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Interactive comment on “Global free tropospheric NO₂ abundances derived using a cloud slicing technique applied to satellite observations from the Aura Ozone Monitoring Instrument (OMI)” by S. Choi et al.

Anonymous Referee #2

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The manuscript by S. Choi et al. presents a method to estimate NO₂ concentrations in the troposphere above clouds from OMI satellite measurements. Tropospheric NO₂ detection above and within clouds from satellite has been attempted before (e.g. Boersma et al. [2005]), but Choi et al. extend the approach to the global scale and also focus on polluted areas and all 4 seasons. The central idea is to compare “nearby” tropospheric columns that have been retrieved under situations of similar cloudiness, but with different cloud heights (“cloud slicing”). Assuming that the free tropospheric NO₂ concentration is constant with altitude, the reduction of column with higher clouds can

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then be used to derive the free tropospheric, above-cloud NO₂ concentration. This technique has been applied extensively for tropospheric ozone retrievals, but is now being applied for the first time on tropospheric NO₂.

The authors describe their method clearly, and evaluate their method by comparing against independent aircraft measurements. That the validation does not prove to be an overwhelming success was to be expected, in view of the small difference signals, and the detection limit of the OMI retrievals. Nevertheless the above-cloud tropospheric NO₂ climatology constructed by the authors appears to be a compelling result.

Major comments

1. The main issue I have with the method by the authors concerns their use of a simple geometric air mass factor (AMF) for converting the slant columns into the above-cloud vertical columns that are at the basis of their method (Eq. (6)). Using a geometric AMF may be a reasonable choice for retrievals of stratospheric NO₂ columns, but it will lead to considerable errors for above-cloud retrievals, because the sensitivity to NO₂ within and also above the cloud is strongly enhanced by the bright cloud. This is clearly indicated in the radiative transfer studies shown in e.g. Hild et al. [2002], Eskes and Boersma [2003], and Boersma et al. [2005]. The authors should therefore revisit their geometrical AMFs and replace these by more realistic AMFs that take into account the increased sensitivity above the effective cloud pressure level (and still discard the NO₂ below as is done in the geometric AMF). See also the study by Beirle et al. [2006]. The more realistic AMFs will certainly be higher than the simple geometric AMFs used here, and their use will improve the agreement between the OMI-derived and GMI modelled mixing ratios, and between OMI and INTEX-B.

2. Rather than just citing a relative bias in the OMI NO₂ slant columns, I propose to also quote the absolute bias in the vertical columns. Various studies (e.g. Belmonte-Rivas et al. [2014]; Krotkov [2012], Boersma et al. [2014]) suggest that the bias in the OMNO2A vertical (stratospheric) columns is rather constant over an orbit after

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converting the slant to vertical columns.

3. The assumption that the NO₂ concentration does not change with altitude is generally defensible, but will lead to errors in case of lightning NO_x production (and aircraft NO_x). The 'profiles' shown in Fig. 6 of the manuscript show that the higher the cloud, the higher the inferred above-cloud NO₂ concentrations. Such patterns have also been reported in the study by Boersma et al. [2005] from GOME, and high above-cloud NO₂ has been observed from various aircraft campaigns near and even within thunderstorm clouds. This immediately shows that (1) a simple geometric AMF is inaccurate for such situations, and that the AMF should take into account the actual vertical sensitivity, and (2) AMFs should account for realistic a priori profile shapes that will be very different in lightning situations. The authors have all the means at hand with the GMI model and state-of-science radiative transfer codes.

4. Section 4.2.2 should refer to the study by Boersma et al. [2005], as there are various parallels to be drawn. In that study, a considerable production of lightning NO_x over the tropical oceans was inferred from cloudy GOME measurements, with spatial patterns similar to those shown in the upper right panel of Figure 4. Similar to the GMI model here, the TM3 model used in their study also failed to reproduce a substantial source of lightning NO_x production over the ocean, pointing at similar misrepresentations in the lightning parameterization in both models. Since these lightning parameterizations are still in use in many CTMs, it is important to point out that these are in need of improvement.

Minor comments

Please provide some quantitative estimates on the cloud pressure errors from the OM-CLDRR and OMCLDO₂ products.

P1565, L 22: 'US/VIS wavelengths' should be UV/Vis wavelengths.

P1579, section 4.4: to my opinion the results from the Belmonte-Rivas study should be

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