Reply to Referee #2

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.

This reviewer brings forward a number of issues (about the influence of a priori information on results, the analysis of profile errors, and the selection of CRF thresholds) that were not mentioned in the original manuscript in the interest of space. The authors are glad to clarify these topics here in the hopes of satisfying his/her concerns.

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:

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

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,

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

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}.

Note that AMF_{above} may vary appreciably with cloud pressure, particularly for low cloud levels over polluted areas. Cancelling the stratospheric contributions like the reviewer suggests may produce large errors under certain conditions, but let us continue.

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) * $n(p_{mid})$,

Note that AMF_{above} (mean sensitivity above the cloud) may differ from the mean sensitivity in the cloud (i.e., strictly between p_{dn} and p_{up}), so that approximating the second term by (1-CRF) * $n(p_{mid})$ may be perilous too, but we understand the reviewer's concerns. Please continue.

Assuming that CRFs and AMFs (above and below the cloud) were similar (which is a very strong assumption, not generally applicable but anyway, stated here as an exercise), then one would get that the vertical column between cloud levels is equal to the first term, i.e. the difference between vertical columns sensed at two cloud levels (assuming cancellation of the stratospheric component), which we could write as:

$c * VMR(p_{mid}) * (p_{dn}-p_{up}) = VCD_{dn} – VCD_{up} =$

$= VCD_{above,dn} - VCD_{above,up} + (1-CRF)*(VCD_{below,dn}- VCD_{below,up})$

Where we take roughly:

$VCD_{dn} = VCD_{above,dn}+(1\text{-}CRF)*VCD_{below,dn}$

$VCD_{up} = VCD_{above,up}+(1\text{-}CRF)*VCD_{below,up}$

Minus the second term, which is the difference between vertical columns sensed below those two cloud levels:

$(1\text{-}CRF)*(VCD_{below,dn} - VCD_{below,up})$ … proportional to $(1\text{-}CRF) * n(p_{mid})$

Which is precisely the correction that we seek to remove form the first term. The a priori information is used to correct for what contamination is expected to arise from underneath the cloud. The difference, however, lies in the fact that the a priori correction in Eq.(2) of the manuscript is applied to the tropospheric slant column SCD (not to the tropospheric vertical column VCD): in this case the contaminating term (and its correction) is preceded by a AMFbelow/AMFabove factor which is typically smaller than unity (since the scattering sensitivity *m(z)* typically decreases towards the surface). There will be contamination from the lower layers in cloud slicing, but this contamination is reduced by the scattering sensitivity profile under the cloud when dealing with SCDs.

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.

The next figure (Fig.R1) shows the magnitude of the annual mean correction SCDbelow relative to the annual mean corrected slant column (SCDabove = VCDabove * AMFabove) at 870 hPa in the CRF 50% case. Over strongly polluting urban centers, this quantity may be as high as 100%, meaning that the model based correction is allowed to remove up to 50% of the original total tropospheric slant column (SCDtrop = SCDabove + SCDbelow) observed by OMI.

