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Supplement of

OMI tropospheric NO_2 profiles from cloud slicing: constraints on surface emissions, convective transport and lightning NO_x

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On the influence of a priori information

A *priori* information from a Chemical Transport Model (CTM) is used in our cloud-slicing approach to estimate and remove the NO_2 contributions to the tropospheric slant column SCD_{trop} that come from underneath the cloud. The correction is articulated in the manuscript as:

$$VCD_{above} = (SCD_{trop} - SCD_{below})/AMF_{above} \quad \text{(S1)}$$

$$SCD_{below} = (1 - CRF) \cdot \sum_{ground}^{CLP} m_{clear}(p) \cdot n(p) \cdot T_{corr}(p) \quad \text{(S2)}$$

Where SCD_{below} is the correction term based on a priori information. Note that the correction is applied to the slant column – not the vertical column - which is advantageous in terms of the size of the correction, knowing that the scattering sensitivity typically decreases towards the surface. Also note that the size of the correction term is controlled by the Cloud Radiance Fraction (CRF) threshold: the lower the cloud fraction allowed, the larger the contribution from underneath the cloud that will need to be corrected for.

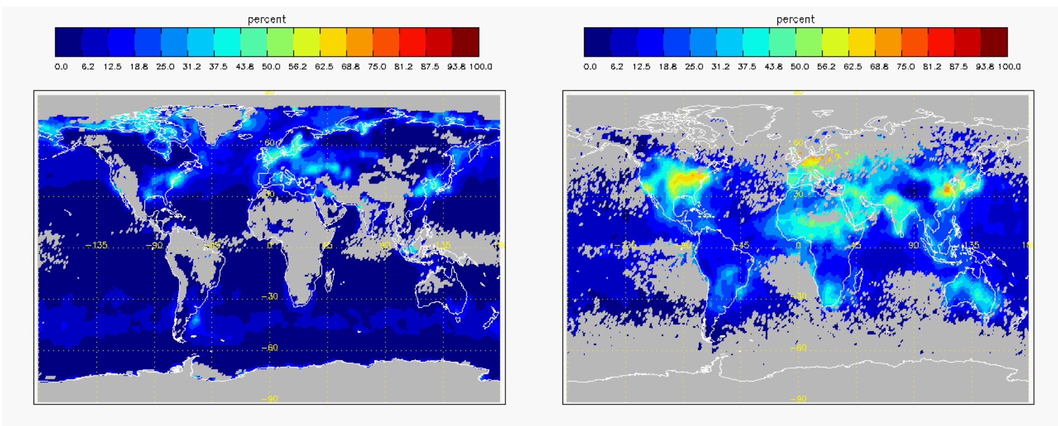
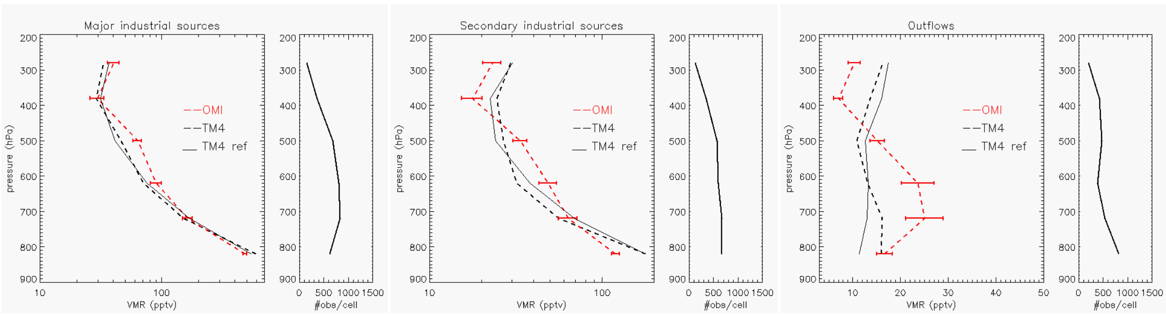


