**Atmospheric
Measurement
Techniques
Discussions**

Interactive comment on "An improved tropospheric NO$_2$ retrieval for satellite observations in the vicinity of mountainous terrain" *by* Y. Zhou et al.

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First of all, we want to thank this reviewer for the thoughtful and constructive comments. We have prepared a revised manuscript that will be submitted shortly

In the following reviewer comments will be in italics, our response in normal typeface.

General comments

The paper by Zhou et al. describes an approach to reduce topography-related errors of tropospheric NO$_2$ from OMI satellite measurements over the Alpine region, and includes comparisons with ground-based in situ observations. This paper is a follow up

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to the study described in Schaub et al. 2007. Although the paper is clearly written and presents some interesting results, there remain important questions (see below) that should be resolved. There are inconsistencies between this study and the results described in Schaub et al. 2007 that are not clearly explained. Therefore, I can not recommend it for publication in AMT in its current stage. The paper may be acceptable after important revisions.

The referee is right that there are significant inconsistencies between this study and earlier work of our group published by Schaub et al. (2007). The main reason for this is we found a bug in the DOMINO retrieval code which produced too low box airmass factors near the ground. It occurred in the process of interpolating the air mass factor (box AMF) between the values at the two neighboring pressure levels of the lookup table when the lower level was located below the surface. Moreover, the magnitude of this error largely depended on the position of the actual surface pressure relative to the reference points in the lookup table. The results by Schaub et al. (2007) were based on a few selected cases only with specific p_{surf} obtained from TM4 model and effective pixel-averaged surface pressure p_{eff} from aLMo model. Unfortunately, the p_{eff} of the limited pixels ($\sim$ 960hPa) analyzed by Schaub et al. (2007) (as listed in their Table 2) were located near the position with largest errors, while errors at p_{surf} are much smaller. This causes a very high sensitivity to surface pressure changes. The results of Schaub et al. (2007), although pointing in the right direction, are therefore biased high. This bug was only detected and eliminated at the beginning of this study by the authors at Empa independently. The shape of corrected box air mass factor profiles presented here are now consistent with work performed earlier for GOME and SCIAMACHY retrievals and documented in a GOME-2 technical report (http://o3msaf.fmi.fi/docs/vs/2005/o3safvsgome-2NO2finalreport061126.pdf). This problem was briefly mentioned in our manuscript but will be more clearly documented in the revised version.

Specific comments

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2.1 Introduction

A general overview of the treatment of topography in DOAS retrievals is missing in the introduction. The impact of the topography on the air-mass factor is a well known issue in DOAS retrieval of trace-gas columns from satellite measurements. To my knowledge, most state-of-the-art retrieval algorithms take the surface height into account for the calculation of the AMF. A very similar approach to reduce topography-related errors as presented here, using effective pixel-average terrain heights based on GTOPO data, has been used for the operational trace-gas column retrieval algorithms for GOME and SCIAMACHY since many years (see for instance: earth.esa.int/pub/ESADOC/GOME/ATBD.pdf, 2004).

Author response: Many thanks for these valuable comments. It is clear that any retrieval needs to deal in some way with the topography and its effects on the airmass factor (AMF). The altitude of the surface affects the pathways of photons and hence the sensitivity of a satellite measurement to NO_2 in a given atmospheric layer. Furthermore, the a priori NO_2 profile assumed in the AMF calculation needs to match the topography data set. Although these effects are well known as the referee correctly points out, the way the topography is treated differs strongly among available retrieval algorithms and, except for the study of Schaub et al. (2007), there had been no quantitative analysis of the sensitivity of the NO_2 columns to the way the topography is treated. We admit that our manuscript did not sufficiently refer to and acknowledge existing approaches to this problem followed by other groups which, however, have not yet been adequately documented in the peer-reviewed literature. In fact, in the meantime most retrieval algorithms appear to employ a high-resolution topography data set in one or another way (e.g with or without accounting for meteorological variability in surface pressure, and with largely different approaches to the modification of the a priori profile). Since GDP4.0 the GTOPO topography data set is also used in the operational GOME (and GOME-2) algorithms (see GOME-2 technical report referenced above). We will therefore adjust the motivation for this study by briefly summarizing

