Belmonte Rivas et al. report the global NO2 volume mixing ratio (VMR) profile climatology for cloudy scenes obtained by applying cloud-slicing technique to OMI NO2 tropospheric column and OMI O2-O2 cloud product. They maximize the number of usable OMI measurements by employing cloud radiance fraction (CRF) threshold greater than 20% for individual measurements and 50% for daily representative value for grid boxes. The authors then compare the OMI cloud-slicing profile climatology with TM4 model results, and suggest possible reasons that may have caused the apparent model shortcomings. There are several major and minor points that need to be addressed before publication in ACP.

Major comments:

1) Contribution of a priori information to the results

From the method presented in the paper, NO2 volume mixing ratio (VMR) for a pressure bin is proportional to:

$$c \cdot \mathrm{VMR}(p_{mid}) = \frac{\mathrm{VCD}_{above}(p_{dn}) - \mathrm{VCD}_{above}(p_{up})}{p_{dn} - p_{up}}$$

where p_{mid} is center of the target pressure bin, p_{dn} is lower threshold and p_{up} is the upper threshold of the bin, VCD_{above} is tropospheric NO2 VCD above a given cloud pressure level, and c is a constant.

And from Eq. (2) in the paper,

$$c \cdot \mathrm{VMR}(p_{mid}) = \frac{\frac{\mathrm{SCD}(p_{dn}) - \mathrm{SCD}_{strat} - \mathrm{SCD}_{below}(p_{dn})}{\mathrm{AMF}_{above}(p_{dn})} - \frac{\mathrm{SCD}(p_{up}) - \mathrm{SCD}_{strat} - \mathrm{SCD}_{below}(p_{up})}{\mathrm{AMF}_{above}(p_{up})}}{p_{dn} - p_{up}}.$$

Here, SCD (no subscript) is the measured NO2 slant column at a given cloudy scene. (p_{dn}) or (p_{up}) means that the measured cloud pressure is p_{dn} or p_{up} at the given scene where the SCD is measured. SCD_{strat} is stratospheric slant column (VCD_{strat} (from model) * AMF_{strat}), and is independent of the target pressure bin. We may neglect stratospheric SCD in calculating free tropospheric NO2 VMR since the stratospheric SCD and AMF_{above} do not vary much between p_{dn} and p_{up}. According to Eq. (3), SCD_{below} is the integrated model profile from the ground to the cloud pressure weighted by the scattering weight, and then multiplied by (1-CRF). Given this information, $VMR(p_{mid})$ can be expressed:

$$c \cdot VMR(p_{mid}) = \frac{\frac{SCD(p_{dn})}{AMF_{above}(p_{dn})} - \frac{SCD(p_{up})}{AMF_{above}(p_{up})}}{p_{dn} - p_{up}} - \frac{\frac{SCD_{below}(p_{dn})}{AMF_{above}(p_{dn})} - \frac{SCD_{below}(p_{up})}{AMF_{above}(p_{up})}}{p_{dn} - p_{up}}$$

$$= \frac{\frac{SCD(p_{dn})}{AMF_{above}(p_{dn})} - \frac{SCD(p_{up})}{AMF_{above}(p_{up})}}{p_{dn} - p_{up}} - \frac{\frac{(1-CRF(p_{dn}))\int_{p_{sfc}}^{p_{dn}} m(p)n(p)\,dp}{AMF_{above}(p_{dn})} - \frac{(1-CRF(p_{up}))\int_{p_{sfc}}^{p_{up}} m(p)n(p)\,dp}{AMF_{above}(p_{up})}}{p_{dn} - p_{up}}$$

where n(p) is the a priori trace gas profile from the model, m(p) is the scattering weight, and p_{sfc} is the surface pressure. Here, the first term consists of the actual contribution from NO2 between p_{dn} and p_{up}, the true information we are looking for. On the other hand, the second term consists of a priori information of below-cloud NO2 profile. If CRF and AMF are similar with respect to pressure in (p_{dn}, p_{up}) range, the second term is simply (1-CRF) * n(p_{mid}), the difference of a priori below-cloud columns for p_{dn} and p_{up} times the ratio of the clear portion to the pixel. Since the CRF threshold is not very high (20% for individual measurements and 50% for daily representative value per grid box), the retrieved VMR contains a priori information, but it is not clear exactly how much.

Based on this fact, the very good agreement between the cloud-slicing and model profiles, particularly in urban regions (the first row of the Fig. 8), is questionable. In polluted urban regions, the major contribution of tropospheric VCD is coming from the boundary layer (mostly below clouds) and thus NO2 VMR is high in the boundary layer (~ppb level) and lower troposphere while very low in middle upper troposphere (<50 pptv), and the model profiles reproduce this feature well (black lines in the first row of Fig. 8). Then how can one be sure that the “good agreement” with the model in urban profiles, particularly in lower-mid free troposphere, is not coming from the (1-CRF) * n(p_{mid}) of the model profile instead of true free tropospheric NO2 VMR?

Authors will need to examine the contribution of a priori information in the results, or should remove profiles that are highly affected by the a priori information.

2) Error discussion:

p8028, l11
The “instrumental error” discussed in this subsection is actually the retrieval error. Please refer to Rodgers (JGR 1990) for proper nomenclature.

First, the retrieval error certainly is not completely random. The error analysis assumes random errors. This should be clearly stated.