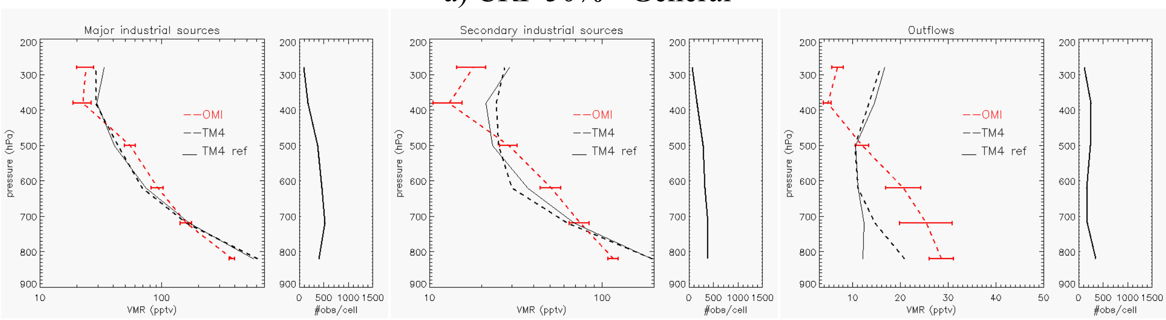
Figure S1 – SCD_{below}/SCD_{trop} ratio for low clouds (left, 870 hPa) and high clouds (right, 280 hPa) for the CRF 50% case According to the TM4 model.

Figure S1 shows the magnitude of the annual mean correction term SCD_{below} relative to the annual mean tropospheric slant column SCD_{trop} for the lowest and highest cloud levels (at 870 hPa and 280 hPa respectively) in the CRF 50% case. Over strongly polluting centers, where the largest contribution to the tropospheric vertical column comes from below the cloud and which constitute the worst case scenarios, this quantity may be as high as 50% for low clouds (820 to 500 hPa), reaching up to 66% for high clouds. Note that the relative a priori contribution increases with cloud height, as the size of the column under the cloud increases relative to the total column. So it is the upper tropospheric NO_2 estimates over strongly polluted areas that become the most influenced by a priori information (up to 66% relative to the information carried by the OMI observation, worst case).

To further clarify the influence of priori information on the resulting cloud-slicing profiles, we completed a separate trial run where the CRF threshold was raised from 50% to 80%, whose main results are reproduced in Figures S2-S4.

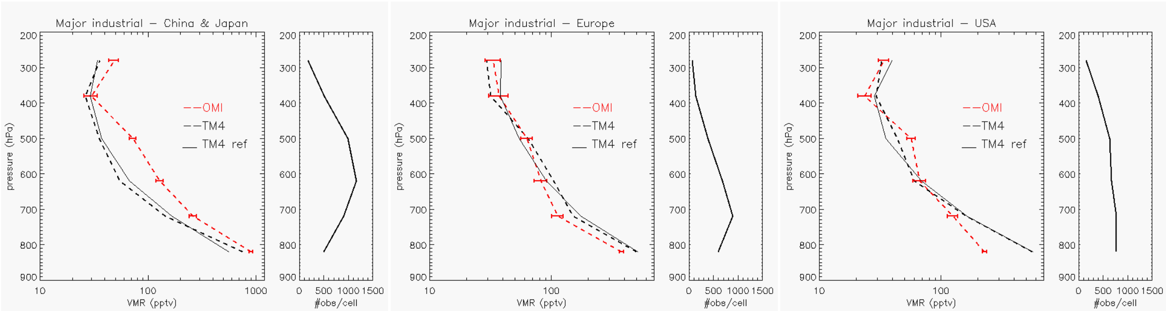


a) CRF 50% - General

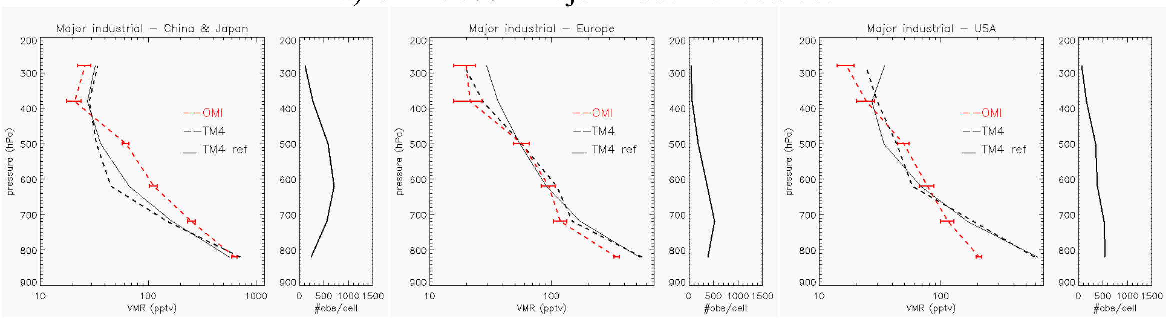


b) CRF 80% - General

Figure S2 – Average tropospheric NO_2 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.

