

General comments:

1. *Unfortunately, the paper remains a bit vague and less conclusive in some parts but refers to an accompanying paper that is still not yet available. As the present paper seems to lay the foundation for future analysis, the experimental section should be more elaborated on (see comments below) because a sound quality assurance of the data is key when looking at trends etc. Moreover, the findings should be more discussed in relation to other available time series at elevated Northern hemisphere measurement sites. This is already the case for the trends in Chapter 3.3 but could be extended to the sections where diurnal and seasonal cycles are presented.*

Response: Thank you for these valuable suggestions. We have added/corrected information in the site and measurements section (including the height of the measurement platform, the detailed processing procedure of the data, etc.), which hopefully will assure the reader of our sound data quality. The diurnal and seasonal cycle of ozone at WLG will be further discussed in relation to the other elevated northern hemispheric sites. However, this study mainly focuses on the long-term trends, which is why the added discussion was kept brief.

2. *The authors often refer to ozone concentrations but use ppb units. Concentrations cannot be given in ppb as numbers in ppb refer to mole fractions or mixing ratios.*

Response: We thank the referee for pointing it out. Indeed, by definition, mixing ratio and concentration is not the same, and the ppb values in the manuscript refer to the mixing ratio of ozone. According to the suggestion, we changed all the “concentration” to “mixing ratio”.

3. *The order of the Figures does not correspond with the appearance in the text. The references to Figs. 4 and 7 come earlier than the one to Fig. 3. Please reorder the Figures.*

Response: We thank the referee for the suggestion. Since the figures were ordered mainly according to the results and discussion section, we did not want to reorder them, for better logicity and readability. However, we deleted the previous

references of Fig 4 and 7 in the data and methodology section, since they were not indispensable.

Specific comments:

1. Abstract:

The abstract is rather long; I suggest shortening it, e.g. by deleting “using a modified Mann–Kendall test and the Hilbert–Huang Transform analysis for the trend and periodicity analysis, respectively.” and “Analysis suggests that there is a season-diurnal cycle in the three-dimensional winds on top of Mt. Waliguan. Season dependent daytime and nighttime ranges of 6 h were determined based on the seasondiurnal cycle in the three-dimensional winds and were used to sort subsets of ozone data for trend analysis.”

Line 22: replace “increasing trend” by “positive trend”

Line 24: delete “relatively”

Lines 25-26: shorten the sentence to “Spectral analysis identified four episodes with different positive trends, with the largest increase ...”

Response: Thanks for your advice, the abstract has accordingly been revised as:

“Tropospheric ozone is an important atmospheric oxidant, greenhouse gas and atmospheric pollutant at the same time. The level of tropospheric ozone, particularly in the surface layer, is impacted by emissions of precursors and is subjected to meteorological conditions. Due its importance, the long-term variation trend of baseline ozone is highly needed for environmental and climate change assessment. So far, studies about the long-term trends of ozone at representative sites are mainly available for European and North American sites. Similar studies are lacking for China, a country with rapid economic growth for recent decades, and many other developing countries. To uncover the long-term characteristics and trends of baseline surface ozone mixing ratio in western China, measurements at a global baseline Global Atmospheric Watch (GAW) station in the north-eastern Tibetan Plateau region (Mt. Waliguan) for the period of 1994 to 2013 were analysed in this study. Results reveal higher surface ozone during the

night and lower during the day at Waliguan, due to mountain-valley breezes. A seasonal maximum in summer was found, which was probably caused by enhanced stratosphere-to-troposphere exchange events and/or by tropospheric photochemistry. Significant positive trends in surface ozone were detected for both daytime (1.5-2.7 ppbv 10a-1) and nighttime (1.3-2.9 ppbv 10a-1). Autumn and spring revealed the largest increase rates, while summer and winter showed weaker increases. Spectral analysis identified four different episodes with different positive trends, with the largest increase occurring around May 2000 and Oct. 2010. A 2-4 year, 7 year and 11 year periodicity was found in the surface ozone mixing ratio. The results are highly valuable for related climate and environment change assessments of western China and surrounding areas, and for the validation of chemical-climate models.”

2. **Introduction:**

Page 30990, line 14: reference to Lin, 2015 is missing

Response: We appreciate your detailed inspection. There has been a mistake in creating a link to the reference, which has been corrected in the revised manuscript.

