Reply to comments of reviewer #1 on the manuscript "Operational total and tropospheric NO_2 column retrieval for GOME-2" by P. Valks et al.

We would like to thank the reviewer for his helpful comments and suggestions. In the following, we will reply to them point by point, including the reviewer's text in italic.

P1619: Add references for tropospheric NOx chemistry.
We have include two references (Seinfeld and Pandis, 1998; Jacob et al., 1996) in Section 1.

P1619 L26: "daily basis": clarify that ERS-2 does not provide daily coverage!
We have improved the description of the spatial/temporal coverage of GOME/ERS-2.

P1621 L27f: make clear that this is the default.
Done.

P1624 L7: So far, the model is fully linear. It could be noted that it becomes nonlinear by shift/squeeze, which is described later.
We have clarified in the 2nd paragraph of P1625 that the fit becomes non-linear by including shift and squeeze parameters.

P1625 2nd paragraph and P1626 1st paragraph both deal with shift. I recommend to have both of them consecutively.
We have reorganised Section 3 as suggested.

P1625 3rd paragraph: add a reference for the determination of the Ring reference spectrum.
To our knowledge, the reference on the determination of the Ring reference spectrum (Change and Spurr, 1997) used in the paper is the most appropriate.

P1625 L27: "to correct": add "partly"
Done. Intensity offset effects can not be fully corrected with this approach.

P1629 2nd paragraph: please comment on possible alternatives - do you plan to use the Kleipool LER in future?
That is correct. We plan to use a new surface albedo climatology in a next version of both the operational GOME-2 NO_2 and cloud algorithms. Besides the OMI LER climatology (Kleinpool et al., JGR, 2008), there is also the possibility to use the MERIS albedo climatology (Popp et al., AMT, 2011). The main advantage of the MERIS albedo data-set is the overpass time of 10:30 LT, which is close to that of GOME-2. Furthermore, the MERIS albedo data-set can be used for the OCRA/ROCINN cloud retrieval in the O2 A-band. The use of a new surface albedo climatology is discussed in the outlook paragraph of Section 9 in the updated paper (see also last comment below).

P1630 L10: This depends on the actual profiles of NO2, aerosols and clouds as well as on the aerosol properties as SSA!
We agree. We have reformulated this sentence (removed second part) and refer to Sect 6.3 instead, for a discussion on the effect of aerosols on the cloud retrieval.

P1631 L5: The first step is the calculation of total VCD via Eq. 2 with a stratospheric AMF.

We agree that this is in fact the first step, and that this sentence in the manuscript can be confusing. A sentence has been added to clarify this point.

Section 5.1: For any stratospheric correction procedure, the stratosphere might be overestimated, which causes negative tropospheric columns. This has to be mentioned, and plots and maps should not hide negative values, if they occur! For instance on 22 Feb 2008 (Fig. 1), the estimated stratospheric column is higher than the observed total column over western Europe, thus also at OHP! Negative tropospheric columns for single pixels and in the monthly mean provide additional information on the uncertainty of the stratospheric correction, see Beirle et al., 2010, AMT.

The reviewer is right that an overestimation of the stratospheric NO_2 column in the stratospheric correction procedure can lead to negative tropospheric NO_2 columns. In general, negative tropospheric columns are a result of (mostly) systematic errors in the stratosphere-troposphere separation and random noise in the NO_2 slant columns (mainly due to photoelectron shot noise).

We have changed Fig 3 and 4 in the paper to better show the areas with negative tropospheric columns (see comment on "*Fig 4"* below). However, the application of tropospheric air mass factors to derive the vertical tropospheric columns, the effect of clouds and the averaging involved in the calculation of monthly averaged values, makes it difficult to interpret negative values in monthly tropopsheric NO_2 distributions (such as Fig. 3 and 4 in the paper). We think that it not within the scope of this paper to include a detailed discussion on negative NO_2 columns.

Concerning Fig.1: we have expanded the discussion on the stratospheric NO_2 distribution on 22 Feb 2008 in Sect. 5.1 and 6.2 regarding the overestimation the stratospheric column over Western Europe and the resulting negative tropospheric columns (see comment on "*P1639 L22"* below)

P1632 L14: This is much lower than the value given in Martin et al., 2002, please comment on that.

