

## ***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:

Major (1) Use of the Geometric Approximation Only P5, 8: Your comment, “: : it has

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lower systematic errors because of the geometrical approximation” needs to be backed up. Do you have a reference, maybe a short explanation of your reasoning? You need to somehow prove to me that it is better to use a geometric approximation of the VCD vs. one of several RTM/inversions approaches. You did not do this here, but rather allude to some other studies. I am not convinced that the geometric approximation is the best? Prove me wrong? 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  $15^\circ$  to avoid surface obstacles on light paths along the line of sight, at the same time, it has lower systematic errors at  $15^\circ$  than at  $30^\circ$ . 2) And we add the discussion of the error budgets for geometric approximation in section 2.3. It also shows the systematic errors at  $15^\circ$  is smaller than at  $30^\circ$  by using the geometrical approximation. Changes in manuscript: Lower systematic errors were achieved at  $15^\circ$  than at  $30^\circ$  by using the geometrical approximation (discussed in Section 2.3 below).

Alternatively, you could provide a comparison of your VCDs with RTM-inversion derived HCHO VCDs. I would also suggest that you provide a comparison of your HCHO VCDs to those measured via satellite. This would give me more confidence in your conclusions Response: Thank you very much for your suggestion. We have considered your advice, and I add the comparison of geometrical approximation with inversions approaches at section 2.3. Changes in manuscript: 2.3 Error budgets The following error sources were considered as the error estimates for the MAX-DOAS results: 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  $15^\circ$  and  $30^\circ$  are usually obtained from the geometric approximation. The

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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  $15^\circ$  and  $30^\circ$  are shown as a function of the effective cloud fractions (eCF), as  $0 < \text{eCF} \leq 1$ ,  $0 < \text{eCF} \leq 0.3$ ,  $0.3 < \text{eCF} \leq 0.7$ , and  $0.7 < \text{eCF} \leq 1.0$ . 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  $\text{EA}=15^\circ$  than at  $\text{EA}=30^\circ$ , with the Diff being mostly smaller than 6% for the  $15^\circ$  elevation angle of and smaller than 16% for the  $30^\circ$  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). b. The fitting error of the DOAS fit is derived from the dSCD fitting error to VCD error by using geometric approximation, as

and the hourly average of the HCHO VCD fitting error was from 3.61% to 27.19% for the entire period. c. Cross section error also constitutes one of the error sources. Some previous research reported that cross section errors of O<sub>4</sub> (aerosols) and HCHO are 5% and 9%, respectively (Bogumil et al., 2003; Meller and Moortgat, 2000; Thalman and Volkamer, 2013; Vandaele et al. 1998 ). Wang et al (2017b) estimated the errors related to the temperature dependence of the cross sections, and the corresponding systematic error of HCHO was estimated to up to 6%. Since the three errors are mainly independent, the total error can be calculated by combining all the above error sources, adding up to about 12% on average.

(2) Emission Totals from 2008 for CAMS model P7, I12: You use emission totals from 2008. Your year of study is 2014, that is a difference of 6 years and a lot can change. Why didn't you use a more recent emission inventory? Is there one? If so, why didn't you use it? Response: Thank you for your question. The simulation work of the model is made by the ECMWF, and we download the data from CAMS real-time products in ECMWF (<http://apps.ecmwf.int/datasets/data/cams-nrealtime/levtype=sfc/>). Besides, the MAX-DOAS data can verify the model. We also make the conclusion that inven-

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tory needs to be updated according to our comparative study. Annual emissions from anthropogenic, biogenic and natural sources and biomass burning for 2008 in Tg for a composition Integrated Forecasting System (C-IFS) (CB05) run at T255 resolution (Flemming et al., 2014). The 2008 global emissions is used as a total amount of emissions to assimilate data in the C-IFS model. Then, for their near real-time data, they will be added to the latest satellite observation data for assimilation.

(3) HCHO VCD error P9, I11-120: It is likely that when the wind comes from the south it is more polluted than when the wind comes from the north. However, an average HCHO VCD of  $7.57 \times 10^{15}$  vs.  $6.64 \times 10^{15}$  is hardly conclusive. This is a 14% difference. WHAT IS THE ERROR OF YOUR VCD? I would estimate that is it a least 10%, likely over 20%. As such, your statistics here are weak. Please define the error of your VCDs and then re-word this section. For example, in Figure 9, you have error bars on your VCDs, but no mention of how you calculate them [they also look very low to me]. Response: Thank you for your advice. Although the uncertainty of HCHO VCD is about 6% for the  $15^\circ$  elevation angle, the uncertainty is comparable to the systematic difference of HCHO under different wind fields. However, uncertainty effects on systematic bias can be averaged as zero for a long-term measurements, therefore the systematic differences of HCHO VCDs still considerably indicate that more pollutants are transported from the southern region.

