Response to reviewer #2

Thank you for your comments and suggestions. We have addressed all the issues that were raised and have made changes accordingly. We have also improved the English language and made other necessary changes. Please see our point-by-point response below.

General comments

This manuscript presents a detailed analysis on the interannual variability and long-term trends of surface ozone at the Mt. Waliguan (WLG) station for the period of 1994-2013. A number of approaches including backward trajectory, chemical transport model simulations, tropospheric ozonesonde dataset, correlations with multiple climate modes, and multi-variable regression are applied to address this issue. The results identify the importance of stratosphere-troposphere exchange to the observed ozone increases at WLG in spring, and increasing influences of anthropogenic pollution from Southeast Asia in summer.

This study provides valuable information to better understand the long-term changes of surface ozone at a background station in western China. I also feel difficult to follow while reading the manuscript, and I understand the attempts to combine together all these different approaches and difficulty in assessing their inconsistency quantitatively.

I have a few comments listed below for helping authors to clarify the manuscript.

Specific comments

1) Page 5, Line 5:

It is not clear how you clustered the trajectory directions into 45-degree bins. It shall be helpful to plot and define these bins on a figure, such as on a panel of Figure 1.

RE: Thank you for the suggestion. Instead of trajectory start-point direction we have calculated average trajectory directions, which have been used for clustering. To clarify the calculation process, we added the following example to show how the average 24h and 168h directions have been calculated as a supplement figure. The 45-degree bins were depicted along with the example.

We have made some changes in the second paragraph of section 2.2: "To study the overall air-mass origin and to determine whether the air-mass collected pollutants from the nearby cities, the average direction of each trajectory relative to the WLG station is calculated both for the 168h and for the 24h trajectory (Figure S1). The 168h and 24h average directions relative to WLG are clustered into bins of 45° and the occurrence frequency in each bin is calculated."

