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Comment on acp-2022-25

Anonymous Referee #2

Referee comment on "Multi-axis differential optical absorption spectroscopy (MAX-DOAS) observations of formaldehyde and nitrogen dioxide at three sites in Asia and comparison with the global chemistry transport model CHASER" by Hossain Mohammed Syedul Hoque et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-25-RC2, 2022

The manuscript by Hoque et al. shows MAX-DOAS measurements of NO_2 and HCHO at three sites in Asia, namely Phimai (Thailand), Pantnagar (India) and China (Japan). The MAX-DOAS measurements are compared with the global chemistry model CHASER simulated concentration in the near-surface layer as well as the profiles. An attempt was made to use the ratio of Formaldehyde and NO_2 concentrations to derive ozone production sensitivity.

While I have mentioned some critical concerns about the significance of this study with respect to the Journal in my short review before the discussion phase, I provide my elaborate review here. Most likely, the short review prior to the discussion phase is not available in the interactive discussion; I append that here and expect it to be addressed.

Broadly the paper covers two separate aspects, namely MAX-DOAS measurements and comparisons with the global model. On the one hand, there are some shortcomings in both aspects of this study; I also find it difficult to motivate the readers, why such a comparison should be made in 2022, and what do we expect to learn from it. For a comprehensive evaluation of the global model, a global dataset (e.g. NDACC) should be used, which are also recently employed to evaluate TROPOMI data products (e.g. (De Smedt et al., 2021; Lerot et al., 2021; Verhoelst et al., 2021)). If the study is focused on south-east Asia, why a regional model with a better spatial resolution is not used?

Several previous studies have used high resolution (few km), global models, for comparison with MAX-DOAS measurements and emphasised the need to even go for higher spatial resolution (sub km). This study, on the other hand, presents the model results at 2.8° resolution in the base case and 1.4° in the improved resolution case, which

in my opinion, is too coarse for comparison with MAX-DOAS measurements.

Concerning the drawbacks related to MAX-DOAS retrievals, I find the vertical grid resolution (1km) too coarse, which limits the usability and interpretation of such data for air pollution-related studies. There are some technical issues related to the measurements as well, but those should be discussed in a detailed review if the editor deems the manuscript suitable for discussion in ACPD.

Detailed review:

Introduction:

- The authors motivate the readers about the current study in a way that MAX-DOAS measurements of near-surface concentrations and profiles are used to evaluate a global model CHASER (lines78-98). A study with such motivation is more suited for GMD (model evaluation papers). At least in the introduction, I could not find motivation for understating the atmospheric chemistry of the region of interest.
- Line 62 – I think it is more accurate to replace "radiation" with "radiance".
- Lines 68-78: In my opinion, MAX-DOAS is, a powerful independent technique for monitoring atmospheric constituents, and I would mention it first before stating that it is complementary to in situ and satellite measurements. Observation, dataset and methods1. In my opinion, the climate classification for Pantnagar should be done in a different way. The current classification does not consider summer as a separate season and is rather partly combined in spring and summer monsoon. The months Apr-June are extreme summer months in the Indo-Gangetic plain, with daytime temperatures above 40°C and an average temperature above 30 °C.

Observation, dataset and methods:

- Figure1: As the study focuses on the evaluation of the model over the south and east Asian region, I would recommend restricting the map boundaries to only relevant regions. The colour codes show the surface volume mixing ratios (VMR) and not concentrations. The colour bar legend should be corrected accordingly.
- lines 139-141: Campaign is used two times in the same sentence.
- MAX-DOAS system: What is the spectral range of the spectrometer used in these measurements. I am keen to know why the higher wavelength window of 460-490nm was chosen for NO_2 retrieval. The instrument used for this study participated in the CINDI and CINDI-2 campaign, and there the fit interval used for NO_2 retrieval was

425-490nm or 411-445nm.

- lines 148-149 Why would you want to minimise the variations in measured signals for various off-axis measurements. According to the DOAS principle, reference measurements should be taken at a 90° elevation angle to account for stratospheric contribution in the dSCDs. If the 90° measurements could not be taken due to any physical restrictions, this should be stated accordingly.
- How would the additional off-axis measurements at elevation angle > 10°reduce the systematic errors in the fitting results. In my opinion, measurements at some elevation angles (e.g. 15° and 30°), provide important information regarding the trace gas and aerosol profiles during inversion and should not be skipped if possible. Moreover, later in this study, the authors analyse profiles at high altitudes (> 2Km), and measurements at high elevation angles are necessary for the accuracy of such retrieval. Even the surface layer used in this study has a thickness of 1km, and measurements at high elevation angles are crucial for this layer as well.
- Lines 170-171 and Figure 2: How does the DOAS fit for O4 look like in the two wavelengths window used in this study. An intercomparison of O4 dSCDs retrieval from the two fit windows should also be shown (at least in the appendix).
- Line 187. It was difficult for me to visualise what the profile shapes look like for different values of F. It would be nice to have example plots showing the profile shapes for some values of F (similar to that shown in Fig 1 of Beirle et al., 2019 for h and s)
- It will be more accurate to save that VMRs are "calculated" using the partial VCDs rather than "converted". Though in this study, the height of the box is fixed, in general, it would be better to also mention that this conversion also considers the height of the box.
- Why the heights of the boxes are chosen to be so wide at 1km. Several studies (e.g. Kumar et al 2020) indicate a strong gradient in NO2 profiles in the lowest 1km. As the MAX-DOAS measurements in this study are used to evaluation of near surface VMRs of trace gases from the global model, higher vertical resolution in the profile retrieval should be more relevant.
- Lines 222-223: In lines 170-171, the authors state that significant O4 absorption in 460-490nm was used to retrieve the O4 ΔSCD. Then why an aerosol retrieval in the same wavelength window is not performed? Rather an Angstrom exponent was used to retrieve the AOD at 470nm.
- Lines 223-223: What is the basis of the assumption of Angstrom exponent = 1. How does the choice of Angstrom exponent affect the retrieval?
- Line 261: Please cite the latest version of CHASER and mention the model version number.
- Lines 262-264: What is the name of the chemical mechanism used for CHASER simulation?
- Please provide specific details of biomass burning emissions. Which product of ECMEF (might be GFAS?)
- Lines 278-285: Please provide an estimate of NOx and VOCs emissions from different sectors in the regions of interest. This is important to understand and confirm the important emissions sectors speculated in the subsequent sessions.
- Line 286: It would be nice to already mention here, what is the purpose of multiple CHASER simulations?

