We thank the reviewer for her/his very encouraging comments regarding the significance of the submitted work and positive feedback with a strong recommendation for final publication. 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.

This paper investigates the temporal variation of the vertical distributions of aerosols, NO2, and HCHO and corresponding AOD and vertical column densities (VCDs) retrieved from MAX-DOAS observations performed at Mohali, north-west Indo Gangetic plain. The measurements presented in the study cover the period from January 2013 to June 2017. The different factors driving the seasonal and diurnal variations of the above parameters are identified and extensively discussed. The MAX-DOAS data sets are also used to validate co-located observations from the OMI and MODIS satellite instruments. This paper is well written, clearly structured, and presents very interesting results which fit well with the scope of ACP. In addition, I would like to say that for me this study is a breakthrough in the evaluation of the air quality in India using ground-based and space-borne remote sensing observations. I therefore strongly recommend the final publication of the manuscript after addressing the following comments:

We thank the reviewer for her/his encouraging feedback and highlighting the importance of our work.

Page 3, lines 97-98: It is written that the two stationary MAX-DOAS measurements over India did not report trace gas VCDs? What do they report then? Only aerosol measurements? The locations of those stationary MAX-DOAS measurements are also not clear.

The previous two stationary MAX-DOAS measurements over India were reported from Pantnagar (29.03° N, 79.47° E), (Hoque et al., 2018) and Barkachha (25.06°N, 82.59°E) (Biswas et al., 2019) respectively. We have modified the corresponding text in manuscript to make the locations of these measurements clearer (lines 65-68 of the revised manuscript):

'The few studies are limited only to four days of mobile measurement around Delhi (Shaiganfar et al., 2011) for estimation of NOx emission from Delhi and satellite validation and more recently at a suburban site Pantnagar (29.03° N, 79.47° E), (Hoque et al., 2018) and a rural site Barkachha (25.06°N, 82.59°E) in the Indo Gangetic plain (Biswas et al., 2019).'

The latter two studies focussed primarily on surface VMRs of NO_2, HCHO and CHOCHO. For example, the study from Pantnagar reports the near-surface mixing ratio of NO_2, HCHO and Glyoxal without providing the trace gas dSCDs and aerosol information. The study from Barkachha reports the dSCDs and "path average" surface concentration of NO_2 and HCHO derived from MAX-DOAS measurements at 3° elevation angle. We have modified lines 97-99 of the original manuscript to include this information (lines 101-103 of the revised manuscript).

"The two stationary MAX-DOAS measurements so far over India focussed primarily on surface volume mixing ratios (VMRs) and have not reported the VCDs of trace gases, and hence lack intercomparison with the satellite observation."

Page 5, lines 160-163: You should add in the legend of Fig D2 to which day those horizon scan results corresponds? Is the steep increase in the measured intensity always centred between 0° and 0.3° during the 4.5 years of measurements?

We have updated Fig D2 of the original manuscript (Fig. F3 of the revised manuscript) to add the day of the horizon scan measurements (17-09-2014). Concerning the second question, we would like to mention that we did not always observe a steep increase in measured intensity centred between 0° and 0.3°. Our method of horizon scans yields the most accurate results in clear sky conditions. In the presence of low lying and rapidly changing clouds, abrupt changes in intensity are observed, whereas in the presence of fog a steep increase in intensity is not observed, resulting in poor performance of this method under such conditions (Donner et al., 2020). During the 4.5 years of measurements, the 5th and 95th quantiles of the variation of horizon position determined by measured intensity variation at 404nm were -0.22° and 0.46°, respectively. Here it should be noted that part of this variability is probably related to changing weather conditions.

Page 6, line 176 (Eq. 1): How do you select SCD90? Do you use the zenith dSCD of the scan for correcting all the off-axis dSCDs, or do you interpolate the zenith dSCD at the time of the off-axis

measurements of a scan by using the zenith dSCDs just before and just after those off-axis measurements? From my experience, it can have
an impact on the resulting off-axis dSCDs, especially in the case of HCHO.

We thank the reviewer for her/his question about this important detail. We have chosen the zenith dSCDs of the off-axis measurements of a scan by interpolating the zenith spectra before and after the elevation sequence at the time of off-axis measurement. We have also added this information in the revised manuscript (lines 183-185).