other approaches and pointing out their differences. Nevertheless, we are convinced that the two main elements of our study, that is the presentation of a viable and proper method for including a high-resolution topography in the retrieval and the detailed analysis of the sensitivity of airmass factors and NO_2 columns to these changes, are still valuable in providing a better understanding of the differences between the different data products for the satellite community. Furthermore, we demonstrate that a proper treatment of the topography is important for specific areas next to mountain ranges such as the Po Valley, and it is unclear how other approaches followed by other groups will affect the NO_2 columns over such areas. In our study we document the effect for different seasons (and cloud cover thresholds) based on a complete reanalysis of two years of OMI observations. We will rework the introduction section accordingly and adjust the title of the article to refer more specifically to the OMI NO_2 retrieval.

P784 22-29: Here it is mentioned that the DOMINO algorithm uses the same coarse resolution data set to obtain the a priori NO2 profiles and surface pressure to ensure consistency. Does this mean that the accuracy in the retrieved NO2 VTC is in general better using this "DOMINO" method? Otherwise, what would be the point of using this "DOMINO" method to ensure consistency, as compared to the use of effective pixel average terrain heights based on GTOPO data, in terms of the NO2 VTC accuracy?

Author response: This consistency simply avoided further computing efforts in adjustment of a priori profiles to match the surface pressure data set. Our study would not make much sense if we thought that this is the best way of dealing with this problem. On the contrary, we demonstrate that this approach can lead to biases over specific areas and therefore needs to be addressed as discussed in this paper. p784, 24-25 is deleted. Changes are made as described in response to the last question.

2.2 Data and methods

P785, P786 1-12 Three different OMI NO_2 products are mentioned, but they are not clearly described. In Boersma et al., 2007, the near-real time NO2 algorithm is called

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DOMINO, but in this paper, DOMINO seems to stand for an improved off-line algorithm. This must be clarified. Why don't you start with a short description of the "standard" OMI near real time product (with reference), then introduce the improvements in the (DOMINO ??) off-line product? Also, it must be clearly described here if the treatment of the surface pressure in the AMF calculation is the only difference between the algorithm used in this study and the (DOMINO ??) off-line algorithm.

Author response: We agree that the definition is somewhat confusing. The near-real time product has been developed to make NO_2 columns rapidly available within only a few hours after measurement. After about 24 hours the near-real time data is overwritten by the more accurate offline product (Boersma et al., 2008b) and served to the scientific community through the TEMIS web site. This offline product should therefore be considered to be the "standard" DOMINO product, as described in detail in the DOMINO Product Specification Document (Boersma et al., 2008b). To avoid confusion, we now mention that the term "DOMINO" refers to the offline product and only briefly describe its relation to, and differences from, the near-real-time product. P785, 24-p786, 12 will be modified in the revised manuscript as follows.

"The near-real time retrieval algorithm has been developed for the rapid generation of NO_2 columns within 3 hours of the actual OMI measurement (Boersma et al., 2007). As opposed to the near-real time product, the Dutch OMI NO_2 (DOMINO) offline product (in the following referred to as the DOMINO product) is a more accurate post-processing data set based on a more complete set of OMI orbits, improved Level 1B (ir)radiance data (collection 3, Dobber et al., 2008), analyzed meteorological fields rather than forecast data, and actual spacecraft data. These improvements make the offline product the recommended product for scientific use (Boersma et al., 2008b). The improved instrument calibration parameters used in collection 3 lead to much lower across-track variability, or stripes, in the OMI NO_2 products and therefore no de-striping is currently applied. Whenever an OMI viewing scene contains snow or ice, this is detected based on the NISE ice and snow cover data set (Nolin et al., 2005) us-

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ing passive microwave observations. The albedo from the TOMS/GOME albedo data set is then overwritten with a value of 0.6 for snow over land. Detailed descriptions of the algorithm for the DOMINO data products are given in Boersma et al. (2008b, 2009). The NO_2 vertical columns studied in this work are basically calculated in the same way as the DOMINO product data (version 1.0.2) available from ESA's TEMIS project (Tropospheric Emission Monitoring Internet Service, www.temis.nl). Deviations will be detailed in Sect. 2.2. Data is available since October 2004."

