Reply to comment of Anonymous Referee # 1

Achim Zien

April 19, 2014

Thank you very much for your comments and your confidence in our research. Your comments have been taken into account in the revised version of the paper and without doubt have pointed towards unclear parts of our method. We hope that the revision made has made the paper more clear to its audience.

1 Cloud and NO_2 profile

In section 2.1, the authors present their approach to retrieve VCDs in partially cloudy scenes, and the limitations of the their approach to calculating AMFs are discussed in Section 4.2. As the authors indicate, the air-mass factor depends on where in the vertical direction the trace gas is situated, and this becomes even more significant when clouds are in the scene. My one major concern is that the authors appear to base their assumption about the vertical distribution of NO2 in cloudy scenes on one single set of observations (the TRACE-P campaign). Therein, it was found that CO tends to be homogeneously mixed inside the cloud, resulting in those cases from the passage of a cold front over an emission region. While this phenomenon may be typical of initiating long-range pollutant transport over China (which is indeed eventually re-confirmed by the algorithm), I wonder how appropriate it is to apply systematically across all cases of cloudy scenes.

The TRACE-P observations were taken right along the Asian Pacific Rim; but the authors of the present paper do not explain why the vertical distribution of NO2 in the presence of clouds will remain constant from the coast to the open ocean. Further in the paper, the authors discuss the issues resulting from the chaotic movement under conditions of strong winds (l. 710); can this alter the appropriateness of the homogeneously mixed profile at cloud height? It may be that the authors cannot find examples of other field data to confirm or deny to existence of other vertical distributions in the presence of clouds, but the fact that their choice is based on a single reference from a single region of the world targeted for specific meteorological dynamics is not given enough attention or discussion. What do modeled NO2 profiles over the oceans look like when clouds are present? While they may not be entirely accurate, their variety may lend some insight into the appropriateness of the single profile that is being assumed in this paper.

We agree with the reviewer that the assumption on the NO_2 profile is critical and that we have taken a very rough approach by assuming a well-mixed profile colocated with the cloud. Unfortunately, there is to our knowledge no measurement of both NO_2 and cloud profiles in long-range tranport events, and only few trace gas and cloud profile measurements in long-range tranports at all. This is, why we used the TRACE-P measurements.

The reviewer suggests using model profiles in order to evaluate the development of NO_2 profiles during long range transport events. At the beginning of our project, we tried to use GEOS-Chem model output as a priori for our retrievals but had to realise that the representation of long range transport events in the model did not match well with observations of both clouds and NO_2 in the satellite data. A model like WRF-Chem will probably perform much better but such data is not available for multiannual global runs as would be needed here. In response to the comment made by the reviewer, we have investigated data from the MACC reanalysis (Inness et al., 2013, doi:10.5194/acp-13-4073-2013) which has a reasonably high resolution of about 1.25×1.25 degrees. For one of the examples discussed in the paper (October 2nd, 2010), the model NO_2 field reproduces the observed export pattern at least qualitatively (see Fig. 1). In Fig. 2, three exemplary NO_2 profiles are shown, indicating that the model predicts a relatively well mixed layer of NO_2 with a layer top altitude increasing with distance from the source. The model clouds (here approximated by the sum of liquid water and ice content) show a very similar behaviour with the exception of the missing values below cloud base. In this case, our assumption of similar behaviour of cloud and NO_2 is justified but the assumption of a layer between 3 and 5 km is not. As discussed in the paper, the vertical position of such a cloud does not have a very large effect on the AMF as long as the NO_2 is well mixed within the cloud.

This rather anecdotal confirmation for our approach could in principle be repeated on other cases but is a very manual and subjective process where individual events have to be identified in the model data, pixels be selected, and profiles be compared visually. We therefore do not think that adding some more random cases would make our argument much stronger and cannot conceive an automated way to analyse the link between cloud and NO_2 profiles in LRT events in model data as a function of transport time.

We do however appreciate the point made by the reviewer and have extended the discussion on the uncertainties introduced by our assumption.