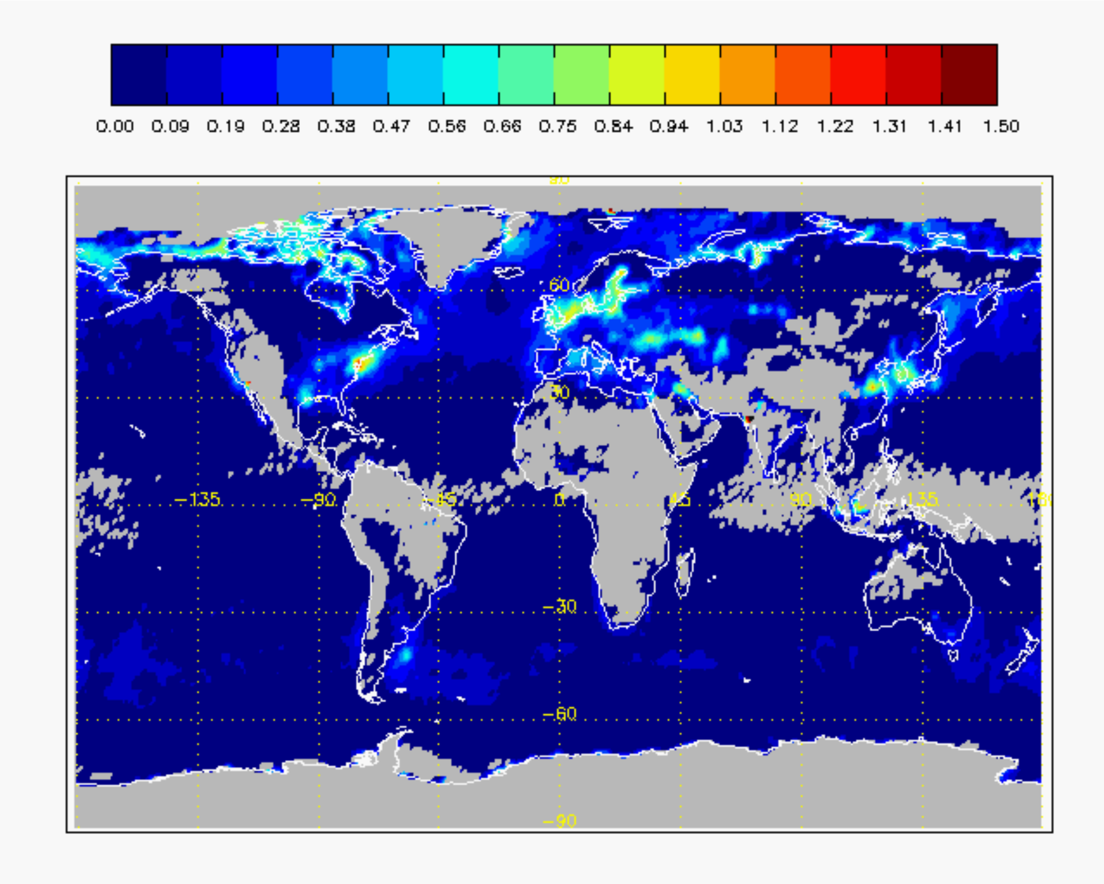


Figure R1 – Ratio between the annual mean correction (SCDbelow) for undercloud leakage and the annual mean tropospheric slant column above the cloud (SCDabove = SCDtrop - SCDbelow) at 870 hPa in the CRF 50% case.

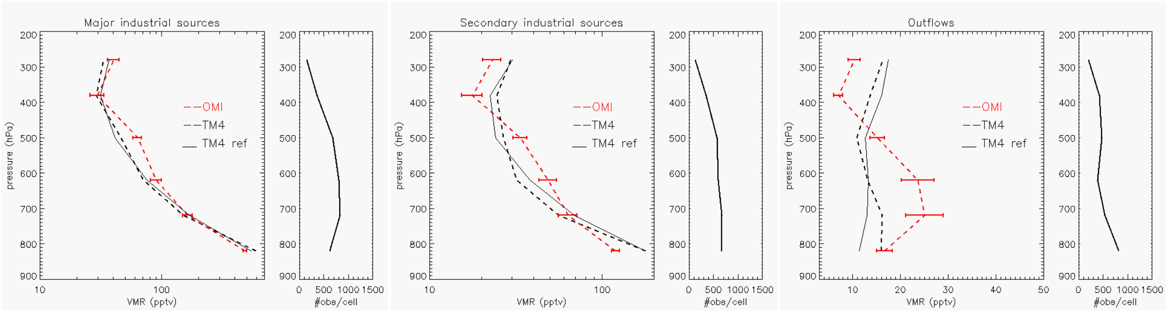
Based on this fact, the very good agreement between the cloud-slicing and model profiles, particularly in urban regions (the first row of the Fig. 8), is questionable. In polluted urban regions, the major contribution of tropospheric VCD is coming from the boundary layer (mostly below clouds) and thus 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.