"For analysing the off-axis spectra measured at time 't', we calculate the FRS at the time of the measurement by interpolating the zenith spectra measured before and after the complete measurement sequence."

Page 14, lines 447-448 and next paragraphs on page 15: are there any measurements of the boundary layer height at Mohali? It could be an added value to the discussion. If no measurements exist, maybe ECMWF Era-interim BLH could be used.

We thank the reviewer for her/his suggestion. Unfortunately, measurements of boundary layer heights are not available at Mohali. Following the reviewer's recommendation, we now use the boundary layer height from ERA5 land reanalysis data. It should be noted that there are various parametrisations for calculation of boundary layer height, and ERA5 uses the bulk Richardson method recommended by Seidel et al. (2012), which is based on datasets from Europe and the United States. In the revised Figure 6, we also show the diurnal evolution of BLH in the four major seasons, as well as the monthly variation of the afternoon time boundary layer height. Additionally, we show the diurnal evolution of the profile heights of aerosol, NO_2 and HCHO in the appendix (Fig. F10). While performing these comparisons, we realised that for HCHO, often unrealistic values at high SZA occur, which is probably related to the spectral interference with ozone absorption. Hence in the revised manuscript, we limit the analysis of the HCHO profiles results to SZA < 60°. We mention this in lines 259-262 of the revised manuscript:

"For the HCHO profile inversion, we observed unrealistic h and s at high solar zenith angles (SZA> 60°), which are probably related to spectral interferences with the ozone absorption within the DOAS analysis. Therefore, we only consider HCHO profile results for measurements with SZA less than 60°."

We have revised lines 446-450 and 461 of the original manuscript as follows to append this new information (lines 407-425 and 436-444 of the revised manuscript)

"The vertical profile of aerosol extinction is expected to be primarily driven by the boundary layer height (BLH) and to some extent, the photochemistry, which eventually drives secondary aerosol formation (Wang et al., 2019). At Mohali, the diurnal evolution of the aerosol extinction profile heights reaches its maximum during afternoon hours. In Fig. 6A, we show the typical diurnal evolution of BLH from the ERA5 reanalysis data for the four major seasons. We observe a growth of the BLH from morning until noon with a maximum at 14:00 L.T. and a subsequent decline. The maximum BLH up to 3 km is observed in summer. Shallow daytime BLH up to 1.2 km are observed in the monsoon period due to overcast sky condition, stronger wind and high surface moisture, and in winter due to low surface temperature and low surface heat flux (Sathyanadh et al., 2017). We observe that the aerosol is trapped in the bottom layers (within 400m) in winter, whereas during the afternoon hours in summer, monsoon and early post-monsoon months, aerosol extinction up to 0.2 km^{-1} is observed even at around 1.5 km altitude. Though the ERA5 BLH is shallow in monsoon, yet we observe similar aerosol profiles during that period as those during summer, which indicates that at Mohali, the vertical distribution of aerosol does not follow ERA5 BLH transition from summer to monsoon. Over India, the monsoon months are characterised by strong convective activity which can bring the surface air aloft to several km despite a shallow ERA5 BLH (Lawrence and Lelieveld, 2010). The convection is rather strong in the Himalayan foothill region (which also includes Mohali) and pumps the surface pollutants even into the UTLS (upper troposphere/lower stratosphere) (Fadnavis et al., 2015). The evidence of pollutant transport associated with deep convection is crucial for PAN formation in the UTLS, which is observed by the modelling studies over the IGP and Himalayan region. Long-lived non-methane VOCs (e.g. ethane) can be transported to the UTLS where both convective transported NO_x from the surface and exchanged from stratosphere serve as fuel for PAN formation."

