Atmos. Chem. Phys. Discuss., 14, C584–C588, 2014
www.atmos-chem-phys-discuss.net/14/C584/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global free tropospheric NO_2 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 #1

Received and published: 20 March 2014

Review of 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.

In this manuscript, data on free tropospheric NO2 is retrieved from OMI observations using data taken at different cloud conditions. The retrievals are validated by comparison with data from airborne in-situ observations and reasonable agreement is found. Using a long time series (3 years), a coarse climatology of upper tropospheric NO2 is

created, showing interesting seasonality in its geographic distribution. As verification, the climatology is compared to results from the GMI model. As a side product, an estimate of the stratospheric column is derived which is compared to the operational OMI stratospheric NO2 product and the GMI model atmosphere.

The paper is clearly structured, well written and reports on a novel satellite data product, free tropospheric NO2 amounts. The technique used has to my knowledge never before been applied to NO2 and the results are interesting as very little is known about spatial distribution and seasonality of NO2 in the upper troposphere. The approach taken and the methods used are sound and nicely described, and a thorough discussion of uncertainties and results is provided. There are however several points which I think need to be improved or re-considered in the manuscript, and I therefore recommend publication in ACP only after my comments listed below have been taken into account.

Major Comments

- A geometric AMF is used for computation of the NO2 vertical columns over clouds. While this is probably a very good approximation above a cloud, it is not a good approximation for NO2 within a cloud. As the cloud top pressure from OMCLDO2 and OMCLDRR give the cloud optical centroid pressure, there always is a contribution from NO2 within the cloud which will be seen with another AMF. This is further complicated by the sampling issue discussed below. I think this needs to be discussed.
- As pointed out in the manuscript, cloudy scenes differ from clear sky scenes in many respects as they are representative of other meteorological situations, photochemical regimes, transport patterns (frontal systems) and vertical NO2 distributions. Other satellite studies using cloudy data have highlighted the occurrence of transport events in cloudy situations (e.g. Stohl et al., 2003 or very recently Zien et al., 2013) as well as lightning (e.g. Beirle et al., 2009, Boersma

et al., 2005) which will have important impacts on the statistical sampling of the free troposphere using cloud slicing. I'd suggest to discuss all these effects in a dedicated section in a qualitative way and to indicate the direction and size of the various effects that you expect. Some of the information is already present in the manuscript but should be collected and discussed in a more consistent way.

- An estimate for zonal stratospheric NO2 columns is derived and compared to the operational product. The good agreement between the two independent estimates is taken as closure validation of the cloud slicing technique. While this looks good at first sight, I think that the two estimates are neither independent, nor does the agreement tell much about the quality of the cloud slicing product. The reason is that for a zonal average, even taking all OMI NO2 slant columns and applying a stratospheric AMF without any correction will lead to reasonable results. In the operational product, regions with known pollution are excluded, making the estimate better. In the cloud slicing product, only cloudy scenes are used, removing most of the BL pollution NO2, which again should result in a good estimate of the stratospheric NO2 without further processing.

  The extrapolation to tropopause pressure (Fig. 1d) will remove the free tropospheric component from the above cloud total columns which at an estimated 30 ppt adds up to about 2E14 molec cm-2. This relatively small correction (which is the cloud slicing component of the stratospheric values shown) is of the same order as the differences between the two OMI stratospheric NO2 columns shown in Fig. 7. Thus the only conclusion I can draw from this comparison is that the free tropospheric columns derived with the cloud slicing method are so small, that they do not matter much for the stratospheric column. I therefore think that the whole discussion of the stratospheric columns needs to be revised (for example by showing the stratospheric estimate using all cloud slicing data but without extrapolation to tropopause height) or completely removed.

- The known bias in the OMI NO2 slant columns is referred to in many places

throughout the manuscript and used as explanation for higher than expected free tropospheric values. However, to my knowledge, the bias in the current OMI NO2 product is not a relative error but rather an absolute offset on the slant columns. As in the cloud slicing method the slope of a set of measurements at different cloud pressure is analysed, such an offset will not contribute significantly to the uncertainty at constant AMF. I therefore do not agree with the repeated statements explaining biases by the OMI SC problems and think they should be removed.

- The comparison between model and OMI free tropospheric NO2 VMR sounds OK in the text but looking at the figures, I hardly see any similarity. Both the spatial pattern and the absolute values are very different, and all the lightning signatures shown in the lower panels of Fig. 4 are clearly not reproduced in the OMI data. I think these discrepancies should become clearer in the text. It might also be worthwhile to mention the impact such differences in vertical distribution might have on tropospheric AMFs.

Minor Comments

- p 1561 l9: It is contributes => It contributes
- p 1566 l27: I do not see why equation 4 is based on any assumptions on NO2 – this is about the cloud scene pressure.
- p 1569, l1: why is the lightning contribution derived using all scenes? Doesn't this create a very different sampling than the satellite data?
- p 1569 and elsewhere: I'd prefer a small *p* for pressure
- p 1577 l12: in the both => in both

- p 1581 l29: over the North America => over North America

References

Beirle, S., M. Salzmann, M. G. Lawrence, and T. Wagner (2009), Sensitivity of satellite observations for freshly produced lightning NOx, Atmos. Chem. Phys., 9(3), 1077-1094.

Boersma, K. F., H. J. Eskes, E. W. Meijer, and H. M. Kelder, Estimates of lightning NOx production from GOME satellite observations, Atmos. Chem. Phys., 5, 2311 –2331, 2005

Stohl, A., H. Huntrieser, A. Richter, S. Beirle, O. Cooper, S. Eckhardt, C. Forster, P. James, N. Spichtinger, M. Wenig, T. Wagner, J. Burrows, and U. Platt, Rapid intercontinental air pollution transport associated with a meteorological bomb, Atmos. Chem. Phys., 3, 969-985, 2003

Zien, A. W., Richter, A., Hilboll, A., Blechschmidt, A.-M., and Burrows, J. P.: Systematic analysis of tropospheric NO2 long-range transport events detected in GOME-2 satellite data, Atmos. Chem. Phys. Discuss., 13, 30945-31012, doi:10.5194/acpd-13-30945-2013, 2013

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 1559, 2014.