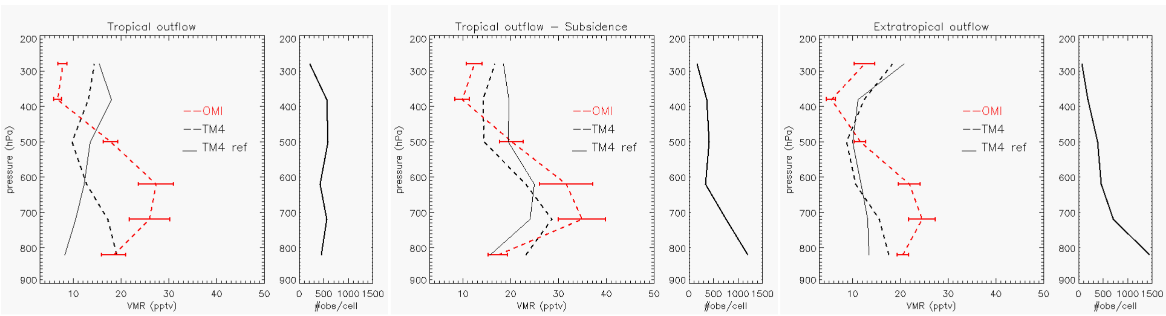


a) CRF 50% - Major industrial sources

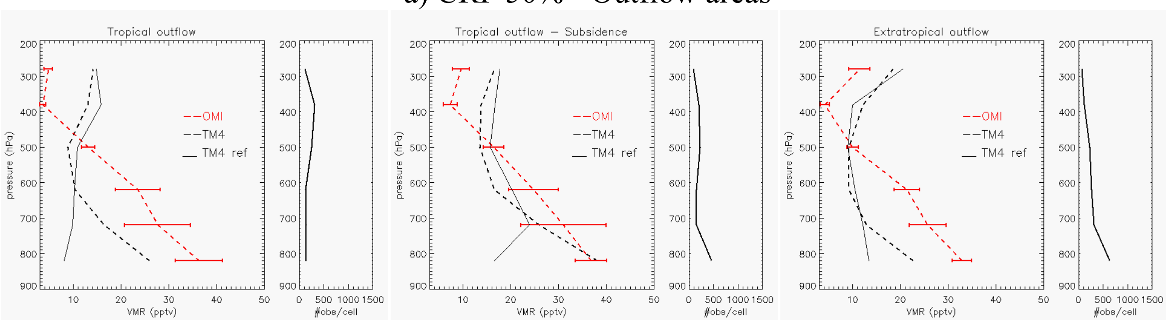


b) CRF 80% - Major industrial sources

Figure S3 – Average tropospheric NO_2 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.



a) CRF 50% - Outflow areas



b) CRF 80% - Outflow areas

Figure S4 – Average tropospheric NO_2 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.

In Figure S2, all major features observed at mid-tropospheric levels persist after changing the CRF threshold from 50% to 80%. The largest change consists in a general reduction of upper tropospheric NO_2 (at 280 and 380 hPa) in the 80% case, along with smaller biases at the lowest level (820 hPa) characterized by decrements 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 reduction of NO_2 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 NO_2 there (i.e. lightning events), resulting in overall lower NO_2 amounts. A critical look at the geographical distributions of NO_2 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 allowed cloud fraction to 50% is not forcing any observation-to-model agreement at upper levels, but increasing the representativity of observations.

