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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ground-based MAX-DOAS observations of tropospheric formaldehyde and comparisons with CAMS model at a rural site near Beijing" *by* Xin Tian et al.

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Dear editors and reviewers, Thank you very much for your constructive comments and advices on our manuscript. Your positive evaluation and comment encourage us and would be great helpful to our research. We have carefully considered every comment, and made corresponding revisions in the revised manuscript and marked every change in red.

Point to point response is following:

General Comments: 1ãĂĄHowever, a problem that detracts from the entire paper is

that the language is not fluent and precise. There are frequent spelling and grammatical errors (fragment sentences, unnecessary words, incorrect verb tense, missing articles, convoluted or run-on sentences). The authors are strongly suggested to engage the help of an English editor. Response: Thank you very much for your suggestions. We have considered your advice, and asked for help from an English language service.

2ãĂĄA few sections would benefit from re-structuring for increased logical flow and clarity. The scientific methods and assumptions are not sufficiently clearly outlined. The methodology section is disorganized and needs additional technical details. The paper does present some novel data and reaches substantial conclusions, but the results are sometimes not enough to support the interpretations and conclusions without further statistical analysis and/or expanded discussion. When discussing results, the authors must consider whether trends and differences in measured values they are interpreting are statistically significant given calculated or expected uncertainties. The authors must also try to place their conclusions within the context of previous literature (e.g., presented in the introduction). Response: Thank you for your advice. The error budgets are added in sec. 2.3. The section 2 has been reorganized. The introduction has also been rewritten.

Specific Comments: 1ãĂĄFor all regressions, the coefficient of determination (R2) statistic may be more appropriate since this value indicates the variance in the dependent variable that is predictable from the independent variable. Response: Thank you very much for your suggestion. We have considered your advice, and all coefficient of determination R are changed to R2 in this paper, including in the figures.

2ãĂĄPage 2 Line 3 What specific results had a good correlation coefficient? VCDs of HCHO? Other? Response: Thank you for pointing out. It's the VCDs of HCHO. It is changed to make it clear. Changes in manuscript: The HCHO VCDs of the CAMS model and MAX-DOAS were generally consistent with a correlation coefficient R2 greater than 0.69.

3ãĂĄPage 2 Line 23 Did the emissions decrease from 100% to these values or are these values the quantity of the decrease? It is not clear Response: Thank you for your advice. These values indicate the proportion of emissions from the corresponding source. It is changed to make it clear. Changes in manuscript: The analytical results of the Chemical Mass Balance (CMB) model showed that the contributions of coal-fired boilers, dust, and motor vehicles to PM2.5 in Beijing were around 2%, 7%, and 30%, respectively, during the APEC summit (Cheng et al., 2016).

4ãĂĄPage 2 In general, since some of the pollutants were measured by multiple papers that you cite, consider sorting this paragraph by pollutant rather than by author. Otherwise, it becomes repetitive and confusing to have to keep referring to the values from the previous papers earlier in the paragraph. Response: Thank you for your advice. We have considered your advice, and it is changed in the paper. Changes in manuscript: Presently, many studies have analyzed the effects of emission reduction measures during the APEC summit. Ground-based observations were taken to investigate the air quality changes associated with a series of stringent emission-reduction measures (Fan et al., 2016; Li et al., 2016; Liu et al., 2016; Tang et al., 2015; Chen et al., 2015; Wang et al., 2016a; Wang et al., 2016b; Wang et al., 2017a). Wang et al (2016a) selected five representative in situ stations in different locations in Beijing and found that average concentrations of SO2, NO2, PM10, and PM2.5 decreased by 61.5%, 40.8%, 36.4%, and 47.1%, respectively, whereas the average concentration of O3 increased by 101.8%, compared with the same period over the last five years (PM2.5 since 2013). O3 in urban and suburban areas of Beijing is mostly in the control area of volatile organic carbons (VOCs). The possible reason for the increase in O3 is that the emission control measures of NOx are greater than the emission control measures of VOCs, which leads to the weakening of the inhibition of O3 formation by NOx, resulting in significant increases in O3 concentration. Although the traffic and urban stations produce a lot of pollution due to motor vehicle emissions, the NO2 concentrations of suburban and regional stations significantly dropped compared with the traffic and urban stations as a result of the control measures. The NO2 emitted by mo-

tor vehicles in the Beijing urban area remained high even under the measures taken to limit the number of vehicles (Wang et al., 2016a). Space observations were also used to evaluate the effect of emission control measures on the changes in NO2 tropospheric vertical column densities (VCD) and aerosol optical depth (AOD) in Beijing and surrounds based on Ozone Monitoring Instrument (OMI) and Moderate Resolution Imaging Spectroradiometer (MODIS) retrieval. The results showed that NO2 VCD and AOD were mostly reduced by 47% and 34% in Beijing, respectively (Huang et al., 2015; Wei et al., 2016; Meng et al., 2015). The analytical results of the Chemical Mass Balance (CMB) model showed that the contributions of coal-fired boilers, dust, and motor vehicles to PM2.5 in Beijing were around 2%, 7%, and 30%, respectively, during the APEC summit (Cheng et al., 2016). Zhang et al (2017) analyzed the characteristics of aerosol size distribution and the vertical backscattering coefficient profile during the 2014 APEC summit using lidar observation. Particles with larger sizes were better controlled during the APEC period, with the number concentration of accumulation mode and coarse mode particles experiencing more significant decreases of 47% and 68% (Zhang et al., 2017). Published studies have focused mainly on the effects of commonly measured gas pollutants, particulate matter, and aerosols, but not HCHO (Cheng et al., 2016; Fan et al., 2016; Huang et al., 2015; Li et al., 2016; Liu et al., 2016; Meng et al., 2015; Tang et al., 2015; Chen et al., 2015; Wang et al., 2016a; Wang et al., 2016b; Wang et al., 2017a; Wei et al., 2016).

