

Reply to comments of reviewer #2 on the manuscript “An improved NO₂ retrieval for the GOME-2 satellite instrument” by A. Richter et al.

First of all, we would like to thank the reviewer for his detailed and critical comments which lead us to some interesting additional tests and retrievals. We have tried to take up as many of the suggestions as possible but – not surprisingly – do not agree with all of them.

The status of the algorithm improvements is unclear for the reader. In some parts of the manuscript they are presented as algorithm improvements, whereas the first sentence of the conclusions presents it as a new product. In the discussion on the solar spectrum it is concluded that it is up to the user if a fixed or daily solar spectrum is used, indicating that no clear choices have been made. The authors should state clearly from the beginning and consistently in the manuscript if they are presenting a new product or an improvement to a (part of the) algorithm. If it is a new it should be clear how it relates to other products and where the data is made available for users. Also if this is a new product a clear description on the air mass factor calculations shall be included.

This paper is intended as a contribution to the continuing improvement of satellite retrievals of atmospheric NO₂. It reports on some new approaches taken in a scientific retrieval performed at the University of Bremen and some of the results obtained. It is not intended as a product description as it would be published by an agency providing operational data products to users. It also explicitly excludes the aspect of air mass factors as this is not the topic of the current study. In this sense, it is an algorithm improvement. However, the algorithm has been applied to 4 years of GOME-2 data resulting in an improved set of slant columns, a first application of which has been shown and in that sense, we thought it is also a report on a new data product.

More details on the recommendation for use of different solar spectra have also been requested by the other reviewer. Our main conclusion from the study is, that a consistent data set covering the complete GOME-2 time series should use the daily solar measurements, However, better accuracy for shorter time periods (up to several months) can be obtained by using a single solar measurement. Which of the two options is the better choice depends on the individual study. Also, for each such study a separate solar spectrum has to be selected, necessitating a completely new run of the analysis. See also more discussion of solar spectra below.

In response to the reviewer’s comments, we have

- * rephrased several sentences avoiding the term “product”,
- * expanded the discussion on use of solar spectra.

The part on the “soil feature” is not convincing and cannot be published in the current way. I conclude that the soil feature in fact is absorption by O₄. The presented map is very similar to maps of the O₄ column showing highest values for reflective low altitude surfaces.

The reviewer is right in pointing out the similarity between the O₄ cross-section and the soil correction term. In fact, this is a worry and has already been briefly discussed in the original paper.

However, we do not believe that the soil signal is just O₄ absorption for several reasons:

Most importantly, the O₄ maps are not “very similar” to those of the soil signal. On the contrary, the highest O₄ columns are observed over cold bright surfaces (snow, ice) where we do not detect any soil signal. This is shown in Figure 1 for one day of not cloud screened

data showing a) the sand signal and b) the O4 slant column from a fit identical to our improved fit but without the sand signature (to avoid a possible correlation as suggested by the reviewer)