"We show the diurnal evolution of characteristic profile heights (H_{75}) in Fig. F10 for the four major seasons. Fig. 6B shows the mean afternoon time characteristic profile heights (H_{75}) for aerosol, NO_2 and HCHO for different months, together with the mean ERA5 BLH. Due to their short atmospheric lifetime (< 6 hours) during daytime, H_{75} for NO_2 and HCHO are lower than those for aerosol. H_{75} for the measured species are observed to be smaller than the typical boundary layer heights. In the monsoon season, we observe H_{75} comparable to those in summer, even though the boundary layer height is shallow and comparable to that in winter. Trace gases and aerosol from the surface are lofted up due to deep convection in the monsoon leading to high H_{75}. This indicates that the vertical mixing of aerosol during the monsoon is not driven by the parameters used to calculate the ERA5 BLH, but rather follows the trend of ambient daytime temperature, which does not show such large difference between summer and monsoon (e.g. Fig. S2 of Kumar et al. (2016))."

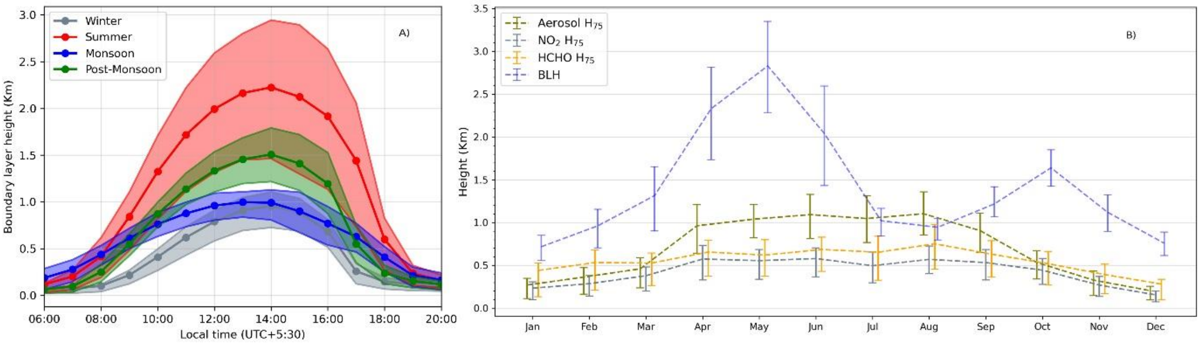


Figure 6: A) Diurnal evolution of hourly means ERA5 boundary layer height (BLH) at Mohali for the four major seasons of the year. B) Mean afternoon time (between 12:00 and 15:00 Local time) profile height (with 75% of the total amount below) for aerosols, NO_2 and HCHO and the ERA5 BLH for different months. The upper and lower vertical error bars represent the monthly variability as 75^{th} and 25^{th} percentiles, respectively.