(4) Figure 12 – Correlation Analysis I understand what you are trying to achieve here. However, I am not sure why you choose the period Oct 26 – Nov 20, 2014? This seems random? Why not use all your data? Response: Thank you for your question. Because in the previous study around APEC, the period used was from October 26 to November 20, 2014, so the analysis here we used the corresponding period. However we have considered your advice, and I use all the data for the correlation analysis. The change is made in the paper. The new correlation analysis indicates that the correlation coefficients between HCHO VCD and NO<sub>2</sub> VCD at rush hour and between HCHO VCD and O<sub>3</sub> during the noon time are slightly reduced. However, the results

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still show high correlation between HCHO VCD and O<sub>3</sub> during the noon time and low correlation between HCHO VCD and NO<sub>2</sub> VCD at rush hour.

The NO<sub>2</sub> VCD is not described. Is it VCD<sub>geo</sub>? Is it data from the same instrument and time? Did you also compare your O<sub>3</sub> with the 7-9 & 16-18 periods? You don't have to show the plot but I would like to know the R of that? Hopefully it is very low to prove your point. Similarly, did you compare the NO<sub>2</sub> VCDs with the HCHO VCDs from 11-14 period. You need a more complete assessment here to really prove your point. Response: Thank you for your advice. The NO<sub>2</sub> VCD is VCD<sub>geo</sub> and the data is from the same MAX-DOAS instrument and time. All the suggested comparisons are added in the paper and please see the following changes. Changes in manuscript: Determining pollution sources is crucial to controlling air pollution. Three time intervals were used for determining the main HCHO sources. The first interval was defined as noontime from 11:00–14:00 and is associated with strong photochemical reactions. The second and third intervals were defined as the morning rush hour from 7:00–9:00 and the evening rush hour from 16:00–18:00. To further determine whether the pollution sources of HCHO at UCAS were primary or secondary formations from other VOCs, the correlations of HCHO with the primary pollutant NO<sub>2</sub> or secondary pollutant O<sub>3</sub> were analyzed (Anderson et al, 1996; Possanzini et al., 2002). Surface O<sub>3</sub> data were obtained from in situ measurements in the UCAS supersite, and troposphere NO<sub>2</sub> VCD data were retrieved from the same MAX-DOAS measurements using geometric approximation. The linear correlations of noontime average HCHO VCD with NO<sub>2</sub> VCD and O<sub>3</sub> from 11:00–14:00 and rush hour average HCHO VCD with NO<sub>2</sub> VCD and O<sub>3</sub> from 7:00–9:00 and 16:00–18:00 are shown in Fig. 14. Direct analysis of the data indicates that noontime average HCHO had a higher correlation coefficient with NO<sub>2</sub> VCD and O<sub>3</sub> than rush hour. This implies that a small amount of HCHO comes from the traffic emissions during rush hour. A good correlation coefficient R<sup>2</sup> of 0.73 was found between HCHO VCD and O<sub>3</sub> during the noontime, which indicates that the main source of HCHO was from secondary photo-oxidation formation at noon. In contrast, a correlation coefficient of 0.38 between HCHO VCD and NO<sub>2</sub> VCD during noontime was

better than during rush hour ( $R^2=0.06$ ), which may be due to the contribution of vehicle emissions to HCHO precursors. A longer NO<sub>2</sub> lifetime with less dispersion efficiency in winter and HCHO from continuously generated photo-oxidation contributed to the higher correlation between HCHO VCD and NO<sub>2</sub> VCD at noon higher than during rush hour. The transport of NO<sub>2</sub> and VOC may constitute one of the causes. The VOCs from transport generate HCHO due to strong photo-oxidation at noon. This result indicates that secondary photo-oxidation formation of HCHO from other VOCs should be the dominant source at UCAS.

Figure 14: Scatter plots and linear regressions (a) of noontime average HCHO VCD measured by MAX-DOAS against O<sub>3</sub> VMRs measured by a stationary ozone monitoring instrument, and (b) rush hour average HCHO VCD against NO<sub>2</sub> VCD measured by MAX-DOAS from October to December 2014.

What happens if the R value for O<sub>3</sub> and 7-9&16-18 periods is also high? I believe you have something here but be careful about how you present it. I also need to know exactly where your O<sub>3</sub> monitor is, is it at ground-level? Response: Thank you for your question. The R<sup>2</sup> value for O<sub>3</sub> and HCHO at 7-9&16-18 periods is 0.03. 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 (O<sub>3</sub>) was measured by UV photometry (model 49i; Thermo Scientific), which is in the UCAS supersite. And we add the corresponding content in the revised manuscript.