P786, 9-11 Why is the surface albedo data-set now based on Koelemeijer et al, 2003? As described by Boersma et al., 2004, the combined TOMS/GOME surface albedo data-set was used in the "standard" OMI near real time product, because this albedo data-set was considered an improvement upon the Koelemeijer et al, 2003 climatology.

Author response: The referee is right. The sentence will be deleted. The surface albedo data set used in DOMINO is from the TOMS/GOME dataset, consistent with the albedo used in the O_2-O_2 cloud retrieval (Sneep et al., 2008).

P787, 8-17 The detailed description of the layer definitions in the TM4 model using hybrid level coefficients is not very informative. Please give more information on the number of layers in the lower, middle and higher troposphere.

The description of the hybrid layers may indeed be considered as a technical detail. However, we believe it is useful to describe as it will allow the reader to fully comprehend our approach. Furthermore, hybrid or sigma vertical coordinate systems are very common and therefore this information will be useful for most other groups using NO_2 a priori profiles from models. The approximate number of levels in the troposphere and boundary layer (below 2 km) will be mentioned in the manuscript.

P788, 21-22 What is exactly plotted in Figure 2? As explained in the text, heff is calculated for every OMI pixel, i.e. it has the irregular OMI spatial resolution. Is the heff used for Fig 2 an average for a certain period?

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Author response: Yes, h_{eff} used for Fig. 2 is the terrain height difference averaged over January, 2006. The heights h_{eff} and h_{TM4} are first determined for each OMI pixel separately and then mapped onto a fine regular grid by averaging over all pixels covering a given grid cell. The terrain height differences shown in Fig. 2 thus reflect the differences at the resolution of OMI rather than the height difference at the full resolution of GTOPO. Note that despite selecting a single month (January 2006) the averaged terrain height difference does not depend on the time period chosen. The caption of Fig. 2 will be changed to better explain this.

P789, 9-12 It would be interesting to known the effect of the additional surface pressure levels in the AMF-LUT on the calculated AMF. I expect that this effect will be rather small.

Author response: Yes, this effect is negligible. P789, 9-12 is therefore deleted.

P789, 13-20 This error in the calculation of the AMF in the DOMINO product, as described here, seems to point to a major problem. Besides possible topography-related errors, this error in the DOMINO retrieval algorithm results in additional errors in the calculated NO2 VTC. How large is this additional error in the NO2 VTC for the polluted Swiss Plateau and Po Valley regions? And how can this error in the DOMINO product explain the much larger topography-related errors (30-40 percent for polluted conditions) reported in Schaub et al., 2007? In the conclusions, the authors write that the error in the calculation of the AMF in the DOMINO product results in too low box AMFs close to the ground (this is correct) and therefore in a too large sensitivity to surface pressure changes. However, looking at Fig 7 and 8, it is not obvious to me how this effect can fully explain the discrepancies between the results reported in Schaub et al., 2007 and this study? Furthermore, I would not describe the elimination of this problem in future versions of the DOMINO product as an "additional improvement".

Author response: The reasons for these differences were mentioned above. The impact of the error detected in the retrieval code on NO_2 VTC differs with location and

time. The averaged relative difference between the NO_2 VTC (2006 and 2007 fall, cloud radiance fraction <50%) before and after elimination of this error is between 10 and 23% over the Swiss Plateau and the Po valley (elimination of the error increases the air mass factors and therefore decreases the NO_2 columns). The impact is more spatially uniform in winter and summer as opposed to spring and autumn, with differences of up to 26% in winter and 14% in summer. For pixels with a cloud radiance fraction larger than 50% the error is smaller since only the a few layers close to the surface are affected. P789, 13-20 will be modified in the revised manuscript.

