Responses to the comments of Reviewer 1

We thank the referee for the helpful comments. We have revised the manuscript accordingly. The responses (blue fonts) are provided after stating the reviewer comments. Figure, Table, and line numbers correspond to the revised manuscript. The highlighted text are corresponding changes in the revised manuscript.

In this paper, formaldehyde (HCHO) and nitrogen dioxide (NO2) vertical profiles are retrieved from MAX-DOAS observations at three sites in Asia from January 2017 through December 2018. The three sites are Phimai in Thailand, Pantnagar in India, and Chiba in Japan. They correspond to rural, semi-urban, and urban conditions, respectively. The NO2 and HCHO concentrations in the 0-4km altitude range show consistent seasonal variations throughout the investigated period, which are interpreted in terms of dry and wet seasons and, in the case of Phimai and Pantnagar, biomass burning episodes. The HCHO to NO2 concentration ratios together with MAX-DOAS ozone retrieval results are also used to infer the ozone sensitivity to NOx and VOCs at the three sites. It is found that reasonable estimates of transition regions between the NOX-limited and VOC-limited ozone production regimes can be derived when the NO2-HCHO chemical feedback is accounted for.

In the second part of the study, the MAX-DOAS observations of NO2 and HCHO are used to assess the CHASER global CTM at the three sites. CHASER shows reasonably good performances in reproducing the abundances of both trace gases in Phimai and Pantnagar but not in Chiba. Comparison results are interpreted in terms of model resolution, emission inventories, and contributions of the different emission sources.

This study fits with the scope of ACP. However, there are a lot of aspects of the work that should be further clarified and/or discussed prior to final publication. Those aspects are detailed below. Moreover, as already raised during the quick review, the overall presentation quality is questionable, largely due to the poor English language used throughout the manuscript but also to repeated errors in the axes and title labeling of several figures. This should be improved in the revised version of the paper.

Response: We thank the reviewer for the comments which helped to improve the quality of the manuscript. In addition to the specific comments, the revised manuscript has been proof-read by a professional proof reader. Moreover, the figures have been improved according to the reviewer comments

Important specific comments:

*Lines 225-235: The VCD retrieval is based on several assumptions that are poorly discussed and justified. For instance, did you check that the dependence of the Abox profiles on the trace gas concentration profiles is indeed minimal? Did you test other a priori VCD values? How valid is assuming an Angstrom exponent value of 1.00?

Response: We thank the reviewer for the comments. Firstly, yes, sensitivity test assuming 30 and 50% uncertainties in the A_{box} profiles were performed and no significant changes in the results were observed. Such values (i.e., 30 and 50%) were estimated empirically from comparison with sky radiometer and LIDAR observations. Rather than the word "minimal" we use the word "low" in the revised manuscript. Because, we can't judge whether the dependence is minimal, despite optimal A_{box} profiles. Secondly, yes, we tested different a priori VCD values. Because the area in the averaging kernel was close to 1, the retrieval was almost independent of the a priori values. Thirdly, the choice of the Angstrom values had non- substantial impact on the retrieval. Uncertainty related to the Angstrom value was smaller than the uncertainties in the A_{box} profiles. The following texts are added in the revised manuscript.

L 254 – 257: The choice of the Angstrom exponent value can induce uncertainty in the retrieved VCDs. However, such uncertainty was found to be non-significant compared to that of A_{box} profiles. Uncertainty in the A_{box} profiles are assumed to as high as 30 to 50%. Such values are derived empirically from comparison with sky radiometer and LIDAR observations (i.e., Irie et al., 2008b).

*Lines 234-236: You should describe how these averaging kernels are calculated. Looking at Figure 3, HCHO and NO2 VCD averaging kernels seem to be close to unity but it is not the case for f1 averaging kernels, and especially f2 and f3 averaging kernels which are close zero. Does it mean that you can basically retrieve only the VCDs from your measurements and that for f1, f2, and f3 the retrieval essentially reproduces the a priori? Also, are similar averaging kernels obtained for the two other stations? These points should be further discussed in the revised manuscript.