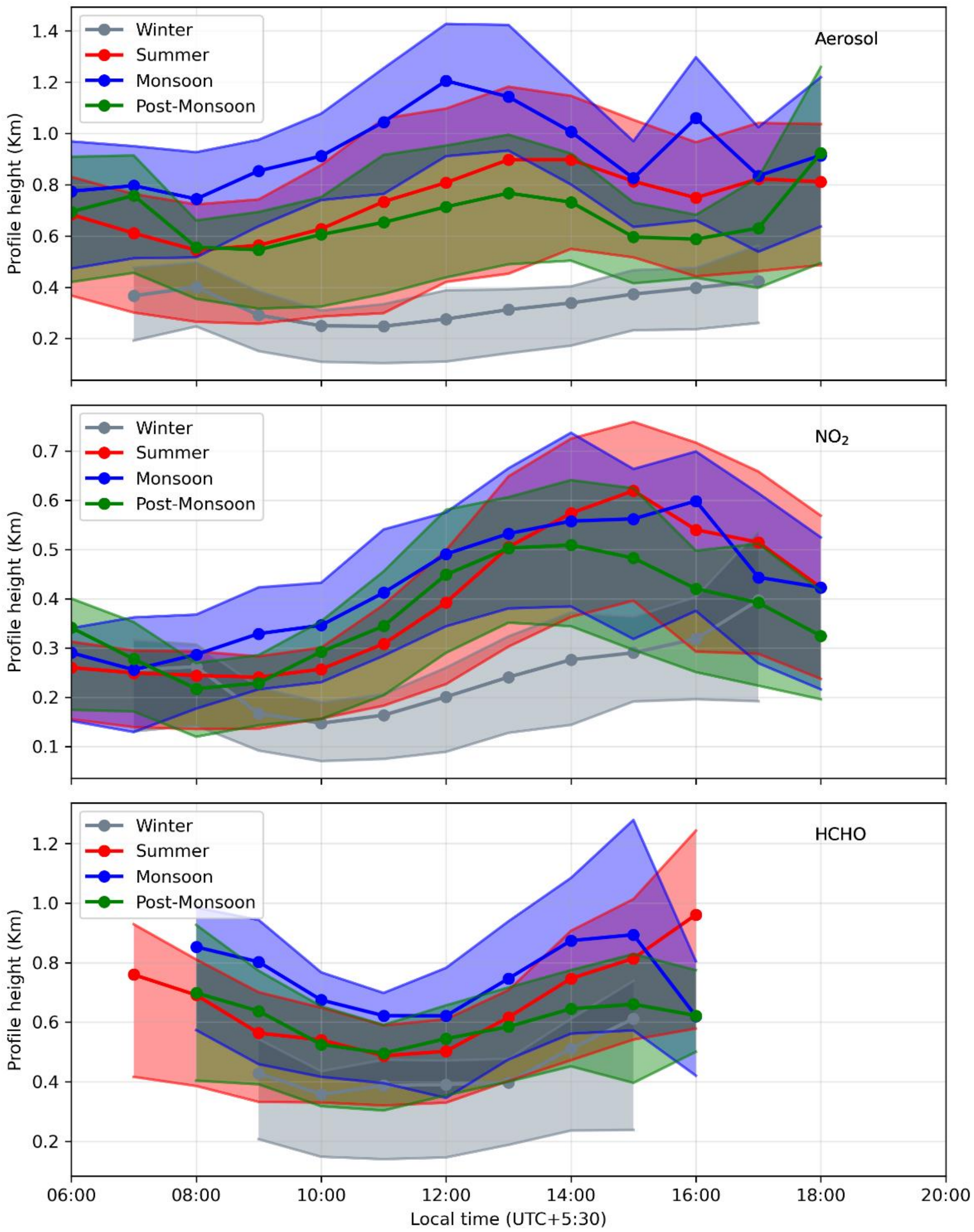


Figure F10: Diurnal variation of characteristic profile heights of aerosol (top panel), NO_2 (centre panel) and HCHO (bottom panel) for the four major seasons.

Page 18, line 562 and page 23, line 723: MAX-DOAS VCD measurements are spatially representative of a few kilometres in the field of view and this horizontal sensitivity strongly depends on the aerosol load. I think it would be useful to have to estimate this horizontal sensitivity for the main sky conditions presented in Figure 3. Numerous studies have also shown that taking into account this horizontal sensitivity in the selection of the co-located satellite data can have an impact on the agreement with ground-based MAX-DOAS observations. This is especially the case in the present study where MODIS data are given on a 1x1 km2 grid, i.e. at a horizontal resolution which is significantly higher than the typical horizontal distance of several kilometres representative of the MAX-DOAS measurements.

We thank the reviewer for her/his suggestion to estimate the horizontal sensitivity of MAX-DOAS. The horizontal sensitivity distance (HSD) of MAX-DOAS measurements can be estimated as the e-folding distance of O_4 differential box airmass factors at the location of the instrument.
For elevation angles smaller than 6°, Wagner and Beirle (2016) have introduced a 2D function of solar zenith angle and relative azimuth angle to derive an estimate of the HSD from the measured O_4 dAMF. Hence using the MAX-DOAS measured O_4 dSCD, we have calculated the HSD for clear sky cases with low aerosol (AOD < 0.85 at 330nm) and clear sky cases with high aerosol (AOD > 0.85 at 330nm) for 2° elevation angle.