Page 30991, lines 4-5: “there are a few representative sites ...”; does this statement refer to the situation in China? Which are the other stations? To my knowledge, the China Meteorological Administration also operates a remote measurement station at Shangri-La at nearly the same elevation than Mt. Waliguan. Are surface ozone observations available from the Shangri-La station?

Response: Yes, this statement was referring to the situation in China, which we will make clear by rephrasing this sentence to “Continuous long-term observations of surface ozone are made only at a few representative sites in China, among which is the Mt. Waliguan (WLG) GAW station”. Continuous surface ozone measurements are also carried out at Shangri-La, Yunnan Province (since the end of 2007) and Akedala, Xinjiang Province (since 2009), which are both sites in western China. Waliguan is the site with the highest altitude and the longest (and

most complete) surface ozone data record.

3. *Section 2.1 Sites and measurements*

This part needs some elaboration. Duplicate ozone measurements seem to be available for most of the time. The authors state that data were used if the two analyzers agree within 5ppb. A quality control criterion of matching data within 5ppb is pretty lax and well above the data quality objectives for key GAW goals (see e.g. the GAW report #209 “Guidelines for Continuous Measurements of Ozone in the Troposphere”; available at <http://www.wmo.int/pages/prog/arep/gaw/gaw-reports.html>). How was the data flow implemented in detail? Was there one master and one backup instrument? How did they compare? Did you experience e.g. a steady bias, a perfect match, a difference as function of daytime, season, temperature, humidity ...? Or random differences? What happened when the master instrument didn't record data but data from the backup analyzer were available? Were the data from the backup instrument used to fill the gaps? Have the backup data been corrected based on a long-term master backup comparison? How many gaps were filled? Maybe an additional figure could help just showing a time series that illustrates which analyzer provided when data for the final data set used for the analysis. The authors mention that a TE49i model is used since 2011. Did this analyzer become the master instrument? How was it ensured that there is a smooth transition when changing the master instrument? Did the old and the new master run in parallel for a certain time? All these things are important information because the observed trends are small and could be also biased by some instrumental artefacts. When looking at Fig. 4a, there seems to be a discontinuity with slightly elevated ozone mole fractions for approximately the last two years. This step roughly coincides with the implementation of the TE49i analyzer. Can the authors comment on that?

Response: We understand the referee's concern about the data quality and agree

that it is fundamental for this and the upcoming study to assure the reader that our measurements are trustworthy. The GAW report #209 suggests for the case of two instruments, to eliminate 1-minute averaged data showing a difference larger than 5 ppb. Since we had data records in 5-minute averages, we eliminated data with differences larger than 3 ppb (Apology for the wrong information in the previous manuscript). The data quality objectives in the GAW report #209 requires the measurement to have an uncertainty of ± 1 ppb or less. The following figure shows the occurrence frequency distribution with the difference of ozone mixing ratio between two instruments. As can be seen in figure 1, 95% of the data pairs show discrepancies within ± 1 ppb and the difference between two instruments shows nearly a random distribution around zero.

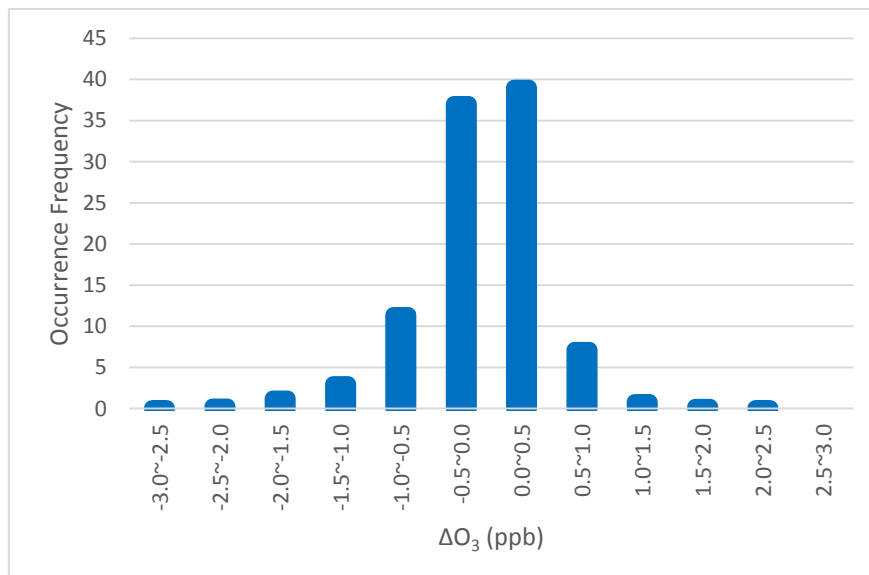


Figure 1 Occurrence count distribution with the difference of ozone mixing ratio between two instruments

The results of the audit reports from the World Calibration Centre for surface ozone should be further proof that our instruments were capable of fulfilling the data quality objectives.