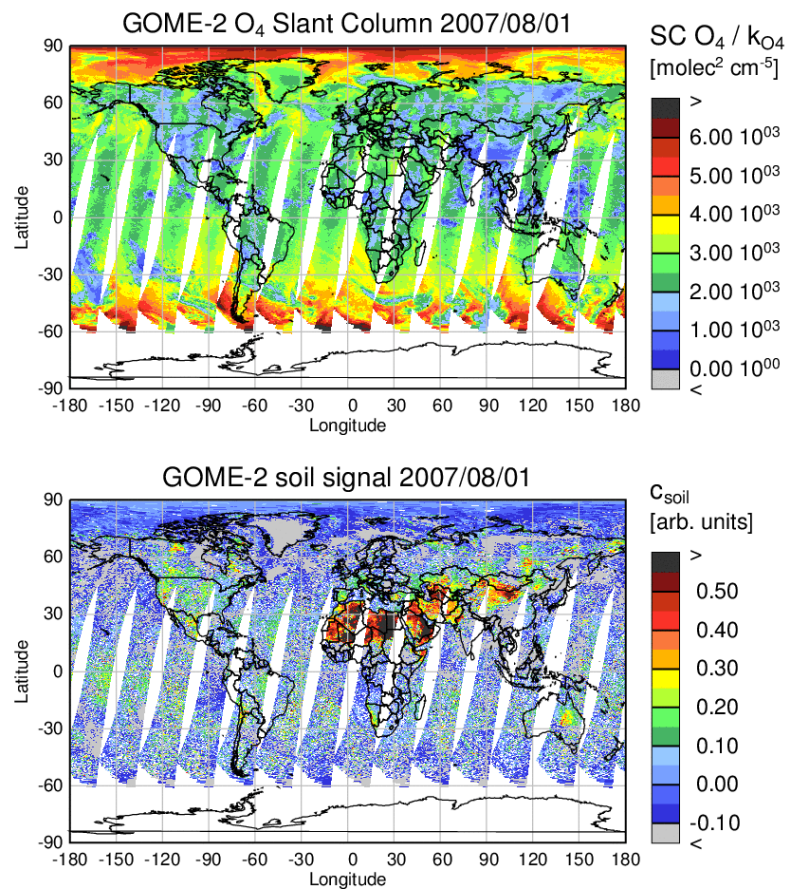


Figure 1: One day of GOME-2 retrievals showing slant O₄ columns (top) and the fit parameter of the soil signature (bottom). In the fit used for the O₄-plot, no soil signature was included.

Second, as discussed below, a change of the O₄ cross-section doesn't change the soil pattern.

Third, there are many incidences of large O₄ columns over water or ice (Greenland), but the soil signal is large only over regions with bare soil.

In the following, we will reply to the individual points made by the reviewer. We have not included the additional figures in the manuscript, but have extended the discussion of the soil feature to reflect the points raised here.

The main reasons that the "soil feature" is not convincing:

-The "soil signal" presented in figure 6 shows very strong correlation with the O₄ absorption in this wavelength region. Note also that an outdated O₄ reference spectrum by Greenblatt is used in the fit.

As discussed above, we agree that the two signals are similar but by no means identical, and therefore do not lead to similar maps as shown in Figure 1.

The reviewer is right that we have used an old O₄ reference spectrum, as in our experience, it leads to good fitting results. In response to the reviewer's comment, we have repeated the analysis on one month of data using the more recent cross-sections from Hermans et al. The results for the soil parameter are offset by about 0.1, but show the same spatial pattern as

before (see Figure 2). This indicates that a) there is some cross-talk between O₄ and the soil effect but b) the large signals over soil are not an artefact of using a specific O₄ cross-section. It should be noted that the fitting residuals are slightly poorer when using the Hermans et al. cross-section.

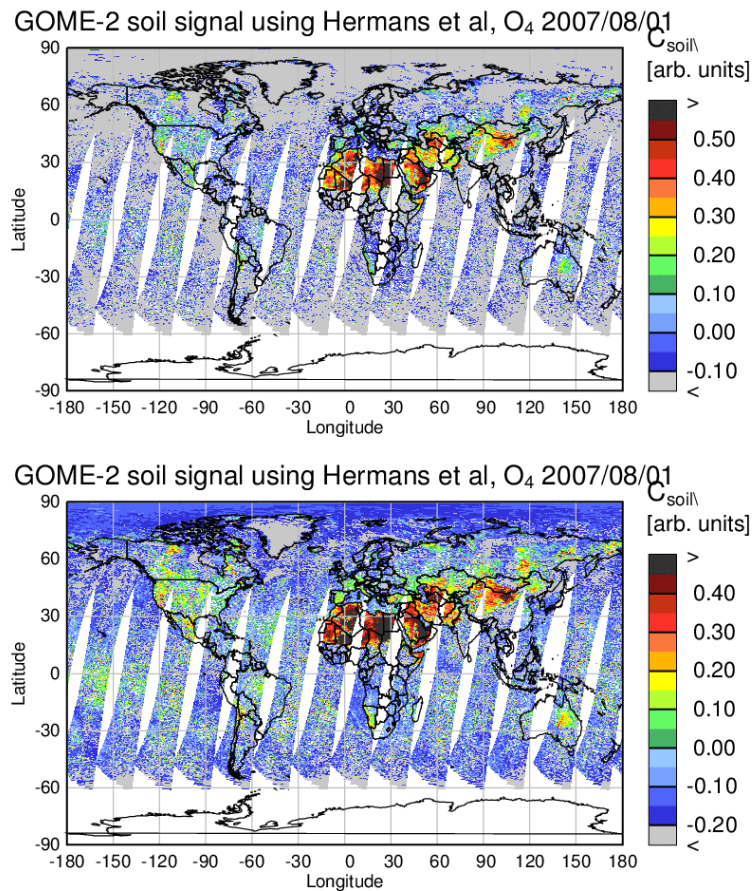


Figure 2: Soil signal retrieved when using the O₄ cross-section of Hermans et al. in place of Greenblatt et al. The top panel uses the same colour scale as Figure 1, the bottom panel is offset for clarity.