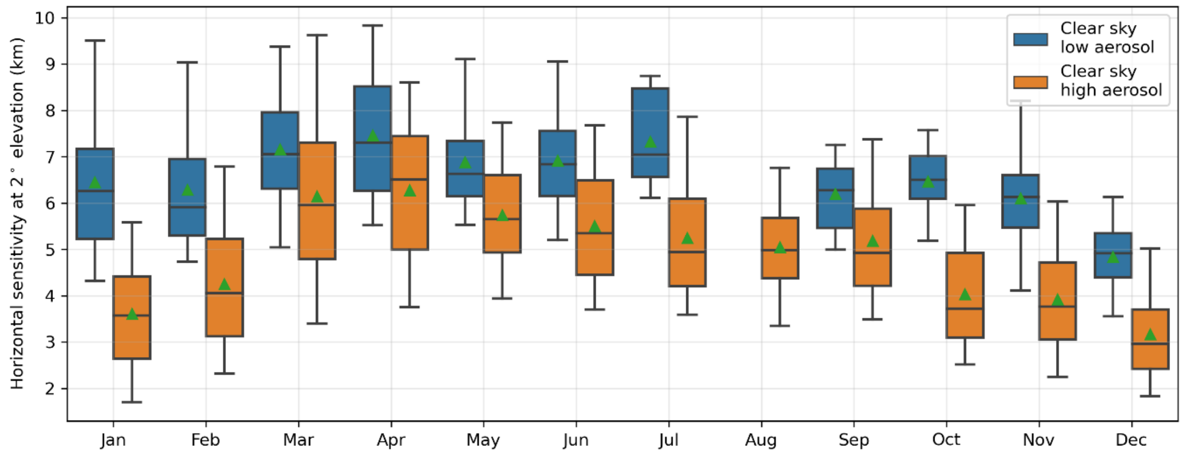
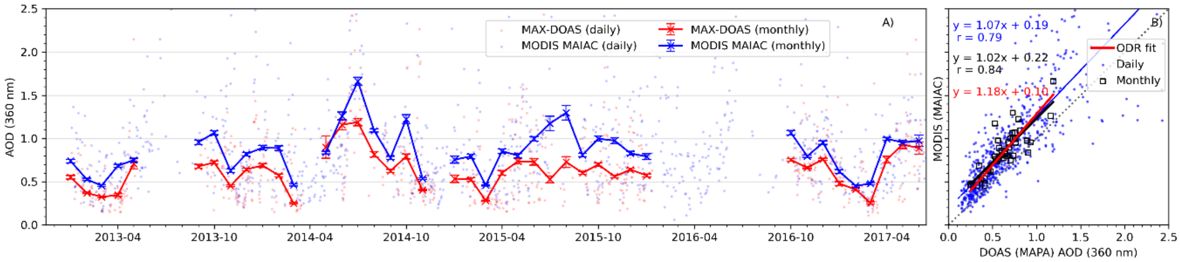


Figure F14: Box and whiskers plot showing the horizontal sensitivity distance of MAX-DOAS measurements during afternoon hours (between 12:00 and 15:00 local time) for 2° elevation angle. The blue boxes represent clear sky conditions with low aerosol load, and the orange boxes indicate clear sky conditions with high aerosol load.

We observe that for clear sky cases with high aerosol load, the mean daytime HSD is in the range 5-7 km, whereas, for high aerosol load cases, it is in the range of 3-6 km. The horizontal sensitivity is maximum in the summer months. Here it is important to note that this estimate is mainly representative for the near-surface layers.
For the comparison with the satellite data sets, the vertically integrated quantities are used. For MAX-DOAS observations, these quantities are mainly constrained by the high elevation angles (≥ 30° degree). For such high elevation angles, the sensitivity range is much closer to the instrument (at distances up to 1 and 2 km for layer height of 0.5 and 1km, respectively (see Fig 6).
Accordingly, we have modified lines 562-563 of the original manuscript as follows (lines 545-554 of the revised manuscript):

"MAX-DOAS measurements are spatially representative of a few kilometres in the field of view, depending on the ambient aerosol load and elevation angle, whereas the ground footprints of individual OMI pixels are 13×24 km^2 in the best case. We have calculated the horizontal sensitivity distance (HSD) of MAX-DOAS for low elevation angles as the e-folding distance of O_4 dAMF from the instrument location (Wagner and Beirle, 2016). Fig. F14 shows that the mean afternoon time (between 12:00 and 15:00 local time) HSD ranges between 5 and 7 km for clear sky condition with low aerosol load and between 3 and 6 km for high aerosol conditions. Here it is important to note that this estimate is mainly representative for the near-surface layers. For the comparison with the satellite data sets, the vertically integrated quantities are used. For MAX-DOAS observations, these quantities are mainly constrained by the high elevation angles (≥ 30°). For such high elevation angles, the sensitivity range is much closer to the instrument (at distances up to 1 and 2 km for layer height of 0.5 and 1 km, respectively) (see Fig 6A)."
And added Figure F14, as shown above in response:

In the original manuscript, we have retained MODIS AOD measurements within 2 km of Mohali for comparison with MAX-DOAS measurements. Incorporating the reviewer's suggestion, we performed a sensitivity study by retaining MODIS measurements within 5 km of Mohali for comparison with MAX-DOAS AOD measurements (see figure below). However, this did not bring a noticeable change in the agreement as shown in the figure below (compared to Fig 7 of the original manuscript).



Page 21, lines 646-648: Since satellite total HCHO AMFs and averaging kernels are missing in the files, you could proceed the other way round for eliminating the difference caused by the non-representative satellite a priori HCHO profiles, i.e. recalculating satellite AMFs using MAX-DOAS vertical profiles and dividing the satellite slant column densities by those new AMFs (see e.g. De Smedt et al., Atmos. Chem. Phys., 15, 12519–12545, 2015). Maybe something worth to try.

We thank the reviewer for her/his suggestion for recalculation of satellite AMFs using MAX-DOAS vertical profiles. This could easily be done if averaging kernels of MAX-DOAS measurements were known (e.g. from profile inversion using optimal estimation method). However, for parametrised profile inversions (e.g. MAPA), as used in this study, averaging kernels are not provided. We have indicated the limitation of our approach in lines 613-616 of the original manuscript.

"A different approach for improved agreement between MAX-DOAS and satellite VCDs is by using the MAX-DOAS NO_2 profiles as a priori profiles for the calculation of airmass factors for the satellite retrieval (Chan et al., 2019). However, such an approach was not possible in our study because for parameter-based profiles inversion (like MAPA), no averaging kernels are provided"

Page 50, Figure D4: How MODIS would compare to the AERONET sun photometer measurements at these two sites? Angstrom exponent could be used to convert MODIS AOD from 470 nm to 360 nm.

We observe very good agreement between AERONET and MODIS AOD measurements at Lahore for all the months of the year. For New Delhi, though the agreement is good for most of the months, a larger scatter in the data is observed due to a smaller sample size of AERONET measurements. AERONET measurements are available at 440nm, and we have used the Ångström exponent to convert MODIS AOD from 470nm to 440nm.

In the revised manuscript, we now also show the seasonal variation of MODIS AOD measurement at these two sites together with AERONET measurements in Figure D4. Additionally, we also show the scatter plot for agreement between AERONET and MODIS AOD observation. Please note that, in the revised Fig F5, now we only use the AERONET measurement between 9:30 and 11:30 and between 12:30 and 14:30 local time to ensure consistency with the MODIS overpass times.