The authors also had some reservations regarding the weight of a priori information in the results. To clarify this matter, we did run a separate trial increasing the cloud fraction

(CRF) threshold from 50% to 80%, whose main results we reproduce here:

CRF 50% - General



CRF 80% - General

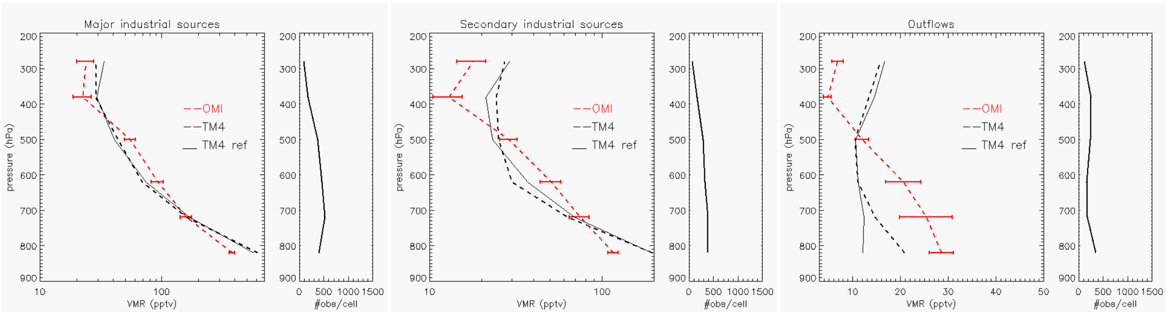
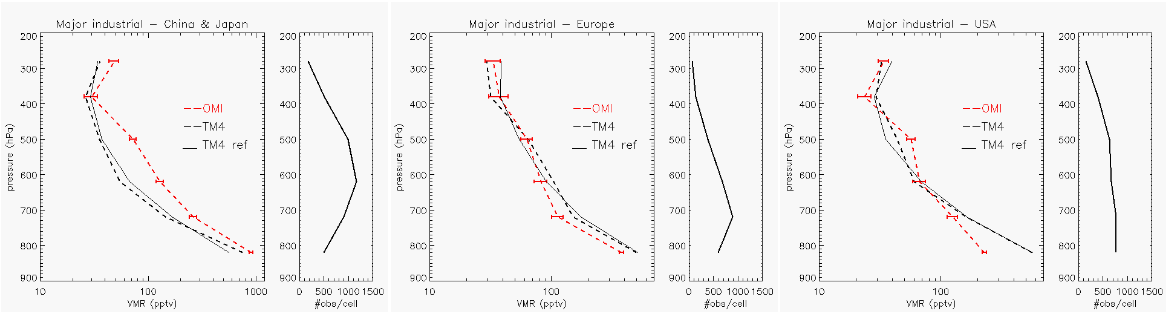


Figure R2 – Average tropospheric NO2 profiles for the year 2006: all primary sources (left), all secondary sources (middle) and all outflows (right). Identical to Fig.9 in manuscript, but for CRF 50% case in top row, and CRF 80% case in the bottom row.

All the main features at mid-tropospheric levels persist after changing the CRF threshold from 50% to 80%. The largest change consists in a general decrease of upper tropospheric NO2 amounts (280 & 380 hPa) in the 80% case, along with smaller biases at the lowest level (820 hPa) with decreases over polluted areas and increases over outflow areas. The overarching question is whether those differences are caused by the influence of a priori information, or by a change in representativity induced by selective sampling. The decreased NO2 amounts at upper tropospheric levels is clearly a sampling effect, which we attribute to a poorer capture of convective activity, which has a known preference for low cloud fractions. Screening the lower cloud fractions at upper levels is screening the very source of NO2 there (i.e. lightning events), resulting in overall lower NO2 amounts. A critical look at the geographical distributions of NO2 from OMI and the TM4 model at high altitudes (see Fig.5a) should persuade the reviewer of the lack of observation-to-model correlation in the 50% case. In our opinion, lowering the cloud fraction is not introducing any artificial observation-to-model agreement at upper tropospheric levels, but increasing the representativity of observations. We discuss the matter of the lowest level separately: let us have a look over the urban regions.