-The GOME-2 “soil signal” shown in Figure 7 shows no signal over the Tibetan plateau, which does show up in albedo maps and visible imagery as a high reflectance region with a lot of bare soil. The high altitude of this region is the cause for low columns of O₄ and thus for the absence of the signal in Figure 7.

We agree that over all high altitude regions, the soil signal is lower (this is also true over Greenland). This is indicative of some cross-talk, and we have added this point in the paper. As shown above, this does not imply that the overall signature is an O₄ correlation.

-I tried to find the soil feature in several of the relevant spectra in the Aster database (<http://speclib.jpl.nasa.gov/>) but I could not find any.

We had searched this data base as well but at least those data sets that we looked at did clearly not have enough signal-to-noise-ratio to see such structures in the 400 – 500 nm region where differential structures in the data base are dominated by noise.

As discussed in the reply to the other reviewer, we have tried to measure the sand signal with a DOAS instrument on the roof of our building using some sand from the Sahara. The results show a structure similar to that deduced from the GOME-2 spectra but without the sharp peak. Use of this spectrum in the GOME-2 data analysis leads to similar maps for the sand-coefficient but significantly poorer fits. As this is based on a single quick measurement,

we would like to do some more measurements using a better set-up and different sand targets to gain more confidence in the results before publishing the data.

The discussion on the fit window selection is incomplete. It describes that several reference spectra have been added to the fit. However, according to Table 1 also the order of the polynomial was reduced from fourth to second order, for reasons not discussed.

Thank you for pointing out this omission. The degree of polynomial in the original fit was quite large as smaller degrees lead to poor fits over some region, most notably over deserts (see Figure 3). We believe that this is because of signals from liquid water absorption and spectral surface reflectance not explicitly accounted for in the original retrieval. By including these cross-sections in the fit, a lower degree polynomial degree could be used, replacing a closure term by physically meaningful terms while maintaining good fitting quality.

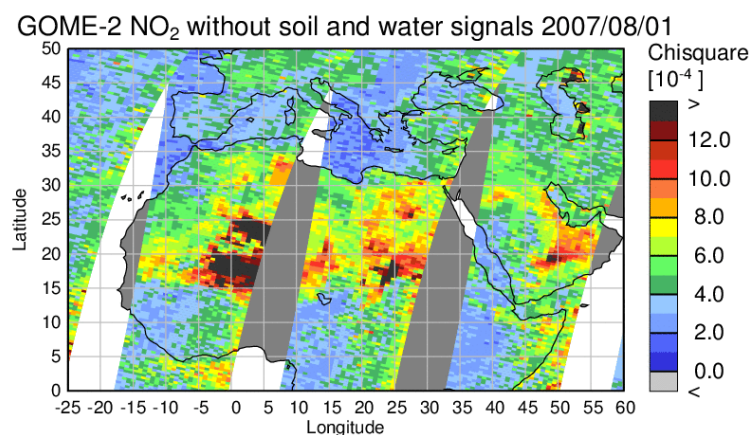


Figure 3: Fitting residual for one day of data using a fit without the liquid water, soil, and calibration cross-sections.

In response to the reviewer's comment, we have added a paragraph on the polynomial.

For new reference spectra it should be shown that the resulting fit parameters give meaningful physical results and that the correlation between fit parameters is reasonable to give independent results.

We agree with the reviewer and have added a figure showing the retrieved fit parameter for liquid water absorption which shows the expected behaviour indicating that it gives meaningful physical results. The fit result for the surface signature was already included in the original paper. We have not included an example of the fit results for the calibration function as this is only a very minor contribution to the overall fit (and in fact has nearly no effect on NO₂ columns):

In addition, other aspects regarding the length of the fitting window shall be discussed in the manuscript:

-Broader fit windows have larger error due to co-alignment errors (the fact that different wavelengths do not see exactly the same area on ground)

This is an interesting point which depends critically on details of the instrument design. We are not aware of any detailed study of this effect in GOME-2 measurements but expect it to be rather small considering the large footprint of 40 x 80 km² which already averages over a large area. We have added a note on this to the manuscript.