Authors “propagate” the instrumental error by assuming that the retrieval errors are random (50% for VCD and 100 hPa for cloud pressure), then compute an estimated VMR error using the summed error ratios divided by square root N (number of profiles retrieved) (Eq. 9). Please provide a reference or mathematical basis for this formulation.

The authors should compute standard error of the retrieved VMR, using the standard deviation of retrieved VMRs (for a grid box, per each pressure level for the desired time period) and dividing the standard deviation by the square root of N. This is the most direct way to obtain the standard error of the VMR, since the standard error of the mean is the standard deviation of the SAMPLE distribution divided by square root of the number of profiles (given that one profile retrieval is one sampling trial).

p8038 l9: “and scaling by the square root of the number profiles collected per grid cell”
Similar to the comment on the error discussion in p8028, putting VMR errors divided by square root N (the number of profiles in a given region) may be too optimistic.

As a result of the issues discussed above, the presented error bars in Fig. 8 and 9 may be unrealistically small. Since the cloud-slicing technique uses a very marginal variation of NO2 VCD depending on cloud pressure (which also has large uncertainties), the errors in the resulting VMRs should be fairly large for individual cases.

There may be more sources of systematic error (other than the pseudoprofile error), including but not limited to the error from uncertainties in a priori profiles and the stratospheric column.

While cloud-slicing NO2 profiles show very good agreement with model NO2 profiles, the authors make a number of statements based on the differences between NO2 profiles from cloud-slicing and TM4 model throughout Sect. 3. Error discussion is an issue in this case because some of the statements are valid only if the cloud-slicing profile errors are smaller than the difference between the profiles from cloud-slicing and the model. I suspect the errors of the cloud-slicing NO2 VMRs are greater than the error bars presented in the paper. OMI VMR errors are correlated with model errors and this needs to be discussed. The magnitude of errors needs to be carefully examined and the discussion also needs to be revised accordingly.

The section 2.1.3 is hard to follow in general. The section heading of Pseudoprofile errors doesn’t well represent the rest of the section that includes retrieval error. The subsection Pseudoprofile (systematic) error really focuses on a correction method. This section should be reorganized and rewritten for clarity.

3) p8027, l15
Authors collect OMI observations where cloud radiance fraction (CRF) > 20% (equivalent to cloud effective fraction > 10%), while using grid cell data with CRF > 50%. Cloud pressure errors need to be considered, because the error of cloud pressure is proportional to 1/CRF. Cloud radiance fraction > 50% for overall measurements would be a proper threshold for cloud slicing technique.

4) p8042, l20-21: “total tropospheric NO2 column from the cloud-slicing technique”
By nature, we can only use partial columns in cloud-slicing technique since this technique uses above-cloud columns only, i.e. from cloud pressure level to tropopause, at least for OMI NO2 column. Then what does the “total cloudy tropospheric NO2 column for OMI” used to produce

the right panels of Fig. 13 and Fig. 14 mean? If the authors separately derived “total tropospheric OMI NO2 column for cloudy condition” (other than the above-cloud column), they should state the method in the manuscript.

In addition, if “total tropospheric NO2 column from the cloud-slicing technique” for OMI is calculated in some way, the calculated “total cloudy tropospheric OMI column” includes a priori information instead of “true” information of tropospheric NO2 below clouds. Then, this comparison might not be a valid consistency check.

5) p8043 l12-18
The left panel of Fig. 14 shows that the OMI NO2 tropospheric column in clear conditions seems smaller than the model column over the northeastern US, Europe and Japan while greater over China, India, Middle East and middle Russia for the year of 2006. But it might not necessarily be caused by the NO2 long term trend, because it can result from uncertainties in the 2006 emission inventory or other inputs/dynamics in the model.

Minor comments:

Overall figures: the authors need to enlarge labels and numbers in the figures so they are readable. The figure should be understandable from the caption and this is not always the case.

p8021, l15
The paragraphs under “OMI NO2 columns” actually describe OMI NO2 and OMI O2-O2 cloud product, so an appropriate heading is needed.

p8025, l2 and throughout the manuscript
“CTP” in the equation 3 seems to mean Cloud Top Pressure according to Fig. 2. However, as explained in p8022 l7-8, the cloud pressure retrieved from O2-O2 product the cloud midlevel pressure and is different from the cloud top. Therefore, it is not appropriate to call it CTP. In addition, any acronym that is used in the manuscript needs to be explained in the manuscript, not only in the figure caption, for clarity.

p8025, l15: “Where AMF is the total airmass factor.”
In this circumstance, AMF here seems to be total tropospheric AMF for mixed cloudy scenes, which is CRF*AMF_{cloudy} + (1-CRF)*AMF_{clear}, where AMF_{cloudy} is the AMF for a fully cloudy scene with a given cloud pressure and AMF_{clear} is the AMF for a fully clear scene. Is this correct? It should be better stated in the manuscript.

p8026, l17-19: “Using OMI’s cloud information to sample the TM4 model amounts to assuming that the model is driven by the same cloud conditions observed by the instrument.”
This sentence is not clear.

p8026, l20: “but we also know that current model cloud fields are able to reproduce the average geographical and vertical distribution of observed cloud amounts reasonably well”
Authors need a proper reference for this statement.

p8032 l9: “total VCD column”
Does this mean “total tropospheric NO_2 VCD”?

p8038 l2: “15”
I see only 11 items in Table 2 and Fig 7b.

p8041 l7: “observation update”
I presume “observation update” means OMI NO_2 VMR cross sections, but it is not explained in the manuscript.