CRF 50% - Major industrial sources



CRF 80% - Major industrial sources

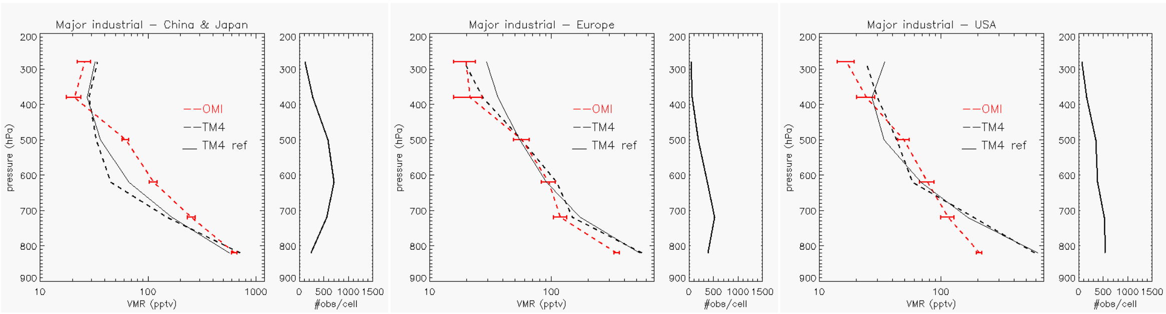
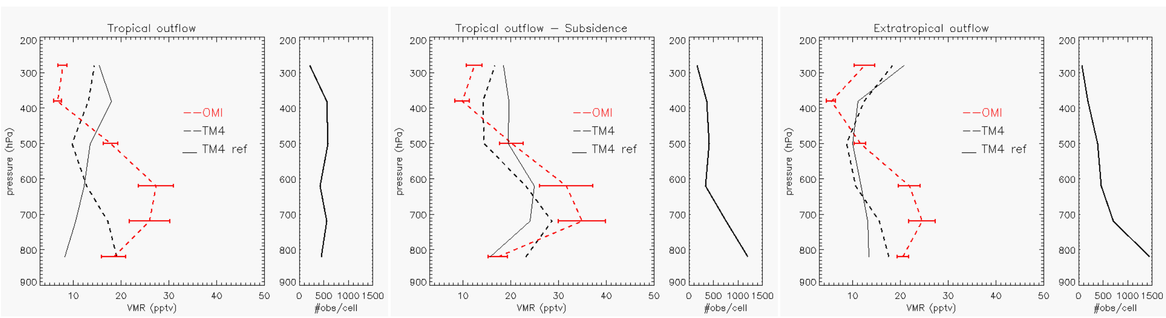


Figure R3 – Average tropospheric NO2 profiles for the year 2006: over China (left), Europe (middle) and the USA (right). Identical to first row in Fig.8 of the manuscript, but for CRF 50% case in top row, and CRF 80% case in the bottom row.

Over industrial sources, passing from 50% to 80% CRF produces a slight decrease in lowest tropospheric NO2 amounts, which does not seem to be consistently driven by a priori information. Changes in NO2 at the lowest level (820 hPa) over Europe or USA are very small. Over China, the deviation from the model increases as we lower the CRF threshold – running counter to the premise of contamination by a priori information. So over urban regions, where a priori corrections would be expected to carry more influence, we do not see any clear signs of a priori information pulling results towards the reference model. Lastly, let us have a look over the outflow regions.