2.3 Data and methods

P790, 1-10 Did the authors use additional snow cover data to make sure that partly snow covered OMI pixels were excluded in the calculation of the monthly mean maps in Fig 3 and Fig 4? The surface albedo climatology used in the retrieval algorithm does not provide information on the actual snow cover, and partly snow covered OMI measurements could be included in the 50% cloud radiance threshold case.

Author response: Yes, we did account for actual snow cover. The snow cover data used in the latest version of DOMINO is based on the near-real-time daily global ice concentration and snow extent product (NISE) (Nolin et al., 2005) using passive microwave data. Albedo values from the TOMS/GOME albedo data set are being overwritten with a value of 0.6 for snow over land as described by Boersma et al. (2008b). Therefore, only pixels with albedo smaller than 0.6 are selected to exclude snow covered OMI measurements. This will be described in the revised manuscript.

P791, 24-25 Please include an explanation why the box AMF near the ground increases when the surface pressure is decreased (Fig 7). This explanation is missing in section 3.2.

Author response: The effects are in fact interesting and should be explained. For a given atmospheric layer at a fixed altitude, the box AMF is reduced over an elevated surface because fewer photons will be scattered from the atmospheric below. For a

layer at a fixed altitude relative to the surface (e.g. 0-100 m above ground), however, the box AMF will be higher over an elevated surface than over a low surface because fewer photons are scattered by the atmosphere above and hence more light reaches the surface. This will be described in the revised manuscript.

P792-P793 As described by the authors, the topography-related error for cloud free pixels is relatively small (<=8%) compared to the partly cloudy case. However, there is a relatively long discussion on this error in Section 3.2. I suggest to shorten this section (or combine with section 3.3) by removing the detailed analysis for different retrieval parameters (Fig. 9 and Table 1). A short explanation of the effect for an averaged summer profile (4%) and winter profile (8%) should be sufficient.

Author response: We agree that the topography-related error for cloud free pixels is relatively small. However the detailed analysis for different retrieval parameters with Fig. 9 and Table 1 provides deeper insight to the problem compared to our previous study. In addition to the a priori profile effect discussed in Schaub et al., 2007, the solar zenith angle is found to contribute significantly to the difference between winter and summer. Furthermore, Fig. 9 illustrates the sensitivity of the topography-related error to the surface pressure change which allows the reader to inter- or extrapolate the results to other situations than those discussed here. Therefore, section 3.2 is shortened as suggested while keeping Fig. 9 and Table 1.

p793, 7-19 will be modified to "A similar analysis was made for the other two retrieval parameters changing strongly with season, solar zenith angle and albedo. As seen in Fig. 9b, for the larger SZA in winter, the relative changes in AMF_{trop}) and NO_2 VTCs are more sensitive to differences between p_{TM4} and p_{eff} than for the smaller angles in summer. This effect thus adds to the differences observed between winter and summer. Fig. 9c shows that, in contrast to the two previous parameters, the sensitivity of relative changes in AMFtrop and NO_2 VTCs to changing surface pressure is almost the same in winter and in summer, even though the largely different albedos have a significant effect on the absolute values of the AMF_{trop})".

P793, 1-3 The results from this study are not consistent with the results reported in Schaub et al., 2007. The topography-related error for cloud free pixels reported in Schaub et al., 2007 is 3 to 5 times larger than the one reported in this study. Is this only due the error in the calculation of the AMF in the DOMINO product, as mentioned in Section 2.2? Here it should also be noted that Fig 13 in Schaub et al. 2007 is not correct: it gives a wrong impression of the topography-related effect (in contrast to figures 7 and 8 in this paper, which are correct). Since this paper presents a follow-up study to Schaub et al. 2007, the problems with this previous study should be clearly described.

Author response: The reasons for this discrepancy were already discussed above and will be more explicitly described in the revised manuscript. We agree that the explanation for the effect as provided by Fig. 13 in Schaub et al. (2007) is oversimplified and not correct. Shifting the surface to a lower altitude does not cause an extension of the sensitivity profile to the lower altitude but rather the sensitivity profile changes as shown in our figures 7 and 8. This was already mentioned in the manuscript but obviously needs to be stated more clearly.

The following changes will be made:

1. p793, 2-3, "in agreement with the findings of Schaub et al. (2007)" is deleted.