(5) Assumption that the HCHO VCD is the correct result On P12, l3 you state that the CAMS model UNDERESTIMATES : : . How do you know this? How do you know the MAX-DOAS result is the correct result and better than the CAMS model? What other VALIDATION do you have? Did you compare it to the satellite data; ground-data extrapolated to a column {see comment 1}? You may be right, but you may also be wrong. I am not convinced, especially without any error analysis of your HCHO VCDs or CAMS

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model. I would say that your CAMS model could be really off since it uses emission totals from 2008. Maybe the emission estimates in the model for 2008 are simply much lower than the 2014 values? You allude to this on P12, I14-15, right? Response: Thank you for your question. According to your advice, we evaluate the systematic error of the geometric approximation by comparing the VCD calculated using the geometric approximation and those retrieved using a PriAM profile inversion algorithm.. The new discussion is added in Section 2.3. The result shows that the systematic error is less than 6% for the elevation angle of 15 degrees. Besides, satellite retrievals of HCHO have more problem than MAX-DOAS measurements. MAX-DOAS is an usual technique to validate the HCHO satellite data (cite: De Smedt, I., Stavrakou, T., Hendrick, F., Danckaert, T., Vlemmix, T., Pinardi, G., Theys, N., Lerot, C., Gielen, C., Vigouroux, C., Hermans, C., Fayt, C., Veeffkind, P., Müller, J.-F., and Van Roozendaal, M.: Diurnal, seasonal and long-term variations of global formaldehyde columns inferred from combined OMI and GOME-2 observations, *Atmos. Chem. Phys.*, 15, 12519-12545, doi:10.5194/acp-15-12519-2015, 2015.). In addition MAX-DOAS retrievals of HCHO have been well proved and evaluated in the previous study. Wang et al., 2017b retrieved tropospheric HCHO VCDs and vertical profile in Wuxi from 2011 to 2014, and the DOAS fit setting derived from the formaldehyde slant column measurements during CINDI: intercomparison and analysis improvement. Therefore MAX-DOAS results of HCHO are valuable and sufficiently confident to be used for validation of model simulations. For the old emission inventory, the inventory is used by the operational CAMS model. We agree it could be lower than the current emission. The conclusion is also our finding by comparing MAX-DOAS measurements with the model data.

P12, I3: What do these ranges mean? Is it due to different grid-sizes? Response: Thank you for your question. These ranges are due to the different grid-sizes. We do some change in the paper to make it clear. Changes in manuscript: On average, the CAMS model underestimated HCHO VCDs by  $1.56\text{--}2.02 \times 10^{15}$  molec cm<sup>-2</sup> and  $1.27\text{--}2.12 \times 10^{15}$  molec cm<sup>-2</sup> compared to the MAX-DOAS measurements at 8:00 LT and 14:00 LT, respectively, due to different grid-sizes.

(6) RMAX-DOAS vs Rmodel P13, I10: You R concept is interesting. Based on this I would think that R(DOAS) should be higher than R(model) for cases when the temp is cold (and secondary is HCHO is lower than predicted via the model), do you see this? Alternatively, if primary HCHO emissions are under predicted in the model R(DOAS) again would be higher than R(model) right? So what does this R concept really tell us? A graph like Figure 15, does not tell me much? However, if you separate out case studies maybe you get some more information. P14, 121-23: If the CAMS model underestimates primary sources of HCHO then  $R(\text{DOAS}) > R(\text{model})$  but “under a situation with a low temperature when the production rate of secondary HCHO is relatively low” won't the CAMS model also underestimate the secondary HCHO production also causing  $R(\text{DOAS}) > R(\text{model})$  as well? What is the assumed temp in the model, or does it use real-time met-data? How do we know what is the problem, is it a problem with the assumed temp, if so can you adjust that to check? OR is it a problem with the emissions inventory (perhaps a bigger issue). Again, the above concept seems to have merit, but you need to develop this and explain it further, because I am somewhat confused. Also, despite your analysis I have no feeling as to how much HCHO is secondary and how much is primary (and isn't that what the R calculations are for?). Response: Thank you for your suggestions. Here are some explanations for your questions. We agree on your conclusion, if there is big bias in the model simulations of the secondary production of HCHO, it can also cause deviations of  $R(\text{DOAS})$  and  $R(\text{model})$ . Following your suggestion: 1) we separate the plots in the periods of October to November and for December (see below). But there is no significant difference between the two periods.

Scatter plots and linear regression of RModel against RMAX-DOAS (see the text) from October to December 2014 (a) and from October to November and for December due to the changing of temperature (b). There are not significant differences between the two periods.