CRF 50% - Outflow areas



CRF 80% - Outflow areas

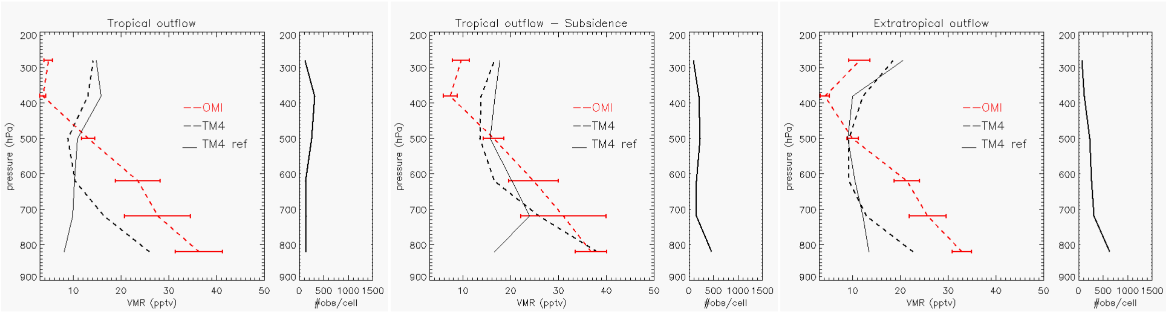


Figure R4 – Average tropospheric NO2 profiles for the year 2006: tropical outflow (left), tropical outflows over subsidence regions (middle) and extratropical outflows (right), for CRF 50% case in top row, and CRF 80% case in the bottom row.

Over the outflow regions, passing from 50 to 80% produces a general increase of NO2 amounts in the lowest level (820 hPa), sometimes away and sometimes towards the model. In this case, and unlike in any of the previous cases, changing the threshold is also changing the TM4 model pseudo-profiles, basically reflecting different sampling conditions (though leaving the pseudoprofile ratio basically unchanged). Note that model pseudoprofile errors (i.e. the difference between the black continuous and dashed lines in Fig.R4) at the lowest level are larger in the 80% case, which comes to say that the less samples, the less representative the result. In summary, we don't see any clear signs of a priori information contaminating the results, but we do see hints of results being influenced detrimentally by the lower sampling densities afforded by a higher CRF threshold. That is why we went for the CRF 50% threshold, which essentially means that at least 50% of the information contained in the radiance at grid level is coming from above the cloud.

Note that all the bias signatures observed in the CRF 80% case appear to be a consistent result of selective sampling: removing the lower cloud fractions induces negative biases at high altitude (when part of the lightning NOX production is removed), negative biases at low altitude over industrial regions (when part of the advection from boundary layer NO2 is removed) and positive biases over the outflow regions (when part of the advection from clean boundary layer air is also removed).

The zonal mean tropospheric NO2 cross sections for the CRF 50% and CRF 80% thresholds are appended next, to corroborate that changing the CRF threshold does not change the general picture appreciably.

