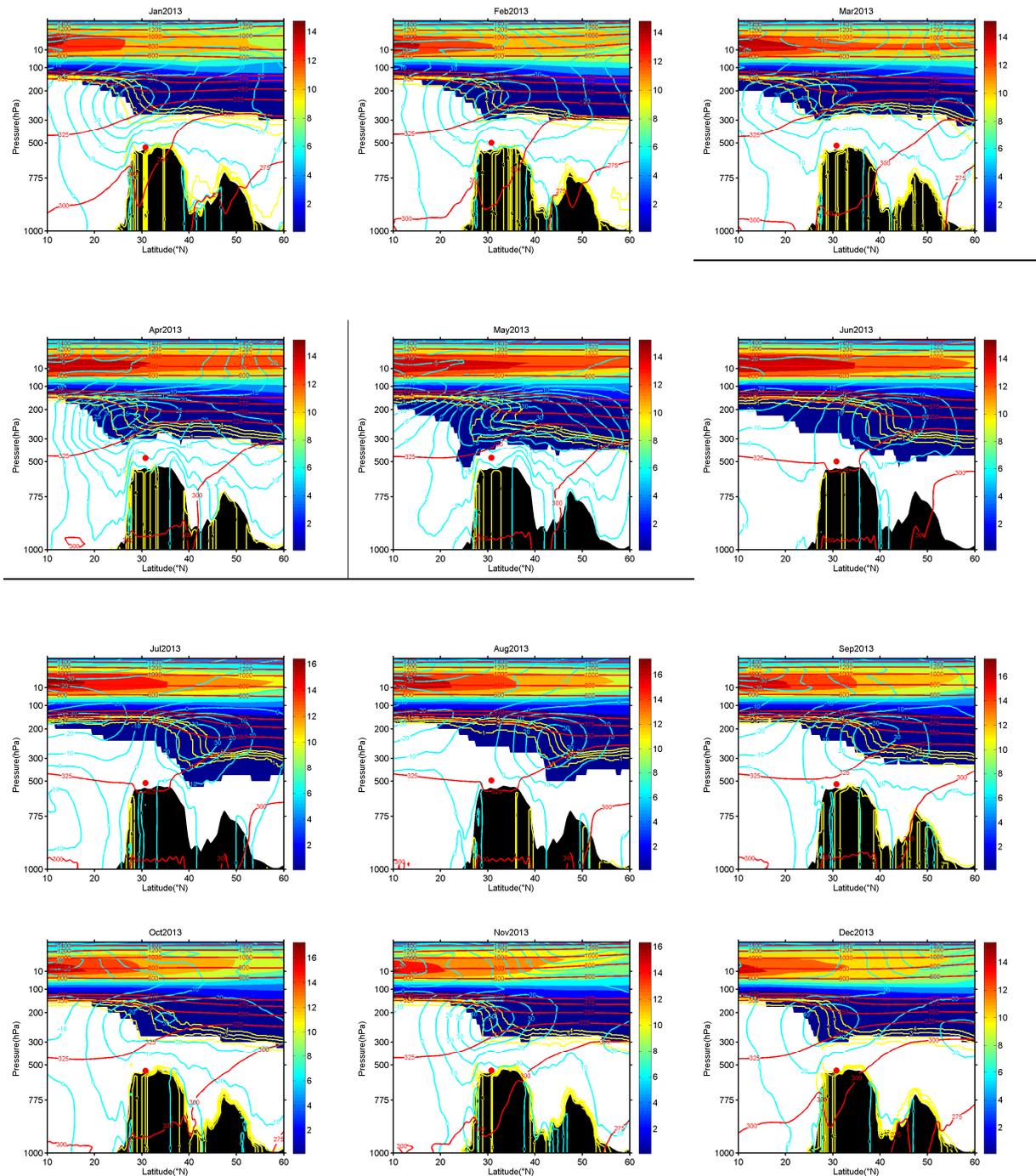
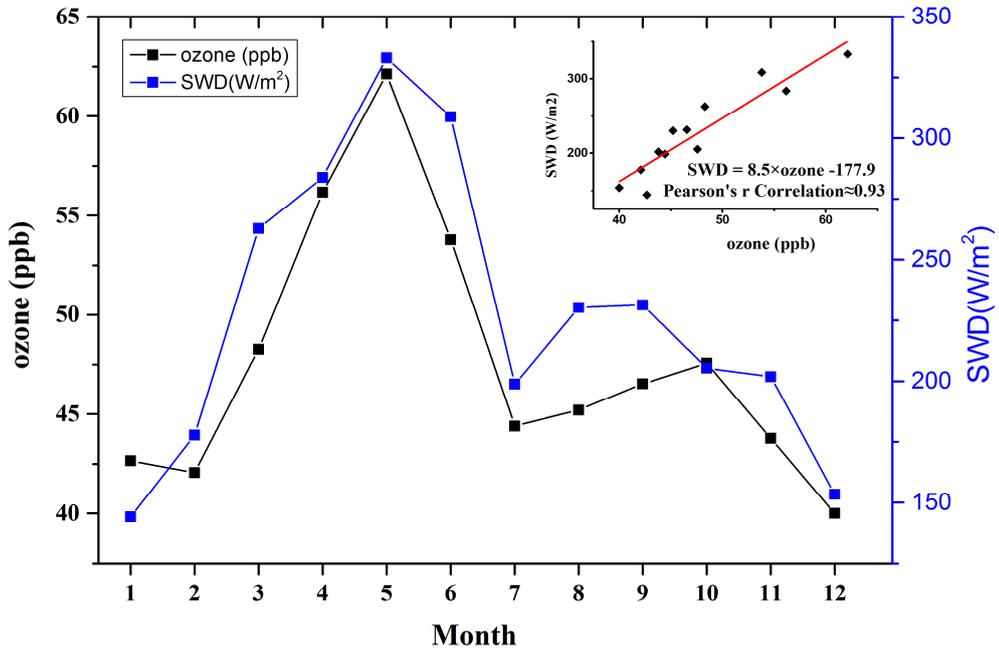


Fig. 5. Monthly mean meridional cross-section at 91°E (over Nam Co Station) at 20:00 UTC+8 in 2013, derived from ERA-Interim data, including zonal winds (cyan contours, m/s), potential vorticity (yellow lines, contours of 1, 2, 3, 4 potential vorticity unit), ozone (solid color, $\times 10^6$ kg/kg) and potential temperature (red contours, K). The color bar shows the scale for the contour plots 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 at Nam Co Station.



