Responses to Anonymous Referee #2

We thank the reviewer for the constructive suggestions and comments. We appreciate the reviewer's comments and these comments are very helpful for improving the manuscript. We understand that the comments are positive on the scientific content of the manuscript while appropriate revisions and clarifications are necessary.

We have addressed the reviewer's comments on a point to point basis as below for consideration.

General comments:

1) In section 2.2.2, how many percent of data are removed using the filter wind directions and SZAs?

Response: The wind direction and SZA filtering criterions removed 5.4% and 15.8% of all data, respectively. This information is now supplemented in the revised manuscript line 122 to 123.

2) The authors use the trace gas profiles and vertical profiles of pressure and temperature from WRF-Chem for AMF calculation. What is the spatial resolution and time resolution of the WRF-Chem profiles? Did the authors use a fixed trace gases profile for all the measurements or use the unique profile dependent on the measurement locations and time?

Response: In this study, the WRF-Chem model (version 3.7) was used to simulate the vertical profile of aerosol and trace gases as well as other meteorological parameters. The simulation domain covers large part of East China (17-49 °N, 95-124°E) with a horizontal resolution of 20 × 20 km and 26 pressure sigma level from ground level up to50 hPa. The time resolution of the model output is set to 1 h. Details of the configuration of the model can be found in (Liu et al., 2016;Su et al., 2017). Atmospheric profiles obtained from the model simulation were then interpolated in both spatial and temporal dimension to MAX-DOAS measurements location and time for AMF calculation. Details of the model simulation and AMF calculation are now included in the manuscript line 171 to 172.

3) The authors use the aerosol extinction coefficients measured by Mie lidar measurements for the AMF calculations. Did the authors use a fixed aerosol extinction profile (average the aerosol extinction coefficient from all the lidar measurements during the campaign) or use the specified

profile dependent on locations, time and the availability of lidar measurements?

Response: Lidar measurement was carried out together with MAX-DOAS most of the time during the campaign. AMF of the MAX-DOAS observations is calculated using hourly averaged Lidar aerosol extinction profile. This information is now supplemented in the manuscript line 172.

4) A fix set of single scattering albedo (SSA) of 0.95, asymmetry parameter of 0.68 and surface albedo of 0.06 is assumed in the radiative transfer calculations. Please explain why use this setting. Any references?

Response: The single scattering albedo (SSA) of 0.95 and asymmetry parameter of 0.68 are chosen according to the sensitivity study by (Chen et al., 2009). For oceans or rivers, the surface albedo is generally low and keep around 0.06 (Pinker et al., 1995). We have added the references in the revised manuscript line 188.

5) In section 2.2.4, there are too many formula and introduction of the determination of the tropospheric VCD. I suggest shortening the section and combining some of the formula.

Response: We followed the reviewer suggestion and shortened the introduction of the determination of the tropospheric VCD and combined some of the formula. Changes are listed in the following:

a) Line 149-150: we put the formula (SCDmeas = SCDtrop + SCDstrat) into the sentence

b) Line 151-152: sentence combination: “it can be assumed that the light path in the stratosphere for zenith and off zenith measurements are very similar, i.e. SCDstrat(α)≈SCDstrat(90°)”

c) Line 157-158: add description of VCDtrop: “and the tropospheric vertical column density (VCDtrop) can be expressed as follows”

d) Line 193-195: remove this paragraph about the description of tropospheric vertical column density (VCDtrop) in the ACPD version

e) Line 207: delete the “tropospheric vertical column density”

6) I agree with the comment from Referee #1 about the high NO_2 VCD on Nov. 29. Please explain the reason of high NO_2 and relative low SO_2 concentrations during this day.

Responses: The measurement ship was sailing around Wuhan on 29 November. Elevated tropospheric NO_2 observed around Wuhan is probably related to high emission, i.e., traffic emissions, in Wuhan as it is the largest city in Hubei. A rather low SO_2 level might due to lower SO_2 emissions located along the Yangtze River around Wuhan, e.g., coal-fired power plants.

7) In section 2.3, USTC's OMI tropospheric NO_2 product is used. I suggest showing more detailed information about the USTC OMI product, e.g., the data source of NO_2 slant density (SCD). In addition, USTC shall be explained when appeared for the first time.