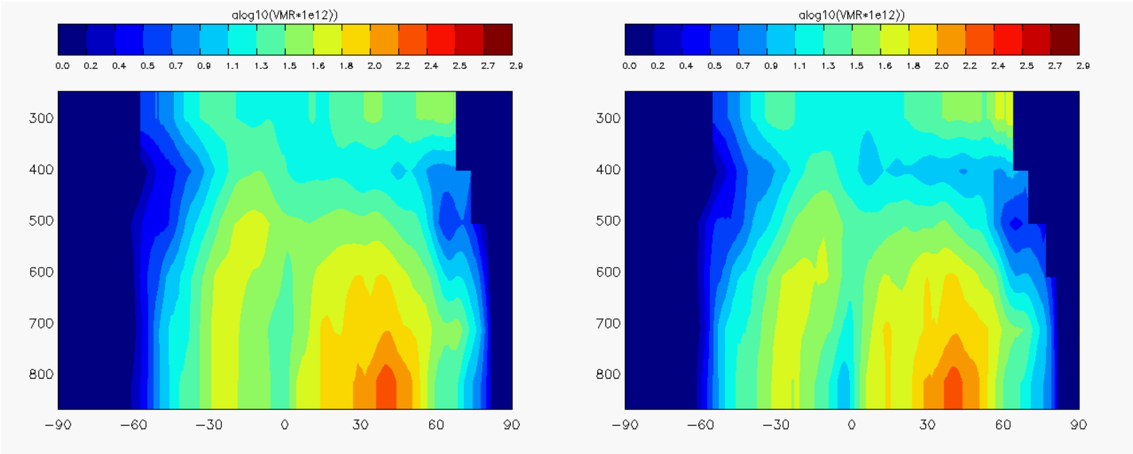


Figure R5a - CRF 50% (left) and CRF 80% (right) zonal means (as in Figure 10a of manuscript)

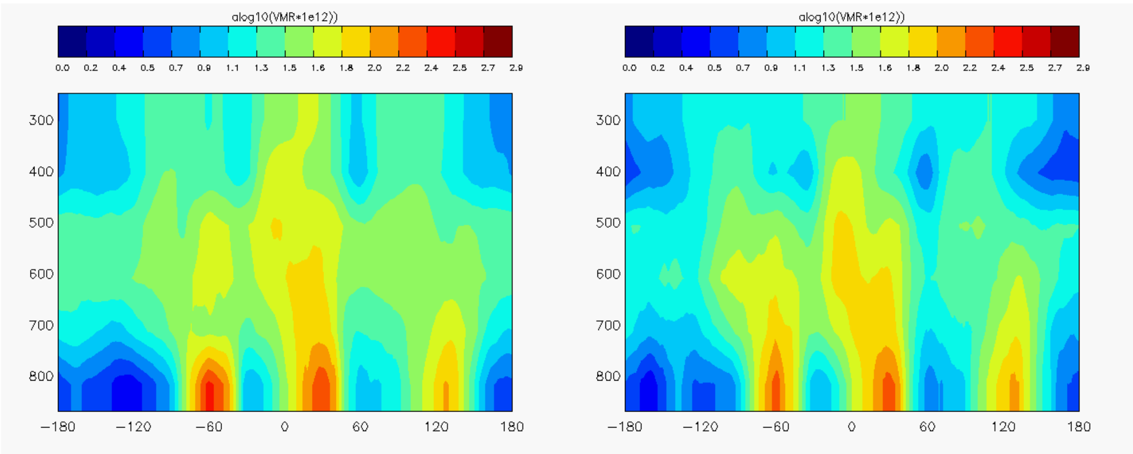


Figure R5b - CRF 50% (left) and CRF 80% (right) tropical cross-section (as in Figure 11 of manuscript)

This state of affairs was summarized in the original manuscript (first paragraph, pp 16) as:

“Results from the CRF>80% trial run include notably diminished cloud frequencies and spatial coverage, seriously thinning the population that produces the annual averages and generally damaging their representativity. This effect is particularly notable in the upper two levels (280 and 380 hPa) and to lesser extent over the large-scale subsidence area in the lowest level, since deep convective and low marine stratocumulus clouds are not particularly extensive but have a preference for low effective cloud fractions. Excluding the contributions from these cloud types in the CRF>80% case does not change the mid-tropospheric NO_2 patterns relative to the CRF>50% case, but it is biasing the OMI aggregates in the upper troposphere low relative to the modeled average, which is not particularly sensitive to this change.”

To which we add: “The CRF>80% trial run does not show any clear signs of a priori information constraining the results, but it shows hints of results being influenced detrimentally by the lower sampling densities afforded by a higher CRF threshold.”

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.

OK. Instrumental error will be referred as to retrieval error in the manuscript.

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

OK. Retrieval errors are assumed Gaussian.

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.

Please see below.

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).