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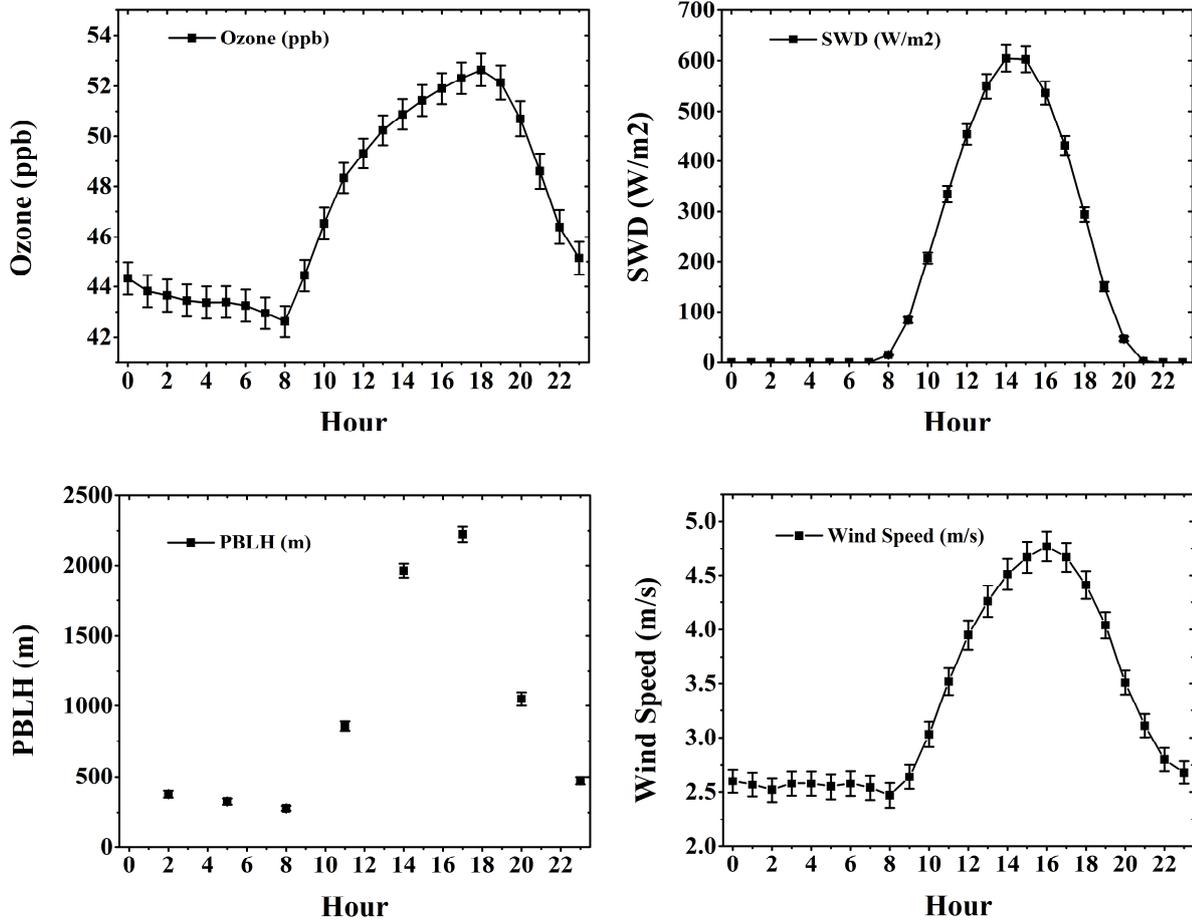
Fig. 6. Comparison between monthly average surface ozone (black) and monthly average SWD (downward shortwave radiation, blue) at Nam Co Station in 2012.

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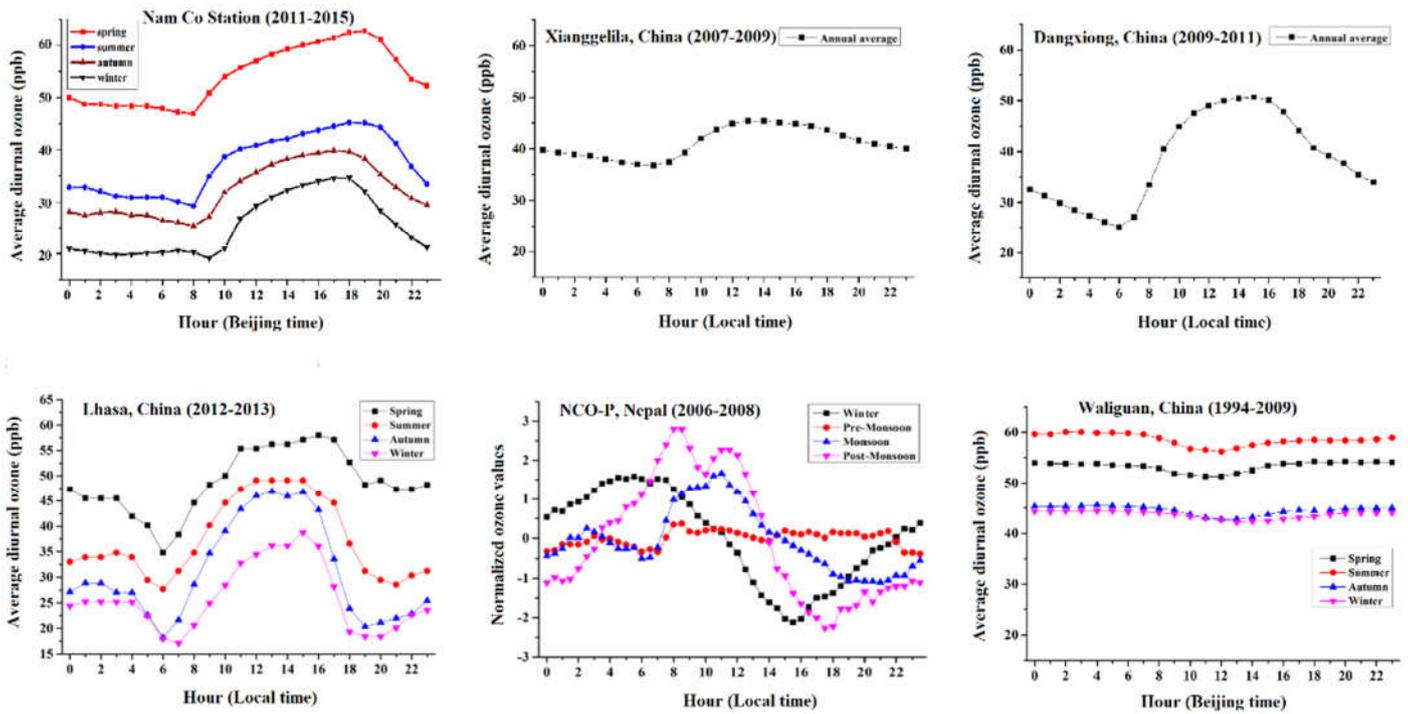
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Fig. 7 Diurnal variations of hourly average of surface ozone, SWD (downward shortwave radiation), wind speed and PBLH (planetary boundary layer height) during the whole measurement period at Nam Co Station. Error bars show the 95% confidence intervals.