5ãĂĄPage 3 Line 6 Three sources are listed despite elsewhere in the paper it is stated that there are two sources, which one is it? Response: Thank you very much for your reminding. There are mainly two sources for troposphere formaldehyde, besides, only a small fraction of HCHO is from direct emissions of biogenic sources (e.g., vegetation). Changes in manuscript: Troposphere formaldehyde mainly originates from two sources.

6ãĂĄPage 3 This discussion may benefit from writing out some of the most important chemical equations for the reader equations for the reader. Response: Thank

you for your suggestion. Accordingly, equations are added in the paper. Changes in manuscript: All of the photolysis equations of HCHO to form OH radical at wavelengths below 370 nm are listed as follows: HCHO + hν→H+ HCO(λ≤370nm)→H2 +CO (1) H +O2→HO2 (2) HCO + O2→HO2 + CO (3) HO2 +NO→OH + NO2 (4)

7ãĂĄPage 3 Line 16 It is unclear whether the importance of quantifying HCHO is to track emissions of VOCs or NVOCs or the generation of OH, or all of these. Response: Thank you for your suggestion. It represents all of these. And it is added to make it clear. Changes in manuscript: Therefore, HCHO can reflect anthropogenic VOC emissions and VOC emissions through the fast production of short-lived NMVOCs. Identifying the major sources of HCHO is essential for quantifying the photolysis sources of OH and their contributions to aerosol formation and for effectively controlling photochemical pollution (Bauwens, et al., 2016; Chang, et al., 2016; Ling, et al., 2017; Ma, et al., 2016; Tanaka, et al., 2016).

8ãĂĄPage 4 Line 5 What unit of HCHO? VCDs? Response: Thank you for pointing it out. It is VCDs. And it is added in the paper. Changes in manuscript: In this study, we used the ground-based MAX-DOAS instrument installed in Huairou District (suburban area) of Beijing to evaluate the effects of the sources and depositions of HCHO VCDs and their relations with emission control measures and meteorological conditions during the period from October 26, 2014 to November 20, 2014.

9ãĂĄPage 4 Line 10 The last paragraph of your introduction may benefit from explicitly stating your research objectives (perhaps as a list). What were all the components included in the spectral fitting for HCHO? Include what cross section reference spectra used (including author), fitting window etc. Response 1: Thank you very much for your advice. It is changed in the paper. Changes in manuscript: In this study, we used the ground-based MAX-DOAS instrument installed in Huairou District (suburban area) of Beijing to evaluate the effects of the sources and depositions of HCHO VCDs and their relations with emission control measures and meteorological conditions during the period from October 26, 2014 to November 20, 2014. Two pollution episodes and

their relationships with meteorological conditions were analyzed during APEC to evaluate the effects of regional transport and local emissions. Afterwards, three episodes, defined as “pre-APEC,” the period of APEC and “post-APEC,” were used to evaluate the influences of emission control measures on the changes in HCHO VCD during APEC. The correlations between HCHO VCDs with NO2 VCDs and O3 were used to determine the main HCHO sources and evaluate the dominant error sources of HONO simulations of Copernicus Atmosphere Monitoring Service (CAMS) model.

Response 2: Thank you very much for your suggestion. The spectral fitting for HCHO including cross section, reference spectra and fitting window is introduced in detail in section 2.1 (MAX-DOAS Methodology). And the information should belong to technique detail, so I think it is more appropriate to be written in section 2 than in the introduction. Changes in manuscript: Fig. 2 shows a structural representation of the MAX-DOAS system. This system comprises a telescope, stepper motor, spectrometer, and computer. Sunlight is focused by the telescope, which is installed outdoors and reaches the spectrometer through an optical fiber. The spectrometer was placed in a temperature-controlled box at 20°C to ensure that the spectrograph could work at a stable temperature under the changing ambient temperature from -15°C to 30°C in China. The spectrometer was produced by Ocean Optics and was named Maya (https://oceanoptics.com/product/maya2000-pro-custom/). The spectrometer covers the range of 290 nm to 420 nm, and its instrumental function is approximated as a Gaussian function with a full width at half maximum (FWHM) of 0.5 nm. MAX-DOAS was routinely operated for 24 h. Due to the intensity of the sunlight, only the daytime measurements were used for analysis. The nighttime measurements could be used to correct the dark current and offset. The azimuth angle view of the telescope was fixed at 0° (North) during the entire observation period. A full MAX-DOAS scan comprises six elevation angles (EA) (3, 5, 10, 15, 30, and 90°) and lasts for approximately 10 min (see Fig. 3). Each measurement had an average of 100 SCANS, and the integration time was adjusted automatically based on the light intensity. Table 1 lists the detailed setup of the MAX-DOAS instrument.

10ãĂĄSection 2.1 Please list the final equation used for calculating VCDs from fitted DSCDs given your geometric approximation. Response: Thank you for your advice. It is added in the paper. Changes in manuscript: Then the tropospheric VCD can be obtained from the following equation:

11ãĂĄReorganize section 2 so that sections 2.1, 2.3, and 2.4 are grouped together for a more logical sequence flow. What is included in the VCD error calculation? Response: Thank you for your advice. It is changed in the paper. The original sections 2.2 “Monitoring locations” and 2.3 “MAX-DOAS instrument and measurement” are grouped together as the new section 2.1 “Monitoring locations and instrument”. The original sections 2.1 “MAX-DOAS Methodology” and 2.4 “DOAS analysis” are grouped together as the new section 2.2 “DOAS Spectral retrieval and determination of tropospshericVCD”. The error budgets is added in the text, see Sec. 2.3 “Error budgets”.