Results and discussion:

- Lines 301 and 306 (and also at several sections of the manuscript): Figure 4 shows volume mixing ratios (not concentrations).

- Figure 4: Please use the same y-axis scale for all the subplots. Also, in lines 302 and 348, it is important to mention that the standard deviations (or error bars) show the variability (not to be confused with measurement uncertainty).
- Lines 326-328: How do these mixing ratios and the seasonality compare with the other studies reported in the Indo-Gangetic plain or other sites in India) (e.g. Biswas and Mahajan 2021, AAQR, Kumar et al 2020, ACP).
- Section 3.1.3.1: In my understanding, the HCHO and NO2 indicator ratios (R_{FN}) indicator proposed by Martin et al., 2004 and Duncan et al., 2010 are based on the tropospheric vertical column densities (VCDs) and NOT concentrations. As the authors work with the MAX-DOAS system in this study, why they have chosen to calculate the ratio based on concentration and not the VCDs?
- Lines 390-391 and Figure 5: What is the person correlation coefficient of the scatter plots shown here. I wonder, how robust are the calculations drawn based on slopes of the scatter plot if the correlation is poor.
- Figure 5: Please show a similar plot colour-coded according to solar radiation (radiance at a selected wavelength). This would enable the authors to evaluate the contribution of chemistry in ozone production independent of available solar radiation.
- How do the R_{FN} values compare to previous studies (based on model, satellite and MAX-DOAS observations) in India (or Indo-Gangetic plain)?
- Line 472: I was wondering if the boundary layer height directly from the model simulations or reanalysis data products (e.g. ERA5) can be used and more suitable.
- Section 3.1.3.2: It is difficult for me to understand the need to calculate the factor "F" (column to surface conversion factor, equation 9) in the context of this study. Authors use and discuss "F" to get column integrated values (i.e. concentration). However, the MAX-DOAS retrieval also provided the vertical column densities, which is a much simpler approach.
- Lines 501ß503 It might be true that there is no relevant literature in the south and south-east Asia presenting "F" values. But there is sufficient literature discussing both the surface concentrations and the vertical column densities, from which "F" can be derived.
- Lines 511-512: Averaging kernels are highly sensitive to atmospheric conditions, and hence these should be applied to individual profiles, and the averaging should be performed rather than using an averaged averaging kernel for a season.
- Lines 515-516: How and between which parameters are the R values calculated? Are the R-Values calculated using individual measurements, daily average or seasonal mean?
- Figure 8: can the authors explain why the application of averaging kernels significantly decreases the column in Phimai, but results in an increase in Chiba?
- Lines 533, 550, 590, 652, 653, 655: The MAX-DOAS profile retrievals are performed at a vertical resolution of 1Km, and hence it is not appropriate to quantitively evaluate the model profiles at intermediate layers (e.g. 0.5 km or 200m).
- Line 546: Please provide appropriate reference justifying the model overestimation of biogenic emissions.
- Lines 554-558: If biogenic emissions are overestimated in the model (as mentioned before), I would expect a higher increase in simulated HCHO than observed between January and August.
- I am surprised to learn that emissions due to wintertime heating is not included in the anthropogenic emission inventory. From the EDGAR website (https://edgar.jrc.ec.europa.eu/dataset_htap_v2#p1), it seems that the sectors "htap_6 Residential" and "htap_3 Energy" include the wintertime heating emissions.
- Sector 3.2.3 could be merged with 3.2.1 and 3.2.2.
- Lines 678-680: It is not clear for me, why observations above 1.8km are compared with the model. Both MAX-DOAS profile retrievals and model simulation are performed above the ground level.
- Lines 685-686: This brings me to the previous comment. Why in the first place, measurements are restricted to elevation angles less than 10°.

- Lines 687-691: What is included in the whole IGP. Please show it on a map. What are the limitations of comparing the measurements at a point (representative of a few Km) to the entire IGP?.
- Figure 11: Please use the same y-axis range for subplots of HCHO and NO2.
- Line 711-712: If the biogenic emissions are overestimated, how come the simulated isoprene concentrations are reasonable?
- Figure 12 and line 735: In my opinion, it will be better to show the time series at the three stations rather than the zonal mean if the inferences are made with respect to observation at the three sites.
- Lines 738:740: 10% is the average, and based on this, one can not infer that the comparison result will improve by at least 10%.
- Line 748: How did the authors estimate that the impact of model resolution is 20%.
- Line 755: What stops the authors from using an updated emission inventory if those are already available.
- Line 782: How are the biogenic emissions optimised?
- Lines 789-791: Please provide an estimate of NOx emissions from different sectors based on the emission inventory used for the simulations.

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