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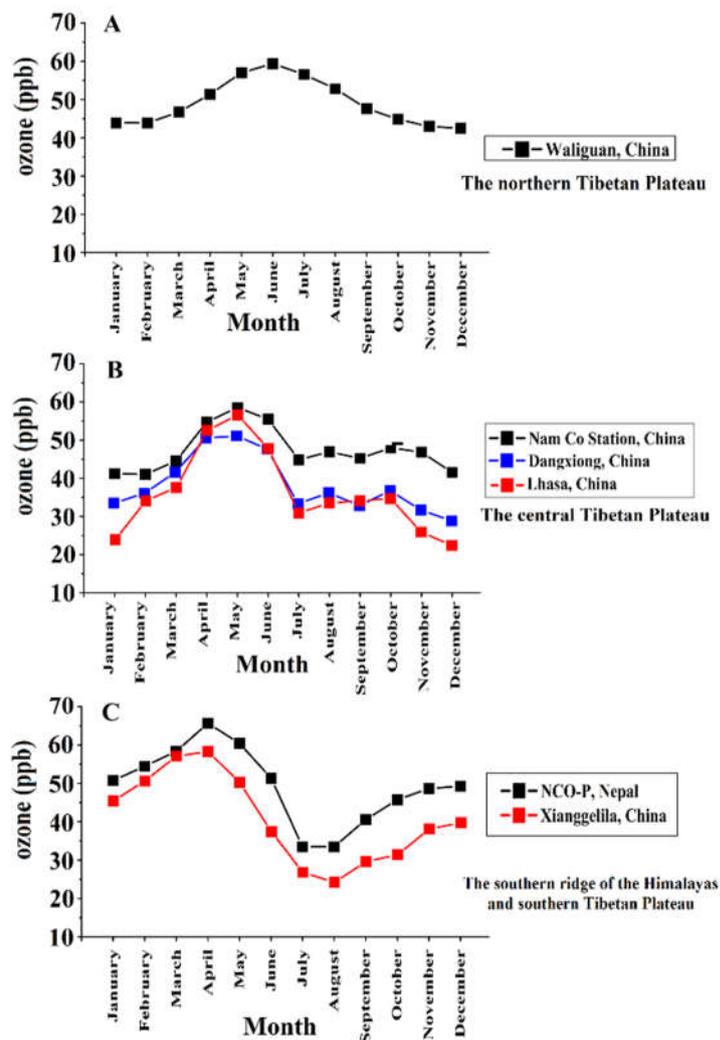
815 **Fig. 8. Comparison of diurnal profiles of surface ozone concentration at different sites in the Tibetan Plateau**
 (referred to Ma et al., 2014; Lin et al., 2015; Ran et al., 2014; Cristofanelli et al., 2010; Xu et al., 2011.)
 Measurement years at different sites are displayed in brackets.

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840 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).

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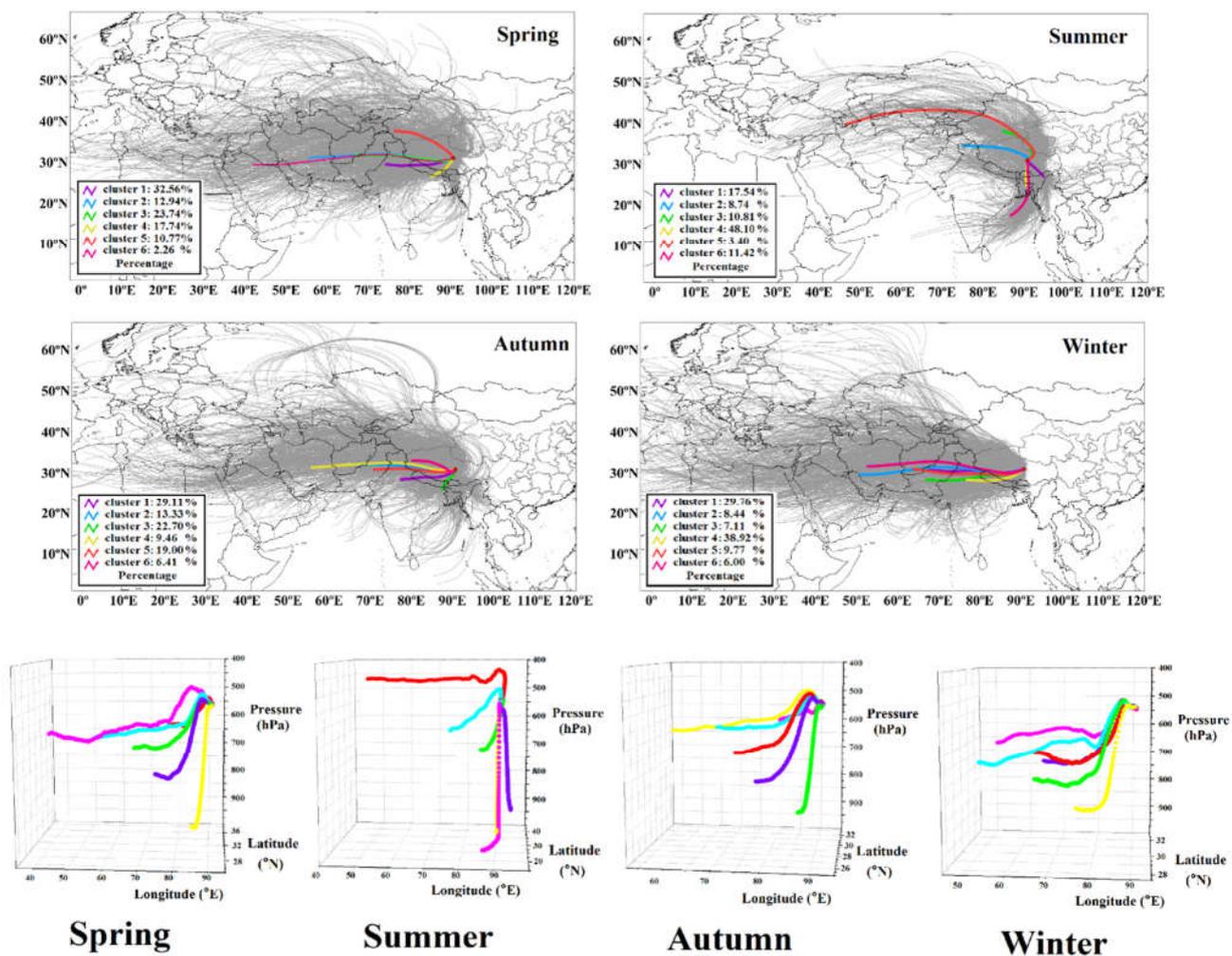


Fig. 10. Backward HYSPLIT trajectories for each measurement day (black lines in the maps), and mean back-trajectory for 6 HYSPLIT clusters (colored lines in the maps, 3D view shown on the right of the maps) arriving at Nam Co Station by season.

