Responses to Anonymous Referee #1

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) The authors use lidar aerosol profile for AMF calculations to convert SCDs to VCDs for trace gases. However, only aerosol profile at the lowest 2 km is used and aerosol above 2 km are ignored. I think it is necessary to estimate the uncertainty due to ignoring aerosols above 2 km. In addition, the lidar has a blind height of 195m, so how the aerosols at the lowest 195m were treated?

Response: A sensitivity study has been conducted to address the influence from aerosol above 2km to the AMF calculation. Tropospheric NO_2, SO_2 and HCHO AMFs are calculated using lidar aerosol profile at the lowest 2km and WRF-Chem aerosol profile above 2km. The results are compared to AMFs calculated with only lidar aerosol profile at the lowest 2km. Comparison results show that AMFs calculated with considering aerosol above 2km are on average 2-4% lower than AMFs without considering aerosol at upper altitudes. The result indicates that ignoring aerosols above 2 km only cause a negligible error on the AMF calculation. In addition, aerosols at the lowest 195m are considered to be homogeneous in the AMF calculation. These information are now supplemented in the manuscript line 174 to 180.

2) As described in Sect 2.2.4, the authors has finally employed the new method for VCD estimation in the mobile measurements, which is recommended by Wagner et al., 2010. Maybe the authors could shorten the introduction of the geometric approximation and standard method for the VCD estimation. Alternatively, I suggest the authors to provide the comparison of the retrieved VCD results between standard and suggested methods? For example, taking one day as an example, to present the time series of the DSCD_mea, DSCD_offset and AMF_trop as used in the E.q. (12).

Response: We followed the reviewer suggestion and shortened the introduction of the geometric approximation and standard method for the VCD estimation. Changes are listed in the following:

a) Line 149-150: we put the formula ($SCD_{meas} = SCD_{trop} + SCD_{strat}$) 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. $SCD_{strat}(\alpha) \approx SCD_{strat}(90°)$"

c) Line 157-158: add description of VCD_{trop}: "and the tropospheric vertical column density (VCD_{trop}) can be expressed as follows"

d) Line 193-195: remove this paragraph in the ACPD version

In addition, we have compared the VCD retrieved with both standard and suggested method. Time series of the DSCD_meas, DSCD_offset, AMF_trop and VCD_trop on 28 Nov 2015 are shown in Figure R1. Fig. R1a shows the time series of NO_2 DSCDs for both elevation angles. Higher DSCDs are obtained from the lower elevation angle. The offset caused by the NO_2 absorption in the Fraunhofer reference spectrum and the stratospheric absorption (see Eq. 7 in the revised manuscript) are determined by a polynomial fit. $DSCD_{offset(t)}$ is then calculated following Eq. 10. The tropospheric VCDs of NO_2 calculated with both approaches (corresponding to Eq. 7 and Eq. 8 in the revised manuscript) are shown in Fig. R1d. Comparing with standard method, the temporal development of tropospheric NO_2 VCDs calculated by the new method is less noisy. Similar results can be found for SO_2 and HCHO in Figs. R2-3.