2) we check the source of meteorological data in the model. The CAMS global real-time

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production system uses all the meteorological observations from the ECMWF numerical weather prediction system, which is extracted from satellite real-time meteorological data. We also compared the temperature in the model with in-situ measurements. The results are shown in the response according to your point “P12, I124” in the minor comment. Generally good agreement can be seen. Therefore the model simulations could predict the secondary formation of HCHO well, but it can't be confirmed.

Based on the two further analysis, we noticed that the diurnal variation of HCHO is a mixed effect of primary emission, secondary formation, and probably also meteorology. It is impossible to gain the conclusion that which is the factor which causing the deviation of R(DOAS) and R(model). Therefore the R comparisons only generally evaluate the quality of model simulations on diurnal variations of HCHO. As you asked, both underestimation of primary emission and overestimation of secondary emission by model simulations can cause the similar fact that R(DOAS)>R(model). We can not firmly conclude which is the reason. And the method can't give quantified conclusion of HCHO source. Therefore we add a clarification in the revised manuscript. Changes in manuscript: It needs to be noted that the diurnal variation in HCHO is the result of the combined influence of primary emissions, secondary formation, and meteorology. We found that RMAX-DOAS was generally larger than RModel. However, it was impossible to determine the factor causing the deviation in RMAX-DOAS and RModel. Therefore, the R comparisons generally only evaluate the quality of the model simulations on diurnal variations in HCHO.

Minor P2, I3: {Q} Is the correlation coefficient (R=0.83)? If so, say (R=0.83, not ~0.83)  
Response: Thank you for your suggestions. Correlation between HCHO VCDs retrieved from the MAX-DOAS measurements and those obtained from the CAMS model at 8:00 LT and 14:00 LT from October to December 2014 in different grids were compared. The correlation coefficient R is more than 0.83, So we use ~0.83. And it is changed in the paper. Changes in manuscript: The HCHO VCDs of the CAMS model and MAX-DOAS were generally consistent with a correlation coefficient R2 greater

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than 0.69..

P2, I14: {Q} How is “APEC blue” defined? Perhaps a brief statement of how the actual reduction strategies were defined and the defined APEC levels would be useful? Is there an APEC-red for example? Response: Thank you for your suggestions. For the sake of guaranteeing the smooth convening of the APEC meeting, China took a series of effective measures which played a prominent role in improving the air condition in Beijing and surrounding regions. As a result, a better quality environment emerged, which we called “APEC-Blue”. This is reported in the Chinese web-site (<https://baike.so.com/doc/7519682-7792600.html>). The actual reduction strategies were added in the paper to make it clear. Changes in manuscript: Since November 1, 2014, parts of the Jing-Jin-Ji region and surrounding areas had begun to implement an emission reduction plan according to the APEC conference air quality assurance policy. Formal emission reduction measures were implemented in the Jing-Jin-Ji region and surrounding areas from November 3 and included limiting the production of factories, shutting down construction sites, implementing traffic restrictions based on even- and odd- numbered license plates, and improving road cleaning (Wang et al., 2016). In response to the possible adverse weather conditions from November 8–10, the “enhanced emission reduction measures” were implemented in the Jing-Jin-Ji region and surrounding areas from November 6. These various efforts coupled with relatively favorable weather conditions than previous years resulted in the emission reduction measures having significant effects. Based on estimations, all types of main pollutants were reduced by over 40% in Beijing and by over 30% in other provinces, through these measures (Wang et al., 2016).

P2, I16-17: {Q} Do you or the authors of the Wang et al. make any conclusion as to why the O<sub>3</sub> rose to 189%? Does this have to do with being in a NO<sub>x</sub>-limited or VOC-limited regime? Response: Thank you for your question. Wang et al., 2016a gave the reason that the O<sub>3</sub> in urban and suburban areas of Beijing is mostly in the control area of VOCs. The possible reason for the increase of O<sub>3</sub> is that the emission

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control measures of NO<sub>x</sub> are greater than the emission control measures of VOCs, which leads to the weakening of the inhibition of O<sub>3</sub> formation by NO<sub>x</sub>, resulting in significant increasing of O<sub>3</sub> concentration. And it is added in the paper to make it clear. (Besides, the introduction is reorganization to make it more logical. ) Changes in manuscript: Wang et al (2016a) selected five representative in situ stations in different locations in Beijing and found that average concentrations of SO<sub>2</sub>, NO<sub>2</sub>, PM<sub>10</sub>, and PM<sub>2.5</sub> decreased by 61.5%, 40.8%, 36.4%, and 47.1%, respectively, whereas the average concentration of O<sub>3</sub> increased by 101.8%, compared with the same period over the last five years (PM<sub>2.5</sub> since 2013). O<sub>3</sub> 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 O<sub>3</sub> is that the emission control measures of NO<sub>x</sub> are greater than the emission control measures of VOCs, which leads to the weakening of the inhibition of O<sub>3</sub> formation by NO<sub>x</sub>, resulting in significant increases in O<sub>3</sub> concentration.

