Atmos. Chem. Phys. Discuss., 13, C11114–C11117, 2014
www.atmos-chem-phys-discuss.net/13/C11114/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Systematic analysis of tropospheric NO_2 long-range transport events detected in GOME-2 satellite data" *by* A. W. Zien et al.

Anonymous Referee #1

Received and published: 15 January 2014

General Comments:

In this paper, the authors develop a methodology to systematically identify long-range NO2 transport events in GOME-2 satellite observations. An approach to retrieve tropospheric vertical column densities in cloudy scenes (which are commonly excluded from analysis) is presented, and an algorithm to automatically identify long-range transport events is developed and evaluated. Following a discussion of representative case studies, a statistical investigation of the whole dataset is performed where plume properties, typical transport routes, and total outflow flux are explored globally and by region.

This work makes a very interesting contribution to the study of trace gas composition by satellite observations. By attempting to retrieve information from cloudy scenes, which is understood as necessary to accurately capture long-range transport events often associated with frontal systems, the work will no doubt provoke an insightful and constructive discussion. Here, the authors make quite a few simplifying assumptions that are in some cases based on very little supporting data (see below) and which some may find to be contentious, but that in my opinion essentially reflect the state of the science and are not unwarranted. With the data from cloudy scenes at hand, and an algorithm to identify long-range transport events developed, the authors are able to establish novel observations into the nature of continental NO2 outflow (e.g. that there does not appear to be a trend in the mass distribution of NO2 plumes; that long-range NO2 transport is a common occurrence; that the export of NO2 to the oceans represents about a permil of global NO2 emissions; etc.) that have otherwise been entirely elusive. While the limitations of the algorithm for identifying long-range transport events must be kept in mind, in my opinion they are addressed adequately by the authors and accompanied by the appropriate caveats.

I do not take any significant issue with the authors' approaches and believe this work will be of interest to other investigators. Overall, the paper is well-developed, generally well-referenced, and written clearly. The methods are adequately laid out, and the presentation of the data in figures and tables adds to the clarity of the text. I therefore recommend it for publication in Atmospheric Chemistry and Physics, after the following comments have been addressed either in discussion or in a revised manuscript.

Specific Comments:

1) 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.

2) 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.

Technical comments

1) 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.

2) l. 1206: spelling of "instead"

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

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 30945, 2013.