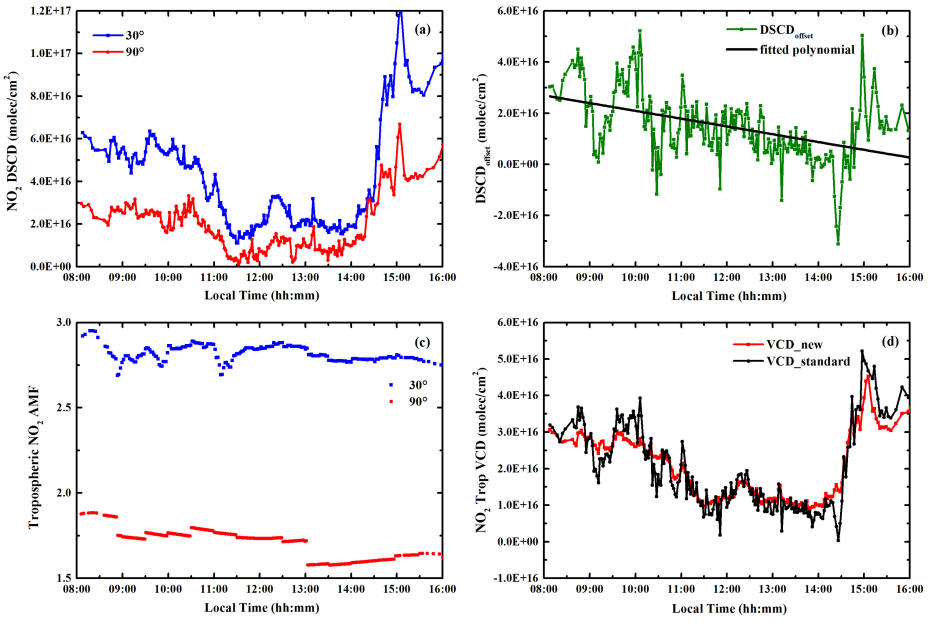

Figure R1. An example of determination of tropospheric NO_2 from the spectra measured on 28

November 2015. (a) NO_2 DSCDs, (b) $DSCD_{offset}$ (see Eq. 10) plotted as a function of time (green points), (c) Tropospheric NO_2 AMFs calculated by the radiative transfer model SCIATRAN, (d) The tropospheric VCDs of NO_2 calculated by new and standard method (corresponding to Eq. 7 and Eq. 8).

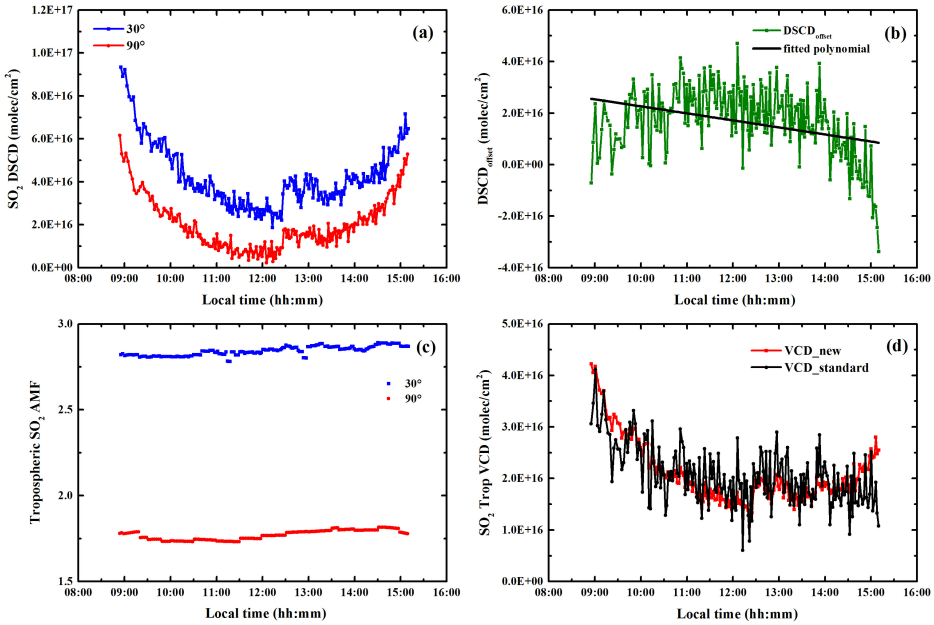

Figure R2. Same as Fig. R1, but for an example of determination of tropospheric SO_2 from the spectra measured on 27 November 2015.