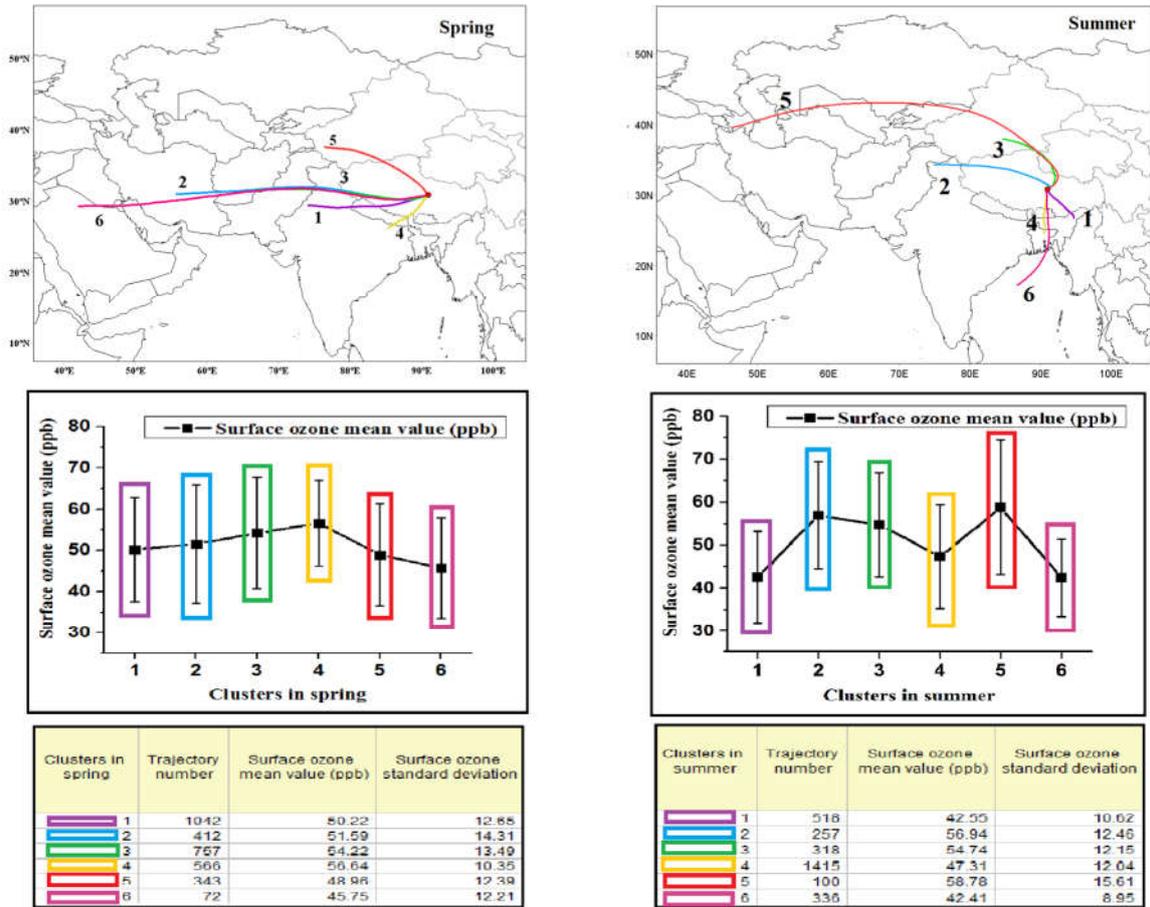


Fig. 11. Mean trajectory of 6 HYSPLIT clusters arriving at Nam Co Station in the spring and the summer, and the range of surface ozone mixing ratios measured at Nam Co Station by cluster.

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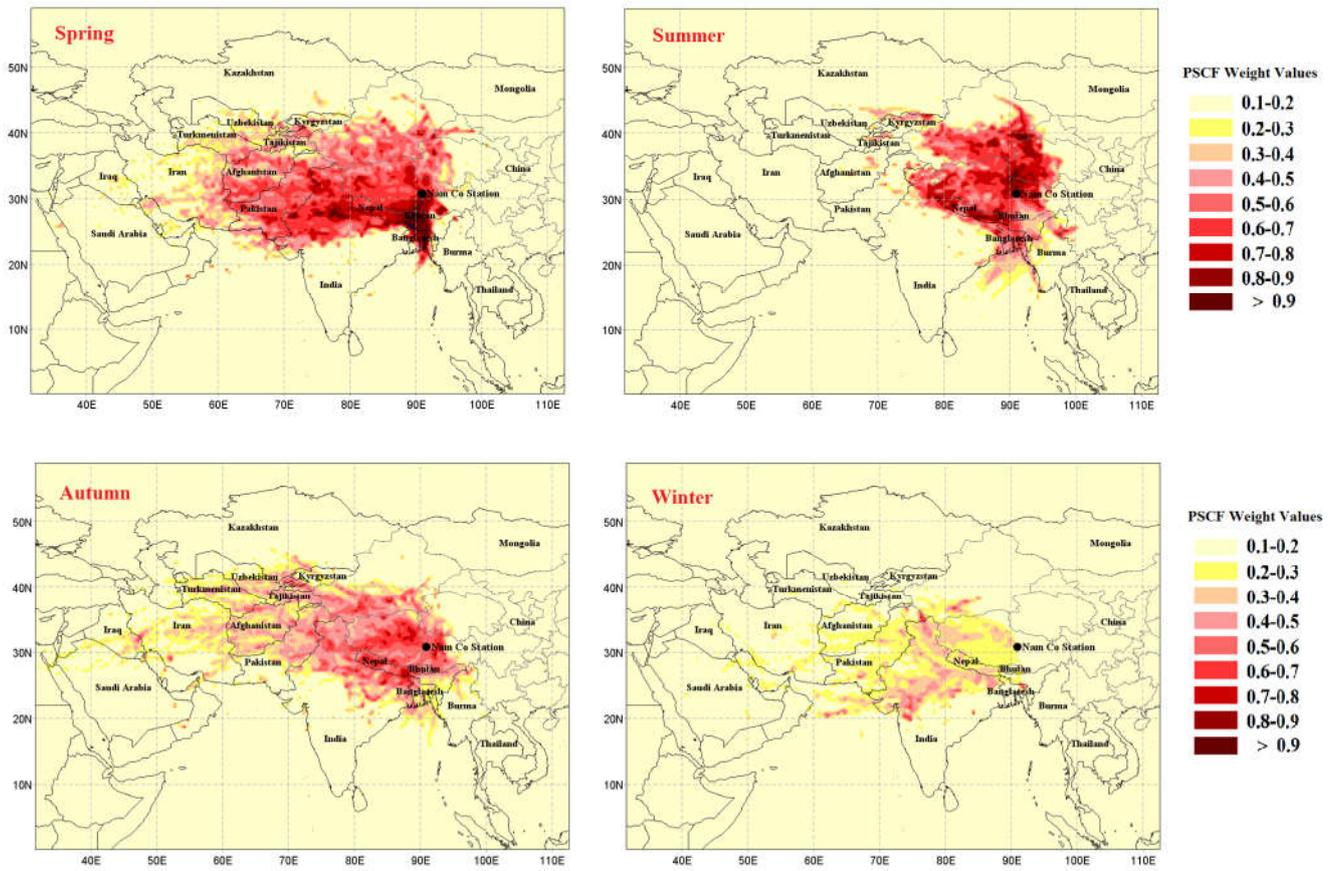