Response: We agree with the reviewer suggestion and now included a more detailed description of the USTC's OMI NO_2 product in section 2.3. In this study, USTC's OMI tropospheric NO_2 product is developed based on OMI's primary product and has proven to be more suitable for the atmospheric conditions in China (Liu et al., 2016;Su et al., 2017;Xing et al., 2017). Slant column densities (SCDs) of NO_2 are retrieved by applying the DOAS fit to OMI spectra (data source: OMI Level 1B VIS Global Radiances Data product (OML1BRVG) (https://disc.gsfc.nasa.gov/Aura/data-holdings/OMI/oml1brvg_v003.shtml)). The information of USTC's OMI product are now supplemented in the revised manuscript line 234-237.

8) Figure 7d is a bit confusing, please describe clearly what the green lines represent? Is it 6, 12, 18 or 24 hour backward trajectories? What is the altitude of the backward trajectories?

Response: In Figure 7d, the green lines represent the 24 h backward trajectories during pollution events. The green lines show the backward trajectories calculated for each hour during the detected episodes. For example, five green lines on 1 Dec represent the backward trajectories ending at 04, 05, 06, 07, and 08 UTC on 1 Dec 2015 (corresponding to gray columns in Fig 4). The green markers indicate the location of air masses 6, 12, 18 and 24 h before arriving to the measurement ship. Considering atmospheric pollutants are mainly concentrated in low altitudes during heavy pollution episodes, the trajectory arrival heights were set to 500 m and assumed to be representative for the entire boundary layer. This information is now included in the manuscript line 300-302.

9) The authors mention that the different spatial patterns detected by MAX-DOAS and OMI on

Dec. 2 might be due to the strong influence by the aerosols. This is an interesting episode because it might show that the effect of neglecting aerosol in satellite AMF calculations on satellite NO_2 retrieval. Please prove this hypothesis using the Lidar measurement data.

Response: We agree with the reviewer suggestion and estimated the influence of ignoring aerosols in satellite AMF calculation on NO_2 retrieval. As lidar measurements only cover limited area, therefore, we have recalculated OMI NO_2 VCDs by taking aerosol information from WRF-Chem simulation into account. Figure R1a shows OMI NO_2 VCDs calculated without consider aerosol in the AMF calculation while Figure R1b shows the OMI NO_2 VCDs recalculated using aerosol information from WRF-Chem simulation. The spatial distributions of NO_2 are changed after including aerosol in the radiative transfer calculation. Significant enhancement of NO_2 VCDs can be observed over some areas. However, the spatial patterns of NO_2 detected by MAX-DOAS and OMI are still quite different. The result indicates the impact of aerosol could not fully explain the discrepancy between MAX-DOAS and OMI on this day. On the other hand, the MAX-DOAS and OMI observations agree better during OMI overpass time (black star symbols) and the agreement decay when the time differences between MAX-DOAS and OMI measurements become larger. This implies a strong temporal variability of NO_2 on this day. We have now supplemented the additional information and explanation in the manuscript line 352-360.

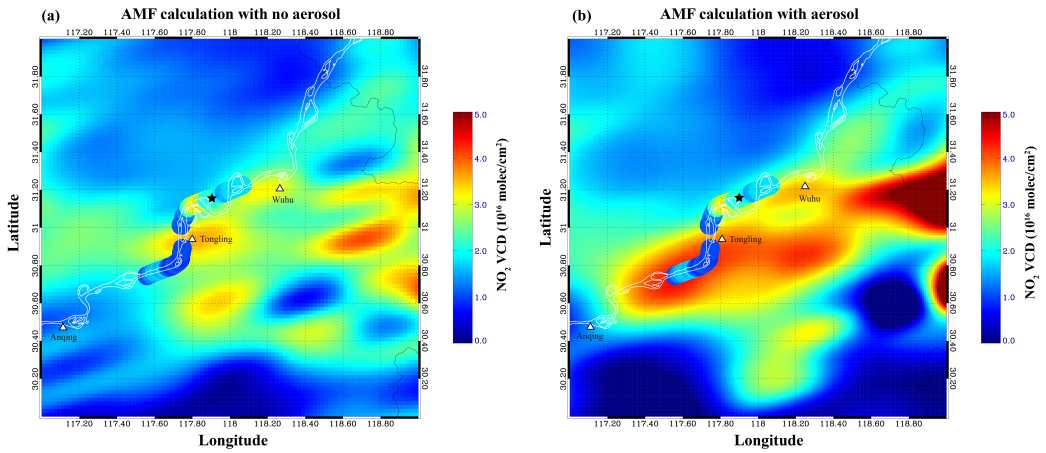

Figure R1. Spatial pattern of tropospheric NO_2 VCD measured by OMI calculated under two different AMFs.

