

We would like to thank the reviewer for her/his encouraging comments regarding the quality of the submitted work and suggestions. We respond to the specific comments below, where the reviewers' comments are marked in blue, our responses are shown in black, and the modification in the manuscript is shown in red.

The paper titled 'Long term MAX-DOAS measurements of NO<sub>2</sub>, HCHO and aerosols and evaluation of corresponding satellite data products over Mohali in the Indo-Gangetic plain' reports long term MAX-DOAS observations of AOD, NO<sub>2</sub> and HCHO from Mohali, a suburban site in the Indo-Gangetic plain covering the period of January 2013 – June 2017. The MAPA algorithm is used to retrieve vertical profile and vertical column densities (VCD) and the results are discussed in detail. Seasonal and annual trends along with diurnal variation of vertical profiles are discussed and inter-comparison of the MAX-DOAS measured AOD, NO<sub>2</sub> and HCHO with satellite observations are reported. Finally, they have compared surface volume mixing ratios of NO<sub>2</sub> and HCHO with in situ observations. Whilst the study is strong on analysis, with the methodology explained in detail, the paper is weak on new results. At present, the manuscript as it stands is more appropriate for a methods journal, for example Atmospheric Measurement Techniques. No novel results are presented in terms of improving our understanding of the chemistry or physics of the region, or of the two chemical compounds and their impacts.

We thank the reviewer for her/his assessment and suggestion. With respect to the suggestion to submit the paper to another journal, we are sorry if the novelty of the results and the new findings got diluted by the emphasis on the technical description pertaining to methodology. To address this concern of the reviewer and strengthen the results of the paper, we have now shifted several technical aspects to the appendix and improved the discussion of the novel results (for details, see below). We would like to add that like many other esteemed papers published in ACP, our paper contains methodological aspects (which are important for a thorough understanding, especially as such studies are lacking from the region) and also many results with relevance for atmospheric chemistry and physical processes. In our opinion, the most important novel aspects of our study are the following:

- a) In addition to the first MAX-DOAS measurements of NO<sub>2</sub> and HCHO vertical column densities from this under-represented yet crucial part of the world, we report in particular the vertical profiles of aerosol, NO<sub>2</sub> and HCHO for the first time from the Indo Gangetic plain (and India). This is important information for several aspects of atmospheric chemistry and physics including atmospheric modelling, satellite retrieval and understanding atmospheric dynamics. The application of vertical profile includes calculation of airmass factors required for conversion of slant column densities (SCD) to vertical column densities (VCD), understanding the atmospheric chemistry at higher altitudes, understanding atmospheric dynamics, medium and long-range transport, evaluation of the vertical distribution of chemical tracers in atmospheric models.

A major fraction of the NO<sub>2</sub> column was found to be located in the bottom-most layer extending from surface until 200 meters in all the seasons. We show that during summer and monsoon seasons, there is a significant fraction of formaldehyde present at intermediate layers (between 200 and 600m altitudes) and sometimes even higher than the surface indicating an active photochemistry at these layers. Following the reviewer's feedback, we now also discuss the vertical distributions with respect to the ERA5 boundary layer height (BLH) and found that the seasonal trends in the derived vertical distributions are not strongly influenced by the BLH. Monsoon season is particularly interesting as the pollutants from the surface are lifted up to higher altitudes due to deep convection even though the ERA5 BLH are shallow. NO<sub>x</sub> and VOCs transported to high altitudes can be transported to a larger area and can also participate in secondary chemistry producing reservoir species such as PAN.

- b) We analyse the annual, seasonal and diurnal profiles of AOD, NO<sub>2</sub> and HCHO. We acknowledge that several AOD measurements and in situ measurements of NO<sub>2</sub> have been reported from this region. However, HCHO (which is primarily a secondary photo-oxidation product and serves as an indicator of photochemical activity and VOCs) has rarely been reported. By performing analyses of seasonal and diurnal trends of AOD, NO<sub>2</sub> and HCHO,

we identify sources and chemical processes that drive their ambient levels. The sources of HCHO are identified to be quite contrasting from majorly photochemical from biogenic and anthropogenic sources in summer, monsoon and early post monsoon to primary anthropogenic in winter and late post monsoon. We show that even though the region around the measurement location has undergone urbanisation, an obvious trend was not observed in AOD, NO<sub>2</sub> and HCHO for more than four years of measurements.

- c) Using the measured HCHO and NO<sub>2</sub> VCDs, we show that ozone production is sensitive to NO<sub>x</sub> and VOCs in winter, but shifts towards NO<sub>x</sub> in summer. This analysis was originally performed for the peak daytime hours, which overlaps with OMI overpass and generally when the maximum in the diurnal profiles of ozone is observed. Following the reviewer's suggestion, we have extended this analysis for the morning and late afternoon hours, which strengthens the observation part of the manuscript.

The main conclusions of the paper are twofold – presenting an inter-comparison with satellite products, which is a methodology based conclusion and second that ozone production is sensitive to NO<sub>x</sub> and VOCs in winter, but more to NO<sub>x</sub> during the other seasons – not a novel result considering the past publications globally and in this region. Even the methods section is not new considering it has been developed in the past by some of the co-authors and has already been used in different parts of the world.

Hence, I would reject the current paper and encourage submission in a methods journal or ask the authors to focus more on the observations rather than the observation methodology to decipher novel results. At present the manuscript, although replete with instrument and retrieval details, does not include significant results on atmospheric chemistry.

- a) We understand the reviewers' concern that intercomparison with satellite observation is primarily method based. In our view, putting the technical rigour for the measurements and intercomparison, which are first of its kind from this region is, however, crucial for the manuscript. Keeping in view that several studies, especially over India, use only satellite observations to draw important conclusions regarding NO<sub>x</sub> and VOC emissions, ozone production control, VOC source identification, long term trend analyses (e.g. Ghude, et al. 2008, Surl, et al. 2018, Chaliyakunnel, et al. 2019, Hilboll, A., et al. 2013), the evaluation of various satellite data products is crucial for improving the understanding of atmospheric chemistry and physics over the IGP. This has not so far been done for the IGP (lines 76 -92 of the original manuscript). Our study provides the first evaluation of three OMI NO<sub>2</sub> data products and two OMI HCHO data products over the IGP.
- b) Quantitative evaluation of the sensitivity of ozone production in different seasons over India for a period longer than one year has been reported by Mahajan et.al., 2013 using SCIAMACHY observations for the mean of the years 2002-2013, and by Kumar et al. 2010 and Sharma et al. 2016 using WRF model for the year 2008 and 2010, respectively. Recognising the rapid urbanisation and industrialisation in the IGP, the sensitivity might change, and our observations provide a crucial update for the same. Moreover, the unique feature of our study is that we calculate the sensitivity using ground-based observations as opposed to the previous studies using SCHIMACY observation (coarse resolution, limited sensitivity close to the ground) and model simulations (which rely on coarse resolution and uncertain emission inventories in the region).