12ãĂĄAre any of the MAX-DOAS VCDs removed from the dataset due to cloud fraction? If so, what was the cut off cloud fraction value? Response: Thank you for your question. In this paper, the effects of different cloud coefficients on MAX-DOAS inversion VCDs and the HCHO VCDs from MAX-DOAS and CAMS model under different cloud coefficients are both compared. It is found that the cloud coefficient has negligible influence on it. During the entire APEC period, it is basically sunny and cloudless weather. It is added in the paper to make it clear. The data is pre-screened. The spectrum with too small a light intensity and an excessive integration time are removed. The spectrum pre-screen is added in the section 2.3. Changes in manuscript: 1) The effects of different cloud coefficients on MAX-DOAS inversion VCDs and the HCHO VCDs from MAX-DOAS and the CAMS model under different cloud coefficients were compared. The results show that cloud coefficient had a negligible influence on the retrieval of HCHO VCDs by MAX-DOAS. Additionally, sunny and cloudless weather generally occurred during the entire APEC period. Thus, all of the data obtained in the different cloud coefficients were used.

2) We excluded data for solar zenith angle (SZAs) larger than 75° because of the

stronger absorptions of stratospheric species and a low signal-to-noise ratio. Data with a large root mean square (RMS) of the residuals and large relative intensity offset were also excluded.

13ãĂĄPage 5 Line 8 It is unclear what causes lower systematic errors. Response: Thank you for your question. This sentence is not very clear so that caused some misunderstandings for you. We make changes in the paper to make it clearer. 1) We mean that: According to Ma et al., 2013 and Wang et al., 2017c, they found the systematic error is larger for larger elevation angles and larger RAA. So this study uses the geometric approximation method to determine HCHO VCDs at an elevation angle of 15° to avoid surface obstacles on light paths along the line of sight, at the same time, it has lower systematic errors at 15° than 30°. 2) And we add the discussion of the error budgets for geometric approximation in section 2.3. It also find the systematic errors at 15° is smaller than 30°by using the geometrical approximation. Changes in manuscript: Lower systematic errors were achieved at 15° than at 30° by using the geometrical approximation (discussed in Section 2.3 below).

14ãĂĄPage 6 Line 18 Why was the FRS from this day chosen to fit all retrievals? Since you are fitting all your measured spectra against one FRS, you must consider the effect of SCD(FRS) (the component of trace gas in the FRS used) and the SCD(Solar Zenith Angle), which is the difference in the stratospheric component of SCD observed due to the difference in SZA between the times of measurement and the FRS time. SCD(SZA) changes with time and change the apparent diurnal trends. Please justify why you did not account for the SCD(SZA) and SCD(FRS). For example, was your FRS was obtained during a very low pollution period and/or are the stratospheric HCHO levels are expected to be trivial. For more information see Wagner, T., Ibrahim, O., Shaiganfar, R. and Platt, U.: Mobile MAX-DOAS observations of tropospheric trace gases, ATMOSPHERIC Meas. Tech., 3(1), 129–140, 2010. Response: Thank you very much for your advice. Maybe my expression is not clear in the text, which makes you misunderstood. First, we use a FRS to retrieve all the spectra and obtain the dSCD. Then

I subtract the dSCD at 90° from the dSCD at off-zenith angles in the same elevation sequence to derive the delta SCD. The procedure can deduct the influence of stratospheric absorption and variation of instrumental properties. We add the description in the paper to make it clear. Changes in manuscript: The geometric approximation was used to convert the dSCD to the tropospheric VCD. In the first step, the differential slant column densities (dSCDs) were derived from the DOAS spectral analysis with a so-called FRS and measured in a small sun zenith angle at 90° elevation around noon (Hermans et al., 2003; Hönninger and Platt, 2002; Kraus, 2006). The SCD includes two parts of the absorption signal of the troposphere and stratosphere. To remove the interference of stratosphere absorption and variation in instrumental properties, dSCD at off-zenith elevation angles were subtracted by the dSCD at 90° elevation angle in the same elevation sequence to derive $\triangle$SCD following the equation below:

15ãĂĄPage 6 Line 23 What is the software reported SCD error in molec/cm2? Response: Thank you for your reminding. The SCD error is 8.14 $\times$ 1015 molec/cm2. Changes in manuscript: HCHO SCD was 7.21 $\times$ 1016 molecules cm-2 with an error of 8.14 $\times$ 1015 molecules cm-2.

16ãĂĄPage 6 Line 24 Are you missing units on this number? Response: Thank you for the asking. The root mean square of the residual error here is the residual spectral structures of optical depth, and there isn't unit for the optical depth. Changes in manuscript: The root mean square of the optical depth of the residual spectral structures was $1.08\times10-3$.

17ãĂĄPage 7 section 2.6 At what height were these meteorological parameters measured? At what time frequency before averaging? Response: The height of these meteorological parameters measured is about 15m. The weather station is about ten meters away from MAX-DOAS. All measured meteorological parameters are recorded of 1 min time intervals. Changes in manuscript: The MAX-DOAS instrument was deployed on the balcony (without a roof) of a classroom on the 4th floor in the laboratory building in the campus of UCAS (116.67°E, 40.4°N). The UCAS supersite is on the top

floor of the laboratory building, which is about 10 m away from the MAX-DOAS instrument. Meteorological parameters, including wind speed (WS), wind direction (WD), temperature (T), and relative humidity (RH), were continuously measured by a MetPak automatic weather station (Gill Instruments Ltd, Lymington, UK) at the UCAS superstation from October 28, 2014 to December 31, 2014 (Fig. 4a). All of the measured meteorological parameters were recorded at 1-min time intervals.