P2, I25-P3, I1: {Q} What were Zhang's conclusions (briefly)? Response: Thank you for your advices. We have considered your advice, and we add the Zhang's conclusions to make it clear. During the APEC conference period, the average concentration of PM<sub>2.5</sub> was  $37.7 \pm 35.4$  mg/m<sup>3</sup>, which was 48% and 54% lower than that of BAPEC and AAPEC period, respectively. Compared with ultrafine particles (<100 nm), the number concentration of accumulation mode and coarse mode particles experienced more significant decreases by 47% and 68%, indicating that particles with larger sizes were better controlled during the APEC period. Changes in manuscript: 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).

P3, I1-2: ADD {REFS} for the published studies here. Response: Thank you for your advice. The REFS were added in the paper. Changes in manuscript: Published studies

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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).

P3, I5: {REF} is not in your final reference list. Response: Thank you for your remind. We are so sorry for making this mistake. The REF is added in the final reference list.. Changes in manuscript: Fried A, Cantrell C, Olson J, Crawford J H. Detailed comparisons of airborne formaldehyde measurements with box models during the 2006 INTEX-B and MILAGRO campaigns: potential evidence for significant impacts of un-measured and multi-generation volatile organic carbon compounds, Atmos. Chem. Phys., 11, 9887-9957, 2011.

P3, I14-15: HO<sub>2</sub> and OH are radicals not ions, please correct this. Response: Thank you for your remind. We are so sorry for making this mistake. It is corrected. Changes in manuscript: As an active gas, HCHO can be photolyzed to generate HO<sub>2</sub> free radicals. HO<sub>2</sub> rapidly and radically reacts with NO to generate OH, which can influence the oxidation ability of the atmosphere.

P3, I22: fix {REF}, you mean Honninger et al., 2004 right? Response: Thank you for your remind. We are so sorry for making this mistake. Yes, it is corrected in the paper. Changes in manuscript: A type of passive differential optical absorption spectroscopy system, called Multi-axis Differential Optical Absorption Spectroscopy (MAX-DOAS), has been used over the past decade to measure tropospheric trace gases (Honninger et al., 2004; Wagner et al., 2004; Sinreich et al., 2005; Wagner et al., 2007; Vigouroux et al., 2009).

P4, 16: I would call it the Beer-Lambert Law Response: Thank you for your advice. It is corrected in the paper. Changes in manuscript: MAX-DOAS, which is an optical remote-sensing technology that records the spectra of scattered sunlight at different elevation angles, can be used to quantitatively measure trace gases based on Beer-

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Lambert Law (Hönninger and Platt, 2002; Bobrowski et al., 2013; Roozendaal et al., 2003; Trebs et al., 2004; Hönninger et al., 2004; Wagner et al., 2004).

P4, I9: {Q} Were clouds a factor? How often was it cloudy? Was the data pre-screened in any way? 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. The data is pre-screened. The spectrum with too small a light intensity and an excessive integration time are removed.

Figure 1: Change the colour red on your figure, it is hard to read. Response: Thank you for your advice. It is corrected in the paper. Changes in manuscript:

P5, I21: : : some point sources (e.g. XX and YY). Add some key examples, factories or power plants? Response: Thank you for your advice. Some point sources here mean stationary sources from the rural settlement. They are not factories and power plants. And it is added in the paper. Changes in manuscript: The site is mainly influenced by emissions from vehicles on China National Highway 111 that runs from the north and south as well as some stationary sources from the rural settlements across the highway (Zhang et al., 2017).

Figure 2: fix the text on your figure (e.g. spectrograph as one word) Response: Thank you for your suggestion. It is changed in the paper. Changes in manuscript:

P5, I24: {C} change stepping motor to stepper motor? Response: Thank you for your advice. It is changed in the paper. Changes in manuscript: This system comprises a telescope, stepper motor, spectrometer, and computer.

P6, I2: {Q} Why was the temp set to 20C? Response: Thank you for your question. The changing ambient temperature in China is from -15 °C to 35 °C in a year. And the

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weather in spring and autumn is a little longer in Beijing with the temperature around 20 °C. So we set the temp as 20 °C to make sure a stable temperature in all seasons. Changes in manuscript: 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.

P6, I10: replace scanning times with SCANS Response: Thank you for your advice. It is changed in the paper. Changes in manuscript: Each measurement had an average of 100 SCANS, and the integration time was adjusted automatically based on the light intensity.

