**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Tropospheric NO$_2$ column densities deduced from zenith-sky DOAS measurements in Shanghai, China, and their application to satellite validation" *by* D. Chen et al.

D. Chen et al.

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First we want to thank this reviewer very much for his positive and constructive review. Before we address his comments in detail point by point, we first give a short overview on the major changes of the manuscript.

a) We inserted a much more detailed error discussion taking into account the effects of different contributing error sources, especially their dependence on SZA and the tropospheric NO2 VCD. The errors are expressed as absolute and relative errors and presented in the new Fig. 5.

b) We included additional sensitivity studies for the determination of the tropospheric AMF taking into account the effects of varying asymmetry parameter and surface

albedo. The results are summarized in Fig. 6 (old Fig. 10).

c) We now use tropospheric AMF for a single scattering albedo of 0.95 for the determination of the tropospheric NO2 VCD from the zenith-sky observations. While reliable information on this parameter is difficult to obtain, we think that a value of 0.95 might be more realistic than a value of 1.0 (purely scattering aerosols). The application of the new value leads to an increase of the tropospheric NO2 VCD by about 2-5% depending on SZA.

d) As also suggested by the other reviewers, we include a new figure (Fig. 15) showing the correlation analysis and time series comparison of the SCIAMACHY data with the surface NO2 concentration.

A:
General Comments:
The manuscript describes in details a method to extract tropospheric NO2 vertical columns from a zenith sky DOAS instrument and present some comparisons with long path DOAS and satellite measurements. The paper is well organized and within the scope of ACP. However the various descriptions and discussions on error budgets need to be revised.

Reply: Many tanks for this positive assessment. We addressed all the points raised by the reviewer as indicated by our detailed response (see below). We updated our error discussion in a comprehensive way (see also point a) above).

B:
Specific comments:
1. Abstract: <zenith sky measurements are more suitable for satellite data validation than the in-situ measurements> What do you mean by in-situ? The term <in-situ> is inappropriate. I suppose that you refer to long path DOAS which are not in-situ but averaged surface concentration measurements on a 507m path. In-situ is generally used for <local> measurements. Same remark on page 16716 line 5.

Reply: Thanks for your clarification. We agree that 'in-situ' is not the correct term to describe long-path DOAS measurement. We replaced 'in-situ observations' with 'surface concentration measurements'.

2. <The offset was removed automatically> Page 16717 line 25. What is the offset? Where is it coming from? It seems not to be the dark current of the CCD? Is it appropriate?

Reply: We replaced 'offset' by 'dark current and electronic offset'. The electronic offset is a term added to the observed signal to avoid negative values before ADC processing.

3. Cross sections at 223K and 293K Page 16718 line 18 What does orthogonalized mean? Are-you analysing with both cross sections separately or simultaneously? How is the partitioning between warm (tropo) and cold (strato) taken into account? Is the orthogonalization useful for ozone? Does-it interfere with NO2 measurement?

Reply: 'Orthogonalisation' is a mathematic method adopted in WinDOAS software to avoid correlation between cross sections of similar shapes. You can find more detailed description of this function in the user's manual of WinDOAS (Fayt, C. and v. Roozendael, M.: WinDOAS 2.1 software user manual, IASB/BIRA Uccle, Belgium, 2001.) In our study, we orthogonalised the cross section at cold temperature with respect to that at high temperature. Thus the NO2 fit result represents the NO2 absorption at the high temperature. We added this information to the text in Sect. 2.1.2.

4. SCD strato Page 16722 line 15 To reduce the uncertainty caused by stratospheric column the authors use various pairs of am and pm values. As it corresponds to an averaged value used during a three to four months period, I think that it is not reasonable to have two digits for these columns. (for ex: use 5.9 instead of 5.93). What is the error bar on each measurement?

Reply: We changed the values to include only one digit.

5. Determination of NO2 SCD in reference spectrum. There is no indication of the

relative values of SCDstrato_ref and SCD tropo_ref. What is the error on these SCD in reference spectrum? An indication of the relative values here will help also understanding the discussion in section 4.1.2.