Fig. 12. Likely source areas of air mass associated with higher surface ozone concentrations at Nam Co Station by season identified using PSCF (Potential Source Contribution Function).

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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

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945 **Table 2. Multi-linear regression model for hourly ozone (2011-2014) for 3 different models and the contribution of groups of input variables to the variance (%) of the ozone time series.**

Model A: Log-transformed model of ozone concentration using CAMx stratospheric tracer to identify STE.

Model B: Linear-transformed model of ozone concentration using CAMx stratospheric tracer to identify STE.

Model C: Linear-transformed model of ozone concentration using ERA-Interim potential vorticity to identify STE.

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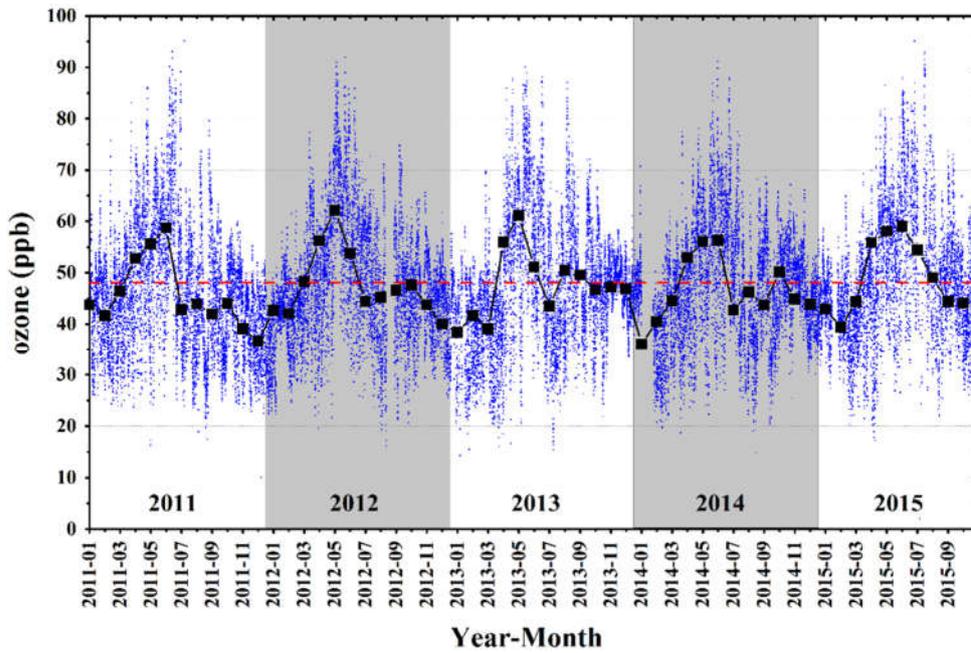
A		B		C	
Number of all hourly data	27310	Number of all hourly data	27310	Number of all hourly data	27310
Number of IRLS hourly data	26005	Number of IRLS hourly data	25934	Number of IRLS hourly data	25985
r (all hourly data calculated)	0.77	r (all hourly data calculated)	0.75	r (all hourly data calculated)	0.75
r (IRLS hourly data calculated)	0.81	r (IRLS hourly data calculated)	0.79	r (IRLS hourly data calculated)	0.80
CAMx Strat Tracers	18.2%	CAMx Strat 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%

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975 **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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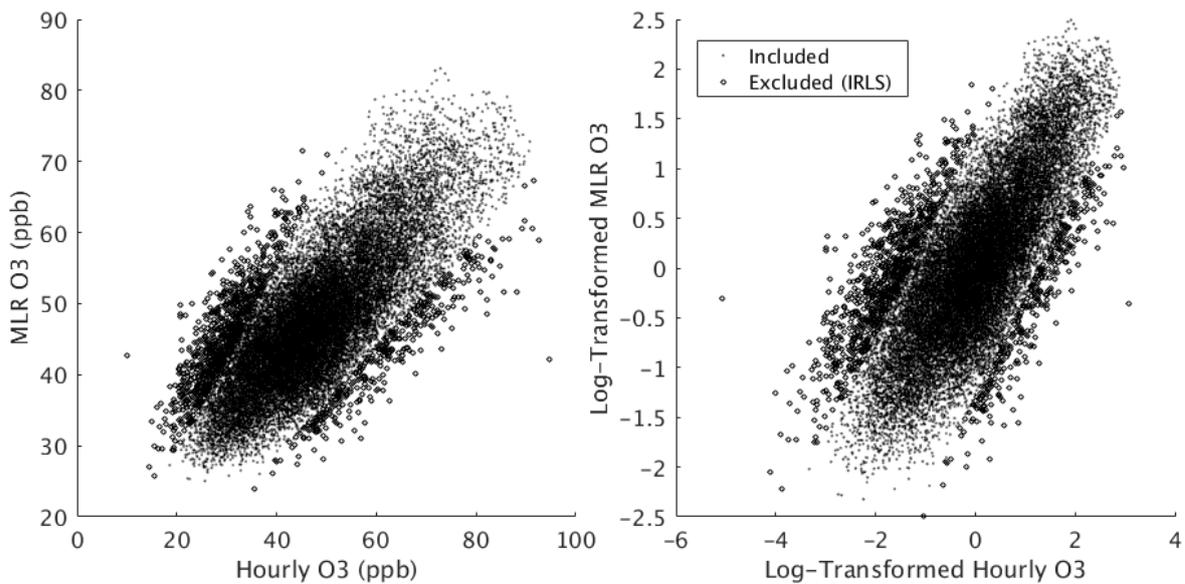