Figure 3: replace a1, a2, etc. with a3 a30 a90 etc. {you don't need to number each one, simply add elevation angles to the alpha directly} Response: Thank you for your advice. It is changed in the figure 3. Changes in manuscript:

Table 1: fix text .. Longitude – one word, {Q} What is the MAYA? Is that Ocean Optics? If so, add that. Response: Thank you for your advice. It is changed in the table. And Maya is a Ocean Optics spectrometer (<https://oceanoptics.com/product/maya2000-pro-custom/>). The briefly introduction of Maya is added in the paper. Changes in manuscript:

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.

P6, 19: replace Doasis with DOASIS Response: Thank you for your advice. It is changed in the paper. Changes in manuscript: The ring structure (Fish and Jones, 2013), which is used to account for rotational Raman scattering effects, was calculated using DOASIS software (Kraus, 2006) based on the FRS and was included in the fit.

P6, I22: replace [derived] with [measured] {as you did in your Figure4} Response:

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Thank you for your advice. I changed the description of figure 4 here. Now figure 4 is changed to figure 5 because some new figure is added. 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.

Figure 6 shows the period of 3-8 November. Why didn't you use the period of 3-12 November (the whole APEC period)? Response: Thank you for your question. This data is used to support the analysis of the transport event. So the meteorological data for the time period corresponding to the transport event is displayed, which is the period of 3 to 8 November, 2014.

P8, l11: You describe 2 peaks on Nov 4 and Nov 7, but what about Nov 3, as seen on Figure 6 that actually has the HIGHEST HCHO VCDs? Response: Thank you for your advice. Two daily averaged HCHO VCD peaks were on Nov 4 and Nov 7, and the rise process of Nov 4 is from the evening of Nov 3.

Figure 7: perhaps replace UTC time with LT for consistency. Response: Thank you for your suggestion. I changed in the paper. Changes in manuscript:

Figure 9: Error bars equal retrieval error. {Q} How is this calculated? Response: Thank you for your question. We have considered your question, and 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 about the retrieval error, we discuss at the section 2.3. Changes in manuscript:

Figure 11: Daily averaged values of HCHO VCDs from October 26, 2014 to November 20, 2014. Error bars denote standard deviations.

P10, l19-20: Is there any way to determine which is more important, the control measures of the meteorology? Perhaps a longer term study? Please comment. Response: Thank you for your suggestion. Comparisons of transports events between from the

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polluted south area and clean north area indicate meteorology condition can vary HCHO amounts by about 50%. However meteorology condition is not under control. Reduction of HCHO emission in the south polluted area can be estimated by ~20% due to control measures of emissions. The significant effects of control measures are important for improving air qualities, especially under a meteorology condition which obstructs depositions of pollutants.

P11, I7-8: Could this have to do with a change in NO<sub>x</sub>-limiting vs. VOC-limiting cases? Please advise. Response: Thank you for your advice. We carefully think about your advice and do some research on previous literature. Wang et al., 2009 found that ozone formation is mainly controlled by VOCs in the near-suburbs of Beijing City and its high-value ozone areas in the downwind direction. In suburban counties and rural areas, the sensitivity of ozone generation to NO<sub>x</sub> becomes important. And the UCAS is located in the outer suburbs of Beijing, in other word, the UCAS belongs to the NO<sub>x</sub>-limiting area. During APEC, the NO<sub>x</sub> concentration gradually decreases due to the control measurement. As a results, the HCHO decreases. After APEC, control measurements are abolished, HCHO concentration is increased with the increasing of NO<sub>x</sub>. There should be not a change in NO<sub>x</sub>-limiting vs. VOC-limiting cases. So we can't draw the exact conclusion. On the other hands, according to the recommendation of reviewer 1, we seriously discussed it. The SNR in evening is low that makes the data not very credible. So I decided to remove this part from the text.

P11, I14: Where was the surface O<sub>3</sub> measurement location exactly? What type of NO<sub>2</sub> VCD was it, geo-approximated, same instrument and location? Please describe. 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. The O<sub>3</sub> was measured by UV photometry in the UCAS supersite. The NO<sub>2</sub> VCD was obtained from the MAX-DOAS observation by using geo-approximated. The description was added

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in the paper. 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. Nitrogen oxide (NO, NO<sub>2</sub>, and NO<sub>x</sub>) was measured by chemiluminescence (model 42i; Thermo Scientific), and ozone (O<sub>3</sub>) was measured by UV photometry (model 49i; Thermo Scientific). These gas analyzers had precision values of 0.5 ppb and 0.4 ppb, respectively. Sec. 3.3 : Surface O<sub>3</sub> data were obtained from in situ measurements in the UCAS supersite, and troposphere NO<sub>2</sub> VCD data were retrieved from the same MAX-DOAS measurements using geometric approximation.