The two instruments were run parallel to each other, with no particular differentiation of master or backup. Generally, they were in very good agreement with each other, and discrepancies larger than 3 ppb typically occurred, when one of the instruments was undergoing a change of particle filter or had a technical

problem such as leaky valves. In that case, the data of the other instrument was used. There was only a small proportion of time, when one of the instruments did not function well.

After the installation of the model TE49i ozone analyzer, our way of running the two instruments (parallel to each other) has not changed and the same criterion for data elimination has been applied. This, together with periodical calibrations using the ozone calibrator at the station, has ensured a smooth transition and no abrupt change in data quality due to the analyzer replacement. The slightly elevated ozone mixing ratio in the last two or three years was not due to the change of an analyzer rather due to other causes as will be discussed in part II.

Page 30993, lines 10-11: "Surface ozone data are recorded every 5 minutes ...". I assume that this statement is misleading as the used ozone analyzers record data in 10 sec intervals. I suppose that the authors want to say that 5 min averages are recorded on the data acquisition. If this is the case, why not saying "Surface ozone data are recorded as 5-minute averages and corrected ..."

Response: The correction suggested by the referee is adopted in the revised manuscript, thank you for the comment.

What was the sampling height above ground for the ozone observations?

Response: The sampling height is 7 m above ground. Thank you for pointing it out, we will add this information to the section on Site and Measurements.

Page 30996, lines 4.5: rephrase the last sentence that it reads "The nighttime window also covers 6 h and is considered to be offset by 12 h to the daytime window."

Response: The correction suggested by the referee has been accordingly made in the revised manuscript.

Section 2.3: Did you use hourly averages for the analysis? Which software was

used for the statistical analysis? Matlab? R? Did you use specific add-ons (packages)?

Response: Thank you for the questions. As shown in Fig. 5, monthly average data were used in the seasonal Mann-Kendall analysis, which we will make clear in the revised manuscript. A fortran program by USGS (Computer program for the Kendall Family of Trend Tests) was used, which we forgot to cite in the former manuscript. A proper citation will be added in the revised manuscript.

Page 31000, line 22: replace “Past researches” by “Previous studies”

Response: The correction suggested by the referee has been accordingly made in the revised manuscript.

Page 31001, lines 10-11: How does the long-term time series of $^{10}\text{Be}/^{7}\text{Be}$ look like. Is it possible to draw any conclusions on changes in STE strength?

Response: Unfortunately, we do not have long-term measurements of $^{10}\text{Be}/^{7}\text{Be}$. We will have to look into other indicators of the STE strength and occurrence frequency.

Page 31001, line 20: “total ozone column”, remove the “,”

Response: Thank you for your careful reading, the correction is made in the revised manuscript.

Page 31001, line 24: mention once more “based on zonal wind information”.

Response: The correction suggested by the referee has been accordingly made in the revised manuscript. This indeed improved the readability of the manuscript.

Page 31003, lines 6-8: this is mainly true for remote locations.

The seasonal ozone peak in the Northern Hemisphere typically occurs in spring, which is believed to be the result of enhanced photochemical production in spring (Monks, 2000; Vingarzan, 2004).

Response: Thank you for pointing that out, we revised this sentence to: “The seasonal peak of the Northern Hemisphere background ozone typically occurs in spring, which is believed to be the result of enhanced photochemical production in spring (Monks, 2000; Vingarzan, 2004)”.

Page 31006: replace “Ds” by “DS” at various locations.

Response: The correction suggested by the referee has been accordingly made in the revised manuscript.

4. ***Summary:*** *the concluding chapter only summarizes the findings presented above. I would like to see some outlook beyond. What will be looked at next? What are the implications of the findings? What does it e.g. mean for efforts to reduce maximum ozone levels in urban agglomerations (e.g. if ozone input due to STE is getting stronger)? Can the results somehow be generalized? What does it mean for the Asian outflow towards the Western US? Is the observed trend in Western US maybe caused by changes in STE input rather than increasing ozone precursor emissions in Asia?*

Response: Thank you for the suggestion. We will add the following few sentences to summarize the implication of our findings and to give an outlook of what to expect in the second paper.