2 Cloud-free and cloudy AMF

> *Along the same lines, it is not clear to me how the total AMF is always calculated by combining the weighted cloud-free and cloudy AMF. It does seem that the only realistic choice is to assume the same NO2 vertical profile is used in calculating the cloudy and cloud-free air mass factors for a single pixel– But I am wondering about situations whether the pixel is partially cloudy. The calculation of the radiance cloud fraction seems to force the authors to assume that in the presence of any cloud fraction, no matter how small, the result is a pixel where the NO2 distribution is homogeneously mixed at the determined altitude of the cloud. What about pixels where the radiance cloud fraction is very small– wouldn't it be better to apply the standard profiles in those cases? Is this done? Is there a radiance fraction cut-off (or should there be?) where the trace gas profile is assumed to be standard vs. homogeneously mixed at one altitude? If there is, it is not clear to me– perhaps I have missed something in the manuscript.*

The objective of this study is to investigate long range transport events, and through the analysis applied, we try to select only those observations which are part of such events. We therefore decided to use dedicated AMFs which are as appropriate as possible for LRT events but will not necessarily be correct for other situations.

The NO_2 is – for the purpose of calculating the AMF – always assumed to be at the same altitude: between 3 and 5 kilometers. Assuming that the NO_2 is elevated (as is neccessary for NO_2 long-range tranport) the changes in AMF are relatively small when mowing the NO_2