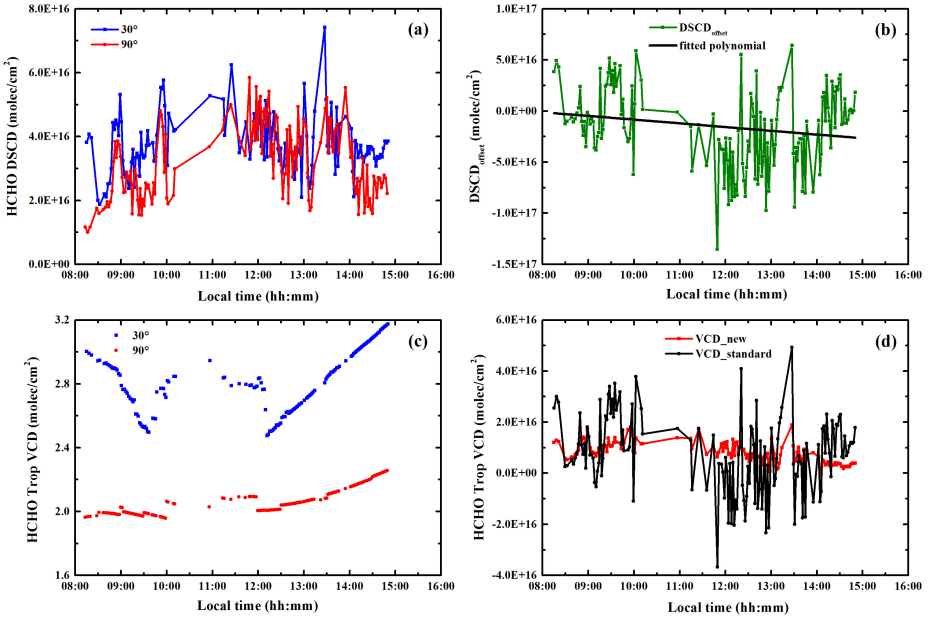

Figure R3. Same as Fig. R1, but for an example of determination of tropospheric HCHO from the spectra measured on 2 December 2015.

3) In this study, OMI VCDs are computed using atmospheric profiles from WRF-Chem simulations. It would be interesting to show how the VCDs are different from other operational products, e.g., NASA product.

Response: In this study, the USTC's OMI tropospheric NO_2 product is used. This product has been reported to be more suitable for atmospheric conditions in China (Liu et al., 2016;Xing et al., 2017;Su et al., 2017). Correlations of daily averaged tropospheric NO_2 VCDs measured by MAX-DOAS with USTC OMI and NASA OMI satellite data are shown in Fig. R4. Compared to the NASA's standard product, the USTC's OMI tropospheric NO_2 VCD agrees better with ground measurements a Pearson correlation coefficient (R) of 0.82 while the correlation between MAX-DOAS and the NASA OMI product is 0.76. The results suggested that accounting for the local atmospheric conditions and use the WRF-Chem model with measured climatology parameter and newest emission inventory to simulate trace gas profile in AMF calculation could improve the accuracy of OMI NO_2 VCD products. The Figure 7 is replaced by Figure R4 in the revised manuscript. These information are now supplemented in the manuscript line 332 to 336.

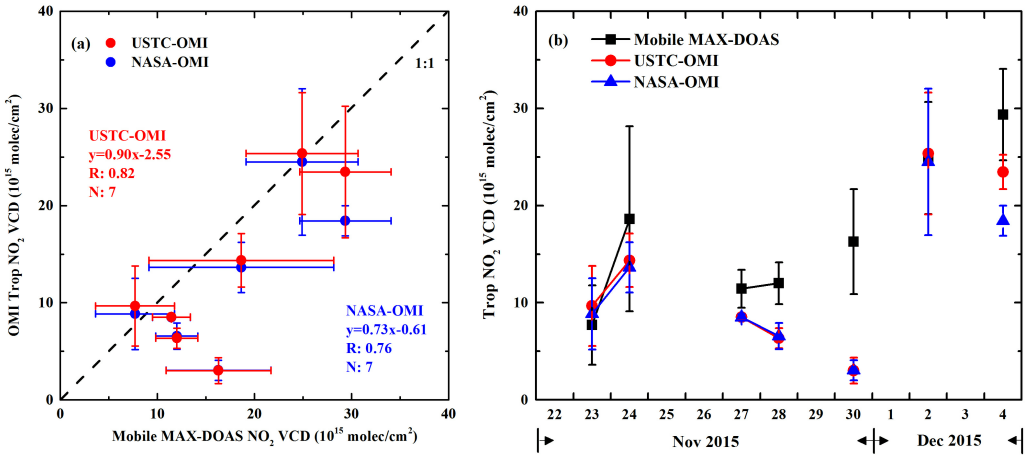

Figure R4. (a) Correlation analysis and (b) time series of tropospheric NO2 VCDs measured by ship-based MAX-DOAS and OMI during Yangtze River campaign. MAX-DOAS data (black markers) are temporally averaged around the USTC OMI and NASA OMI overpass time (red and blue markers, respectively), while the OMI data are spatially averaged within 20 km radius around the ship's averaged position. The error bars show the 1σ standard deviations of ship-based MAX-DOAS and OMI data.