Fig. S2: Scatter plot of model surface ozone mixing ratio against observed surface ozone mixing ratio at Nam Co Station for the Multiple Linear Regression (MLR) model. The dots are points that are included in the regression; the circles are points that were excluded as outliers by the Iteratively Reweighted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

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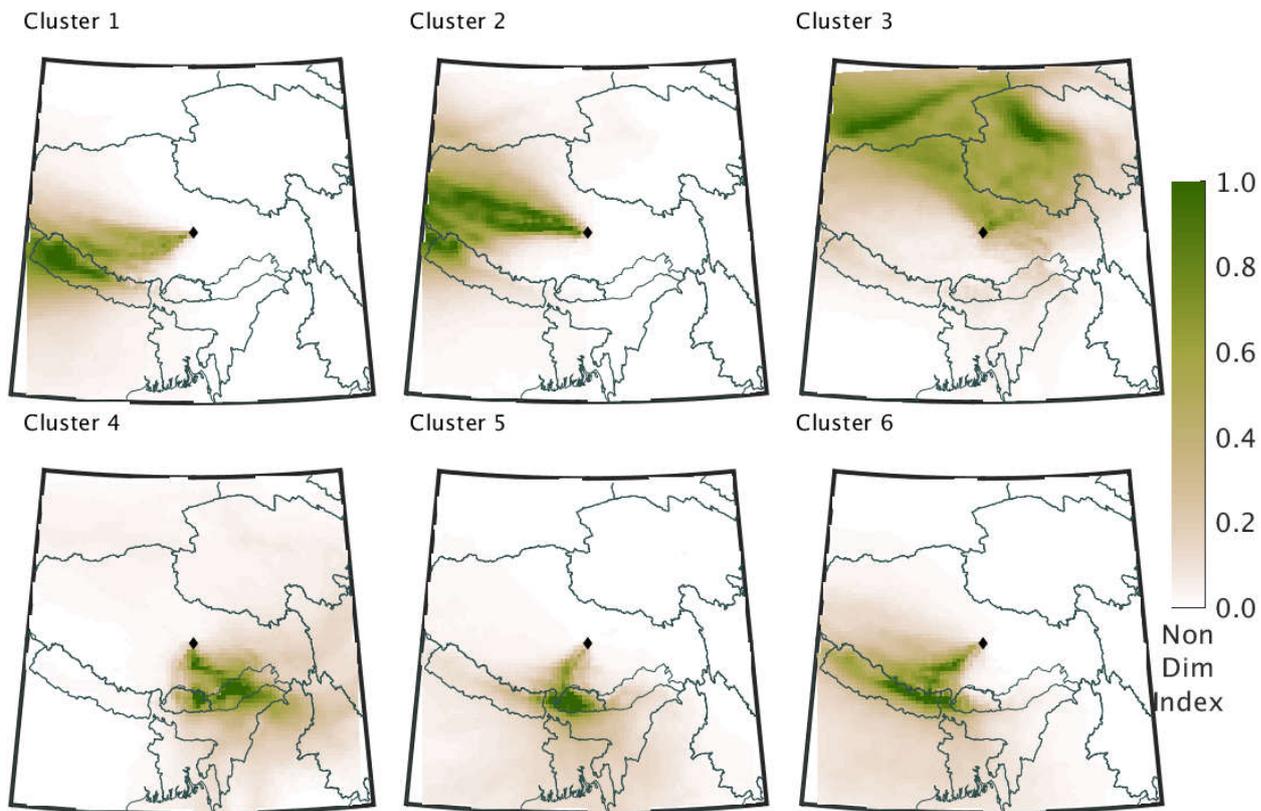
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Fig. S3. Average Residence Time Analysis grids for each WRF-FLEXPART trajectory clusters at Nam Co Station. The black diamond represents the sampling site.

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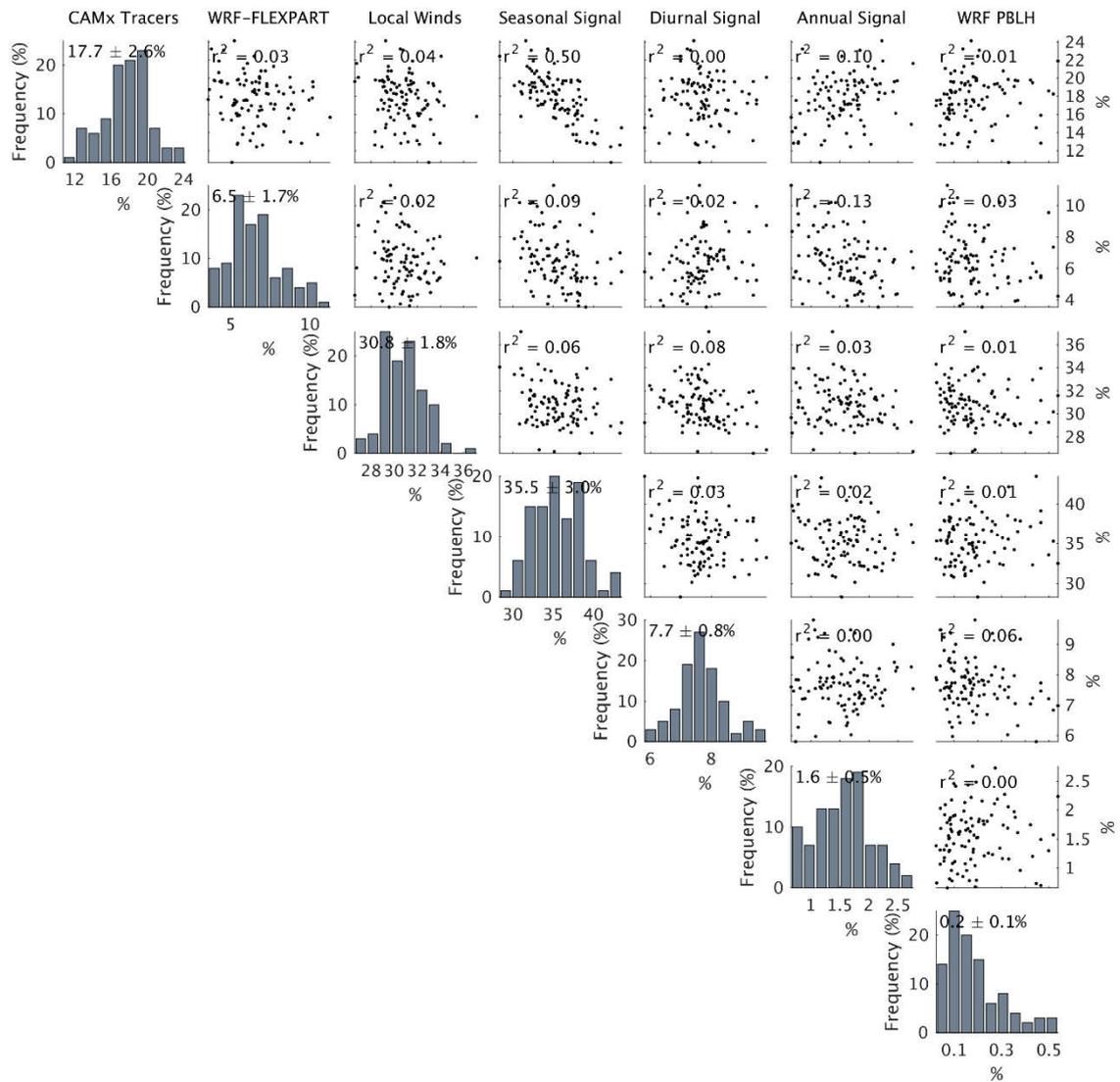