“As WLG is a high altitude mountain-top site in a remote region, measurements of surface ozone and other species can well represent a large scale situation. Previous air mass origin studies and modelling studies suggest that WLG is mostly under the influence of transport from the north-west direction, hence the upward trend in ozone might be a reflectance upon transport from Europe (Zhang et al., 2011; Li et al., 2014). Since Eastern China is in the downwind direction, our results imply that under rising background ozone conditions, even more effort needs to be put in reducing ozone precursors. In the second part of our study, the impact of different air-mass origins and the long-term variations of their occurrence frequencies on the surface ozone mixing ratio and its trend at WLG will be shown. The

anthropogenic impact of the nearest major population centers on the ozone trend will be discussed. The long-term variation of STE and its link to surface ozone at WLG will be displayed. The possible connection of changes in atmospheric circulation oscillations and solar activities with the inner-annual and periodical variations of ozone at WLG will be studied.”

The referee posed a question on the influence of rising ozone at WLG on western US air quality. WLG is located downwind of Europe and upwind of East Asia, one of the most polluted region of the world. If it should have any influence on Western US, it won't be as big of an influence as the outflow of East Asian ozone precursor emissions.

5. *References:* *Add urls to the Zellweger et al. audit reports, if online available.*

Response: URLs have been added for the audit reports, thank you for the suggestion.

6. *Figures:*

Figs. 2 and 3: is it confusing to have two different sets of white dots and dashed lines in Figs. 2 and 3. Since the differences in the seasonal-diurnal variations are discussed in Section 3.1, I suggest to add the daytime range based on the zonal wind (white dots from Fig. 2) in Fig. 3 and to draw the white dots in Fig. 3 based on minimum ozone in a different color. This makes it easier for the reader to compare the different features. The +/- 3h band is maybe even not needed here.

Response: We thank the referee for the good advice, the correction suggested above has been accordingly made and the discussion on Fig. 3 will be adjusted to the following new figure:

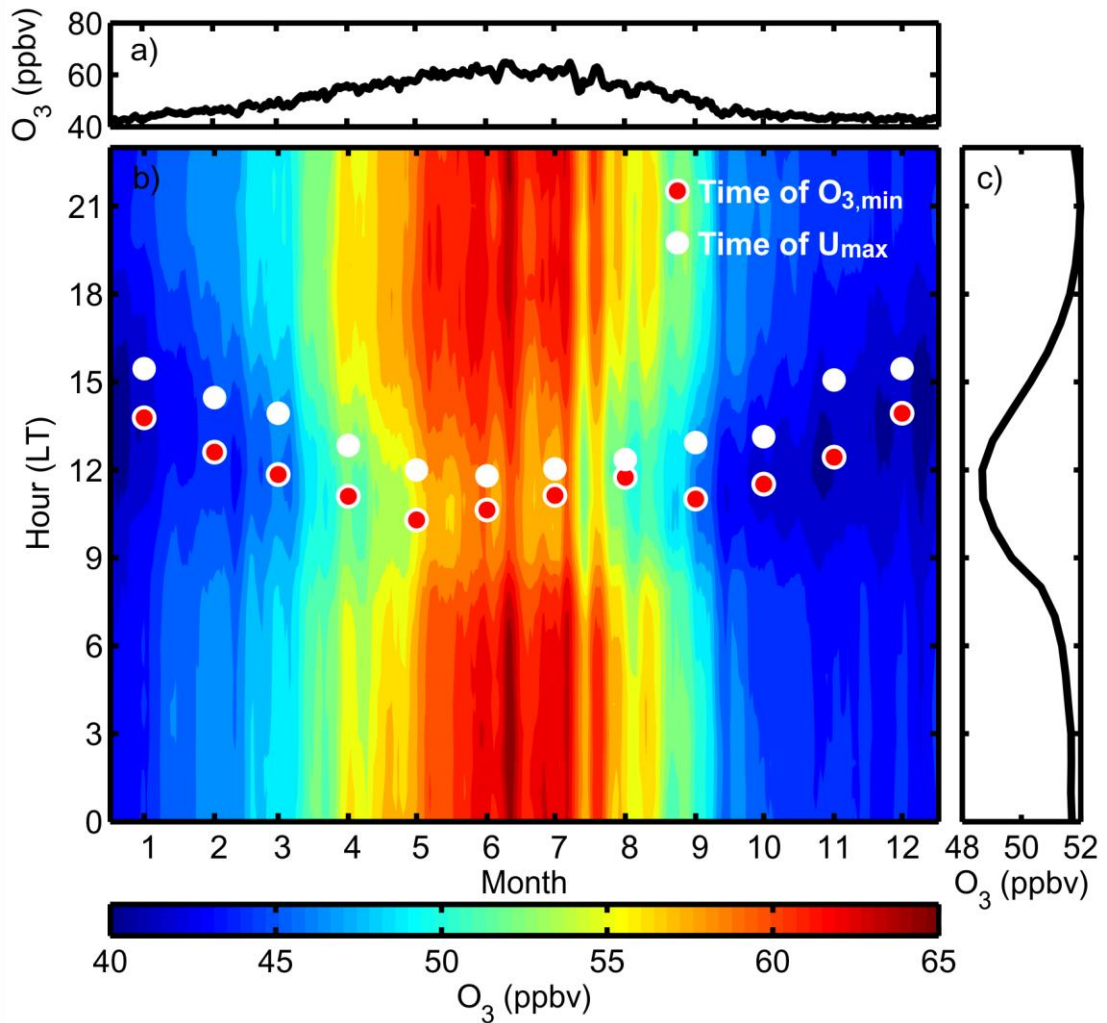


Figure 3 The average seasonal variation (a), season-diurnal variation (b) and diurnal variation (c) of ozone during 1995 to 2013. White and red dots stand for the monthly average local time associated with the diurnal maximum zonal wind and minimum ozone, respectively.

Fig. 5: add on the right hand side of panels a5 to b5 “all”, “daytime”, “nighttime”;
add below the x-axis of the c-row “all”, “spring”, “summer”, ...

Response: The correction suggested by the referee has been accordingly made in the revised manuscript. The new Fig. 5 is shown below:

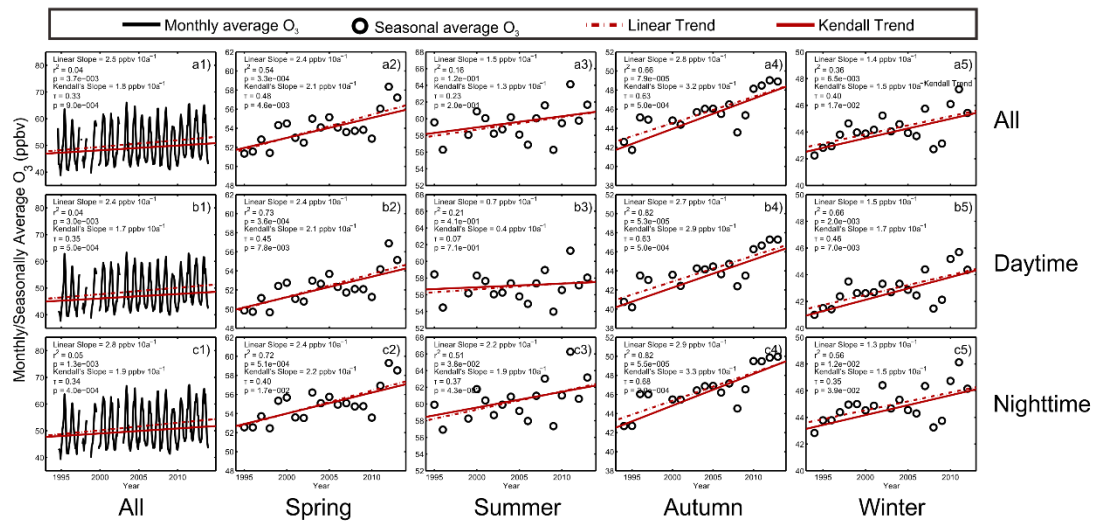


Figure 5. 1) Monthly, 2) spring (MAM), 3) summer (JJA), 4) autumn (SON) and 5) winter time average all day (a), daytime (b) and nighttime (c) surface ozone mixing ratio during 1994 to 2013 (black solid line or black circles) and its variation trend (red lines: dotted line stands for the linear variation and solid line stands for the Kendall's variation slope).