We discuss the matter of the lowest levels next: let us have a look at profiles over strongly polluting centers in Figure S3. Over industrial sources, passing from 50% to 80% CRF produces a slight decrease in lowest tropospheric NO_2 amounts, which does not seem to be consistently driven by *a priori* information. Changes in NO_2 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, which is 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. Finally, let us have a look at profiles over typical outflow regions in Figure S4. Over outflow regions, raising the CRF threshold from 50% to 80% produces a general increase in NO_2 amounts at the lowest level (820 hPa), sometimes away and sometimes towards the model. In this case, 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.S4) 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 detrimentally influenced by the lower sampling densities afforded by a higher CRF threshold. This is the reason why we have finally opted for the CRF 50% threshold, which ensures 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 NO_X production is removed), negative biases at low altitude over industrial regions (when part of the advection from boundary layer NO_2 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 NO_2 cross sections for the CRF 50% and CRF 80% cases are appended next in Figures S5a-b to corroborate that changing the CRF threshold does not change the general picture appreciably.

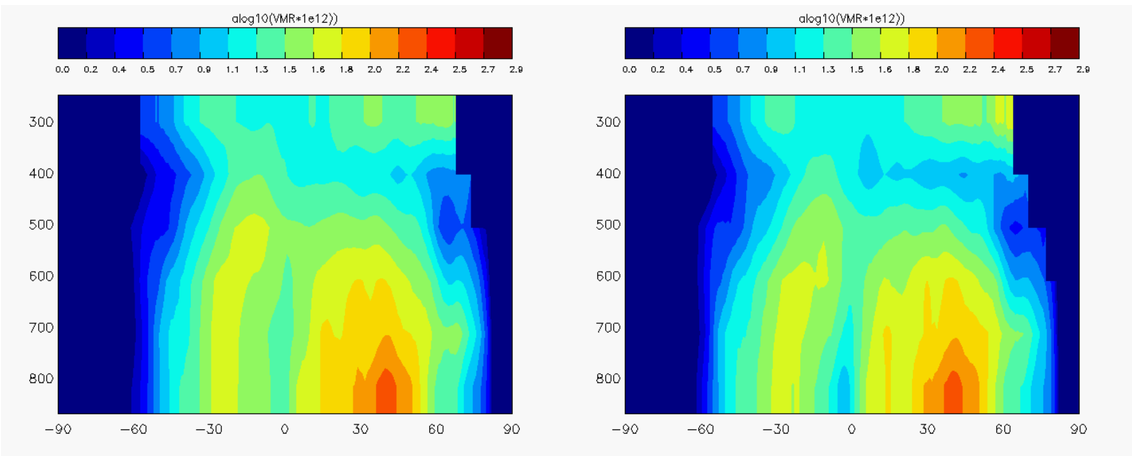


Fig. S5a - CRF 50% (left) and CRF 80% (right) zonal means (as in Fig. 10a of manuscript)

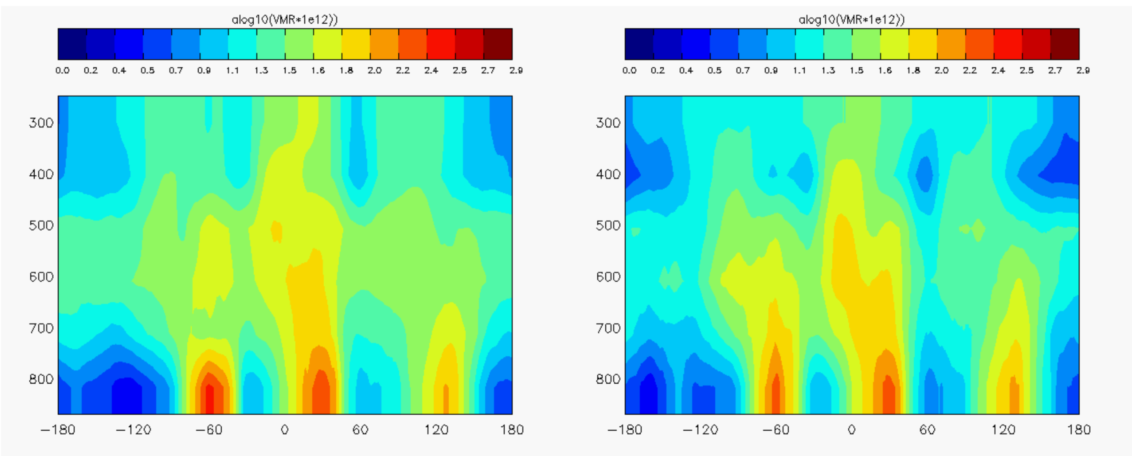


Fig. S5b - CRF 50% (left) and CRF 80% (right) tropical section (as in Fig.11 of paper)

In summary, 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 levels, 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. 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.