Fig. S4. Uncertainty and covariation of the Multi-Linear Regression estimates of the contribution to ozone variance by group, based on 100 realizations of the model using block-bootstrapping. Histograms show the distribution of the contribution estimates and the scatter plots show the cross-correlation 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.

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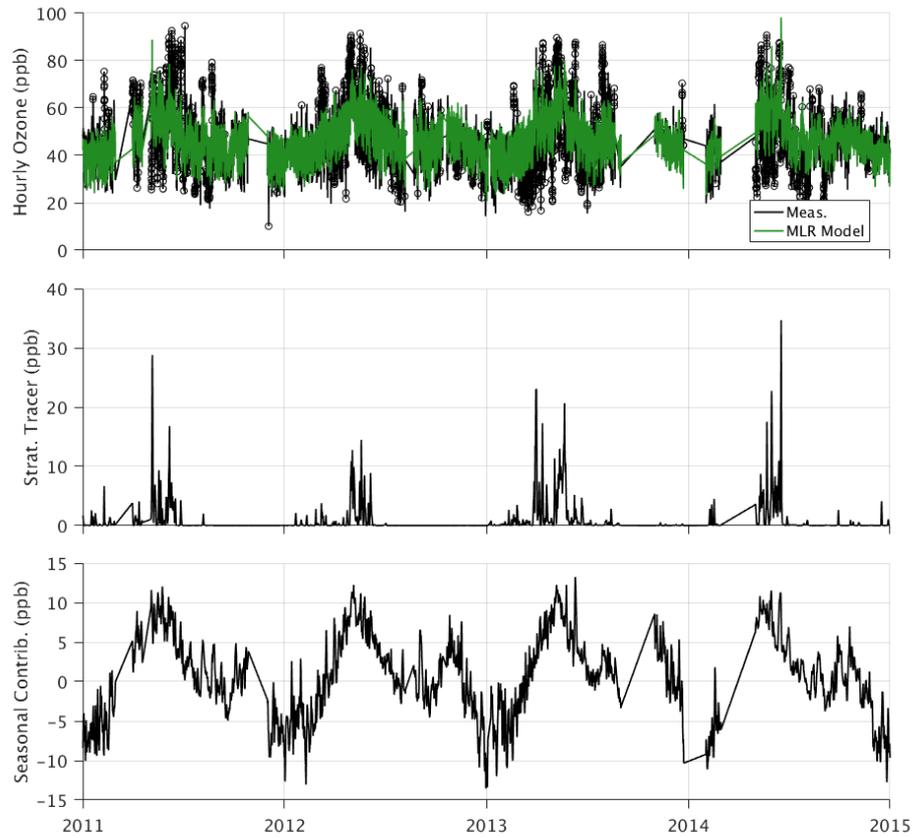


Fig. S5. Same as Fig. 4 but for the linear model. Because the model is linear, the contributions of the stratospheric tracer and the seasonal signal are in concentrations (ppb) rather than scaling factors.

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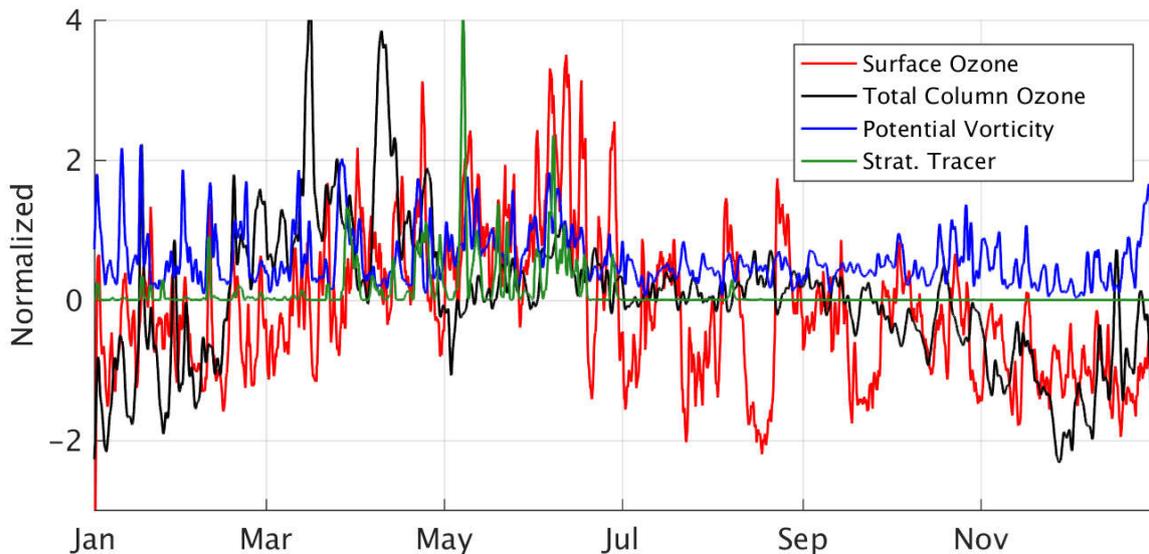


Fig. S6. Time series of 24-hour running average of surface ozone measured at Nam Co for 2011; ERA-Interim Total Column Ozone above the Himalayas, ERA-Interim Potential Vorticity at 350 hPa above the Himalayas (units of PVU); and stratospheric ozone tracer simulated by CAMx (units of 0.1 * ppb).