Response: We thank the reviewer for the comments. Firstly, the F values determine the profile shape. For example, an a priori F1 values of 0.6 means than 60% of the aerosol/trace gas is located below 1 km. If the F value is close to 1, it means the 100% of the aerosol/trace gas is located below the specified height. Thus, even at lower F values, a realistic profile can be derived in our retrieval. The impact of uncertainties in the F values has been discussed in Irie et al. 2008. Fig S2 (supplementary information) shows retrieved aerosol profiles with different F values. Secondly, averaging kernels for the other sites are shown in Fig in the supplementary information. The following changes were included in the manuscript.

L 263 – 270: Figure 3 presents the mean averaging kernel (AK) of the HCHO and NO_2 retrievals during the dry season at Phimai. The area (Rodgers, 2000) provides an estimate of the measurement contribution to the retrieval. The total area is the sum of all the elements in the AK and weighted by the a priori error (Irie et al. 2008a). The areas for VCD and *f1* of NO_2 retrieval are 1 and 0.6, respectively. The *f2* and *f3* values are much smaller. Consequently, at first, the a priori profiles were scaled, and later f values determined the profile shape. The VCD area is close to unity, and therefore, the retrieved VCD is independent of the a priori values. Irie et al (2008) conducted sensitivity studies of choice of the *f* values and reported the effect on the retrieval negligible.

*Line 284: Anthropogenic emissions used in the CHASER model were based on the HTAP_v2.2 2008 inventory. Why didn't you use more recent inventories like the REAS v3 one (see https://acp.copernicus.org/articles/20/12761/2020/acp-20-12761-2020.pdf)? How can it affect the results and conclusions of your study, especially for Pantnagar and Chiba?

Response: We thank the reviewer for comments. We haven't yet updated the inventories due to some technical issues and currently we are trying to resolve the problem. An old inventory will have some impact on our results. We have revised the discussion on emission inventory impact in section 3.3.4. Moreover, we have provided evaluation of simulations against satellite observations in section 3.2.

*Figure 9: How do you explain such a large effect when model profiles are smoothed with the MAX-DOAS averaging kernels, especially in the altitude range (0-2km) where the MAX-DOAS retrievals have a maximum of sensitivity.

Response: We thank the reviewer for the comments. The large effect of the smoothing of the model profiles are likely related to the apriori values used for smoothing. Because the apriori data are the taken from the measured SCD and retrieved VCD values, it is sensitive in the 0-2 km layer, similar to the observations. Utilizing apriori values other than from observations will potentially impact such sensitivity. Figure 9 is Figure 11 in the revised manuscript. The following discussion has been included in the revisions:

L 846 -857 : The AKs improved the partial column and profiles significantly, reducing the MBE by more than 50%. However, the smoothed profiles and partial columns between the 0 - 2 km layer, differ significantly from the simulations, suggesting that the a priori values strongly affect the smoothed profiles. Consequently, the smoothed NO_2 profiles at Chiba (Fig. S5) are biased to the a priori values, similarly to that of at Phimai (Fig. S5). NO_2 smoothed profile sensitivity to a priori values might be attributable to our retrieval procedure. The a priori data are taken from the measured SCD and retrieved VCD values. As a result, the values are sensitive in the 0 - 2 km layer, similarly to the observations. Using a priori values other than those obtained from observations can affect such sensitivity. The smoothing sensitivity to a priori values is stronger for NO_2 than HCHO. The NO_2 profile gradient is higher than that of HCHO (Figs. 10 and 11), which means that, within 10 km (MAX-DOAS horizontal resolution), the NO_2 mixing ratio and a priori variability (sources and sinks) is higher than those of HCHO, leading to a stronger a priori effect on the smoothed profiles.

*Figure 10 and related discussion: What would be also interesting to show are model profiles at both 2.8°x2.8° and 1.4°x1.4° resolution smoothed by the MAX-DOAS AVK. I think only this comparison allows to discuss quantitatively the effect of the model resolution on the CHASER/MAX-DOAS agreement. Since the 2.8°x2.8° and 1.4°x1.4° model profiles have a significantly different shape, we can expect a different impact when those profiles are smoothed with the AVKs.