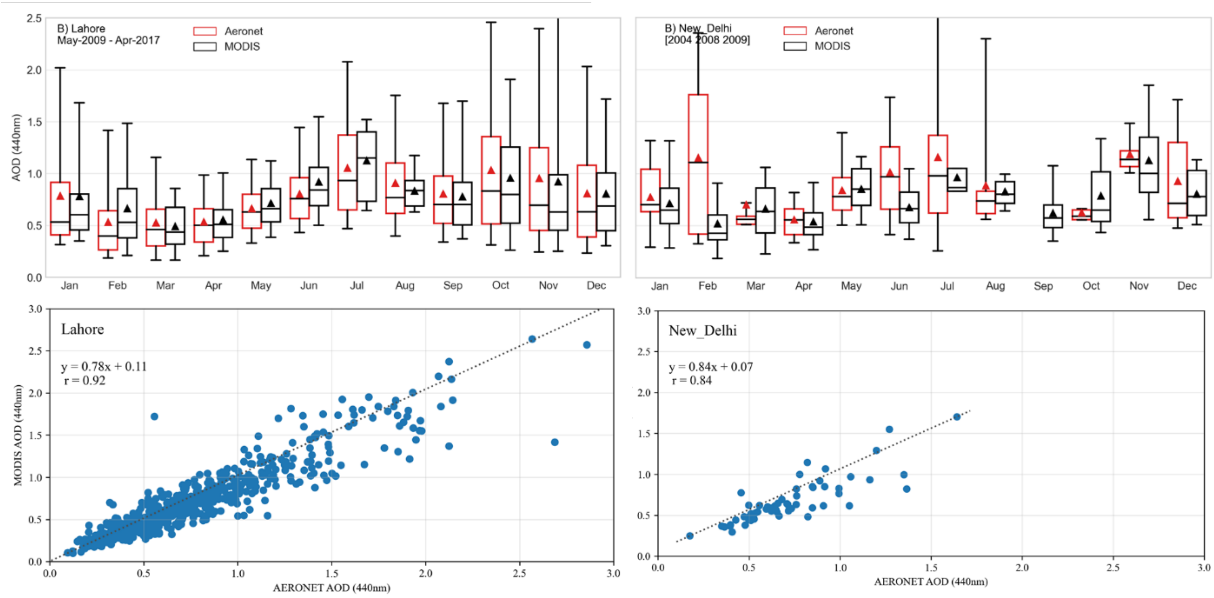


Figure F5: Monthly variation of the AOD (at 440nm) as observed by AERONET sun photometers (red boxes) and MODIS (black boxes) at two sites (A. Lahore and B. New Delhi), which are the nearest stations to Mohali in the Indo-Gangetic Plain. The bottom panel shows the corresponding scatter plots indicating the agreement in the daily MODIS and AERONET measurements.

Technical corrections:

Page 1, line 11: 'We investigate the temporal variation and the vertical profiles. . .'. I find the sentence a bit misleading since you also investigate the temporal variation of the vertical profiles (-> see Figure 5). Maybe some rephrasing is needed here.

Many thanks for this hint. We have rephrased line 11 of the original manuscript to the following (lines 11-13 of the revised manuscript):

"We investigate the temporal variation of tropospheric columns, surface volume mixing ratio (VMR) and vertical profiles of aerosols, NO_2 and HCHO and identify factors driving their ambient levels and distributions for the period from January 2013 to June 2017."

Page 2, line 59: 'ground based' -> 'ground-based'; There are some other places where this should be corrected too.

We thank the reviewer for this suggestion for consistency. We have corrected it at lines 59 and 306. At other places in the manuscript, it was written as "ground-based" in the original manuscript.

Page 5, line 138: Multi Axis Differential Optical Absorption Spectroscopy (MAX-DOAS).

Done.

Page 14, line 444: 'diurnal trends' -> 'diurnal variations' ?

Done.

Page 28, Figure 1: The names of the cities and the x and y axes labels are difficult to read. Maybe you could use a larger font size.

Done. We have increased the font size and axis label sizes from 10 to 14.

Page 37, Figure 13: in the legend, it should be 'Daily mean HCHO mixing ratios. . .' and

Thanks for spotting this. We have corrected it in the revised manuscript.

References:

Donner, S., Kuhn, J., Van Roozendael, M., Bais, A., Beirle, S., Bösch, T., Bognar, K., Bruchkouski, I., Chan, K. L., Dörner, S., Drosoglou, T., Fayt, C., Frieß, U., Hendrick, F., Hermans, C., Jin, J., Li, A., Ma, J., Peters, E., Pinardi, G., Richter, A., Schreier, S. F., Seyler, A., Strong, K., Tirpitz, J. L., Wang, Y., Xie, P., Xu, J., Zhao, X., and Wagner, T.: Evaluating different methods for elevation calibration of MAX-DOAS (Multi AXis Differential Optical Absorption Spectroscopy) instruments during the CINDI-2 campaign, Atmos. Meas. Tech., 13, 685-712, 10.5194/amt-13-685-2020, 2020.

Wagner, T. and Beirle, S.: Estimation of the horizontal sensitivity range from MAX-DOAS O4 observations, Tech. rep., QA4ECV, 2016