In order to focus more on the observations and their interpretation, we have addressed the specific comments of both the reviewers and restructured the manuscript in the following way:

1. We have made section 2.5 more concise and moved technical details about the various satellite data products in the appendix.

2. We have modified Figure 6 to also include boundary layer height from ERA5 and included its discussion with respect to characteristic profiles heights and vertical distribution of aerosol in section 3.2.
3. We have included a comparison of MAX-DOAS surface VMRs of NO<sub>2</sub> and HCHO to available previous works from India.
4. We have included the HCHO/NO<sub>2</sub> ratio for the morning and late afternoon hours in Figure 11 and the relevant discussion in section 3.6.
5. We have restructured section 3.7 to focus more on the interpretation of the retrieved surface concentrations of HCHO and NO<sub>2</sub> and moved technical details about the intercomparison with in situ observations to the appendix. We have also shown the seasonal variation of the surface VMR of NO<sub>2</sub> and HCHO in the insets of Figures 12 and 13.
6. We have modified the abstract to focus more on the novel findings.

#### Specific comments:

##### Measurement technique:

1. Why did the detector temperature have to be adjusted for different seasons? If the temperature was not stabilised for different seasons, would the diurnal temperature change not lead to the same issue? Was the DC and offset measured for different temperatures and removed from the spectra according to the ambient temperature? (line 151-152)

The detector temperature **was stabilised** using a Peltier cooler and set to values such that the following two conditions (lines 149-151 of the original manuscript) are met:

1. The detector temperature is lower than the ambient temperature.
2. The difference between the ambient temperature and detector temperature is not more than 20 °C.

The complete mini-MAX DOAS instrument was installed in the open, and we had to consider large variation in ambient temperature to ensure a manageable workload on the Peltier cooler. The ambient temperature in Mohali ranges from less than 5 °C in winter to up to higher than 40 °C in summer, but the amplitude of diurnal temperature variation is typically less than 20 °C. Hence, **we did not need to adjust the detector temperature to account for the diurnal temperature change**. The dark current and offset measured for the different temperatures were removed from the spectra according to the respective detector set temperatures. This information was provided in the original manuscript in lines 151-155. However, to make it clearer for the readers, we have changed to the following in the revised manuscript (lines 157-158):

“The dark current and offset spectra were recorded every night, and while performing the spectral analysis, these were subtracted from measured spectra recorded at similar detector temperature.”

2. ‘During the period of measurement, the horizon in the viewing direction was determined by a residential building with a height of about 40m at a distance of 3 km.’ - This part is not clear. Does it mean the line of sight has an obstacle within 3 km? Is this why a 1 degree elevation angle was not used? (line 164-165)

One of the crucial steps for setting up a MAX-DOAS measurement is the elevation calibration. We performed the elevation calibration using the horizon scan method as described by Donner et.al., 2020. One of the pre-requisites of this method is the knowledge of the approximate horizon. Several residential buildings of the city of Mohali and Chandigarh lie in the viewing direction of the MAX-DOAS instrument, and hence the first estimate of the horizon was calculated using a tall building in the field of view. The MAX-DOAS instrument is installed at an altitude of 20 m above ground level. Hence, a 40m high building at 3 km distance would correspond to an angle of 0.38°.

We realise that by mistake, in the manuscript, we write the angle of the visible horizon to be about 0.2°. We apologise for it and correct it in the revised manuscript in line 168. Please note that, even after this correction, the visible horizon and that determined from using the horizon scan are close to each other, and further correction is not required.

The reviewer is right that this is also the reason why the measurements at 1° elevation angle were not used. As can be seen from Fig. F3, the field of view (FOV) of the instrument is rather large, and typically the RMS of the spectral analysis for the measurements at 1° elevation is substantially larger

than those for the higher elevation angles. This indicates that these measurements are still affected by the reflected light from the surface. Therefore, we excluded measurements at 1° elevation angle from further processing.

We have modified lines 145-146 of the original manuscript and added lines 170-172 in the revised manuscript to include this information:

“The scattered sunlight spectra were recorded for elevation viewing angles 1°, 2°, 4°, 6°, 8°, 10°, 15°, 30° and 90° at a total integration time (number of scans × acquisition time for one scan) of 60 seconds each.”

“We also see from Fig. F3 that the field of view (FOV) of the instrument is rather large (> 0.7°), and typically the RMS of the spectral analysis for the measurements at 1° elevation is substantially larger than those for the higher elevation angles. Hence, we excluded the measurements at 1° elevation angle from further analyses.”

### 3. What was the calibration process for the instrument? Did the authors perform any spectral calibration?

The spectral calibration is performed with respect to a high resolved Fraunhofer spectrum. This information is provided in lines 157-160 of the original manuscript.

“Wavelength to pixel calibrations were performed in QDOAS software (<http://uvvis.aeronomie.be/software/QDOAS/>: last access 05.03.2020) (Danckaert et al., 2012) every time the detector temperature was changed, by matching the structures in a measured spectrum in the zenith direction at around noontime with those in a highly resolved solar spectrum”

### 4. ‘Based on the measured radiances at 360 nm, colour index (ratio of measured radiances at 330 and 390 nm) and measured O4 airmass factors (O4 SCD/ O4 VCD), we can classify the sky conditions into the following seven categories.’ The difference between the upper and lower wavelengths for the classification is small. Why have the authors have not used radiances from ends of the measured window? (Line 204-205)

In principle, a wavelength pair with a larger difference between the lower and upper wavelength could be used for the cloud classification (e.g. 320 nm/440nm) which is close to the ends of the measured window. However, the chosen wavelength pair has two advantages (Wagner et. al., 2016):

1. The absorption effect of atmospheric ozone is smaller at a longer wavelength, and hence a longer wavelength (e.g. 330 nm) is more robust to variability in ozone.
2. The variability of the surface reflectance is smaller for shorter wavelength (e.g. 390 nm) as compared to longer wavelengths (e.g. 440 nm). Hence a global threshold is more robust for 330nm /390 nm wavelength pair.
3. The signal to noise ratio of the measured spectra is rather high, while the changes of the CI caused by clouds and aerosols are rather strong. Thus, the limitation of the spectral range is not critical.