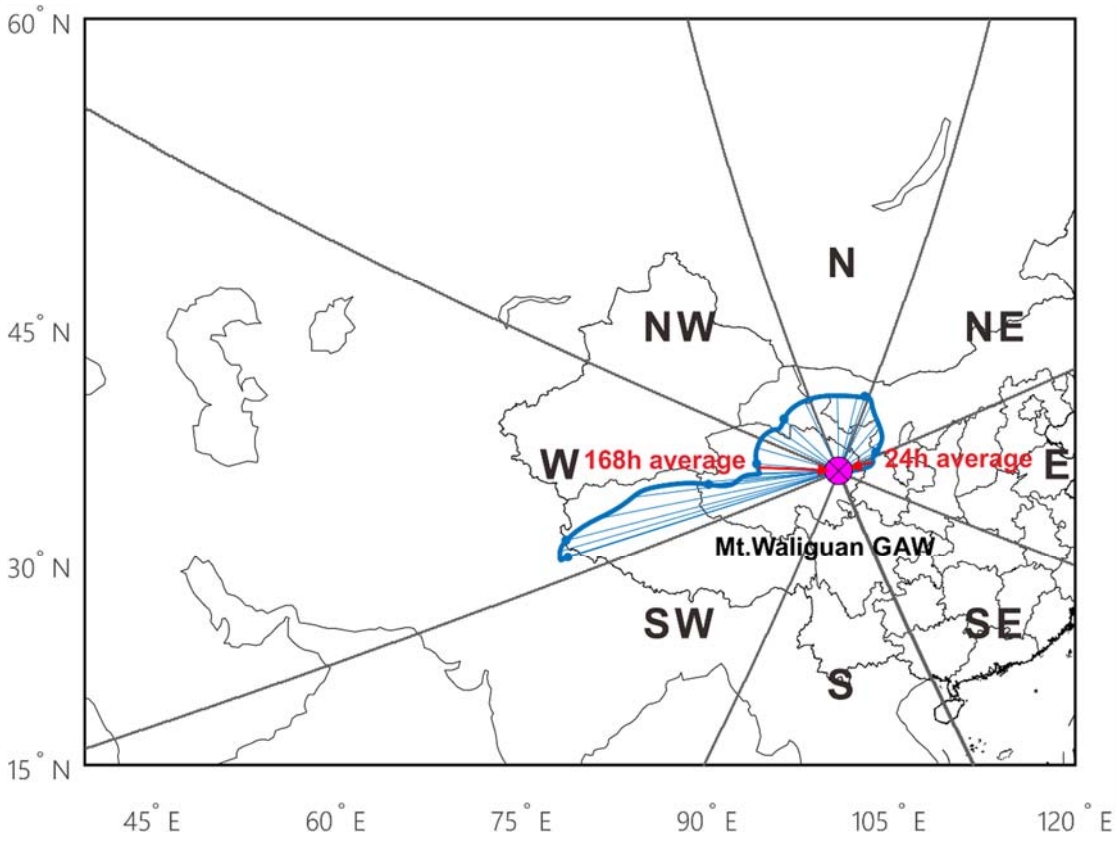

Figure 1 Schematic showing an example of the calculation process of 24h and 168h average trajectory directions and the 45-bins the trajectories were clustered into. The blue line shows a 7day trajectory example that bends from W to E, accounting for all the 168 hours, the average direction is westerly, while accounting only for the first 24 hours, the direction is easterly.

2) Page 8, Line 5:

For the statement "During summer, when air-masses from the east occur most frequently, the entire eastern sector reveals low PSCF", I suggest add "(as will be shown in Figure 2)" after "from the east occur most frequently", so that readers understand how you make the statement.

RE: Thank you for this suggestion. This sentence has been changed to "During summer, when air-masses from the east occur most frequently (as will be shown in Figure 2), the entire eastern sector reveals low values of high ozone PSCF, hardly showing signs of anthropogenic influence on WLG."

3) Page 8, Line 26:

Why do you state "the anthropogenic influence is negligible in all seasons except summer"? From Figure 1, we can also see high anthropogenic influences from Sichuan in spring and fall.

RE: Thank you for pointing out this. Page 8 lines 26-27 has been rephrased as:

"From the t=-168h average trajectory direction frequencies, it can be seen that the anthropogenic influence is strongest in summer, followed by autumn, and almost negligible in winter."

4) Page 11, Line 17-18:

You have argued above that the ozone trend in spring at WLG is driven by stratosphere troposphere-exchange. If so, shall we expect filtering for the East Asian anthropogenic influences, i.e., air masses with lower stratospheric influences, would show a lower trend? However, the results here show nearly no change in the springtime trend. Can you explain?

RE:We have explained this in the revised manuscript:

"We do not expect a decrease in springtime ozone when filtering the model for the East Asian anthropogenic influence, i.e., air masses with lower stratospheric influence, owing to the offsetting effects of increasing East Asian emissions."

5) Page 11, Line 20-30:

This section has showed that stratospheric influences explained two thirds of the ozone trend in spring. How about the rest one third? Would changes in anthropogenic emissions be the cause?

RE: Good suggestions. We have clarified in the revised manuscript:

"The stratospheric influence can explain two thirds of the total ozone increase at WLG in spring, with increases in Asian emissions contributing the rest one third"

6) Page 12, Sect. 3.3:

The TOST dataset are monthly averages from 1994 to 2012. Does that mean the dataset already account for ozone changes associated with increases in East Asian anthropogenic emissions? And then the direct tropospheric ozone transport as calculated in this section (Figure 8 and 9) has considered the tropospheric ozone changes associated with increases in precursor emissions. Please clarify.

RE: The TOST dataset is based on trajectory-mapped ozone soundings (Liu et al., 2013). The monthly averages of ozone in each grid should contain signals of background ozone and ozone produced with the grid from precursors emitted by anthropogenic and natural sources. Therefore, all the mean values in the TOST dataset already account for ozone changes associated with increases in East Asian (and other regions) anthropogenic emissions. One of the key issues in producing the TOST dataset was the impact of ozone production along the trajectories, which might cause errors in the mapped ozone data. A careful assessment indicates that the errors are mostly small and insignificant, as shown in Fig. 2 in Liu et al. (2013). Our approach of using TOST data is similar to the forward mapping in Liu et al. (2013), with the

difference that we focus on the impacts on ozone in the WLG grid from the surrounding grids. Therefore, it is likely that the impact of ozone production along the trajectories on our results is small, as in the case of Liu et al. (2013).

To clarify this we have added the following two paragraphs in section 3.3:

"Different from the GFDL-AM3 FIXEMIS simulation discussed in Section 3.2, the TOST approach discussed in this section does not eliminate the impacts from increases in Asian anthropogenic emissions. The TOST dataset is based on trajectory-mapped ozone soundings (Liu et al., 2013). The monthly averages of ozone in each grid should contain signals of background ozone and ozone produced within the grid from precursors emitted by anthropogenic and natural sources. Therefore, mean values in the TOST dataset account for not only ozone changes due to transport but also ozone changes associated with varying global-to-regional anthropogenic and natural emissions."