18ãĂĄPage 7 Line 24 What does a "static" weather situation mean? Response: The static weather means the wind speed less than 3.5 m s-1. Halfacre et al., 2014 defines that the wind speeds (mode of $\sim$3.5ms-1) is under relatively calm conditions (Halfacre, J.W., Knepp, T.N., Stephens, C.R., Pratt, K.A., Shepson, P., Simpson, W.R., et al.: Temporal and spatial characteristics of ozone depletion events, Atmos. Chem. Phys., 14: 4875–4894, doi:10.5194/acp-14-4875-2014, 2014.). It is defined in the paper to make it clear. Changes in manuscript: Halfacre et al (2014) defines the relatively calm conditions with wind speeds of less than 3.5 m s-1 as the static weather situation. The static weather situation frequently occurred during the observation with a wind speed of less than 3.5 m s-1, and wind speeds of more than 3.5 m s-1 usually appeared under northwest and west winds.

19ãĂĄBetter organization and flow in section2 may be achieved by describing MAX-DOAS methodology in this order: general description of the MAX-DOAS instrument, description DSCDs fitting, determination of VCDs from DSCDs, measurement sequence, and then viewing azimuth and location. Response: Thank you for your advice. It is changed in the paper. The original sections 2.2 Monitoring locations and 2.3 MAX-DOAS instrument and measurement are grouped together as the new section 2.1 Monitoring locations and instrument. The original sections 2.1 MAX-DOAS Methodology and 2.4 DOAS analysis are grouped together as the new section2.2 DOAS Spectral retrieval and determination of the tropospheric VCD. Changes in manuscript: See section 2.

20ãĂĄFigure 6 . manuscript: oposphericVCDtion2.2 hen viewing azimuth and loca-

tion.ion of Phys Response: All the days measured are quality controlled. The quality control criteria are added in the paper. Changes in manuscript: We excluded data for solar zenith angle (SZAs) larger than 75° because of the stronger absorptions of stratospheric species and a low signal-to-noise ratio. Data with a large root mean square (RMS) of the residuals and large relative intensity offset were also excluded.

21ãĂĄPage 8 Line 13 Please quantify in some way the relative change in solar radiation and temperature compared to the days were peaks were not apparent. Response: Thank you for your advice. It is added in the paper. Changes in manuscript: The daily averaged intensity of solar radiation and temperature on November 4 and 7 were compared with data from the two periods of November 4 to 7 and October 1 to December 31, 2014 (Fig. 8). The differences in averaged solar radiation and temperature on November 4 and 7 compared to the period from October 1 to December 31, 2014, were 2.22%, 2.47%, 34.4%, and -23.5%, respectively.

Figure 8: Averaged intensity of solar radiation and temperature in the four periods of November 4, November 7, November 3 to 8, and October 1 to December 31, 2014. Error bars denote the standard deviations.

22ãĂĄPage 8 Line 21 Please define (or find a better descriptor for) “good dispersion conditions”. Response: Thank you for your advice. It is defined in the paper. Changes in manuscript: The value on November 6, 2014 was probably caused by the good dispersion conditions under the northwest winds with speeds of more than 3.5 m s-1, with the air mass mainly originating from the clean northwest area.

23ãĂĄPage 9 Line 14 What type of relevant pollution sources do these cities have? Primary and/orsecondary? Are there many industry and/or vehicular sources? Response: Thank you for your question. The type of relevant pollution source Tangshan, Baoding, Shijiazhuang and Tianjin is primary source. And there are a lot of industry in those cities. For Beijing, the main source is the vehicular source, especially in the urban area (Lin et al., 2009; Lin et al., 2012; Shao et al., 2006; Tang et al., 2015; Wei

et al., 2016). The reference is added in the paper (sec. 2.1). Changes in manuscript: Relevant pollution sources in Tangshan, Baoding, Shijiazhuang, and Tianjin is primary pollution sources. In Beijing, vehicles are the predominant pollution source, especially in the urban areas (Lin et al., 2009; Lin et al., 2012; Shao et al., 2006; Tang et al., 2015; Wei et al., 2016).

24ãĂĄPage 9 Line 16 You may want to state explicitly that there are fewer, smaller or less polluted cities in the Northern region here. Are the lower VCDs due to just dispersion or is it also chemical aging, et Response: Thank you very much for your advice. There are less polluted cities in the Northern region. The northern is clean with low VOC emissions, so there are few precursors of HCHO transported to the measurement station. The lower HCHO VCDs under such conditions are mainly due to less VOC precursors of HCHO. Changes in manuscript: The northern cities are clean with low VOC emissions, and thus few precursors of HCHO were transported to the measurement station in the north wind. The lower HCHO VCDs under such conditions are mainly due to fewer VOC precursors of HCHO. In summary, the wind from this area prominently contributes to the dispersion of the pollutants.

25ãĂĄPage 9 Line 17 and 18 Please explain the dependence of the VCDs on wind speed under different wind directions. Response: Thank you very much for your suggestion. It is added in the paper. Changes in manuscript: In terms of the dependence of HCHO on wind speed, the HCHO VCDs decrease along with the increasing wind speed under the northerly fast and clean wind, which results in the rapid dissipation of the pollution. Under the southerly wind, the HCHO VCDs increase with increasing wind speed. Thus, transport from the south polluted air to the observation site occurs more easily under southerly winds with relatively high wind speeds.

26ãĂĄPage 10 Line 6 What are the errors on each of the VCD values. Are they statistically different? Response: Thank you very much for your advice. According to the advice of review 2, we think it is more reasonable to use the standard deviation to represent the error bars. It is changed in the paper. And the figure 9 is changed to figure