Response: We thank the reviewer for the comments. An additional with smoothed NO_2 profiles at 1.4 resolution has been included in the revised manuscript. However, due to the strong impact of the apriori values, the differences are non-significant. Figure 10 is Figure 12 in the revised manuscript. The following discussion has been included.

L 892 – 894: Figure 12(f) shows the smoothed NO_2 profiles at both resolutions. Although the profile shapes are different, the smoothed profiles are almost identical, which demonstrates that, smoothed NO_2 profile sensitivity to a priori choice is mostly independent of the model resolution.

*Section 3.2.3: Given the coarse horizontal resolution of the CHASER model (2.8°x2.8°), how valid is the assessment of NO2 and HCHO from this model for the Pantnagar station which is located in a region (Himalayan foothills) with highly varying topography? I would suggest to remove Pantnagar from the model evaluation since the topography is not properly taken into account in your analysis.

Response: We thank the reviewer for the comments. We have removed Pantnagar from the model evaluation.

Minor comments:

*Line 125: You should indicate here which types of industries are located in the Pantnagar region.

Response: We have added the following texts:

L136 – 139 : Rudrapur (~12 km south-west of Pantnagar) and Haldwani (~ 25 km north-east of Pantnagar) are the two major cities near Pantnagar, where industries (Fast moving Consumable Goods, electroplating, plywood, pharmaceuticals, automobile and allied industries (Banerjee and Srivastava 2009)) are located.

*Lines 151-152: The use of the 70°EL instead of the 90°EL for the reference spectra should be better justified. How the use of 70°EL (instead of 90°EL) can minimize variations in the measured signals. Also what do you mean by ‘variations in the measured signals’?

Response: Firstly, we used a spectrometer with a fixed integration time throughout the day. The intensity of the spectra usually depends on the elevation angle (EL) within a 15-min interval (time for a complete scan for all EL). Thus, the variation range of intensities measured at all ELs can be large, which occasionally leads to intensity saturation at the reference angle. To avoid such phenomenon, the refence measurement were conducted at 70˚ instead of 90˚. All the ELs were considered in the differential air mass factor calculation to retrieve the vertical profiles. Thus, the choice of reference EL (70˚ or 90˚) is not a critical issue. Secondly, " variation in the measured signals" has been replaced with " to avoid saturation of intensity". The changes/addition in the revised manuscript is as follows:

L163-167: The sequences of the ELs at all the sites were repeated every 15 min. The reference spectra are recorded at EL of 70° instead of 90° to avoid saturation of intensity. Because all the ELs were considered in the box air mass factor (A_{box}) calculation to retrieve the vertical profile, the choice of reference EL (70˚ or 90˚) is not an important issue for this study.

*Line 196: You should give here the AEC value at 100km you used, as well as the scaling height of your exponentially decreasing a priori profile.

Response: The following changes corresponding to this comment has been included in the revised manuscript.

L 211- 216: The AEC profile from 3 to 100 km is derived assuming a fixed value at 100 km and exponential AEC profile shape with a scaling height of ~1.6 km. The k value at 100 km was estimated from Stratospheric Aerosol and Gas Experiment III (SAGE III) aerosol data (λ=448 and 521 nm) taken at altitudes of 15–40 km. The non-substantial influence of such assumptions on the retrievals in the lower troposphere has been demonstrated in sensitivity studies reported by Irie et al (2012).

*Lines 201-203: The parameterization of Irie et al. (2008a) does not provide information on the vertical resolution and measurement sensitivity. Then it is said that 'The retrievals and simulations conducted by other groups for similar geometries (i.e., Frieß et al., 2006) are used to overcome such limitations. I don't understand this latter sentence. Do you mean that you used previous studies based on the optimal estimation method to estimate the vertical resolution and sensitivity of your own parameterized retrieval? Could you please clarify?

Response: We thank the reviewer for the comments. The limitation of our retrieval is that the vertical sensitivity can't be derived instantly. Thus, we estimate the vertical sensitivity from other studies using the similar geometry. Our retrievals using such an approach has been validated with other ground-based observations (Irie et al 2012; Irie et al. 2015). Moreover, multi-component retrievals adopting a similar approach has been reported in details by Irie et al. (2008a).