P11, 118: Too many significant figures! Response: Thank you for your advice. It is changed in the paper. Changes in manuscript: Direct analysis of the data indicates that noontime average HCHO had a higher correlation coefficient with NO<sub>2</sub> VCD and O<sub>3</sub> than rush hour. This implies that a small amount of HCHO comes from the traffic emissions during rush hour. A good correlation coefficient R<sup>2</sup> of 0.73 was found between HCHO VCD and O<sub>3</sub> during the noontime, which indicates that the main source of HCHO was from secondary photo-oxidation formation at noon. In contrast, a correlation coefficient of 0.38 between HCHO VCD and NO<sub>2</sub> VCD during noontime was better than during rush hour (R<sup>2</sup>=0.06), which may be due to the contribution of vehicle emissions to HCHO precursors. A longer NO<sub>2</sub> lifetime with less dispersion efficiency in winter and HCHO from continuously generated photo-oxidation contributed to the higher correlation between HCHO VCD and NO<sub>2</sub> VCD at noon higher than during rush hour. The transport of NO<sub>2</sub> and VOC may constitute one of the causes. The VOCs from transport generate HCHO due to strong photo-oxidation at noon.

P12, I124: What is the assumed temp in the model for Dec 1, 2014 then? Response: Thank you for your asking. We download the temp data of model at 2 meter and compare with the temp from in-situ instrument. The results show that the temp in the model also plummeted in December 1, 2014, and fell below 0 °C.

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Figure: Hourly averaged temperature in CAMS model (grid of  $0.125^\circ \times 0.125^\circ$  and  $0.25^\circ \times 0.25^\circ$ ), at 2 metre and in-situ observations at 8:00 (a) and 14:00 LT (c) from October 29 to December 31, 2014.

P13, I16-20: Briefly state what associated errors clouds could pose. In I19 you say a slight variety (variation), give an error estimate please. Response: Thank you for your suggestion. It is added in the paper. Changes in manuscript: First, clouds can affect atmospheric radiative transport and thus influence optical paths. Furthermore, the atmospheric absorber densities [by (photo-)chemistry or convective transport] are potentially altered due to the changes in optical paths (Grats, ea et al. 2016). Second, AMFs calculated by geometrical approximation could be significantly biased from the reality under cloudy conditions (Brinksma, et al., 2008). REF: Gratsea, M., Vrekoussis, M., Richter, A., Wittrock, F., Schonhardt, Anja., Burrows, J., Kazadzis, S., Mihalopoulos, N., Gerasopoulos, E.: Slant column MAX-DOAS measurements of nitrogen dioxide, formaldehyde, glyoxal and oxygen dimer in the urban environment of Athens, Atmos. Environ., 135,118-131,2016.

P14, I16: Where does this number come from and what dates? It is not the same as Figure 12 and it is not mentioned anywhere else in your paper. Is it a typo? Please advise. Response: Thank you for your remind. There is mistake. This number (0.87) is the correlation coefficient  $R^2$ , and the 0.934 in figure 12 is the correlation coefficient  $R$ . We redraw the figure 12 by using all the data from October to December, 2014. And the figure 12 is changed to figure 14 due to some new figures were added. The new correlation coefficient  $R^2$  of average HCHO VCDs with  $O_3$  is 0.73. It is changed in the paper. Changes in manuscript: A good correlation coefficient  $R^2$  of 0.73 was found between HCHO VCD and  $O_3$  during the noontime, which indicates that the main source of HCHO was from secondary photo-oxidation formation at noon.

P14,I20: Why the range? Grid sizes, I assume? Response: Thank you for your question. The range is mainly due to two different time periods(8:00 and 14:00 LT) and three different grid points. In order to make it clear, I calculate the averaged value

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and change it in the paper. Changes in manuscript: The CAMS model underestimated HCHO VCD by about  $1.63 \times 10^{15}$  molec cm<sup>-2</sup> on average compared to the MAX-DOAS measurements.

Thank you for taking care of our manuscript.

Kind regards, Xin Tian E-mail: [xtian@aiofm.ac.cn](mailto: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-AC2-supplement.pdf>

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

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The NO<sub>2</sub> VCD is not described. Is it VCDgeo? Is it data from the same instrument and time? Did you also compare your O<sub>3</sub> with the 7-9 & 16-18 periods? You don't have to show the plot but I would like to know the R of that? Hopefully it is very low to prove your point. Similarly, did you compare the NO<sub>2</sub> VCDs with the HCHO VCDs from 11-14 period. You need a more complete assessment here to really prove your point.