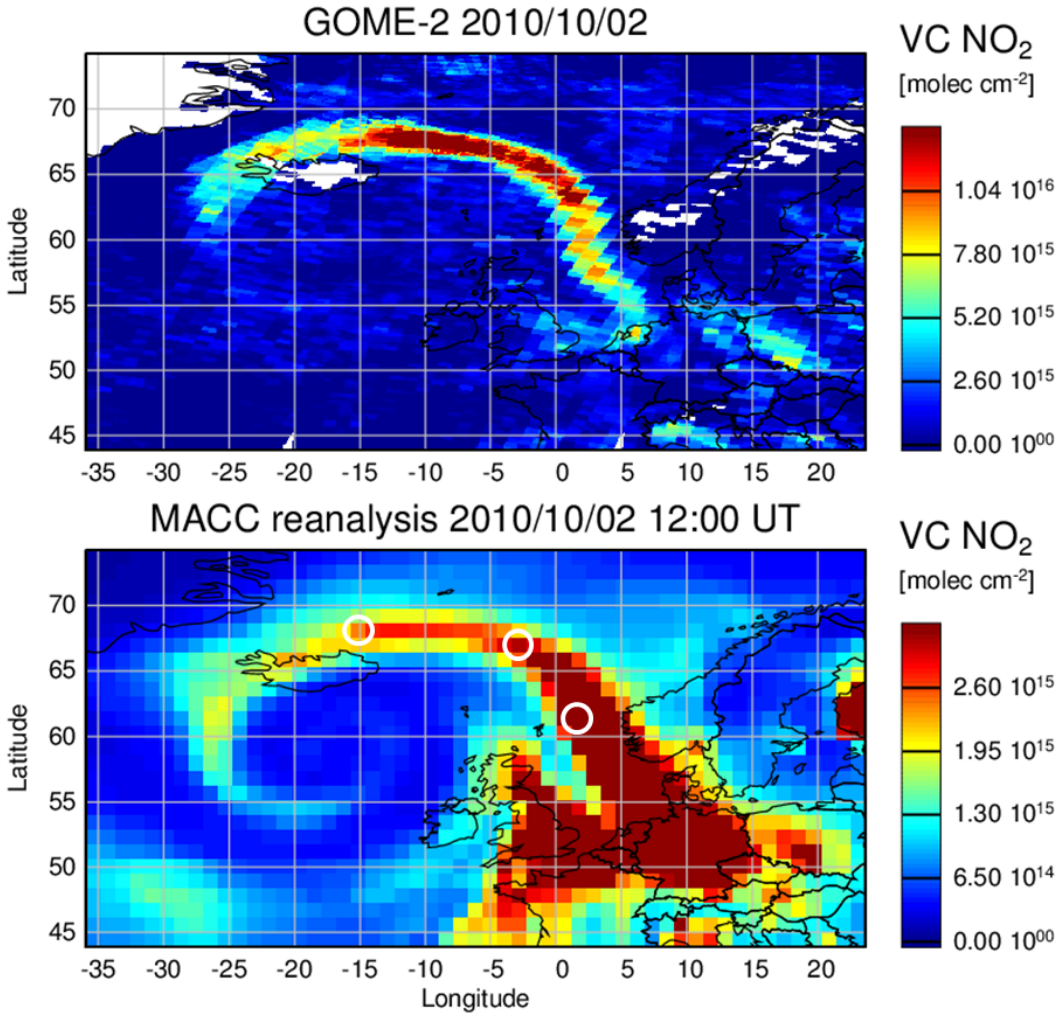

Figure 1: Long range transport event on October 2, 2010 as seen in GOME-2 data (top) and the MACC reanalysis (bottom). Please note the different colour scales. The three circles in the lower plot indicate the pixels for which vertical profiles are shown in Fig. 2.

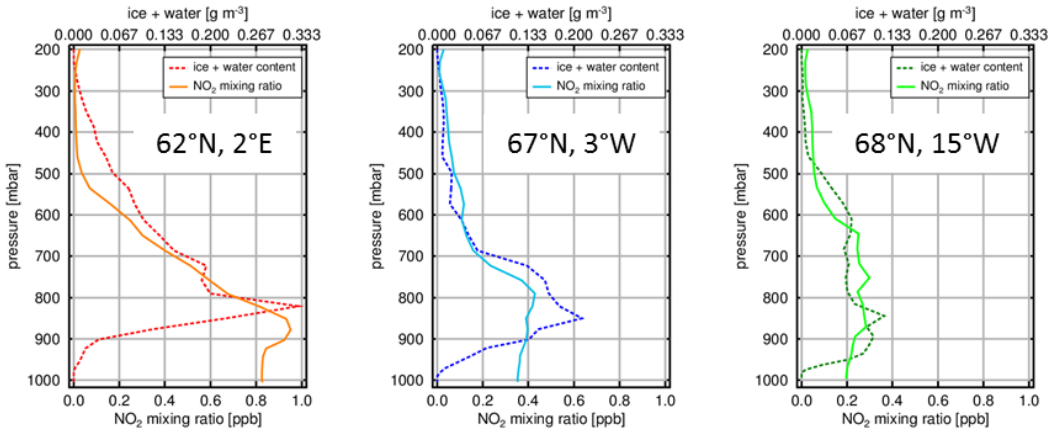

Figure 2: Vertical profiles of NO_2 and cloud liquid water and ice content for three geolocations in the MACC reanalysis output for October 2, 2010.

further up or down. For the cloudy AMF, the cloud is also simulated at an altitude of 3 to 5 kilometers. Thus we assume that the NO_2 in a partially cloudy pixel is homogeneously distributed. In our opinion, this is the most consistent assumption although – as the reviewer correctly points out – it will not be appropriate in pixels not affected by LRT. However, we hope that such pixels will be eventually removed from the analysis which tries to isolate LRT events.

The text has been updated to clarify our approach. In response to the first comment of the reviewer, the impact of locating the NO_2 at the wrong altitude is now discussed in more detail in the paper.

3 Technical comments

l. 882-884: Is this statement (regarding NO2 and glyoxal from bush fires) based on the present work, or presented in the literature? Please include references if necessary.

References for the detection of glyoxal from satellite are:

- Wittrock, Folkard, Andreas Richter, Hilke Oetjen, John P. Burrows, Maria Kanakidou, Stelios Myriokefalitakis, Rainer Volkamer, Steffen Beirle, Ulrich Platt, und Thomas Wagner. 2006. „Simultaneous Global Observations of Glyoxal and Formaldehyde from Space“. Geophysical Research Letters 33 (16): L16804. doi:10.1029/2006GL026310.
- Vrekoussis, M., F. Wittrock, A. Richter, und J. P. Burrows. 2009. „Temporal and spatial variability of glyoxal as observed from space“. Atmos. Chem. Phys. 9 (13): 4485–4504. doi:10.5194/acp-9-4485-2009.

l. 1206: spelling of instead

This has been fixed in the text.

l. 1651-1661: This reference made it out of ACPD into ACP long ago– why not reference the ACP published version?

All ACPD references have been updated, if possible.