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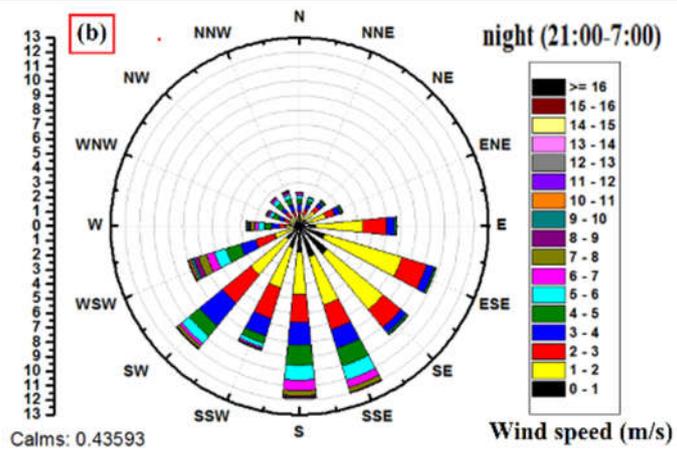
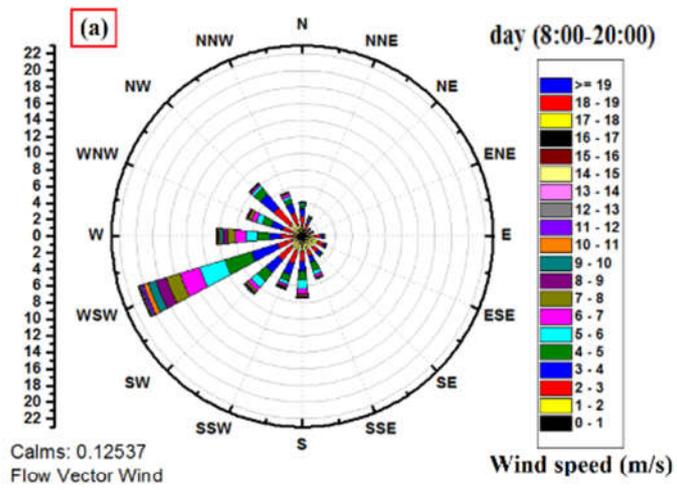


Fig. S7 Wind rose at Nam Co Station during the day (a) and at night (b).

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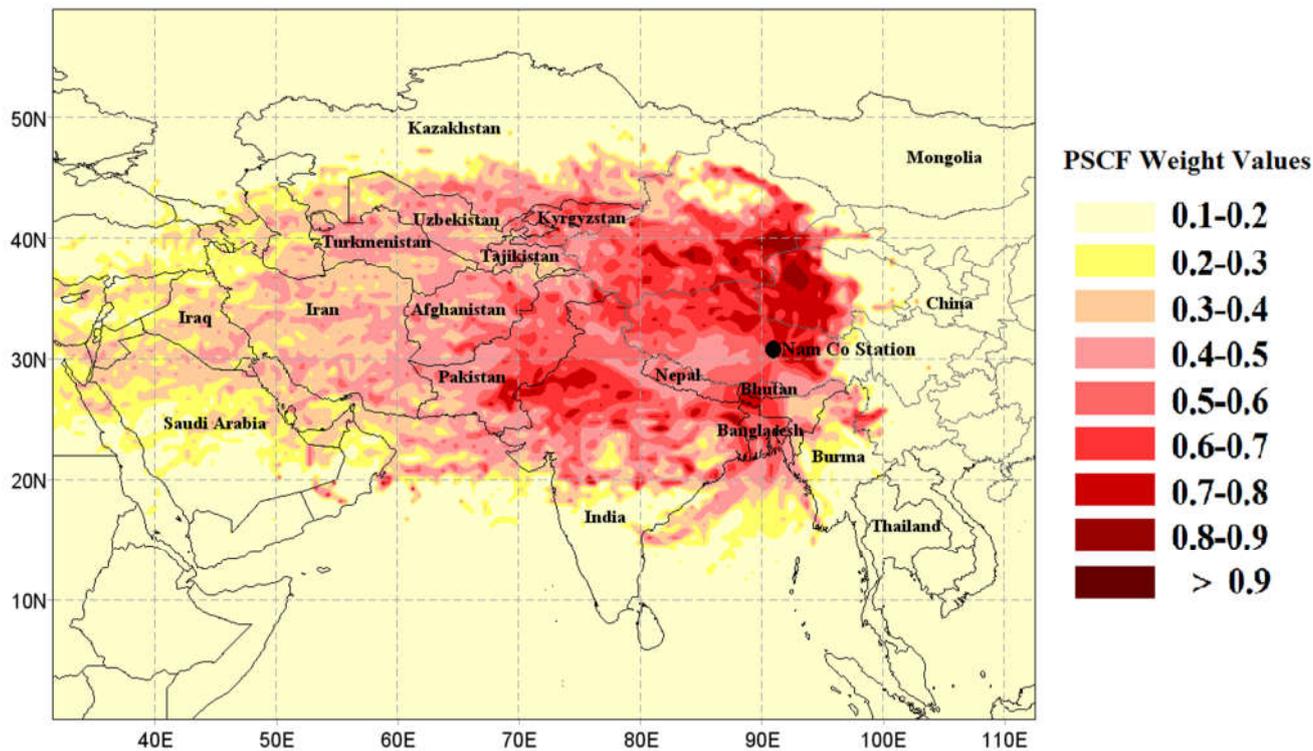


Fig. S8. Likely source areas of air masses associated with higher surface ozone concentrations at Nam Co Station during the whole measurement period identified by PSCF (Potential Source Contribution Function).

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Table S1. Multi-linear regression model by season using log-transforms and CAMx stratospheric tracers (corresponding to Model A in Table 2) and the contribution to ozone variance (%) by group in different seasons.

Statistical Metrics	Spring	Summer	Autumn	Winter
Number of all hourly data	6043	7992	6157	7118
Number of IRLS hourly data	5750	7615	5878	6759
r (all hourly data calculated)	0.83	0.77	0.68	0.74
r (IRLS hourly data calculated)	0.86	0.81	0.73	0.79
CAMx Strat 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%