### 5. The sky conditions defined by colour indexes (CI) were not supported by any other supporting information. Were there any other methods like visual inspection, or comparison with sky images done for validation? Without validation how can the CI be used for cloud classification – especially since it is based only on radiances? What were the thresholds used for the classification – what determines a cloud hole as compared to broken clouds? Although the authors have cited some past work, that is from different parts of the world, with very different aerosol loading and SZAs – this information is missing from appendix B. (Line 206-207)

Comprehensive validation of the cloud classification scheme was performed in earlier studies (Wagner et al., 2014, 2016; Wang et al., 2015). In these publications, the detailed description for the calibration of the thresholds of the CI and their dependencies on elevation angles and time are also given, which are applied in this study.

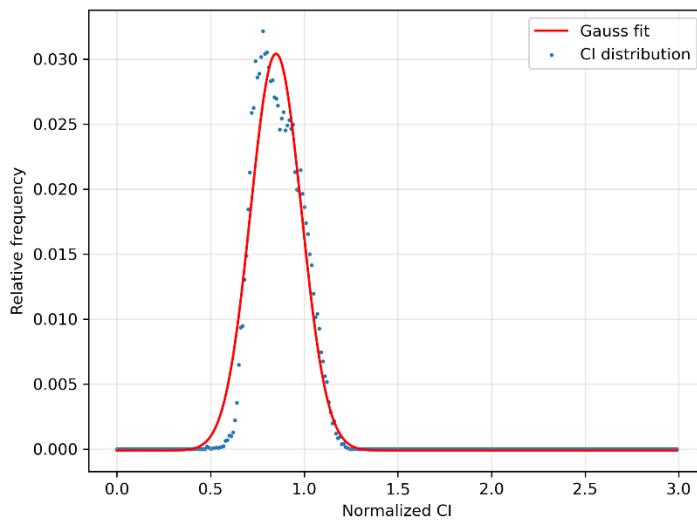
We think that the detail of the method need not be repeated in our study, because they are well documented in Wagner et. al., (2016). But we added some more information about the general idea of the algorithm (see below).

The cloud classification scheme is based on the measured radiances at 360 nm, colour index (ratio of measured radiances at 330 and 390 nm) and the measured O<sub>4</sub> airmass factors (O<sub>4</sub> SCD/ O<sub>4</sub> VCD) (for details see Wagner et al. (2016)). Besides the absolute values of these quantities, also their temporal variation and their elevation dependencies are considered. For the analyses presented in this manuscript, identification of thick clouds and fog was most important, as we retain DOAS measurements corresponding to sky condition without thick clouds and fog. The identification of thick clouds and fog are performed according to measured radiance and O<sub>4</sub> AMF, respectively. The thresholds for normalised measured radiances are calculated specifically for our site (Appendix B). The thresholds for the spread of O<sub>4</sub>, normalised CI, the spread of the CI and the temporal variation of the CI are calculated using SZA dependent polynomials provided in Wagner et al. (2016) (Table 1). We agree with the reviewer that absolute values of the radiances and CI would vary for different parts of the world depending on several factors which include aerosol conditions and spectrometer characteristics. In order to account for the spectrometer characteristics, calibration of CI is performed to get a proportionality constant  $\beta$ , which relates measured CI ( $CI_{meas}$ ) to the calculated CI ( $CI_{cal}$ ).

$$CI_{cal} = \beta \cdot CI_{meas}$$

Only after taking  $\beta$  into account, measured CI is compared to the threshold for cloud classification.

Furthermore, Wagner et al., 2016 have shown that the minimum CI varies only slightly with the atmospheric properties (e.g. AOD). Hence, we first normalise the measured radiance with respect to the corresponding simulated SZA dependent minimum CI. This generally also removes the SZA dependence of CI for SZA < 60°. In the next step, the frequency distribution of the normalised CI for SZA < 60° is plotted. Occurrence of a clear accumulation point (in our case shown in the figure below) similar to Wagner et. al., 2016 shows that CI can be used for cloud classification for our location also.



The maxima of the frequency distribution represent the inverse of  $\beta$ , which can be used to derive the calibrated CI. The calibrated CI can be directly compared to the thresholds. Please note that we use SZA dependent threshold values which also accounts for the variability in the SZA.

The procedure of CI calibration is already depicted in detail by Wagner et. al., 2016 in section 2 and we think that describing the calibration procedure again into our manuscript is beyond the scope of the article considering the reviewer's recommendation to focus more on the interpretation of results rather than technical details.

The reviewer is, of course, right, in that the aerosol type is probably different for the measurement of our study compared to other places. As a consequence, the classification of aerosols by the cloud

classification might be slightly different compared to other places. But this is not critical here, because the main aim – the cloud classification – is hardly affected by these differences.

Following the reviewer's concerns, we modify lines 211-213 of the original manuscript to provide further details about the cloud classification and add lines 228-230 in the revised manuscript to mention the effect of aerosol properties:

“The cloud classification scheme is based on the measured radiances at 360 nm, colour index (ratio of measured radiances at 330 and 390 nm) and the measured O<sub>4</sub> airmass factors (O<sub>4</sub> SCD/ O<sub>4</sub> VCD) (for details see Wagner et al. (2016)). Besides the absolute values of these quantities, also their temporal variation and their elevation dependencies are considered. The thresholds for these quantities (the spread of O<sub>4</sub>, the normalised CI, the spread of the CI and the temporal variation of the CI) are parametrized as polynomials of the SZA as provided in Wagner et al. (2016).”