10) In section 3.1, Line 307-308: “In contrast, lower HCHO VCDs were observed mainly on rainy,

cloudy and haze days". Could the authors explain more about the possible reasons for this phenomenon?

Response: Lower HCHO VCDs were observed mainly on rainy, cloudy and haze days, which might be due to stronger wet deposition and weaker solar irradiation during these days. The possible reasons for this phenomenon are supplemented in the revised manuscript line 316-318.

11) In section 3.4, please explain how to convert HCHO VCD to HCHO concentrations (ppb).

Response: For the estimation of primary and secondary sources of HCHO, the measured HCHO used in the regression model is ground mixing ratios. Usually, surface HCHO mixing ratios can be obtained from the HCHO vertical profiles (e.g., 0-200 m layer) (Wang et al., 2014). As the viewing elevation angles of the MAX-DOAS measurements only include 30° and 90°, therefore, there is not enough information to retrieve HCHO vertical profiles. In this study, we use a simplified formula introduced by (Lee et al., 2008) to convert mean HCHO DSCDs to mixing ratios (ppbv).

$$M(\text{ppb}v) = 1.25 \times \frac{DSCD(molecule\ cm^{-2})}{dAMF} \times \frac{1}{2.688 \times 10^{16}(molecule\ DU^{-1})} \times \frac{1}{\Delta P(atom)}$$

where M is the mixing ratio, DSCD is the difference between the SCDs of the measured spectrum and that of the Fraunhofer reference spectrum, dAMF is a differential air mass factor (dAMF=AMF(α=30°)-AMF(α=90°)), and ΔP is the pressure difference between surface and 500m height of boundary layer. The AMFs for this study were calculated using the radiative transfer model SCIATRAN 2.2 as described in Section 2.2.4. These information are now supplemented in the revised manuscript line 433-434.

12) In section 3.4, Line 430: "As other factors can also affect the atmospheric HCHO concentration". Please describe the "other factors" in more detail.

Response: In addition to primary emission and secondary formation of HCHO, meteorological conditions, e.g., solar irradiance, could also affect the atmospheric HCHO concentration. This information is now included in the revised manuscript line 451.

Technical corrections:

1) Line 321: ‘Dec 1 and 3’> ‘Dec 2 and 4’

Response: Corrected.

2) Line 344: ‘aerosol‘ > ‘aerosols‘

Response: Corrected.

3) Text on Figure 2: change “dSCD” to “DSCD”, and please uniform the units of DSCD (molec/cm^2).

Response: We have changed “dSCD” to “DSCD” on Figure 2 and uniform the units of DSCD (molec/cm^2) in the revised manuscript.

4) Explain the caption ‘RTM’ in Figure 3-5. Or remove it.

Response: We have removed the caption ‘RTM’ in Figures 3-5. We have merged Figs.3-5 so that the time series of three pollutants (NO_2, SO_2, and HCHO) in each panel with a continuous X-axis (Fig. 3&4 in the revised manuscript).

5) Text on Figure 6 is too small to read. Please use larger font size and put the colorbar on the right.

Response: We have enlarged the fonts in Figure 6 and put the colorbar on the right (Fig. 5 in the revised manuscript).

6) Improve the quality of Figure 10 (too many stripes).

Response: We have do our best to improve the quality of Figure 10, but too many stripes in Fig. 10 is inevitable which caused by the different satellite orbit.

References:

Chen, D., Zhou, B., Beirle, S., Chen, L. M., and Wagner, T.: Tropospheric NO_2 column densities deduced from zenith-sky DOAS measurements in Shanghai, China, and their application to satellite validation, Atmospheric Chemistry And Physics, 9, 3641-3662, 2009.

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Liu, H., Liu, C., Xie, Z., Li, Y., Huang, X., Wang, S., Xu, J., and Xie, P.: A paradox for air pollution controlling in China revealed by "APEC Blue" and "Parade Blue", Scientific reports, 6, doi:10.1038/srep34408, 2016.

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Su, W., Liu, C., Hu, Q., Fan, G., Xie, Z., Huang, X., Zhang, T., Chen, Z., Dong, Y., and Ji, X.: Characterization of ozone in the lower troposphere during the 2016 G20 conference in Hangzhou, Scientific reports, 7, 17368, 2017.

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