2. Title of section 5 changed from "Conclusions" to "Discussion and Conclusions"

3. p801, 8-13 changed to "These findings differ from those of our previous study published by Schaub et al. (2007), which estimated the topography-related error to 13-38% for cloud free pixels over the Swiss plateau."

4. The discussion section will be modified as follows: The main reason for this discrepancy is the error detected in the interpolation of box AMFs from the lookup table described earlier. The results by Schaub et al. (2007) were based on a few selected cases only with specific p_{surf} from TM4 model and p_{eff} from aLMo model. Unfortunately,

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the pressures p_{eff} of the selected pixels ($\sim$ 960hPa) were in a range where interpolation errors in near-surface box AMFs maximized, while errors at p_{surf} were much smaller. This lower part of the box AMFs profile is particularly important in determining the AMF$_{\mathrm{trop}}$ following Eq. (1), since the a priori NO$_2$ profile x_a has highest values in the boundary layer close to the ground as shown in Fig. 8. Moving the surface down into a region where the errors were much larger therefore resulted in too large changes in AMFs and NO$_2$ VTCs in their sensitivity study. Schaub et al. (2007) tried to explain this high sensitivity by assuming that with the surface level shifted to a lower altitude the profile of box air mass factors would be a simple extension of the original profile to the lower altitude. This would result in a strong reduction in box airmass factors near the surface and hence a strong sensitivity of the AMF$_{\mathrm{trop}}$ to surface pressure changes. However, this explanation is oversimplified and not correct since the profiles are not extended but rather rescaled to the new surface altitude as shown in our Fig. 7 and 8, which results in a more moderate sensitivity.

P794, 2-4 Why has a fixed cloud fraction of 15% been selected? It would be interesting to know how the effect on the AMF changes for this particular pixel when varying the cloud radiance fraction from 0% to 50%.

Author response: Thanks for this good point. There is no special reason for choosing a cloud fraction of 15% (which corresponds to a cloud radiance fraction of 38% for this particular pixel), except that it is not an extreme value within the considered cloud radiance fraction range (0-50%). A discussion on the effect of a varying cloud radiance fraction (0-50%) on the sensitivity of the change in NO$_2$ VTC to a change in surface will be added in the revised manuscript.

P794 The retrieval error due to an inaccurate surface pressure as described in section 3.3, should be related to the error in the retrieved OMI cloud parameters, especially the error in the cloud top pressure. Figure 12 shows that for a cloud top located inside the polluted boundary layer (at 900 hPa), the retrieval error due to an inaccurate surface pressure is large (30%), even if the error in the surface pressure is only 20 hPa!

However, the uncertainty in the OMI cloud-top pressure is probably larger than 20 hPa, especially for small cloud fractions (0-20%). How large is the uncertainty in the OMI cloud-top pressure for small cloud fractions? How does the uncertainty in the OMI cloud-top pressure effects the error analyses presented in this section? This issue should be discussed as well in this section.

Author response: The uncertainty in cloud parameters is clearly an important error source in current trace gas retrievals. This uncertainty exists irrespective of errors in surface pressure and is therefore not a focus of the present work. In our study we point out that air mass factors are particularly sensitive to surface pressure errors if cloud tops are located inside the boundary layer. The same will be true for errors in cloud top since, depending on the estimated cloud pressure, a larger or smaller fraction of the polluted boundary layer is shielded by the cloud. The referee is right that a short discussion of this would be helpful. This will be added in the revised version. The uncertainty in cloud top pressure is clearly larger than 20 hPa, in particular for small cloud fractions (see e.g. Sneep et al., 2008). However, as mentioned before, this uncertainty exists irrespective of the surface pressure error and should therefore not be prevent us from treating the surface topography in an adequate way. On the contrary, it highlights the requirement that the surface topography should be treated in a consistent way by both the cloud and trace gas retrieval. This is the case for our new method since a high resolution topography data set has already been used by the cloud retrieval.

