Atmos. Chem. Phys. Discuss., 6, S1150–S1153, 2006
www.atmos-chem-phys-discuss.net/6/S1150/2006/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comparison of GOME tropospheric NO_2 columns with NO_2 profiles deduced from ground-based in situ measurements" *by* D. Schaub et al.

D. Schaub et al.

Received and published: 8 June 2006

Responses to Referee comments in ACPD (acpd-2005-0453)

I would like to thank the anonymous referees for their helpful reviews and hints which allow us to improve our paper. Below are the responses to the specific comments.

Anonymous Referee #1 (comments from 4 May 2006)

We agree with the referee that the description of the two comparison methods - as a "direct comparison" and a "comparison including averaging kernel information" - as well as the last phrase in the abstract could confuse the reader. The related text passages (in the abstract, but also in the text) will be formulated slightly differently to describe the

two approaches more clearly.

Pages/lines 2197/10 and 2207/25: for estimating the fractions of the signal (reaching the satellite) coming from both the PBL and from above the PBL we in fact used our NO2 profiles determined from in situ measurements up to 3500 m asl and interpolated to a constant mixing ratio at 8 km. The NO2 above is neglected. (The same profiles that are used for the different comparisons; no TM4 profiles are used). The result of around 50% of the signal at the spectrometer originating from above the PBL does from our point of view not disagree with neglecting the NO2 in the topmost troposphere: 1. The coincidence between still relatively high NO2 concentrations in the layers directly above the PBL (say between 1000-2000 m asl) and a higher sensitivity there explains a large part of the signal from above the PBL. 2. It is known that the NOx mixing ratio in the troposphere can exhibit a C-shape with again relatively high NOx mixing ratios near the tropopause. However, due to the small pressure at this altitude the absolute number of NOx molecules there is still very small compared to the number of molecules in the lower troposphere. Furthermore, the C-shape is not seen for NO2: the NO/NO2 ratio changes from 0.25 near the ground to around 1 at higher altitudes (note that this will also be changed in section 1). Thus, the above argumentation of low absolute molecule numbers in the topmost troposphere even stronger applies for NO2.

Section 4.3.2: The referee asks about the treatment of snow in the retrieval and suggests to add some discussion on this issue given the mountainous terrain in the study area. We agree with the referee that a short discussion should be added. The treatment of snow has also been stressed in Boersma et al. (2004). There, it is argued that the FRESCO algorithm is able to handle snow covers to some extent. However, for the present work, we suggest this not to be a major issue, because the focus has been on GOME pixels covering mainly the lower part of Switzerland (Swiss Plateau) and surroundings: GOME pixels have been chosen for comparison if their centre coordinates were located within the “frame” given in Figure 1. This frame was chosen in order to reject GOME pixels that are mainly located over the Alps, as shortly mentioned

ACPD
6, S1150–S1153, 2006

EGU

in section 3.1.1 (due to potential retrieval problems including snow cover).

Page 2191, line 25: We fully agree and will change this in the text! (See also the above argumentation about the neglecting the NO2 in the upper troposphere and the 4th comment of referee #2).

Page 2209, line 16: We agree with the referee that Moxim et al. (1996) referred to NOx rather than NO2. This will be changed. This part of the paper describes the fact that frequently (under anticyclonic clear sky conditions) high spring NO2 in both the GOME and the ground-based columns have been found. Rather than explaining this, we cite some other literature that found or simulated NOx maxima during this season. The referee argues that Moxim et al. analysed a broad region that must not be relevant for explaining local observations. We will additionally stress the spatial focus of the Moxim paper. However, the spring NOx maxima found in the lower troposphere by Moxim et al. agree with the NO2 maxima found at elevated measurement sites in the Alpine region. The composition of air masses at such altitudes is known to be influenced not only locally but regionally or even on the hemispheric scale. Due to the GOME columns being an integral over the whole troposphere we would suggest that also columns selected locally can be influenced by larger scale phenomena. TM4 does - in average - not show a spring maxima in NO2. It should, however, be noted that we i) focus on clear sky anticyclonic days only and that ii) not all spring NO2 columns show high values (taking an average only might not show the spring maximum as clear; also Moxim et al. described the episodic character of high-magnitude NOx).

We agree with both referees that extremely large GOME NO2 columns $>$ 300 x 1015 molec cm-2 (up to 500 x 1015 molec cm-2) are unrealistically high. This will be mentioned in the text. Referee #1 further argues that such high columns might be too large for the AK approach to be useful and - rightly - refers to an earlier text passage stating tau $<$ 0.005 is required for weak absorbers. This text passage is wrong and will be changed. In fact, for being a weak absorber, tau $<$ 1 is required (tau $<$ 0.005 is a typical optical thickness for NO2 in the 400-450 nm spectral range). Even for an NO2

ACPD
6, S1150–S1153, 2006

EGU

column of 300 x 1015 molec cm-2 we estimate an optical thickness of 0.15 and the weak absorber limit is still not too bad. It should furthermore be noted that the high column values are an artefact of the very small air mass factor as explained in the text. The measurement itself, or the slant column, is much smaller and this corresponding slant column well falls within the weak absorber regime. Finally, we suggest to keep the “case study” about the extremely high GOME columns on 17 February 2001 in the text, because it is a very nice example of retrieval problems that can occur during frontal passages. Following the referees comments, we will point out the “unrealistically high GOME column” that is found for this particular case.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 2189, 2006.