4) It is not clear how the pollution events are identified. From Figure 3, the NO_2 VCD on 29 of

Nov is also very high, however, this day is not identified as pollution event. The authors should provide more information on the criterion in selecting pollution events.

Response: Pollution events were identified with both NO_2 and SO_2 VCDs reached or above the threshold value of 4.0×10^{16} molec/cm^2. Although the NO_2 VCD on 29 November is also very high, but the SO_2 VCD is relatively low. Thus, 29 November is not included in the pollution events analysis. The threshold value for identifying the pollution events is supplemented in the manuscript line 278.

5) In section 3.1, trajectories are calculated to assess the pollutant transportation. However, it is not clear that these backward trajectories are calculated at which altitude level. It would be much better to show the height of the backward trajectories as well, so that readers could better interpret how pollutants are transported.

Response: 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. In this study, 24 h back-trajectories were calculated by the HYSPLIT trajectory model. This information is now included in the manuscript line 302.

6) It's novel and interested to identify the industrial and vehicle contribution by E.q. 13 and 14, which provide the new insights for the sources appointment. As one of the most developed area in China, there were some more studies focusing on the emission inventories and source appointment for YRD areas. Maybe the authors can review previous results and compare with this study a little bit more in the discussion.

Responses: We have updated the section referring to a recent publication (Xia et al., 2016), which show a dramatic growth of the number of vehicle plays an increasingly significant role for regional NO_2 pollution over past years. For Hubei and Jiangxi provinces, the number of power plant is less than Jiangsu province. Therefore, the contribution from vehicle emission to ambient NO_2 level is expected to be more pronounced with the dramatic growth of vehicle number. Changes are applied to line 415 to 418.

7) For the estimation of primary and secondary sources of HCHO, I have two questions: 1) which

kind of the measured HCHO concentrations was used in the regression model? Since the HCHO levels were determined by MAX-DOAS as VCDs, however, the ambient HCHO concentrations were used in the model. How the authors obtain the ground surface concentration HCHO from the VCDs? Otherwise, why the authors can make regression of the HCHO VCDs with in-situ CO, Ox?

2) For the diurnal pattern, the authors inferred that secondary formation of HCHO shows a peak value during noon time (11:00-14:00), however, there was also another peak of relative contribution of secondary sources around 10:00 LT. How to consider this phenomenon?

Response: (1) 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 500 m 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.

(2) We agree with the reviewer that there is another peak of relative contribution of secondary sources at around 10:00 LT. The 24 h backward trajectories on 3 December (Fig. 6d in the revised manuscript) suggested that rapid transport of air masses carries significant amount of pollutants including formaldehyde precursor from polluted area in northern China. Thus, the peak of relative contribution of secondary sources around 10:00 LT is probably resulted from the transportation of formaldehyde precursor. The explanation of the diurnal pattern has been modified in the revised manuscript (Line 474-479 in the revised manuscript).

Technical corrections:

1) Introduce abbreviation on the first time used in the manuscript, e.g., line 169 WRFChem.

Response: Abbreviation (WRF-Chem) is now introduced at the first time used in the manuscript (Line 170).

2) Text on Figure 3-6 is too small to read. Considering the similarities of Fig.3 to Fig.5, I suggest to merge them together to show the time series of these three pollutants in each panel with a continuous X-axis.

Response: We have merged Fig.3-5 so that the time series of three pollutants (NO_2, SO_2, and HCHO) in each panel with a continuous X-axis. Considering that a single plot covering all data is a bit difficult to read, so we have now separated it into 2 figures (Fig. 3&4 in the revised manuscript) for two periods (from Nov 22 to Nov 28 and from Nov 29 to Dec 4). In addition, we have enlarged the fonts in figure 6.

3) Put error bars on Figure 13 and 14.

Response: We have now included error bars in Figure 14 (Fig. 13 in the revised manuscript).

4) Please uniform the units of VCDs. It's different in Figures (molec/cm2) and manuscript text (molec cm-2) now.

Response: We have uniformed the units of VCDs (molec/cm^2) both in Figures and manuscript text.

References:

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