2.4 Validation

P796, 1-7 It is not clear to me if molybdenum converters are sensitive to NOx (NO2+NO) or to NO2 only (besides the sensitivity to other odd nitrogen species such as PAN, HNO3 and organic nitrates)

Author response: Thanks for the good point. Chemiluminescence detectors can only measure NO. Other species like NO_2 therefore need to be converted to NO before

detection. The converter thus only converts NO_2 (and eventually other species) but the chemiluminescence analyzer behind the converter then measures the sum of NO + converted NO_2. An additional measurement of NO only is obtained by bypassing the converter. NO2 is then the difference between the measurements with and without converter. The text will be edited accordingly to avoid confusion.

P798,8 Are additional actual snow cover data used to exclude (partly) snow covered OMI measurements, or is the selection only done via the cloud fraction and the surface albedo climatology used in the OMI retrieval algorithm?

Author response: As mentioned above, actual snow cover data were used.

P798, 15-17 It is understandable that very high correlation coefficients (> 0.7-0.8) are not to be expected when combining OMI NO2 VTC and in-situ measurements. However, I do not understand why measurements with correlation coefficients of 0.3-0.5 are already considered "well correlated".

Author response: These values of 0.3 - 0.5 refer to the coefficient of determination r^2 and not to the correlation coefficient r as used by others (Ordóñez et al., 2006; Bucsela et al., 2006; Lamsal et al., 2008). Unfortunately, we did not use these terms in a proper way. A value of r^2 = 0.3 corresponds to r = 0.55 which in other studies (e.g. Lamsal et al., 2008) was already considered to be a "good" correlation even though only 30% of the variance can be explained. To be more consistent with other studies we will use r instead of r^2 and revise the text and Figures accordingly. In our study, for most of the studies, r is within the range of 0.6 - 0.82.

P800, 8-10 Please also explain why the slope and r2 for Motta are improved with peff, while the corresponding monthly mean comparison is not improved.

Author response: Thanks for the good point. Sentences are added at P800, 13 for explanation. "This may be explained by the smaller weights in the regression analysis for the points with strong overestimation due to their high uncertainties."

General remark

Neither this paper nor Straub et al., 2007 give information about possible errors related to the other important surface parameter; the surface albedo. Why is there no discussion on possible surface albedo related errors for this region in this paper? How large is the impact of surface albedo uncertainties on the NO2 VTC for this region, compared to the topography related errors?

Author response: We agree that albedo is another and clearly more critical parameter for the retrieval. Discussions on possible surface albedo related errors are added in the discussion and conclusions section. However, a quantitative study of the impact of surface albedo on the retrieval is beyond the scope of this study but it is the topic of our current work. The work on surface pressure should be understood as one element of our work on improving the external parameters used for the retrieval and is only the first step in this process. We will point this out more clearly and mention that surface albedo will be analyzed in a follow-up study. More changes about the further work can be found in the response to the question 2 from referee 2.

Minor Comments

P783, 9-13 Please add the satellite platforms of the instruments mentioned here.
Will be added.

P783, 17 It is better to write "the gradually improving spatial resolution"
Changed

P784, 3-5 Aerosol have a large impact on the AMF as well, and therefore aerosol properties are also key input parameters for the AMF calculation.

The sentence will be changes as follows: "Key input parameters for the calculation of the tropospheric air mass factor are cloud fraction, surface albedo, aerosol, and a priori NO2 profile shape, each having its own uncertainty."

P784, 5-6 Is it really a new type of error that was identified by Boersma et al, 2007? To

my knowledge, these types of errors were already discussed for the TOMS instruments. Therefore, it is better to write: "Boersma et al. 2007 described the type of errors . . ."

Changed to "Boersma et al. 2007 described an additional error source".

P788, 7-8 The cloud fraction (fcl) and the cloud top pressure (pc) are retrieved, not the cloud top height.

Sentence will be changed as suggested.

Reference

Sneep, M., De Haan, J. F., Stammes, P., Wang, P., Vanbauce, C., Joiner, J., Vasilkov, A. P., and Levelt, P. F.: Three-way comparison between OMI and PARASOL cloud pressure products, J. Geophys. Res., D15S23, doi:10.1029/2007JD008694, 2008.

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