11 due to some new figures were added. And we add the standard deviation on each of VCD values. It is added in the paper. Changes in manuscript: The average HCHO VCDs were 9.65×1015, 5.99×1015, and 8.65×1015 molec cm-2 before, during, and after APEC, with fitting errors of 9.39%, 10.12%, and 9.74%, respectively. 27ãĂĄ-Page 10 Line 17. This sentence is too vaguely written. Also, depending on whether the differences between the peak values are statistically significant, depending on the expected errors and the significance of the wind direction change, you may not have sufficient evidence to support this conclusion. Also consider that during APEC time the conditions were not only northerly winds but also higher wind speeds, which you state earlier in the paper tends to reduce the VCDs (which should be explained for clarity). Same comment for the sentence on lines 19 and 20. Response: Thank you very much for your advice. According to the question 43, we plot the VCD data against the wind speed and direction for the pre-APEC, during-APEC, and post-APEC periods, seperately. And we re-organized our discussion in the section 3.2. Please see the modified paragraph in the following . Changes in manuscript: As the measurement station is located in the northern suburban area of Beijing, the effects of the control measurements, which were mainly implemented in the urban areas, on HCHO were only observed at the station when dominant southerly winds occurred. We thus plotted the dependence of HCHO VCDs on the wind speed and directions in Fig. 12d–f for the pre-APEC, APEC, and post-APEC periods. Fig. 12d–f indicate that the averaged HCHO VCDs under south winds during APEC were about 6.46 × 1015 molec cm-2, which was considerably lower than 10.29, 6.46, and 9.20 × 1015 molec cm-2 in the pre-APEC and post-APEC periods. In addition the peak values due to transport from the south urban area on November 4, 2014 and November 7, 2014 during APEC shown in Fig. 11 were 25.75% and 18.3% lower than the peak values under similar wind fields in the pre-APEC and post-APEC periods. In general, the HCHO values under the dominant southerly wind field were considerably lower during APEC than the pre-APEC and post-APEC periods. The phenomenon implies that the control measures had a certain effect on reducing the concentration of HCHO. This suggests that the implementation

of control measures during the APEC summit reduced the concentrations of NO2 and aerosols (Liu et al., 2016; Zhang et al., 2017).

Figure 12: Wind roses in (a) the “pre-APEC”, (b) the APEC, and (c) the “post-APEC” periods. Dependence of HCHO VCDs (1015 molec cm-2) on wind directions for different wind speeds in the pre-APEC (d), during the APEC (e), and post-APEC (f) periods.

28ãĂĄPage 10 Line 25 Some basic equations on HCHO chemistry in the introduction section would be very helpful for the reader by the time they get to this point in the paper. Response: Thank you very much for your suggestion. The equations are listed in the introduction, as the response to question 6. Changes in manuscript: Atmospheric photochemical reactions are related to the intensity of solar radiation as indicated in equation 1.

29ãĂĄPage 11 Line 3 Are you suggesting that this peak in the diurnal variation is due primarily to secondary production of formaldehyde rather than direct emissions? Since the most light is available mid-afternoon and local direct emissions are relatively smaller compared to secondary production? Please make this clearer to the reader. Response: Thank you for your question. Yes, we conclude this peak in the diurnal variation is due primarily to secondary production of formaldehyde rather than direct emissions. It is described in the paper. Changes in manuscript: Since most light is available in the early afternoon and local direct emissions are relatively smaller compared to secondary production, the secondary production of formaldehyde primarily caused the peak at 14:00.

30ãĂĄPage 11 Are your conclusions that the diurnal variability is driven by variation in light levels rather than diurnal variations in emissions? If light measurements are available, you could try correlating the light intensity with the VCDs. Response: Thank you for your advice . Anderson, et al., 1996; Lee, et al., 2015; Pinardi, et al., 2013 report that the increased HCHO at early afternoon implies that photo-oxidation of VOCs was very rapid due to the peak solar irradiance at this point in the day. We also compare the

correlating of the light intensity with the VCDs, but the result shows poor correlation. The reason should be that the lifetime of HCHO and the photochemical reaction rate of VOC to generate HCHO contribute to non linear dependence of HCHO VCDs on light intensity. In order to clarify this point, we modified the manuscript accordingly. Please see the modifications below.

Changes in manuscript: Since most light is available in the early afternoon and local direct emissions are relatively smaller compared to secondary production, the secondary production of formaldehyde primarily caused the peak at 14:00. The diurnal variation in VOC emissions could also play a role in the diurnal variation of HCHO. However, the typical life time of VOCs can reach several days. The diurnal variations in VOC emission are unlikely to change the abundance of atmospheric VOCs. Therefore, diurnal variation in photo reaction rate could be a dominant driving factor. Other smaller peaks appeared in the evening during another period of busy traffic (16:00–18:00 LT), which might be caused by primary pollution sources, e.g., exhaust fumes from vehicles. Thus, the diurnal variations in HCHO during all three episodes were similar to the typical patterns of secondary sources as reported in Anderson, et al (1996), Lee, et al (2015), Pinardi, et al (2013).

31ãĂĄPage 11 Line 5 Where can the reader see evidence of similar diurnal trends in the secondary sources? Response: Thanks for your question. Anderson, et al., 1996; Pinardi, et al., 2013 show typical HCHO diurnal variation of secondary sources, and my results are similar to what they reported. Changes in manuscript: Thus, the diurnal variations in HCHO during all three episodes were similar to the typical patterns of secondary sources as reported in Anderson, et al (1996), Lee, et al (2015), Pinardi, et al (2013).

32ãĂĄPage 11 Line 6 Many of the VCDs in the during, before and after APEC periods are equal within error. Are you referring to only the afternoon peaks HCHO values? The peak during APEC value appears to be equal within error with some of the highest post APEC values. Response: Thank you very much for your question. Many of the

VCDs in the before and after APEC periods are equal, but those all are higher than the value during the APEC. Please notes that the error bars in Fig. 11(Fig. 13 now) denote the standard deviation of HCHO VCDs, but not errors.

33ãĂĄPage 11 Line 7 What are the actual values with associated errors and are they statistically different? Response: Thank you very much for your question. The averaged VCD fitting errors of evening rush hours after and before APEC due to DOAS fit error here are 9.64% and 9.80%. Systematic error of the HCHO VCDs calculated by the geometric approximation is mostly smaller than 6% for the 15° elevation angle.