“While the classification of aerosols might be slightly affected by the specific properties of the local aerosol, the cloud classification is robust to the variability of aerosol properties. However, this is not critical here, because the main aim – the cloud classification – is hardly affected by these specific aerosol properties.”

Broken clouds refer to few cloudy patches in the clear sky, while cloud holes refer to clear sky between clouds. Both cloud holes and broken clouds are detected by a rapid temporal variation of the observed for normalised CI. In the cloud classification algorithm, the normalised measured CI is smaller than the threshold CI for broken clouds, and the inverse is true for cloud holes.

#### 6. What was the threshold for RMS used for filtering the QDOAS analysis? Was there a reason for using this filter?

We thank the reviewer for raising this important question. We have filtered out the O<sub>4</sub>, NO<sub>2</sub> (UV and VIS) and HCHO dSCDs corresponding to a DOAS fit RMS greater than 0.002. Additionally, we filter out all the measurements at solar zenith angles greater than 85°. The RMS threshold was considered according to the recommendation from Wang et al., 2019 and removed most of the obvious outliers. More precisely, this threshold removes 1.1%, 1.4%, 0.7% and 1.3% of the O<sub>4</sub>, NO<sub>2</sub>(UV), NO<sub>2</sub>(VIS) and HCHO dSCDs respectively, for the elevation angles considered for our analyses.

This information was not present in the original manuscript, and we have added the following in the revised manuscript (lines 175-180 of the revised manuscript):

“The typical values (peak of the frequency distribution) of the root mean square (RMS) of the DOAS fit residuals are around  $5 \times 10^{-4}$ ,  $7 \times 10^{-4}$ ,  $6 \times 10^{-4}$ ,  $6 \times 10^{-4}$ , for O<sub>4</sub>, NO<sub>2</sub> (UV), NO<sub>2</sub> (VIS) and HCHO, respectively. In order to retain analyses results corresponding to good quality fits, we have excluded the O<sub>4</sub>, NO<sub>2</sub> and HCHO dSCDs corresponding to a RMS greater than  $2 \times 10^{-3}$  and solar zenith angles higher than 85° (Wang et al., 2019). The RMS threshold removes 1.1%, 1.4%, 0.7% and 1.3% of the O<sub>4</sub>, NO<sub>2</sub> (UV), NO<sub>2</sub> (VIS) and HCHO dSCDs, respectively, of all the measured dSCDs at solar zenith angles less than 85°.”

#### 7. ‘This value was derived as the mean of the Ångström exponent (AE) between 470- 550 nm measured by MODIS for the measurement period, where we do not observe a strong intra-annual variation (Fig. D3)’ The calculation of the AE value from MODIS poses two concerns: The wavelength range is not same and satellite instrument viewing geometry is different than ground based observation. The overpass times of the satellite will also determine what AE is measured, which will change drastically in places with high aerosol loading, or in seasons of biomass burning etc. Why not to use any ground based AOD observations if available – if not, what is the sensitivity of MAPA to the AE values used? (Line 246-247)

Unfortunately, ground-based AOD measurements are not available around the measurement site. These measurements are only available at ~ 250 km from the measurement site (Lahore and New Delhi) (lines 378-381 of the original manuscript). We had foreseen this limitation and checked the

sensitivity of MAPA to the AE values for a smaller subset of our data. We mention the inference of this sensitivity study in lines 249-253 of the original manuscript:

*“We also investigated the effect of the choice of Ångström exponent on the profile inversion for a smaller subset of our data spanning 15 days. We found that AE values of 1.25 and 1.75 (minimum 5th percentile and maximum 95th percentile in Fig. D3) resulted in same number of valid retrievals and the difference in the mean NO<sub>2</sub> VCD was less than 0.1%. The surface NO<sub>2</sub> concentration were slightly higher (4%) for AE value of 1.25 and were 3% lower for AE value of 1.75 as compared to those for an AE value of 1.54.”*

8. Were there any radiosonde or BL height measurements available? This would add to the discussion on the differences in the in situ and MAX-DOAS profiles and also to the seasonal variation.

Unfortunately, radiosonde or boundary layer height measurements are not available at Mohali. We have added discussion about boundary layer height from ERA5 data. Please see the response to reviewer #1 corresponding to the question regarding Page 14, lines 447-44 for a detailed discussion.

Chemistry:

9. ‘Fig. 11 shows the afternoon time (12:30-14:30) monthly mean HCHO/NO<sub>2</sub> ratio calculated using the MAX-DOAS observations. We observe that in winter months, mean daytime HCHO/NO<sub>2</sub> ratios between 1 and 2 are observed, which represents sensitivity towards both NO<sub>x</sub> and VOCs.’ - The HCHO/NO<sub>2</sub> ratio was calculated only for 12:30-12:30 hrs. What was the ratio during rest of the day – emissions would show a diurnal profile, affecting this ratio. (line 650-655)

The HCHO/NO<sub>2</sub> ratio provides a metric which discerns the sensitivity of ozone production towards NO<sub>x</sub> or VOC. This ratio was calculated for 12:30-14:30 hours, which is crucial for two reasons:

1. The daytime maximum of ozone is usually observed during this time window (Kumar et al., 2016).
2. OMI overpass (and also that of the recent TROPOMI instrument) usually happens in this time window. The HCHO/NO<sub>2</sub> ratio can also be calculated from the OMI data product and hence can be evaluated against similar metric calculated using ground-based observation.

We thank the reviewer for highlighting that the ratio might be affected because of emissions which vary on a diurnal scale. We now also calculated this indicator for 09:30-11:30 and 15:30-17:30 hours local time representing morning and late afternoon condition, respectively and revised Fig. 11 and the relevant discussion accordingly.