*Lines 206-208: You should describe in a table the settings (pressure and temperature profiles, wavelength, surface albedo, etc) you used for the calculation of your box air mass factors LUT.

Response: Instead of a table we have mentioned the parameters in the revised manuscript as follows:

L227-235 : Then, a lookup table (LUT) of the box air mass factor (A_{box}) vertical profile at 357 and 476 nm is constructed using the radiative transfer model JACOSPAR (Irie et al., 2015), which is based on the Monte Carlo Atmospheric Radiative Transfer Simulator (MCARaTS) (Iwabuchi, 2006). The values of the single- scattering albedo (s), asymmetry parameter (g), and surface albedo were, respectively, 0.95, 0.65 (under the Henyey-Greenstein approximation), and 0.10. The U.S. standard atmosphere temperature and pressure profiles were used for radiative transfer calculations. Uncertainty of less than 8% related to the usage of fixed values of s, g, and a were estimated from sensitivity studies (i.e., Irie et al 2012). Results obtained from JACOSPAR are validated in the study reported by Wagner et al. (2007). The optimal aerosol load and the A_{box} profiles are derived using the A_{box} LUT and the O_4 ΔSCD at all ELs.

*Lines 244-245: For the estimation of the systematic errors, uncertainties of 30% and 50% on the retrieved AOD are assumed. Where these uncertainty values come from?

Response: The uncertainties of 30 and 50% has been derived empirically from comparison with sky radiometer and LIDAR observations (Irie et al., 2008).

*Lines 246-247: Did you try to estimate the presence of an EL bias e.g. by performing horizon scans on a regular basis?

Response: We thank the reviewer for the comments. The bias in the ELs were estimated from retrievals using additional A_{box} calculations, assuming ±0.5˚ shifts in the ELs. The detail has been explained in Hoque et al (2018).

*Lines 252-254: The criteria used for the cloud screening should be justified. How do you determine them?

Response: The following text has been added corresponding to the comment.

L287-288: The threshold values were determined statistically corresponding to the mode plus one sigma (1σ) in the logarithmic histogram of relative residuals.

*Lines 289-290: Where these emission values come from? References or justification are needed here.

Response: Emission values are taken from the biogenic emission inventory VISIT used in the model. The sentence has been revised:

L326-327: Isoprene, terpene, acetone, and ONMV emissions estimates in the VISIT inventory during July were 2.14×10^{-11}, 4.43×10^{-12}, 1.60×10^{-12}, and 9.93×10^{-13} $kgCm^{-2}s^{-1}$.

*Lines 397-398: In Figure 5, only the O_3 concentrations for SZA < 50° are used to minimize stratospheric effects. Does it mean that only HCHO and NO2 data corresponding to SZA lower than 50° have been selected for these plots? If not, this means that HCHO and NO_2 retrieval results does not timely coincide with the O_3 concentrations. This point should be clarified.

Response: We thank the reviewer for the comment. Yes, SZA < 50 criterion has been applied for all the three datasets.

*Line 398: It is stated that the JM2 O3 product showed good agreement with ozonesonde measurements. Has such verification been done at the three stations involved in the present study? Also, the Irie et al. (2021) reference is missing in the list.

Response: Firstly, appropriate ozonesonde measurements are not available for the sites used in the study. Because, the retrieval settings are similar for all the sites (including that of mentioned in Irie et al . (2021)), we expect a similar quality of the retrievals. Secondly, the missing reference has been included in the reference list.

*Figure 7(a): Even if they both correspond to high O3 concentration conditions, I am surprised to see that the Rfn vertical profiles at Phimai and Pantnagar have both the same shape. Could you comment on this point? Also, why the Rfn vertical profiles from the CHASER model are not included in Figures 7(a) and (b)?

Response: We thank the reviewer for the comments. The high O_3 concentrations in Phimai and Pantnagar occurs due to biomass burning, and thus a similar R_{FN} profile is observed. The R_{FN} profiles from the CHASER model are not included because – (1) we only focused on the R_{FN} profiles obtained from the observations, and (2) to discuss the R_{FN} profiles, the O_3 simulations should be included, which is out of the scope of the current work. However, such comparisons will be discussed in our future studies.