34ãĂĄPage 11 Line 8 Please explain your reasoning. Response: Thank you very much for your suggestion. After consideration, we decided to delete the conclusion that " The absolute HCHO values during the APEC period are obviously lower than those in the pre-APEC and post-APEC periods. The averaged HCHO during evening rush hours after APEC was higher than that before APEC. This finding is an interesting phenomenon, which may be related to some measures taken before the APEC.". Because this difference is small within the uncertainty range, the explanation is not reasonable.

35ãĂĄPage 11 Line 14 Where were the in-situ ozone measurements located relative to the MAX-DOAS measurements? Put this information in methodology. Response: Thank you for your advice. The MAX-DOAS instrument was deployed on the balcony (without a roof) of a classroom on the 4th floor in the laboratory building in the campus of UCAS (116.67°E, 40.4°N). And the UCAS supersite is on the top floor of the laboratory building, which is about ten meters away from MAX-DOAS. Ozone (O3) was measured by UV photometry (model 49i; Thermo Scientific), which is in the UCAS supersite. And I added the corresponding content in the article. Changes in manuscript: Sec.2.1: The MAX-DOAS instrument was deployed on the balcony (without a roof) of a classroom on the 4th floor in the laboratory building in the campus of UCAS (116.67°E, 40.4°N). The UCAS supersite is on the top floor of the laboratory building, which is about 10 m away from the MAX-DOAS instrument. Nitrogen oxide (NO, NO2, and NOx)

was measured by chemiluminescence (model 42i; Thermo Scientific), and ozone (O3) was measured by UV photometry (model 49i; Thermo Scientific). These gas analyzers had precision values of 0.5 ppb and 0.4 ppb, respectively.

36ãĂĄ Section 3.3 would benefit from a reorganization. Perhaps put information about primary versus secondary sources and the correlations first before making conclusions about diurnal trends. Response: We carefully think about the suggestion, but we think our current typesetting is more logical. Firstly, the source of HCHO was implied by the observation of diurnal variation of HCHO, and then the correlation analysis was used to further support the speculation.

37ãĂĄSection 3.4 How are VCDs calculated from the model output (i.e., what vertical height interval was integrated from the modeled vertical profile?) Section 3.4 Explain in more detail why the model poorly captures the local emissions. Could the lack of heterogeneous reaction in the model be contributing to the underestimation of the low HCHO values? Page 12 Line 14. Since the grid size seems to have little impact on the quality of the model output, is the "worse constraint" due to poor or outdated emission inventories local sources in this area? Are the highway emissions included in model calculations? How accurate is the emission inventory of the highway if it's included in the model? Response: The vertical discretisation uses 60 levels up to the model top at 0.1 hPa (65 km) in a hybrid sigma-pressure coordinate. The vertical extent of the lowest level is about 17 m; it is 100m at about 300m above ground, 400–600m in the middle troposphere and about 800m at about 10 km in height (Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, Geosci. Model Dev., 8, 975–1003, doi:10.5194/gmd-8-975-2015, 2015.). HCHO VCD is calculated by an integration of the modeled vertical profiles of HCHO. The model doesn't consider the heterogeneous reaction. So the lack of heterogeneous reaction in the model could contribute to the

underestimation of the low HCHO values. The actual emission totals for 2008 inventory including anthropogenic, biogenic and natural sources and biomass burning. So highway emissions are considered in the 2008 inventory. Due to the establishment of the UCAS from 2013 and the holding of APEC meeting in 2014, the economy near the UCAS has grown rapidly, and the traffic flow has increased significantly in recent years. Thus, it could underestimate the highway emissions by using the 2008 inventory. Changes in manuscript: The underestimation of the low HCHO values by the CAMS model compared to the MAX-DOAS measurements could be attributed to the lower constraint of local emissions in the model near the UCAS measurement station, and the lack of heterogeneous reactions in the model could also contribute to the underestimation of the low HCHO values. The China National Highway 111 is nearby and runs from north to south. The actual emission totals for the 2008 inventory included anthropogenic, biogenic and natural sources, and biomass burning, thus, highway emissions were considered in the 2008 inventory. However, due to the establishment of the UCAS from 2013 and the holding of the APEC meeting in 2014, the economy near the UCAS had grown rapidly, and the traffic flow had increased significantly in recent years. Thus, the use of the 2008 inventory could underestimate the highway emissions.

38ãĂĄPage 13 Line 8 When you say "the primary HCHO is dominant" do you mean that the dominant contribution to the HCHO VCDs is the "local" primary emissions of HCHO? Edit for further clarity. Response: Thank you very much for your advice. Yes, "the primary HCHO is dominant" mean that the dominant contribution to the HCHO VCDs is the "local" primary emissions of HCHO. It is changed in the paper to make it clear. Changes in manuscript: Thus, when the secondary source of HCHO is reduced, namely the "local" primary emissions of HCHO predominantly contribute to the HCHO VCDs, the difference between the MAX-DOAS observation and CAMS model is obvious.

39ãĂĄPage 13 Line 14 Your conclusion is not necessarily sufficiently supported given the small R2 value and "reasonably" (too vague) would have to be defined before it is

clear whether the data support this statement sufficiently. Response: Thank you for your advice. R represents the ratio of HCHO VCDs in the morning (8:00LT)and noon (14:00LT). If R from the model is close to obtained from the MAX-DOAS, it indicates that the trend of diurnal variation of HCHO from the model simulation and MAX-DOAS observation is consistent. In other word, therefore the model can reasonably simulate the systematic diurnal variation of HCHO. Changes in manuscript: R represents the ratio of HCHO VCDs in the morning (8:00 LT) and noon (14:00 LT). If RModel is close to RMAX-DOAS, it indicates that the trend in diurnal variation of HCHO from the model simulation and MAX-DOAS observation is consistent, suggesting that the model can reasonably simulate the systematic diurnal variation in HCHO.