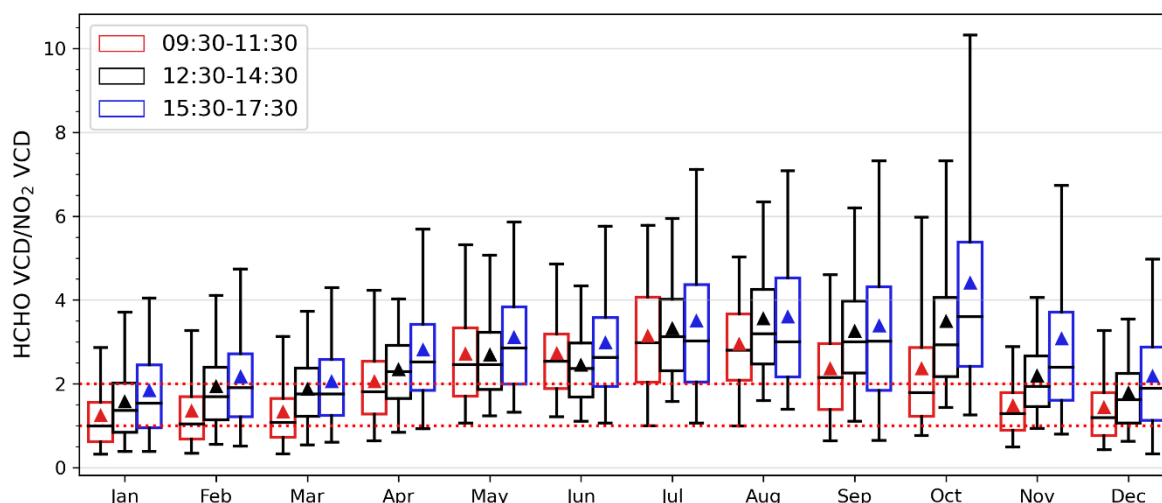


Figure 11: Monthly mean HCHO VCD/NO<sub>2</sub> VCD ratios (triangles) calculated from MAX-DOAS measurements for the morning (09:30-11:30 L.T., red), noon around the OMI overpass time (12:30-14:30 L.T., black) and late afternoon (15:30-17:30 L.T., blue) over Mohali. The lines at the centres of the boxes represent the median; the boxes show the interquartile ranges whereas the whiskers show the 5<sup>th</sup> and 95<sup>th</sup> percentile values.

We have modified lines 650-653 and 661-666 of the original manuscript to the following:

“Martin et al. (2004) recommended the use of the ratio of the formaldehyde and  $\text{NO}_2$  columns from satellite observations as an indicator for the ozone production regime.  $\text{HCHO}/\text{NO}_2$  ratios less than 1 represent a VOC sensitive regime, whereas values greater than 2 indicate a  $\text{NO}_x$  sensitive regime. Intermediate values of the  $\text{HCHO}/\text{NO}_2$  ratio indicate a strong sensitivity towards both  $\text{NO}_x$  and VOCs. The threshold for this indicator was initially calculated for afternoon time (between 13:00 – 17:00 L.T.), but was later extended to also include morning period by Schofield et al. (2006). However, Schofield et al. (2006) also indicated that the upper limit of the intermediate regime might vary spatio-temporally. Nonetheless, higher  $\text{HCHO}/\text{NO}_2$  indicate that reduction in  $\text{NO}_x$  emissions would be more effective for ozone reduction.”

“Fig. 11 shows the monthly mean  $\text{HCHO}/\text{NO}_2$  ratio calculated using the MAX-DOAS measurements for the morning (09:30-11:30 L.T.), noontime around the OMI overpass (12:30-14:30 L.T.) and late afternoon (15:30-17:30 L.T.). We observe a stronger (smaller) sensitivity towards  $\text{NO}_x$  during the late afternoon (morning) as compared to noontime similar to other urban locations in the USA (Schroeder et al., 2017). VOCs contribute to ozone production via their oxidation by OH radicals and subsequent formation of peroxy radicals. During the build-up hours of ozone (between sunrise until noontime) at Mohali, radicals' abundance is also expected to be limited. Hence, the ozone production is more sensitive to VOC (or “radicals”) during morning which shifts towards  $\text{NO}_x$  later during the day. In winter months, mean daytime  $\text{HCHO}/\text{NO}_2$  ratios between 1 and 2 are observed, which represent sensitivity towards both  $\text{NO}_x$  and VOCs. The sensitivity of the ozone production regime changes towards  $\text{NO}_x$  with the onset of summer and stays like that until the end of the post-monsoon season. Over the Indo-Gangetic plain, the strongest ozone pollution episodes are observed in the summer and post monsoon months during the afternoon hours between 12:00 and 16:00 L.T. (Kumar et al., 2016; Sinha et al., 2015). Surface ozone measurements from Mohali have shown enhancement in its ambient concentrations during the late post monsoon as compared to the early post monsoon even though the daytime temperature drops by 6 °C. During summer, enhanced precursor emission from fires lead to an increase in ~19 ppb ozone under similar meteorological conditions. Considering the stronger sensitivity of daytime ozone production towards  $\text{NO}_x$ , the ozone mitigation strategies should focus on  $\text{NO}_x$  emission reductions.”

Additionally, following the general comments by the reviewer, we have modified lines 655-658 in the original manuscript to highlight the importance of this analysis.

“Mahajan et al. (2015) evaluated the ozone production regime over India using the ratio of HCHO and  $\text{NO}_2$  VCDs observed from SCIAMACHY for the mean of years 2002-2012. Over the north-west IGP, the  $\text{HCHO}/\text{NO}_2$  was observed to be less than 1 in the winter months and between 1 and 2 in all other months. From our intercomparisons in the previous sections, we note that while the OMI  $\text{NO}_2$  VCDs are generally underestimated, the HCHO VCDs are generally well accounted for. Hence the true  $\text{HCHO}/\text{NO}_2$  will be smaller than those indicated by satellite observations, which indicates that the estimated sensitivity of the ozone production regime towards  $\text{NO}_x$  should be smaller and shifted towards VOCs. Using WRF-CMAQ model simulation at  $36 \times 36 \text{ km}^2$  resolution model over India for 2010, Sharma et al. (2016) have evaluated the ozone production to be strongly sensitive to  $\text{NO}_x$  emissions throughout the year and recommended reduction in transport emissions which account for 42% of the total  $\text{NO}_x$  emissions. However, with an increase in transport and powerplant emissions (strong  $\text{NO}_x$  sources) over India, the regimes are susceptible to shift away from  $\text{NO}_x$  limited and need to be re-evaluated.”



10. 'However, these are comparable to previous *in situ* NO<sub>2</sub> VMR measured for a period of more than one year at urban and suburban locations of India (e.g. Mohali 8.9 ppb, Pune ~9.5ppb and Kanpur 5.7ppb)(Gaur et al., 2014;Kumar et al., 2016).'What about other locations from India such as Debaje and Kakade (2009) and Beig et al. 2007. There are many other observations of NO<sub>2</sub> and HCHO from urban and suburban regions in India. Please update the citations.