Please note that we do not compute VMRs on daily or orbital basis (since one does not achieve the necessary cloud height diversity in 2x2 degree cells but in exceptional circumstances), but from the difference of annual mean VCDs. There is not a collection of VMRs per grid cell that we can use as “sample distribution” but one mean annual VMR computed from the pressure derivative of one mean annual VCD. The derivation of Eq.9

follows:

$$VMR = factor \cdot (VCD1 - VCD2)/(p1 - p2)$$

Where VCD1, VCD2, p1 and p2 are all mean annual quantities. The error propagation:

$$\delta VMR = factor \cdot \left(\frac{\delta(VCD1 - VCD2)}{p1 - p2} + \frac{(VCD1 - VCD2)}{(p1 - p2)^2} \delta(p1 - p2) \right)$$

$$\delta VMR = factor \cdot \left(2 \cdot \delta(VCD)/(p1 - p2) + \frac{(VCD1 - VCD2)}{(p1 - p2)^2} \cdot 2 \cdot \delta(p) \right)$$

Which is identical to Eq.(9) after taking into account that:

$$\delta(VCD_{annual}) = 0.5 \cdot VCD/\sqrt{N}$$

$$\delta(p_{annual}) = 100\ hPa\ /\sqrt{N}$$

That is, the standard error of the mean annual VCD is the standard error of the single VCD retrieval (assumed 50% for the OMI vertical column density, and 100 hPa for the O2-O2 cloud pressure) divided by the square root of the number of retrievals *N* per cell per year:

$$\delta VMR = factor \cdot \left(VCD/(p1 - p2) + \frac{(VCD1 - VCD2)}{(p1 - p2)^2} \cdot 2 \cdot 100\ hPa \right) /\sqrt{N}$$

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.

Our approach does not use single VMR or cloud pressures instances. We use the pressure derivative of the annual mean VCD along with the annual mean cloud pressure instead.

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 authors agree with the reviewer that error analysis is an issue. The retrieval error bars are indicative of what the instrumental/retrieval precision for single columns is relative to the resulting pseudoprofile error, suggesting that systematic errors dominate due to the sparse sampling nature of the cloud slicing technique. Other sources of systematic error may also intervene, as the reviewer points out, including uncertainties in a priori corrections and errors in the stratospheric column. The effect of uncertainties in the a priori corrections is difficult to estimate, since we take the model that performs the corrections as reference as well, lacking a better ground truth, although the CRF 80% trial run demonstrates that their effect is not appreciable (and certainly not as large as sampling related errors). The effect of errors in the OMI stratospheric column are expected to be small, since stratospheric columns only show a small additive bias (Belmonte Rivas et al., "Intercomparison of daytime stratospheric columns", AMT, 2014) that is bound to cancel via the pressure derivative. One could also include errors from the collocation of model and OMI clouds in this category, which was also mentioned earlier in the manuscript – these errors refer to the fact that we assume that cloud altitudes and fractions in the model are identical to those observed by OMI, which is not entirely correct – but we have no means to estimate its magnitude, safe for assuming that they are small in a statistic sense. Lacking any external validation means, all we can do is describe the nature of these errors, how to bypass them when possible, and expect that the final picture afforded by observations is solid and convincing enough to motivate further studies and a global validation campaign. We do make a number of statements based on the comparison of observations against the TM4 model, but we are aware that they remain on the level of plausible until cloud-slicing profiles are validated. All in all, section 2.1.3 on profile errors in the manuscript is revised to include these comments.

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.

Agreed. Section 2.1.3 as been rewritten and reorganized as outlined above, also following commentary from Reviewer #3. The section title is changed from “Pseudoprofile errors” to “Error analysis”.

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.

As a pre-processing step, we use the CRF 20% threshold when collecting observations into grid cells to ensure that all bins are as densely populated as possible (thus avoiding spatial representation issues with the lower resolution model, whose cells cannot discriminate between low and high cloud fractions). Then a final CRF 50% threshold is applied at grid level (both to model and observation cells), to ensure that only those cells whose aggregated or mean CRF is above 50% are included in the analysis. Thus the final or effective CRF threshold is 50%. The aggregated or mean cloud pressure in the cell may be less accurate when including lower CRFs in the cell, but the alternative, i.e. raising the CRF threshold for observations that go into the cell appears to be biasing the sample distribution of the observation cell relative to that in the model. We found that applying an overall 50% threshold before gridding was screening many of the convective events at high altitude (which have a known preference for low cloud fractions), negatively biasing the upper tropospheric NO2 amounts relative to the model and deteriorating the overall representativity of the observation cell. So that option was discarded.

