

Interactive comment on “Long-term trends of surface ozone and its influencing factors at the Mt. Waliguan GAW station, China – Part 1: Overall trends and characteristics” by W. Y. Xu et al.

Anonymous Referee #2

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Major Comments:

The manuscript presents long-term changes of surface ozone measured at Mount Waliguan in western China. The authors conducted a spearman's linear trend analysis and the Man-Kendall's trend test to determine the slopes of the time series and their 95% confidence intervals (Table 1 and Figure 5). Diurnal and seasonal variations of the slopes are discussed. Spectral analysis is used to determine the time scales of ozone variations (Figures 6 to 8). The scientific approach and applied methods in the manuscript are overall valid. High-quality ozone measurements are sparse in China, and thus the long-term ozone data at Mount Waliguan are highly valuable. However,

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some discussions presented in the current manuscript are vague or sometimes inaccurate. The present manuscript does not provide conclusive evidence on the causes of seasonal ozone trends measured at Mt. Waliguan. The record clearly shows large interannual variability (e.g., the 2011-2012 high-ozone anomalies in spring), which can substantially influence the slope of the linear regression (Fig.7b), but there are no thorough discussions on what are going on. For instance, are there any changes in large-scale circulation patterns during 2011-2012: shifts in the location of the jet stream, anomalies in 500 hPa geopotential height, variability of STE or regional pollution transport? There is a citation to the Part II paper in prep on the influencing factors. The referee suggests that the authors try to condense the discussions and combine the two manuscripts into one concise, thorough, and well-structured paper, which is better than two incomplete papers and will result in better citations in the future.

The manuscript also needs be carefully proofread for the correct use of English Language. There are quite a few errors.

Specific comments:

1. Abstract, Line 16-18: Since this manuscript is NOT about the seasonal cycle of ozone at Waliguan, I don't think you need to get into what causes the summertime ozone maximum in the abstract. The seasonal cycle has been extensively discussed in the literature (e.g. Zhu et al., Ma et al., Ding et al.) as the authors noted in the main text.
2. Abstract, Line 19-20: "Seasonal-dependent daytime and nighttime ranges of 6h . . ." Awkward wording.
3. Abstract, Line 22-23 and Figures 6-8: What does the range of the slope represent? It is more appropriate to report the trends with its 95% confidence intervals in the format of $x \pm x$ ppbv yr⁻¹. The daytime trend for JJA is statistically insignificant at the 95% confidence level (Table 1 and Figure 5b3). I would suggest in the abstract reporting the nighttime trends in $x \pm x$ ppbv yr⁻¹ for annual mean and for each season, which is the

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most useful information for the future readers of the paper. Observed conditions during nighttime at the 3.8 km altitude of Mount Waliguan represent downslope influence of free tropospheric air. Thus nighttime measurements are more representative of baseline conditions compared to daytime measurements. Related to this comment, I would suggest restricting the spectral analysis in Figures 6-8 to nighttime data that are representative of large-scale conditions. Daytime data are influenced by local boundary layer air, particularly during summer, as evidenced by the large differences in daytime and nighttime trend for JJA (Table 1).

4. Abstract, Line 27: "with the largest increase occurring around May 2000". Where do you see this? In Figure 6f? But it does not show up in the 7-year trend (Figure 6e and Figure 7b). Aren't the changes in the ozone increasing rates (slope) just the manifestation of the interannual variability?

5. Somewhere in the abstract, please denote the altitude of Mt Waliguan.

6. Page 30989, Line 15-28, awkward wording

7. Page 30989, Line 20: It is important to clarify that the STE influence on surface ozone is most relevant at alpine sites. Thus, please change "local surface ozone concentrations" to "surface ozone concentrations at high-elevation sites".

8. P30990, Line 1-3: Also cite Parrish et al. (2012, ACP) and Logan et al. (2012, JGR, D09301).

9. P30990, Line 9-10: → "... in causing high-ozone events at western U.S. alpine sites during spring (e.g. Langford et al., 2009; Ambrose et al., 2011; Lin et al., 2012a; Lin et al., 2015)".