We thank the reviewer for indicating the additional works. We have included several additional NO<sub>2</sub> measurements from India for comparison. The two references suggested by the reviewer, however, report the total NO<sub>x</sub> and not NO<sub>2</sub>. In order to compare these to our measurements, we have used NO<sub>2</sub>/NO<sub>x</sub> ratio of 0.9 (Kunhikrishnan et.al., 2006) to estimate mean NO<sub>2</sub> VMR.

Accordingly, lines 685-687 of the original manuscript has been modified as follows:

“However, these are comparable to previous *in situ* NO<sub>2</sub> VMR measured for a period of more than one year at urban and suburban locations (distant from traffic) of India (e.g. Mohali: 8.9 ppb, Pune: ~9.5ppb/8.7ppb and Kanpur 5.7ppb) (Gaur et al., 2014;Kumar et al., 2016;Beig et al., 2007;Debaje and Kakade, 2009), but smaller than near traffic urban measurement (e.g. New Delhi: 12.5ppb/18.6 ppb, Agra: 15-35 ppb) (Saraswati et al., 2018;Tiwari et al., 2015;Singla et al., 2011). Please note that we have used a NO<sub>2</sub>/NO<sub>x</sub> ratio of 0.9 to estimate NO<sub>2</sub> VMR for comparison with the previous measurements which reported NO<sub>x</sub> VMR and hence have a larger uncertainty (Kunhikrishnan et al., 2006).”

Concerning formaldehyde, we have found three previous ambient measurements from India, one using MAX-DOAS and other two employing offline techniques. Out of these two offline measurements, Ghosh et.al. 2015 reported mean ambient HCHO VMR of 217ppb, which is very high and does not represent ambient concentration in our opinion. In the revised manuscript, we added the following line:

“The measured HCHO VMRs are comparable to previous MAX-DOAS measurements from India (Pantnagar: 2-6 ppb), but much lower than those measured previously in India using offline techniques (e.g. North Kolkata:16 ppb, South Kolkata: 11.5ppb) (Dutta et al., 2010;Hoque et al., 2018)”

11. The explanation for higher mixing ratios in the MAX-DOAS as compared to *in situ* observations is not satisfactory. The fact that both the species show higher values for the MAX-DOAS are indicative of an instrumental bias, rather than source regions as speculated in the paper. If the authors are convinced that the power plant, or VOC degradation at higher altitudes contributes to this effect, it can be checked using air mass back trajectories.

The two important factors contributing to the higher NO<sub>2</sub> mixing ratios for MAX-DOAS as compared to *in situ* observations are:

1. The measurement location is relatively cleaner than the surroundings. In contrast to the *in situ* measurements, MAX-DOAS measurements are not only sensitive to the trace gas mixing ratios at the measurement location, but also to the trace gas mixing ratios in the viewing direction upto a distance of several km. The MAX-DOAS instrument is pointing towards the city of Chandigarh which also shows higher NO<sub>2</sub> VCD (Fig. 1.)
2. MAX-DOAS surface VMR are influenced by higher altitudes (which are more representative of the larger area with higher NO<sub>2</sub> mixing ratios). Due to the coarse vertical resolution of MAX-DOAS profiles, the MAX-DOAS surface VMRs are also influenced by the NO<sub>2</sub> at higher altitudes (e.g. that from powerplant plumes)

The Rupnagar power plant (PPI) powerplant is located ~45 km, 340 °N from the measurement site and was operational until the end of 2014.

To further confirm the possible role of PPI towards high surface VMR observed by MAX-DOAS, here we show the hexbin plot showing the frequency of the ratios of MAX-DOAS and *in situ* NO<sub>2</sub> vs *in situ* NO<sub>2</sub> VMR separately for the years 2013-2014 (left) and 2015-2017 (right). We observe that for the year 2013-2014, the ratio was > 1 for a large fraction of data.

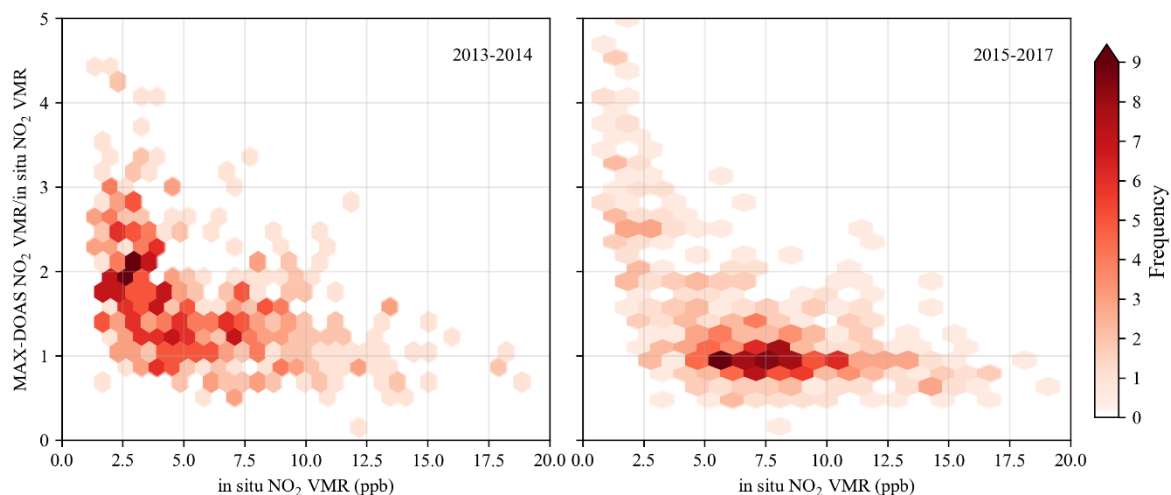


Fig: Hexbin plot showing the frequency of the ratio of MAX-DOAS and NO<sub>2</sub> surface VMRs against in situ NO<sub>2</sub> VMRs for 2013-2014 (left panel) and 2015-2017(right panel).