"One of the key issues in producing the TOST dataset was the impact of ozone production along the trajectories, which might cause errors in the mapped ozone data. A careful assessment indicates that the errors are mostly small and insignificant (Liu et al., 2013). Our approach of using TOST data is similar to the forward mapping in Liu et al. (2013). Therefore, it is likely that the impact of ozone production along the trajectories during their residence time on our results is small, as in the case of Liu et al. (2013).As the bottom layer of the grid in which WLG resides is excluded in our calculations, direct impacts on our results from regional emissions in the grid containing WLG can be ruled out."

Liu, G., Liu, J., Tarasick, D. W., Fioletov, V. E., Jin, J. J., Moeini, O., Liu, X., Sioris, C. E., and Osman, M.: A global tropospheric ozone climatology from trajectory-mapped ozone soundings, Atmospheric Chemistry and Physics, 13, 10659-10675, 10.5194/acp-13-10659-2013,2013.

7) Page 13, Line 16-22:

In this paragraph, CO measurements at WLG are used to analyze the influences of anthropogenic emissions. The results show statistically significant increasing trends only in summer. How about the trend in autumn? The previous section showed that the ozone trend in autumn was driven by anthropogenic pollution, but this did not seem to be supported by the CO analysis. Can you please clarify? As for the contribution from precursor emissions, can the model simulation with fixed anthropogenic emissions provide a better estimate?

RE: The CO data used in the manuscript are monthly CO data from weekly flask sampling (at 5m height) and analysis. A study by Zhang et al. (2011) indicates that the concentration of CO at WLG is subject to influences of regional-scale pollution, particularly in summer. Therefore, our summer CO measurements are less representative of large-scale conditions. In Section 3.2 we study the impacts from

anthropogenic emissions on a large-scale using GFDL-AM3 modeling. Our results are based on comparison of time-varying (BASE) and constant anthropogenic emissions (FIXEMIS).

8) Page 13, Line 23:

I feel confused about the discussion on ozone trends based on different trajectories in this paragraph. It reported the largest ozone trend associated with the SE direction, and the lowest trend with the NW direction. However, back on Page 9, Line 25-30,the trajectory analysis showed that the NW trajectories associated with high ozone concentrations had increasing occurrence frequencies, while the SE trajectory frequencies were decreasing. Are they consistent?

RE: These two results are not contradictory. The ozone concentrations associated with the NW trajectories are high in comparison to other sectors, but they show weak increasing trends, suggesting ozone coming from the NW have not changed much. The increase in NW trajectory frequency is what leads to the result that we experience these high ozone values more often, hence observe an increase in the ozone level. On the other side, ozone concentrations associated with the SE sector are not as high, but they show an increasing trend, highly possibly due to the change in precursor emissions in that sector.

9) Page 26, Table 5:

How do you estimate the ozone transported from East Asia, Europe, and North America? Please clarify.

RE: The GFDL model models CO-like-tracers from East Asia (EACOt), Europe (EUCOt) and North America (NACOt). For each month, we calculate the average ozone value associated with the upper 33 percentile EACOt, EUCOt, NACOt, to represent the ozone transported from East Asia ($O_{3,ea}$), Europe($O_{3,eu}$), and North America($O_{3,na}$). Then we use the East Asian Summer Monsoon Index (EASMI) to filter out those ($O_{3,ea}$,$O_{3,eu}$ and$O_{3,na}$) associated with the lower and upper 15 percentile of EASMI. The relative change induced by the East Asian Monsoon is then calculated with the equation: $(O_{3,\,EASMI}<=15^{th}\text{-}O_{3,EASMI}>=85^{th})/\bar{O}_{3}$.

We have clarified it in the revised manuscript by adding the above information to the header of Table 5.Thank you for pointing it out.

Some other comments

1) Page 3, Line 30:

“GOES-Chem” should be “GEOS-Chem”?

RE: Thank you, we have corrected this typo.

2) Page 8, Line 25:

“and least so in summer”. Need to remove “so”?

RE: Thank you for the correction, we have made according change in the revised manuscript.

3) Page 9, Line 19:

Here alpha is used to denote statistical significance, while in a few other places, such as Page 10, Line 12, ‘p’ is used. Please make them consistent.

RE: Thank you for the suggestion, we have made it consistent throughout the manuscript.

We have tried to address the issues raised by both referees and improved the English language. We have also made other changes where necessary. The title of this paper has been changed to “Long-term trends of surface ozone and its influencing factors at the Mt. Waliguan GAW station, China – Part 2: The roles of anthropogenic emissions and climate variability”.