Atmospheric
Chemistry
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Discussions

ACPD

Interactive
comment

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.

Anonymous Referee #1

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General Comments:

The paper does address relevant scientific questions within the scope of ACP. The authors give proper credit to related work and indicate their own new/original contribution. How their work is original could be more clearly emphasized. The abstract, number and quality of references, and the amount of supplementary material are all satisfactory. However, 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

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Discussion paper

of an English editor. A 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).

Specific Comments:

For all regressions, the coefficient of determination (R^2) statistic may be more appropriate since this value indicates the variance in the dependent variable that is predictable from the independent variable. Page 2 Line 3 What specific results had a good correlation coefficient? VCDs of HCHO? Other? Page 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. Page 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. Page 3 Line 6 Three sources are listed despite elsewhere in the paper it is stated that there are two sources, which one is it? Page 3 This discussion may benefit from writing out some of the most important chemical equations for the reader equations for the reader. Page 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. Page 4 Line 5 What unit of HCHO? VCDs? Page 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. Section 2.1 Please list the final equation used for calculating VCDs from fitted DSCDs given your geometric approximation. Re-organize 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? Are any of the MAX-DOAS VCDs removed from the dataset due to cloud fraction? If so, what was the cut off cloud fraction value? Page 5 Line 8 It is unclear what causes lower systematic errors. Page 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 strato-spheric 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 tropo-spheric trace gases, ATMOSPHERIC Meas. Tech., 3(1), 129–140, 2010. Page 6 Line 23 What is the software reported SCD error in molec/cm2? Page 6 Line 24 Are you missing units on this number? Page 7 section 2.6 At what height were these meteoro-logical parameters measured? At what time frequency before averaging? Page 7 Line 24 What does a "static" weather situation mean? Better organization and flow in sec-tion 2 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. Figure 6 – Are all the days measured or the ones that met some quality control crite-ria? Page 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. Page 8 Line 21 Please define (or find a better descriptor for) "good dispersion conditions". Page 9 Line 14 What type of relevant pollution sources do these cities have? Primary and/or

secondary? Are there many industry and/or vehicular sources? Page 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, etc. Page 9 Line 17 and 18 Please explain the dependence of the VCDs on wind speed under different wind directions. Page 10 Line 6 What are the errors on each of the VCD values. Are they statistically different? 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. Page 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. Page 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. Page 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. Page 11 Line 5 Where can the reader see evidence of similar diurnal trends in the secondary sources? Page 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. Page 11 Line 7 What are the actual values with associated errors and are they statistically different? Page 11 Line 8 Please explain your reasoning. Page 11 Line 14 Where were the in-situ ozone measurements located relative to the MAX-DOAS measurements? Put this information in methodology. Section 3.3 would benefit from

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a reorganization. Perhaps put information about primary versus secondary sources and the correlations first before making conclusions about diurnal trends. Section 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? Page 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. Page 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. Page 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 10o and 30o spectra may be a good measure of the validity of your use of the geometric approximation. Figure 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? Figure 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? Figure 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? 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.

Technical Corrections:

Be careful always to include the correct type of units when quoting numbers, where applicable. Title: Consider adding VCDs after the word formaldehyde. Consider also including the APEC study to the title. General technical comment: when listing VCDs to route, please include the error values. Page 1 Line 22 Abstract: what are the units of HCHO and O3? VCDs? Mixing ratios? Page 2 Line 8 What were the specific dates of the conference? Page 2 Line 18 It is unclear what "traffic" and "regional" stations are? Page 2 Line 20 Missing subscript. Page 2 lines 20 and 21 This statement is too vague as to be informative Page 2 Line 21 What are the units of the measurements of these gases? Page 3 lines 10 and 11 The meaning of this sentence is unclear. Page 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. Page 4 Line 18 Consider changing to "derived from the DOAS spectral analysis [of the measured spectra]" Page 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). 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? Page 10 Line 4 the sentence needs editing for greater clarity and to appropriately describe figure 9. Page 13 Line 24 HCHO were also studied before and after APEC, were they not? Table 1 There are small spacing and English errors. Clarity of Figure 9 may be improved by lines or boxes that indicate the afternoon period. Figure 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. Supplement: 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.?

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