Changes in manuscript:  
figure 1

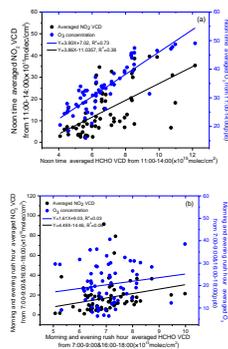
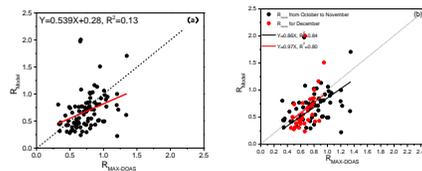


Figure 14: Scatter plots and linear regressions (a) of non-time average HCHO VCD measured by MAX-DOAS against O<sub>3</sub> VMRs measured by a stationary ozone monitoring instrument, and (b) rush hour average HCHO VCD against NO<sub>2</sub> VCD measured by MAX-DOAS from October to December 2014.

Fig. 1.

(6) RMAX-DOAS vs Rmodel  
figure:



Scatter plots and linear regression of  $R_{\text{Model}}$  against  $R_{\text{MAXDOAS}}$  (see the text) from October to December 2014 (a) and from October to November and for December due to the changing of temperature (b). There are not significant differences between the two periods.

Fig. 2.

Figure 1: Change the colour red in your figure, it is hard to read.

Changes in manuscript:  
figure



Fig. 3.

Figure 2: fix the text on your figure (e.g. spectrograph as one word)

Changes in manuscript:

figure:

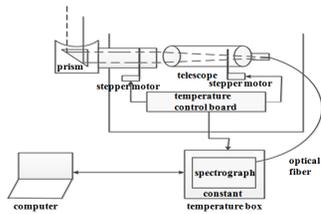


Fig. 4.

Figure 3: replace  $\alpha_1, \alpha_2$ , etc. with  $\alpha_3$  to  $\alpha_{90}$  etc. (you don't need to number each one, simply add elevation angles to the alpha directly)

Changes in manuscript:

figure:

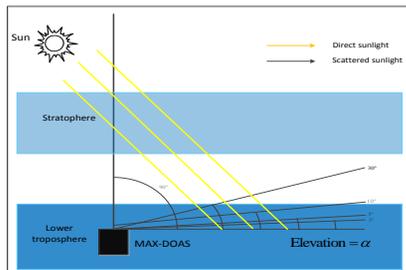


Fig. 5.

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Discussion paper



Table 1: fix text ... Longitude – one word. (Q) What is the MAYA? Is that Ocean Optics?  
If so, add that.

Changes in manuscript:

Table

Spectrometer		Azimuth	Elevation	Temperature	Location		Measuring time
Name	Maya (Ocean Optics)	0 °	3 ° 5 ° 10 ° 15 ° 30 ° 90 °	20 °C	Site	Yanxi Lake campus of UCAS	6:30-18:30
Spectral range	290– 420 nm				Longitude	116.67 °E	
FWHM	0.5 nm				Latitude	40.4 °N	

Fig. 6.

Figure 7: perhaps replace UTC time with LT for consistency.

Changes in manuscript:

figure:

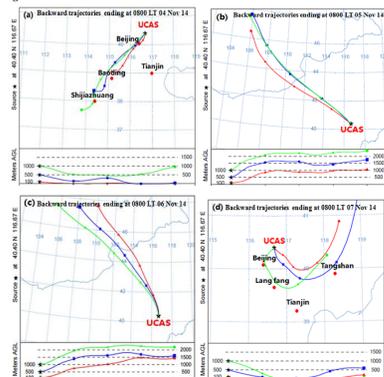


Fig. 7.

Figure 9: Error bars equal retrieval error. (Q) How is this calculated?

Changes in manuscript:

figure:

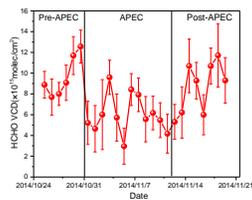


Figure 11: Daily averaged values of HCHO VCDs from October 26, 2014 to November 20, 2014. Error bars denote standard deviations.

Fig. 8.

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PI2,1124: What is the assumed temp in the model for Dec 1, 2014 then?

Response:

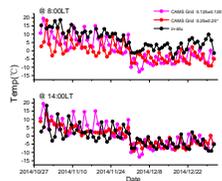


Figure: Hourly averaged temperature in CAMS model (grid of  $0.125^{\circ}\times 0.125^{\circ}$  and  $0.25^{\circ}\times 0.25^{\circ}$ ); at 2 metre and in-situ observations at 8:00 (a) and 14:00 LT (c) from October 29 to December 31, 2014.

Fig. 9.