10. P30990, Line 10-15: The discussions of the results from Lin et al. (2015b) are not quite accurate. They found statistically insignificant ozone trend for the short record of 1995-2008 but the trend is significant for the longer time period of 1995-2014. Consider revising the text as follows:

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"A recent study by Lin et al. (2015b) found that although rising Asian emissions contribute to increasing springtime baseline ozone over the western U.S. from the 1980s to the 2000s, the observed western US ozone trend over the short period of 1995-2008 previously reported by Cooper et al. (2010) has been strongly biased by meteorological variability and measurement sampling artifacts. Nevertheless, the impact of Asian pollution outflow events on western US surface ozone is evident (e.g., Lin et al., 2012b)."

11. The last sentence in P30990: Revise "NCP, YRD and PRD. Observed ozone ..." to "NCP, YRD and PRD, where observed ozone ..."

12. P30992, Line 2: "a larger scale" compared to what? You can just say "on a large scale".

13. P30991, Line 19-30: It is not clear why you bring up the discussions on ENSO and its influence on western ozone. I think the connection is that both WLG and western US are high-elevation regions prone to the STE influence, which can be modulated by climate variability such as ENSO events. Also, Voulgarakis et al. (2011) did not say that changes in dynamics after el nino events hardly leads to changes in stratospheric ozone. In fact, the influence of el nino events on lower stratospheric ozone at mid-latitudes are well known (see introduction and changes in mean ozone aloft sections in Lin et al. [2015a] and references therein). Please consider revising the text as follows:

QBO (...) and ENSO (...) have been shown to influence total ozone burdens over the Tibet (Ji et al., 2001; Zou et al., 2001). This influence could extend to the lower troposphere via STE and thus affect ozone variability measured at the 3.8 km altitude of WLG. A few studies suggested that the change in dynamics after El Niño events can promote the cross-tropopause ozone exchange and lead to a rise in global mean tropospheric ozone concentration (e.g., Voulgarakis et al., 2011). Over western U.S. high-elevation regions prone to deep stratospheric intrusions, however, Lin et al. (2015b) found that the increased frequency of deep tropopause folds that form in upper-level

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frontal zones following strong La Niña winters exerts a stronger influence on springtime ozone levels at the surface than the El Niño-related increase in lower stratospheric ozone burden. The Tibetan Plateau has also been identified as a preferred region for deep stratospheric intrusions (Skerlak et al., 2014, ACP). To extent to which ENSO events, jet characteristics, and STE modulate interannual variability of lower tropospheric ozone at WLG requires further investigation.

14. P30992, Line 3-5: Need to clarify that the debates are on the causes of the ozone season cycle at WLG.

15. P31001, Line 15-20: again here discussions on interannual variability and the influence of QBO is vague (see Major comments and Comment 13 above).

16. P31003, Line 1-5: The daytime and nighttime trends during JJA have overlapping confidence limits (second column in Table 1); do you conduct statistical testing if they are significantly different at the 95% confidence level? If not, try to avoid using wording like “significantly distinct . . .”. To me, “significant” implies statistical results.

During JJA when boundary layer mixing peaks seasonally, daytime measurements at the 3.8 altitude of WLG are influenced by boundary layer air via an upslope flow. Thus daytime measurements at WLG during JJA are NOT representative of baseline conditions on a large scale, which could possibly explain the lack of significant daytime ozone trend at WLG during JJA. For the other seasons there is little difference between daytime and nighttime trends because boundary layer mixing is shallower compared to JJA and WLG is always located in the free troposphere. You can discuss these features without expanding to another paper.

17. P31003, Line 8-10: But the differences in spring and autumn trends at WLG are very small. I think you point is “the largest increase in ozone concentration was found in spring and autumn when seasonal mean ozone concentrations are lower than summer”?

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18. P31003 to P31004: Again, the description of the results from Lin et al. (2015b) is not quite accurate. Please make sure that you carefully read all papers cited in the manuscript and portray past literature accurately. Given limited time, the referee only checked a few papers.

19. P31004, Line 8-10, “From past literature we can discern that, both strong increasing and decreasing trends were mostly caused by the variation in ozone concentrations in the 1990s”.

This statement is not necessarily true for any region in the world. For instance, the largest ozone decreases over the eastern United States occur in the 2000s when U.S. NOx emission controls were implemented.

20. P31004, about Line 25-30: Please also add the description of c1 to c5 time scales in the caption of Figure 6.

21. P31006, about Line 5-8: But the highest ozone values are found in 2011-2012 (per the time series shown in Figure 5), not 2008 and 2013. I don't find the analysis shown in Figure 8 useful at all.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 30987, 2015.

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