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?

The total tropospheric NO2 column (TTC_{NO2}) from the cloud-slicing technique is calculated as the sum of partial vertical column densities across cloud height layers using the annual mean VMR pseudo-profiles as:

TTC_{NO2} (lat,lon) = $SUM_{i=1, ..., 6}$\{ VMR_i (lat,lon) * ($<p_{i+1}>$ - $<p_i>$) / C\}

Where C is the same constant defined in Eq.8. Absent VMR grid values (like at high altitudes over tropical subsidence regions or low over the African continent) are ignored without provision of any new a priori information. The manuscript is revised to include the expression above.

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.

No.

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.

The total tropospheric NO2 column from cloud slicing (i.e. the total cloudy tropospheric column) is calculated as indicated above, without provision of any a priori information – other than that used to perform the undercloud leak corrections when forming the VMR pseudoprofiles.

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.

That is the point. The emission inventory in this CTM is prescribed by the POET database, which is typical of the years 1990-1995. So the anomaly (clear sky tropospheric OMI to TM4 in 2006) indicates that the inventory is outdated, indirectly reflecting changes that over time are consistent with known NO2 long-term trends. Since the anomaly is most notable at the level closest to the surface over urban centers, we consider the effects of other inputs/dynamics as secondary.

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.

Captions and figure labels have been revised, one by one, for readability and clarity.

p8021, l15

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

The heading is changed from "OMI NO2 columns" to "OMI NO2 products".

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.

Agreed. The term Cloud Top Pressure and its acronym CTP are changed into CLP for Cloud Level Pressure where relevant across the manuscript.

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.

AMF is the total airmass factor (first variable in the Temis NO2 data field) used to compute VCD = SCD/AMF. It is different from the tropospheric airmass factor (fourth variable in the Temis NO2 data field) used to compute VCD_{trop} = (SCD - SCD_{strat})/AMF_{trop}. The reference is in Boersma et al., "Dutch OMI NO2 (DOMINO) data product v2.0: HE5 data file user manual". A clarifying note is inserted 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.

The sentence is rephrased into “Using OMI’s cloud information to sample the TM4 model amounts to assuming that cloud altitudes and fractions in the model are identical to those observed by OMI.”

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.

The TM4 model uses cloud fields interpolated from the ECMWF model. An analysis of the model cloud fields from ECHAM5 (branched from an earlier version of the ECMWF general circulation model) against CALIPSO and CloudSAT data attests to our statement (see reference to [Nam et, al, 2014] in the manuscript). Also, a new reference to:

Boersma, K.F., Vinken, G.C.M., and Eskes, H.J.: Representativeness errors in comparing chemistry transport and chemistry climate models with satellite UV/Vis tropospheric column retrievals, Geoscientific Model Development Discussions, in press, gmd-2015-134, 2015.

Has also been inserted, which includes an explicit comparison between OMI and TM5 cloud fields.

p8032 l9: “total VCD column”
Does this mean “total tropospheric NO2 VCD”?

Yes. Corrected.

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

The sentence is rephrased into: “for all the 11 classes (15 classes when the primary and secondary industrial regions are subdivided into China, USA, Europa subclasses) defined in Table 2 and Fig. 7b are shown next “

p8041 l7: “observation update”

I presume “observation update” means OMI NO2 VMR cross sections, but it is not explained in the manuscript.

Agreed. In the manuscript: “Note that in order to bypass pseudoprofile errors, the observed NO2 pseudoprofiles are scaled in this section by the model profile-to-pseudoprofile ratio as in Eq. (13).” The sentence is appended with: “… forming what is called the observation update”.