*Section 3.2.1: I think it would be useful to show the seasonally-averaged MAX-DOAS AVK corresponding to the climate classifications of each site in the Supplement. This would support the discussion here.

Response: We thank the reviewer for the comments. We have provided the seasonal averaged AVKs for the Phimai and Chiba site in the Supplementary information (Fig S4). The discussion on the Pantnagar site has been discarded, thus, the AVKs for Pantnagar are not included.

*Figure 8: given the very large error bars on the MAX-DOAS vertical profiles, I think it is important to say that the CHASER with AK – MAX-DOAS differences are not statistically significant.

Response: We have included the following sentence.

L797 – 798 :Overall, the differences between the observations and smoothed profile are statistically insignificant.

*Section 3.2.3: Why no CHASER versus MAX-DOAS profile comparisons are shown for NO2 and HCHO for Pantnagar? This is not consistent to what is presented at the Phimai and Chiba stations.

Response: The discussion on the Pantnagar site has been discarded due to the complex topography of the site.

*Line 744: Is it 1.1° or 1.4°?

Response: It is 1.1˚ according to Sekiya et al., (2018)

Technical corrections:

*Line 24: ‘variation’ -> ‘variations’

Response: The word was corrected appropriately.

*Line 29: ‘good performances reproducing’ -> ‘good performances in reproducing’

Response: The sentence was corrected appropriately.

*Line 48; ‘the lifetime’ -> ‘the lifetime of HCHO’

Response: The phrase was corrected appropriately.

*Line 78: ‘satellite retrieval’ -> ‘satellite data retrievals’

Response: The wording has been corrected

*Lines 97-98: ‘in three atmospheric environments’ -> ‘in three different atmospheric environments’.

Response: The wording has been corrected

*Figure 1, page 6: I would use ‘concentration’ instead of ‘concentrations’ in the legend of the color bar.

Response: We have replaced concentration/concentrations to volume mixing ratio following reviewer 2’s comments.

*Line 144: ‘campaign’ -> ‘campaigns’

Response: The wording has been corrected

*Line 147: ‘consist’ -> ‘consists’

Response: The wording has been corrected

*Line 164: ‘following equation.’ -> ‘following equation:’

Response: The punctuation has been fixed

*Lines 174-175: ‘cross section data’ -> ‘cross section data sources’

*Line 181: ‘using the optimal estimation method (Irie et al., 2008a; Rogers, 2000)’ -> ‘using the approach developed by Irie at al. (2008a) which is based on the optimal estimation method (Rogers, 2000).’

Response: The sentence has been revised

*Line 182: ‘In this approach, the measurement vector y….are defined as’

Response: The sentence has been revised

*Line 188: ‘window’ -> ‘windows’

Response: The wording has been corrected

*Line 192: ‘compromise’ -> ‘includes’

Response: The wording has been corrected

*Figures 5 and 6: It is not clear to me why the y-axis scales of the three plots are not the same in both figures. Please comment. Also, to my opinion, only the transition lines should change between figures 5 and 6, so one unique figure including the three transition lines should be fine.

Response: We have merged the figures which is figure 5 in the revised manuscript. A consistent y-axis scale has been used.

*Line 458: ‘clarify’ -> ‘support’

Response: The wording has been corrected

*Page 554: ‘imitate’ -> ‘reproduce’

Response: The wording has been corrected

*Figure 9: ‘HCHO’ should be changed to ‘NO2’ in the x-axis label of all plots.

Response: Fig.9 is Fig.11 in the revised manuscript and the axis—label has been corrected.

*Figure 10(b): I guess the blue and green curves should be inverted (green curve should be in blue and the blue curve in green).

Response: Figure 10(b) has been corrected and is Fig 12 in the revised manuscript.

*Figure 11: the same x-axis scale should be used in the four plots.

Response: The comparison discussion on the Pantnagar site has been discarded. Figure 11 is Fig. 13 in the revised manuscript.

*Line 822: ‘Biogenic’ -> ‘biogenic’

Response: Appropriate corrections has been included in he revised manuscript

*Legend of Figure 14(b): ‘no anthrpogenic’ -> ‘no anthropogenic

Response: The legend has been revised. Fig. 14 is Fig 16 in the revised manuscript.
