

Interactive comment on “Tropospheric NO₂ 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 Howard Roscoe 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

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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 comment:

This manuscript is mostly thorough and the subject matter important, though one could debate its degree of originality in scientific as opposed to technical content. Although it is clearly deserving of publication after revision, the authors should consider whether it might be better placed with the new EGU online atmospheric journal for work of a technical nature.

Reply: Many thanks for the positive assessment. We addressed all points raised by the reviewer as indicated by our detailed response (see below). Concerning the suggestion to publish our article in AMT, we would in general agree. However, given the advanced status in the publication process, we like to follow our aim of publication in ACP. Note also, that besides the technical aspects, also important information on the chemical composition in one of the most polluted regions is provided by our article.

B.

Specific comments:

1. The error estimates in Section 3.1.4 contain assertions of small errors at the end of each subsection, but there are few details to show how they are derived. In sub-

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section 3, there are no details at all. Such errors are fundamental to the main thrust of the paper, and for a manuscript otherwise so full of technical detail they are strange omissions.

Reply: We updated our error analysis to include in detail all relevant error sources and give more information on the dependence of the absolute and relative errors on various parameters (see also point a) above).

We are also not told the typical error in PBL height in Section 3.2, incongruous because the effect of an error in PBL height is later explored, but without the reader knowing whether the values used are representative.

Reply: The uncertainty of the PBL height is difficult to assess, because accurate information on the PBL height is difficult to obtain. Thus the assumed profiles can only be seen as a rough estimate. Fortunately, the tropospheric AMF does only weakly depend on these assumptions. We added this information to Sect. 3.1.2, where the seasonal BL variations are first introduced.

2. Surely, the positive intercepts in Figs7 & 9 are unphysical? When there is zero NO₂ at the surface, we should expect the amount in the free troposphere to give an amount in the zenith-sky view, resulting in a negative intercept as these graphs are cast. Or have I misunderstood something? In any case, the fact of intercepts at all is worthy of comment in the text.

Reply: This is an interesting point! In principle we agree with the reviewer's argumentation. However, given the fact that the intercepts are rather small, we think that this effect is usually not important. Nevertheless, we added a short comment in the revised version of our manuscript (at the end of Sect. 4.1).

3. The manuscript is long and sometimes repetitive. Most sections have an introduction which adds little to the sub-section headings, and could easily be cut. Elsewhere, detailed cuts can easily be made, two examples picked from p167373 are: (a) line16:

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cut <over the urban site> from the sentence <Since our experimental site suffers from heavy traffic pollution, . . . > (b) line26: shorten <The light data are . . . (Nighttime lights are for the year 2003).> to <The light data are . . . for 2003.>

Reply: Those sentences have been revised accordingly. Also at other parts, the manuscript has been restructured to make it better readable. For example, the general error discussion and the sensitivity studies for the tropospheric AMFs were combined in two subsequent sections (3.1.4 and 3.1.5).

4. p16714 line26 seems to assert that tropospheric NO2 contributes to radiative forcing. This would be astounding if true and should be backed up with references. If the authors mean it contributes locally via its effect on ozone, then it cannot be true locally in the boundary layer, because changes in composition there occur at almost the same temperature as the surface, so there is little change in net upwelling radiation. To achieve significant radiative forcing, changes in composition must occur at a temperature significantly different to the surface, e.g. in the upper troposphere.

Reply: We have added *Solomon et al, 1999* as the supporting reference, in which the author demonstrated that NO₂ contributes to radiative forcing by directly measuring the absorption of downwelling visible radiation by NO₂. This effect results from local pollution and production by lightning in convective clouds.

5. The assertion in Section 4.1 that high thin clouds can decrease tropospheric absorption is counter-intuitive, the authors should provide some explanation and a reference.

Reply: The effect of high thin clouds on tropospheric absorption has already been explained in the two referred paper, *Wagner et al. (1998)* and *Pfeilsticker et al. (1998)*. When high thin clouds exist, a greater fraction of the observed photons would have passed through the atmosphere on a vertical rather than on a slant path. Thus, for tropospheric species, the absorption would be at least slightly decreased.

6. Section 4.1.2 ignores changes in PBL height during the course of the day. Although

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sunrise and sunset can often be similar, their difference from early pm can easily be 100%.

Reply: We agree that the PBL can vary strongly during the day. Therefore we changed the original sentence

“The different extent of agreement in each group strongly indicates the validity of their PBL height settings in certain period of time”
into

“The different extent of agreement in each group indicates the systematic variation of PBL height during the course of the day”

And as indicated the last paragraph of Sect. 4.1.1 also brings up an idea that we can get some valuable information about the diurnal change of PBL height by making comparison between the VCDtropo_surface and VCDtropo_zenith.

7. What is meant by orthogonal regression in Fig13 caption? Is this some special form of regression?

Reply: The orthogonal regression in old Fig.13 caption is the same as the regression analysis mentioned in other parts of paper. As described in Sect. 4.1, we adopted a weighted bivariate least-squares method, which considers the errors in both y- and x-variables, and minimizes the perpendicular distances between the fitted line and the data. We called such regression analysis as the “orthogonal regression” here.

C.

Technical corrections:

p16715 line21: replace NDSC by NDACC

p16728 line6: replace and by to

p16731 line15: replace AMF by AMFs

Fig6 caption: they are comparisons, not groups of comparisons

Reply: We have made corresponding revisions in the paper.

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