40ãĂĄPage 14 Line 8 You may want to add that, in contrast, correlation with NO2 was lower and what that implies. If VCDs are calculated from the 10° and 30°spectra, how do the values compare to the 15° spectra VCDs? Given that the geometric approximation becomes worse under high aerosol conditions and these VCDs would be expected to diverge in that case, comparison with the 10° and 30°spectra may be a good measure of the validity of your use of the geometric approximation. Response: Thank you very much for your advice. In order to constrain the systematic error of the grometric approximation, we compare HCHO VCDs calculated with the geometric approximation with those retrieved using PriAM profile inversion algorithm. The discussion is added in the manuscript as following. Changes in manuscript: a. The systematic error of the HCHO VCDs calculated by the geometric approximation depends on the layer height of the TGs and aerosols. To evaluate the systematic error of the geometric approximation, we calculated more exact tropospheric HCHO VCDAMF using the PriAM inversion algorithm (Wang et al, 2017b). HCHO VCDgeo at elevation angles at 15° and 30° are usually obtained from the geometric approximation. The relative differences (Diff) between VCDAMF and VCDgeo for HCHO were calculated by Eq. (9):

In Fig. 6, the average relative differences for elevation angles of 15°and 30° are shown as a function of the effective cloud fractions (eCF), as 0<eCF $\leq$1, 0<eCF$\leq$0.3, 0.3<eCF

$\leq$0.7, and 0.7<eCF$\leq$1.0. The cloud fractions (eCF) are downloaded from the ECMWF CAMS model. It can be seen that the biases caused by the use of the geometric approximation are generally much smaller at EA=15° than at EA=30°, with the Diff being mostly smaller than 6% for the 15° elevation angle of and smaller than 16% for the 30° elevation angle in all periods. The bias for Diff caused by using the geometric approximation is about 2% (Ma et al., 2013; Wang et al., 2017c).

41ãĂĄFigure 11 Can you explain why the standard deviation of the pre-APEC time is so much smaller than the post-APEC period despite similar values? Response: Thank you very much for your question. As we can see in figure 11, there are two obvious pollution process after APEC. The HCHO concentration shows a significant lifting process, which makes the standard deviation large.

42ãĂĄFigure 9 Since you show average values, how did the standard deviations of the averages compare to the retrieval errors? Are the larger of the two plotted as error bars? Response: Thank you for your question. The standard deviations of the averages is larger than the retrieval errors. The fitting error is also added in the figure. Changes in manuscript:

Figure 13: Averaged diurnal variation in HCHO VCDs measured by MAX-DOAS in three episodes around APEC. The short cap width of the error bars denotes the one sigma standard deviations around the mean analysis values. The long cap width of the error bars denotes the fitting error.

43ãĂĄFigure 8 Why do moderate wind speeds appear to produce similar VCD values for all wind-directions. Also, why do southerly conditions appear to result in maximum VCDs occurred under the highest wind speeds given that you stated that high wind-speeds tend to reduce VCDs? Response: Thank you for your question. I mean that transport from the south polluted air to observation site is easier under southerly winds with high wind speed. Changes in manuscript: Under the southerly wind, the HCHO VCDs increase with increasing wind speed. Thus, transport from the south polluted air

to the observation site occurs more easily under southerly winds with relatively high wind speeds.

Can you divide the VCD data into wind-speed and time of day and then see if there is a statistically significant reduction of the VCDs under non-Southerly wind conditions during APEC compared to before and after? That may help to determine how much the emissions controls impacted the VCDs independent of wind-direction. Response: Thank you for your advice. We made the new plots following your suggestion (see attached figure) as Fig. 12 in the revised manuscript. We do not see the significant reduction of HCHO VCDs under non-southerly winds during APEC compared to before and after. However it is understandable. Because our station is in the north suburban area of Beijing city. The control measurements were mainly operated in the Beijing urban area. Therefore in order to evaluate the effects of control measures in the city, we need to compare HCHO observed at the suburban station under the southerly winds between different APEC periods, because pollutants in the city center can be transported to the measurement site under the southerly winds. Accordingly we modified the paragraph in Section 3.2 which has been given in the response to your point 27.

Figure 12: Wind roses in (a) the “pre-APEC” , (b) the APEC and (c) the “post-parade” periods. Dependence of HCHO VCDs (1015 molec cm-2) on wind directions for different wind speeds in the pre-APEC (d), during the APEC (e) and post-APEC (f).

Technical Corrections: 1ãĂĄTitle: Consider adding VCDs after the word formaldehyde. Consider also including the APEC study to the title. Response: Thank you very much for your suggestion. We have considered your advice, and it is added in the title. Changes in manuscript: Ground-based MAX-DOAS observations of tropospheric formaldehyde VCDs and comparisons with the CAMS model at a rural site near Beijing during APEC 2014

2ãĂĄGeneral technical comment: when listing VCDs to route, please include the error values. Response: I don’t understand your meaning. Do you mean adding the error

after the VCDs in the text?

3ãĂĄPage 1 Line 22 Abstract: what are the units of HCHO and O3? VCDs? Mixing ratios? Response: Thank you very much for your question. The units of HCHO are VCDs, and the units of O3 are Volume mixing ratio. It is added in the paper. Changes in manuscript: Peak values of HCHO vertical column densities (VCDs) around noon and a good correlation coefficient R2 of 0.73 between HCHO VCDs and surface O3 concentration during noontime indicated that the secondary sources of HCHO through photochemical reactions of volatile organic compounds (VOCs) dominated the HCHO values in the area around UCAS.

4ãĂĄPage 2 Line 8 What were the specific dates of the conference? Response: Thank you very much for your suggestion. The specific dates of the conference are from November 5 to November 11, 2014. High emissions in Beijing and surround area were required to stop or limit their production during 3–12 November 2014. Changes in manuscript: The 2014 Asia-Pacific Economic Cooperation (APEC) conference was held in the Huairou District of Beijing from November 5–11, 2014.