-For broader fit windows the assumption in the AMF calculation that the albedo is independent of wavelength becomes increasingly inaccurate.

We agree with the reviewer and actually had looked into this point using the Kleipool et al. surface reflectance climatology before. We added a comment on this to the manuscript.

-For broader fit windows the assumption of a wavelength independent effective cloud fraction (or radiance cloud fraction) becomes increasingly inaccurate due the variation in Rayleigh scattering.

We agree in principle but believe that this is a small effect compared to many other inaccuracies in the treatment of clouds. We have added a comment on this in the manuscript.

The spike removal procedure is an ad-hoc method that is mathematically questionable. In the manuscript the authors also conclude that this procedure can bias the data noting that “the spike removal approach always needs to be carefully monitored to avoid biasing the data” (page 224, line 14). (Note that no information is given how the careful monitoring is done in practice).

In our opinion, any algorithm removing data points from a measurement has the potential to bias the results. An algorithm that cannot change the results is hardly useful. The critical point here therefore is to make sure, that changes in the results occur only where we have reason to believe that they are improving the results while other parts of the measurements should remain unchanged. This is also the way to monitor the results – one checks if the NO₂ results remain constant in regions outside the SAA. We have added this information to the manuscript.

There are other procedures that should be applied to remove the spikes before the data is adjusted to the model, which is what the proposed method does. The duration of the spikes in the SAA is very short, meaning that for one detector pixel they are only present in one measurement and not in the previous or the next. Therefore procedures have been developed that use the variation in time for the detector pixels to filter out these spikes. Such procedures have been successfully applied to other satellite instruments. This method should be used also for GOME-2 (preferably in the Level0-1B processing step), before using the ad-hoc method described in the paper. The authors should investigate the application of the time variation procedure for GOME-2 and present results in the manuscript.

First of all we would like to thank the reviewer for making us aware of the unpublished algorithm used for de-spiking of OMI data. Judging from the results, it works very well on OMI data and we think these algorithms should be published to make them available for other instruments to use.

Following the recommendation of the reviewer, we have implemented a simplified version of the OMI algorithm in our analysis and applied it to some orbits. There are some parameters which can be varied and we have tried to find a good choice but cannot be sure that this is in line with what was chosen for OMI. In Figure 4, we show the results for the original small window fit using no correction, the OMI type correction and our own correction. As can be seen, both algorithms are quite successful in removing outliers, but our algorithm produces slightly better results.

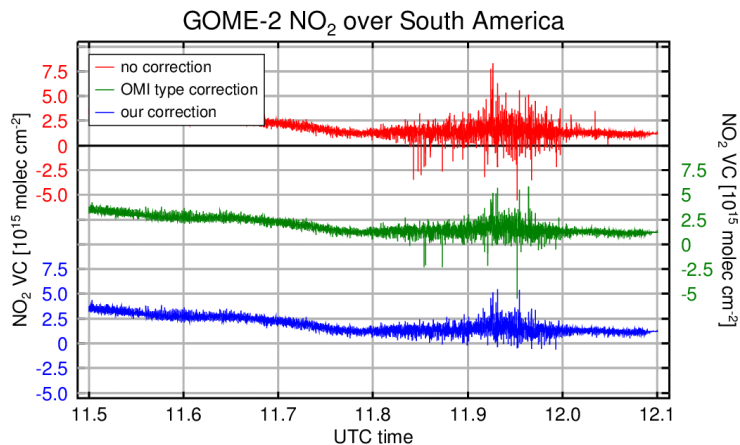


Figure 4: Comparison of the effect of the two spike correction algorithms on one orbit of GOME-2 data passing through the SAA. The fits are using the standard NO₂ fitting window from 425 – 450 nm and the settings given in the paper.