We acknowledge the reviewer's recommendation to also check the airmass back trajectories to confirm if plumes from PP1 approached the field of view of the MAX-DOAS. Figure 4 of Pawar et.al., (2015) have previously shown back trajectories of air masses arriving at Mohali for a period of 2 years (2011-2013). Except for monsoon, more than 80% of the back trajectories were among the clusters 'westerlies', 'local' or 'calm', all of which include the location of PP1. In monsoon, these clusters accounted for more than 50% of the total. The GDAS (global data assimilation system) meteorological inputs used for calculating the back trajectories using HYSPLIT are available at 0.5° (~56 km along latitudes) resolution in the best case. Hence, we feel that local wind vectors, measured at Mohali, would provide a more robust validation of our hypothesis than the air mass back trajectories for distances of this scale.

Hence, we show the wind rose plot for the four major seasons overlaid on mean TROPOMI NO<sub>2</sub> maps around Mohali similar to that in Figure 1 of the manuscript. We observe that in all the seasons except monsoon, the major fetch region includes PP1 (which also lies in the viewing direction of MAX-DOAS). It should be noted that the NO<sub>2</sub> maps were generated using TROPOMI measurements for the period Dec 2017-Oct 2018 and hence PP1 does not stand out as a strong NO<sub>2</sub> source

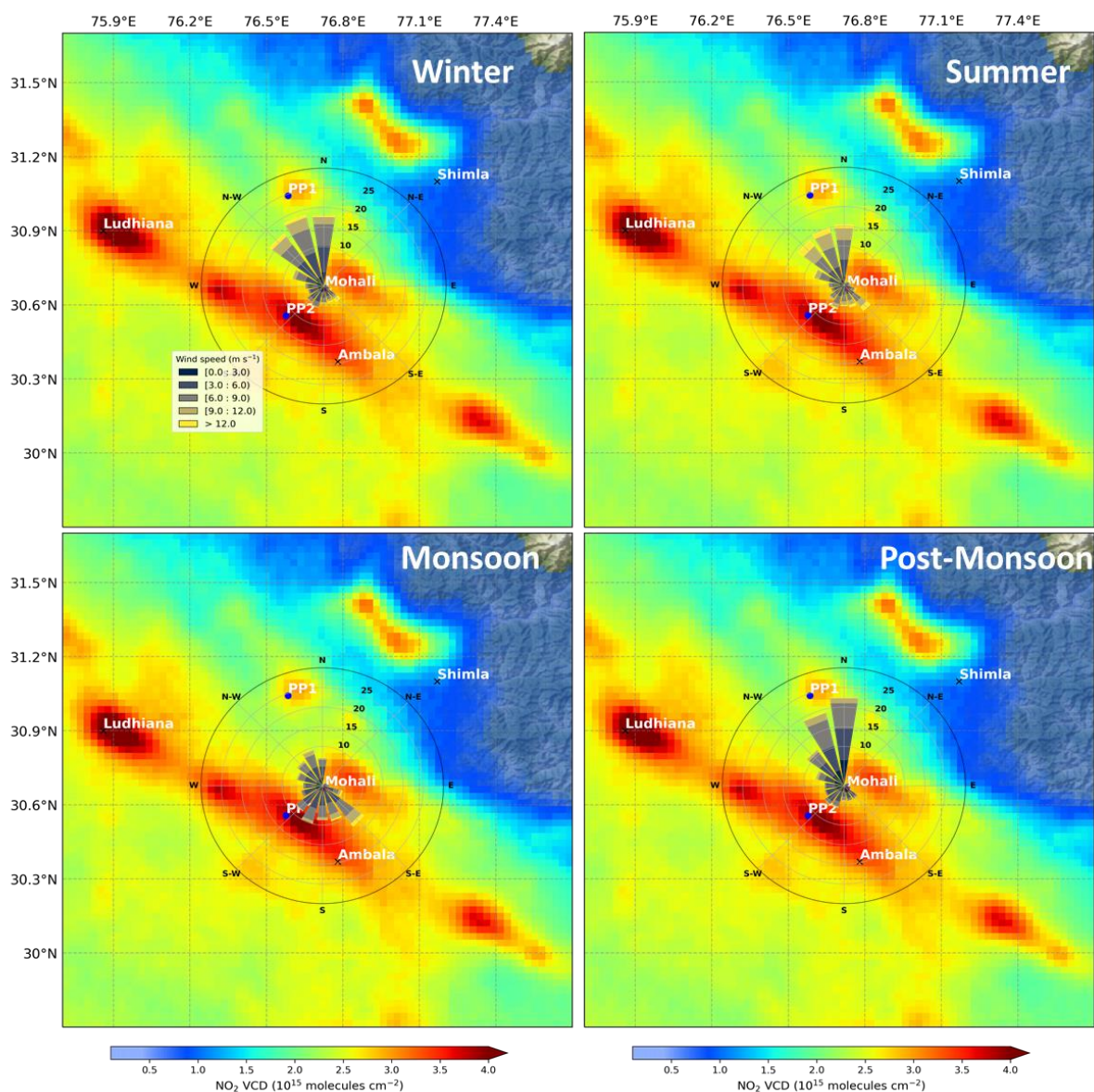


Fig: Wind rose plots overlaid on the mean TROPOMI NO<sub>2</sub> map around Mohali showing the prevalent wind speed and direction during the four major seasons of the year.

In the revised manuscript, we show the wind rose plots (not overlaid on the TROPOMI map shown in Figure 1) as Fig F1, modify lines 130-132 of the original manuscript and add the following text:

Lines 138-140

Fig. F1 shows the wind rose plots indicating the wind speed and wind direction frequencies around Mohali in the four major seasons over the measurement period.

Lines 1168-1170

Pawar et al. (2015) have previously shown back trajectories of air mass arriving at Mohali for a period of 2 years (2011-2013). Except for monsoon, more than 80% of the back trajectories were among the clusters 'westerlies', 'local' or 'calm', all of which include the location of PP1. In monsoon, these clusters accounted for more than 50% of the total. From the wind rose plot of Fig F1, we also observe that in all the seasons except monsoon, the major fetch region includes PP1.