5ãĂĄPage 2 Line 18 It is unclear what “traffic” and “regional” stations are? Response: Thank you very much for your question. Wang et al., (2016a) selected five representative in-situ stations in different locations of Beijing, which represents different emission types and backgrounds. The traffic station is located in the Xizhimen Station of city center in Beijing with heavy traffic and traffic flow. The regional station is located in the suburbs of Beijing to reflect the impact of urban development on the suburban environment.

6ãĂĄPage 3 lines 10 and 11 The meaning of this sentence is unclear. Response: Thank you very much for your remind. It is changed to make it clear in the paper. Changes in manuscript: Being a short lifetime oxidation product, long-living VOCs, such as methane (CH4), contribute to the background levels of HCHO (Pinardi et al., 2013; Stavrakou et al., 2009; Vrekoussis et al., 2010).

7ãĂĄPage 3 Line 24 I believe this should say tropospheric column densities, surface mixing ratios, and vertical profiles of aerosol extinction and trace gas mixing ratios. Response: Thank you very much for your remind. It is changed in the paper. Changes in manuscript: The information obtained from MAX-DOAS measurements includes tropospheric column densities, surface mixing ratios, and vertical profiles of aerosol extinction and trace gas mixing ratios.

8ãĂĄPage 4 Line 18 Consider changing to "derived from the DOAS spectral analysis [of the measured spectra]" Response: Thank you very much for your advice. It is changed in the paper. Changes in manuscript: In the first step, the differential slant column densities (dSCDs) were derived from the DOAS spectral analysis with a so-called FRS and measured in a small sun zenith angle at 90° elevation around noon (Hermans et al., 2003; Hönninger and Platt, 2002; Kraus, 2006).

9ãĂĄPage 6 Line 22 In this sentence and figure 4 you use different terms for the blue and red lines: blue (measured, derived) red (calculated, retrieved, fitted). Response: Thank you very much for your suggestion. It is changed in the paper. Now figure 4 is changed to figure 5. Changes in manuscript: Figure 5: Example of a DOAS fit of a spectrum to retrieve the slant column densities of HCHO; the red and blue curves indicate the fitted absorption structures and the derived absorption structures from the measured spectra, respectively.

Pick one term for each and ensure that the meaning of the term is clear. What about the contribution of the residual to the blue line? Response: Thank you very much for your remind. The residual represents the remaining structure after the measured spectrum (blue line) minus the fitted absorption structures (red line). The smaller the residual, the better the spectral fit

10ãĂĄPage 10 Line 4 the sentence needs editing for greater clarity and to appropriately describe figure 9. Response: Thank you very much for your suggestion. It is changed in the paper. Changes in manuscript: The result shows a "fluctuating effect"

with the HCHO VCDs increasing abruptly over several days and dropping sharply for a few days during the APEC summit.

11ãĂĄPage 13 Line 24 HCHO were also studied before and after APEC, were they not? Response: Thank you very much for your suggestion. It is changed in the paper. Changes in manuscript: We studied the tropospheric HCHO VCDs at the UCAS site in Huairou District, Beijing around the APEC summit based on the MAX-DOAS measurements from October 1, 2014 to December 31, 2014.

12ãĂĄTable 1 There are small spacing and English errors. Response: Thank you very much for your remind. It is changed in the Table 1. Changes in manuscript:

13ãĂĄClarity of Figure 9 may be improved by lines or boxes that indicate the afternoon period. Response: I think maybe you give the wrong number of the figure.

14ãĂĄFigure 6 If relative humidity is not discussed in the results or discussion, perhaps remove it from the figure to have more space to expand the more relevant data. Response: Thank you very much for your suggestion. We have considered your advice, and it is removed in the Figure 6. Changes in manuscript:

15ãĂĄSupplement: More helpful analysis may be achieved by dividing the regressions into bins that do not all include zero cloud fraction. For example, are different trends observed for eCF 0-0.3, 0.3-0.5, 0.5-0.7 etc.? Response: The figure followed shows the result of dividing the regressions into bins of eCF. And the figure also supports the conclusions in our text. The figure is added in the supplement. Changes in manuscript:

Figure S3: Correlation between HCHO VCDs retrieved from the MAX-DOAS measurements and those obtained from the CAMS model data for 0<eCF $\leq$1 (a), 0<eCF$\leq$0.3(b), 0.3<eCF $\leq$0.7 (c), and 0.7<eCF$\leq$1.0 (d) at 8:00 LT from October to December 2014.

Figure S4: Correlation between HCHO VCDs retrieved from the MAX-DOAS measurements and those obtained from the CAMS model for 0<eCF $\leq$1 (a), 0<eCF$\leq$0.3(b),

0.3<eCF $\leq$0.7 (c), and 0.7<eCF$\leq$1.0 (d) at 14:00 LT from October to December 2014.

Thank you for taking care of our manuscript.

Kind regards, Xin Tian E-mail: xtian@aiofm.ac.cn

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Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-440/acp-2018-440-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-440, 2018.

Response to comment 21:

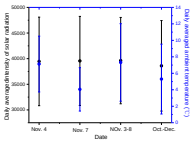

Figure 8: Averaged intensity of solar radiation and temperature in the four periods of November 4, November 7, November 3 to 8, and October 1 to December 31, 2014. Error bars denote the standard deviations.

Response to comment 27:

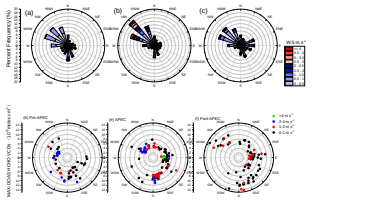

Figure 12: Wind roses in (a) the "pre-APEC", (b) the APEC, and (c) the "post-APEC" periods. Dependence of HCHO VCDs (10^{15} molec cm^{-2}) on wind directions for different wind speeds in the pre-APEC (d), during the APEC (e), and post-APEC (f) periods.

Fig. 1.