Reply: Since we applied different reference Fraunhofer spectra for spectra recorded during different periods (but with one reference spectrum used for a long period), here we take the reference Fraunhofer spectrum measured in 26 Feb 2007 as an example, and list the values of SCDstrato_ref and SCDtropo_ref for readers' reference. The various values are as below:

SCDref = (8.0±1.0) $\times 10^{15} molecules cm^{-2}$

SCDstrato_ref = (4.5±0.5) $\times 10^{15} molecules cm^{-2}$

SCDtropo_ref = (3.5±0.5) $\times 10^{15} molecules cm^{-2}$

We added this information to the text in Sect. 3.1.3.

However, it should be noted that the error on these SCD in reference spectrum has only a very small effect on the tropospheric NO2 VCD, considering that the reference spectrum was taken at local noon with small AMF and low surface NO2 concentration.

6. 3.1.4 Error estimation. This section is supposed to provide a summary of the error. But it is not clear how the respective error have been estimated.

For example: page 16725 line 20, the error on stratospheric slant column is estimated to be about 10% for SZA <85.

For example: page 16726 line 5. the error caused by tropospheric AMF are <15% for most cases.

I think that the evaluation of the errors should be discussed in each paragraph separately. Then, a summary of the error estimates can be summarised in 3.1.4.

Again same remarks on error evaluation in section 3.2. There is no indication of how

the error have been evaluated. A 40% value is suggested on page 16728 line 29. Where is-it coming from?

The error discussion should be completely rewritten as it is an important aspect to be convinced by the section: results and discussion.

Reply: We followed the suggestion of this reviewer and completely rewrote the error discussion. It now contains all relevant error sources in a comprehensive way and shows also the dependencies on SZA and VCDtropo_zenith.

7. page 16733 line 26 error estimate ?

Reply: We now provide more information on the error determination of the SCIA-MACHY data. See Sect. 4.2.1.

8. discussion on page 16735 line 25 and after. It is not obvious looking at figure 8 that the PBL over Shanghai is relatively stable at the satellite overpass time. It seems that the PBL is changing rapidly around 10:30.

Reply: We agree with the reviewer's comment and removed the respective statement. Further research is needed to investigate the details of the development of the boundary layer height during the day. We reformulated the text at the end of Sect. 4.2.2 to: "The quality of satellite validation using surface concentration data will stronly depend on the time of the day and will be best for satellite instruments with overpass times during noon. "

9. page 16735 line 16 ratio between Sciamachy and GB data: I think that two digits for the ratio 1.73 suggests that this value is very accurate. I suggest to write 1.7 + 0.7 Same remark on page 16739 line 12.

Reply: We agree and changed the numbers accordingly.

10. page 16737 line 23 and figure 14 (figure caption) I was lost by the term <light pollution around Shanghai>. At first I thought that it was concerning light (limited,

small) pollution. It would be better to use: <nighttimes light pollution>

Reply: Yes, it will cause confusion to use the word 'light' here. We have replaced it by 'nighttime light' in both the text and figure caption.

C:
Technical corrections:

1. Page 16725 line 1 Display the same variation, suppress <trend>

Reply: The word 'trend' has been deleted.

2. figure 10 in the text page 16730, line 12. replace < the aerosol layer extends lower (case 1 and 2) and higher (case 3 and 4) > by <the aerosol layer extends lower (case 1 and 4) and higher (case 2 and 3)>

Reply: We replaced the sentence by "Figure 6a shows the tropospheric AMFs deduced under the assumptions that the aerosol layer extends lower (case 1) and higher (case 3) than the tropospheric NO2, respectively. "

3. page 16732 line 8 typing error: pollution in lower atmosphere is < light> not <slight>

Reply: We changed 'slight' into 'light'.

4. page 16736 line 12 : satellite observations are more <strongly> affected not <stronger>

Reply: We have made revision accordingly.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16713, 2008.