In the current version of the GDP, a simple correction is applied for background NO_2 in the free troposphere, by using a fixed background NO_2 column (0.1×10^{15} molec/cm^2). This an averaged tropospheric vertical NO_2 column for the (unpolluted) Pacific region derived from MOZART model data. Martin et al., 2002 mention mean tropospheric slant columns of ~ 0.35×10^{15} molec/cm^2 for the (unpolluted) tropical Pacific derived from GEOS-CHEM model data. Assuming an AMF of ~2.2, this corresponds to a mean tropospheric vertical column of ~0.15×10^{15} molec/cm^2. This is indeed higher than the value used in our study, but considering model uncertainties and the overestimation of the GEOS-CHEM NO_2 concentrations over the Pacific compared to aircraft observations as described in Martin et al., 2002, the value of 0.1×10^{15} molec/cm^2 used in our study seems to be a reasonable estimate. In the future, the stratospheric correction could be improved by taking the variability in the background NO_2 column into account.

P1634 5.3: Global cover is lost for GOME, but particularly Europe is still covered. Thus I recommend to compare GOME and GOME2 for the same month over Europe.

It is correct that there is still good coverage of Europe and North America with GOME in 2007. However, there is little coverage over Asia and no coverage of the Pacific region. Therefore applying the stratospheric correction method to GOME over the northern mid-latitudes will lead to systematic errors, since large parts of the covered areas are masked-out in the spatial filtering approach. Therefore, GOME/ERS-2 data for the period 1997-2000 are used in this paper.

P1639 L22: Please show the Mozart fields for the day shown in Fig. 1 (22 Feb 2008) (instead or additionally) to see whether the model would be able to avoid/reduce the stratospheric overestimation over Europe.

Fig.1 below shows the IFS-MOZART field for 22 Feb. 2008 and the one retrieved with the spatial filtering method, using synthetic slant column derived from IFS-MOZART model data as input. Comparing this figure with Fig.1 in the paper shows that the IFS-MOZART model provides a very nice representation of the stratospheric field on this day. It is also clear from both figures that the spatial filtering/masking approach used in the GDP overestimates the stratospheric NO_2 column over Western Europe in this situation. The main problem for the spatial filtering/masking approach here is the large gradient in the stratospheric column over a polluted region (which is masked-out). There seems no other way than using on-line model results (or limb measurements) to get the stratosphere right in these situations.

We have included this picture in the discussion on the regional and seasonal variability in the stratospheric column uncertainty in Section 6.2 (see next comment), as an example where the spatial filtering method can result in large errors in the stratospheric column.

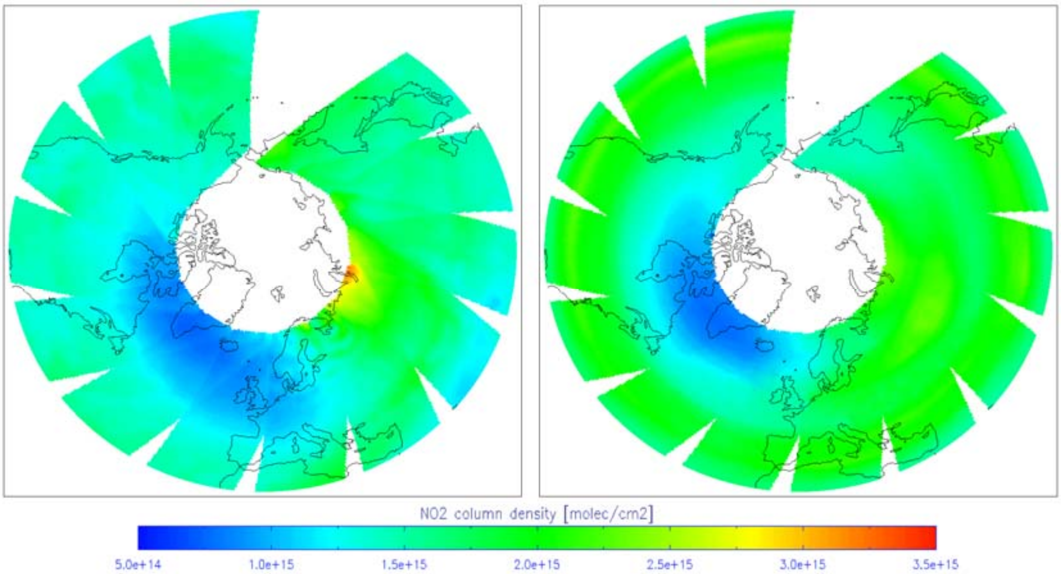

Fig. 1. Stratospheric NO_2 columns from the IFS-MOZART reanalysis model for 22 February 2008 (Left) and those retrieved with the spatial filtering method (as used in the GDP 4.4), using synthetic slant column derived from IFS-MOZART model data as input (Right).