We believe that the OMI algorithm will not work as well on GOME-2 data as on OMI data for two reasons. First, OMI uses a 2-dimensional array and therefore spikes can be identified in both the direction of rows and columns while GOME-2 only has a column. Second, and more importantly, the large full well depth of the diode array used in GOME-2 implies that the relative size of a spike in the SAA is much smaller for GOME-2 than on the CCD employed by OMI. It therefore often is of a similar size as measurement-to-measurement changes induced by the Ring effect and much less easy to identify. In fact, we routinely use an algorithm as that applied to OMI data on our CCD-equipped ground-based spectrometers where it works very well but not on our instruments using diode arrays.

In summary, we agree that it would be good to implement an OMI-like algorithm to the GOME-2 level-0 to level-1 processing. However, we also believe that our algorithm is even more successful in removing spikes in the SAA as in the absence of structures from changing Ring-effect, it can identify smaller spikes. We have mentioned the OMI algorithm in the revised paper but do not present our detailed results as our point here is not to compare our algorithm to (our version of) the OMI algorithm but to show that with our algorithm, we can significantly reduce the scatter of our results.

The authors describe investigations using a fixed or daily solar spectrum in their analyses. As they clearly show an impact of the choice, they do not indicate what the probable causes in the solar spectra are. For example are these caused by spectral features from the diffuser that interfere with the absorption cross sections, or the fact that the random noise in the solar spectrum turns into systematic errors when it used to normalize all the radiance measurements? Even a speculation of the authors would be valuable.

We believe that a correlation between spectral structures induced by the diffuser or the mesh in the solar irradiance light path and absorber cross-sections is the reason for the effects observed. We have added this speculation in the revised paper.

In the description of the daily vs fixed solar spectrum it seems that everything calls for a fixed spectrum, except for the long-term stability of the instrument. A logical solution would be to use a running mean solar spectrum (e.g. a month or longer), to benefit from more than daily measurements, but to capture the long-term variations. The authors should address this in the manuscript and show results why this will or will not work.

We agree that using a running mean over solar spectra would smooth out the effects of changes in solar spectrum and reduce the loss in fitting quality. However, it would not really

solve the problem but just make it a bit smaller. We do not think that anything can be learned from showing the results of using an averaged solar measurements as this is an additive effect and averaging the input (solar irradiance) leads to the same results as averaging the results (offset of the NO₂ slant columns relative to a reference).

On request of both reviewers, we have extended our discussion on the solar spectrum and have now also included the suggestion made by the reviewer.

The focus of the manuscript is on the NO₂ slant column fit. Therefore the authors shall present and discuss relevant details of the fitting procedure, including the fit function.

As we have been using a standard retrieval approach, actually the same code as used in our first publication on GOME BrO in 1998, we don't think that it makes sense to add more than a few words on this.

The OMI NO₂ retrieval algorithms have used a wide fit window since launch for the same reason as it is now introduced for GOME-2. The authors should make a reference to the relevant papers in section 3.

Good point. We have now included references to the OMI NO₂ fit.

In section 5 the new GOME-2 data are compared to SCIAMACHY and the conclusion is that they compare very well. However, the fitting procedures between the new GOME-2 and SCIAMACHY algorithms differ, due to the presented. This contradicts to the approach presented on page 217, line 8. In addition, no information is provided on the air mass factor part of the retrieval. Given the fact that different fit windows are used, the surface albedo's should also differ. How do these differences affect the comparison?

On page 217, line 8, we said that the starting point of the GOME-2 retrieval was to use the same settings as for SCIAMACHY. The whole point of this paper is to present results of an improved retrieval, and to us, it then makes sense to compare that to SCIAMACHY data. We do not see any contradiction here.

With respect to the AMF, we used a purely stratospheric AMF as stated in the text. As stratospheric NO₂ is located at relatively high altitude, the effect of surface albedo is small in regions with low tropospheric column content as is expected as over the Pacific. We have added this point to the revised paper.

The final statement of section 5 is very weak. The conclusion seems to be that the two approaches (daily or fixed solar spectrum) are both non-ideal. Instead of this statement it should be concluded that more effort is needed to come to better solution.

As mentioned above, we have extended this section and tried to make it more conclusive.

According to the text figure 6 is the natural logarithm of the ratio of the spectra (p221, line 16). In the caption of Figure 6 this is not mentioned. Which one is right, the text or the caption?

Thank you for pointing this out. The text was correct and we have changed the figure label and caption accordingly.