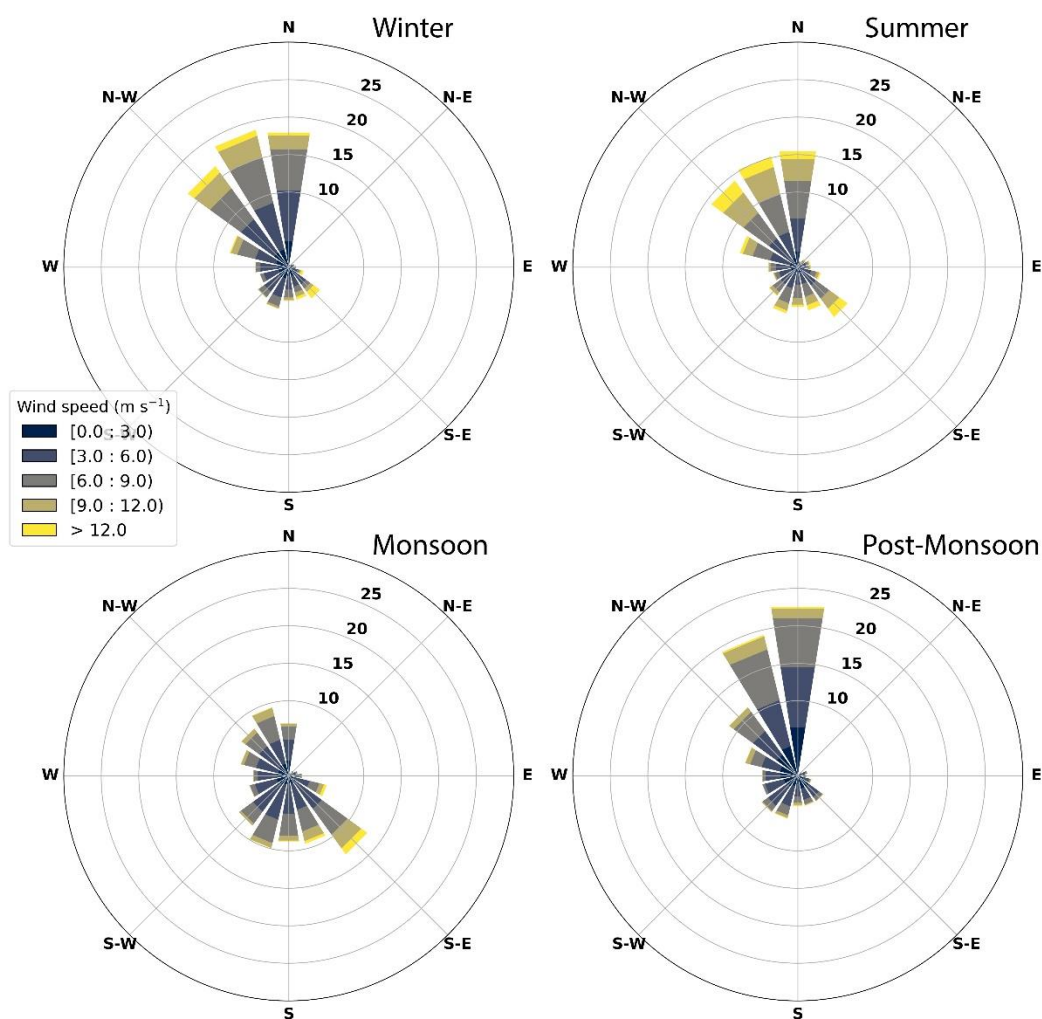


Figure F1: Wind rose plot showing the major fetch region of air mass arriving at Mohali for the four major seasons of the year

For HCHO, however, we do not speculate about any particular source region to be responsible for the observed higher MAX-DOAS surface VMRs. We propose, one plausible explanation based on the vertical profiles of HCHO (as shown in Fig 5 and also previous works e.g. Fig 8 of Kaiser et.al., 2015). HCHO is formed from its precursors (e.g. alkenes, isoprene) during the course of their vertical mixing in the boundary layer (e.g. lines 757-760 of the original manuscript). In situ measurements, which are more sensitive close to the inlet location, do not sample the HCHO which is formed at higher altitudes. As the surface VMR from MAX-DOAS are influenced by the values from higher altitudes, these are accounted for in the mean MAX-DOAS surface VMR in the lowest 200m layer.

## 12. Figures D6 and D7 need to be discussed in more details in terms of the chemistry leading to large diurnal differences between the in situ and MAX-DOAS observations.

We observe similar diurnal profiles of NO<sub>2</sub> surface VMR from in situ and MAX-DOAS measurements. The slightly higher absolute values in MAX-DOAS NO<sub>2</sub> VMR are related to the fact that the diurnal profiles are calculated by binning the raw time series data according to the hour of the day and then calculating the statistics. Hence, any bias in the raw time series data will also be propagated to the diurnal profiles. This difference is more pronounced in winter, because of shallower layer heights and the presence of NO<sub>2</sub> at altitudes higher than the inlet of in situ analyser (Fig 13c and lines (738-740 of the original manuscript). Additionally, there was a noticeable difference in the occurrence of the morning peak between the two measurements, which we have explained in lines 726-740 of the original manuscript.

For HCHO, indeed, we observe a larger difference in the diurnal patterns. However, these differences are also seen in the raw time series data (e.g. Fig 13 of the original manuscript). As the reviewer indicated, the chemistry might lead to these differences, we have mentioned the following in lines 1206-1214 of the revised manuscript:

“Secondary photochemical production is the major source of atmospheric formaldehyde. The photo-oxidation of primarily emitted VOCs occurs during the course of their mixing up in the boundary layer, and hence, a significant amount of formaldehyde is observed at altitudes up to 600 m or even higher in some cases. The surface VMRs from MAX-DOAS shown in Fig. 13 represent the mean in the lowest 200 m layer of the MAPA output, which might also be influenced by higher altitudes due to limited vertical resolution of MAX-DOAS. Surface VMRs from the PTR-MS measurements are sensitive to the inlet height (~15m). Hence, a higher VMR from MAX-DOAS measurement was expected. This is further supported by our observations in Fig. F8, where we observe that for the periods when the emissions of precursors of HCHO are higher (e.g., from crop residue fires in May, June, October and November and from burning for domestic heating in Dec. and Jan.), the bias between the MAX-DOAS and in situ VMRs is also higher.”

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