P1640 L7-15: The given uncertainty is quite small, but I expect that it has a strong seasonal and regional dependency. Especially in winter close to the polar vortex, it will be much higher, which has an impact of several regions of general interest for NO2 analysis, i.e. Europe and the US eastcoast. Thus, please also mention regional/seasonal maximal uncertainties. In addition, it is obvious that the uncertainty of the stratospheric column is higher over polluted regions that have been masked out and interpolated. Thus, I recommend to calculate the uncertainty of the stratospheric column for the polluted regions alone, as this is relevant for the tropospheric product for most studies.

As already mentioned in the paper, the average uncertainty in the stratospheric NO_2 column varies from ~0.15×10^{15} molec/cm^2 for the low-latitudes and the largely unpolluted SH, to ~0.26×10^{15} molec/cm^2 for the polluted northern mid-latitudes. We agree with the reviewer that the seasonal variation should be discussed in the paper as well. The monthly average uncertainty for the northern mid-latitudes varies between 0.22×10^{15} molec/cm^2 (August) and 0.30×10^{15} molec/cm^2 (Jan). When the uncertainty is calculated for polluted regions alone, the

error increases by about 0.03×10^{15} molec/cm^2 for the northern mid-latitudes, as well as for the low-latitudes and SH.
It is clear that the maximum error on 22 Feb 2008 (Fig 1 above) is larger (~0.50×10^{15} molec/cm^2) than the maximum monthly error mentioned above. However, this a “worst case scenario” and the spatial filtering approach usually performs much better.
In the manuscript, we have extended the discussion on the regional and seasonal variability in stratospheric columns uncertainty accordingly.

P1644 L5: Please specify how far the MAXDOAS measurements help to address the validity of cloud porperties and AMF calculations.
We agree with the reviewer that cloud properties can hardly be validated using MAXDOAS measurements, however MAXDOAS data may provide useful ancillary information on the vertical distribution of both NO_2 and aerosols, which both play an important role in driving the accuracy of the AMF calculations. The sentence has been reformulated as follows: “Although not exploited in the present study, MAXDOAS instruments can provide vertically resolved information on both tropospheric NO_2 and aerosols in the lower troposphere. This may be used to the benefit of advanced validation studies where the sensitivity of satellite AMFs to uncertainties on the a-priori information on NO_2 profile shape and aerosol could be investigated.”

P1645 L23: Also the high stratospheric dynamics causes higher std of slant columns in winter.
This has been included in the text.

P1646 L13: "remain sensitive": In case of NO2 above the cloud, sensitivity is even increased!
Indeed. The sentence has been reformulated.

Table 2: CTP error of 3%: This is a highly non-Gaussian error! For low CTP, i.e. high shielding clouds, the 40 hPa will have virtually no effect on Mt, but for high CTP, a small change of CTP can have a very strong impact on Mt, if the cloud is within the NO2 layer. Please also comment on errors due to the simplification of a reflecting cloud and neglecting multiple scattering.
We agree. This have clarified in the text that the error in the AMF due to the CTP error is highly variable, and added a footnote to Table 2.
It is important to note that the cloud model used in the NO_2 retrieval is consistent with the cloud model used in the OCRA/ROCINN cloud retrieval, in the sense that both use the independent pixel approximation representing clouds as opaque Lambertian surfaces. Research on the operational use of an improved cloud model in the trace-gas column and cloud retrieval, where clouds are presented as scattering layers (which also allows full treatment of aerosols if detailed information on aerosol optical properties is available) is ongoing (Spurr et al., 2009).
In the manuscript, we have extended the discussion in Section 6.3 concerning the cloud model used in the NO_2 retrieval.

Spurr R., Zimmer W., Loyola D., Coldewey-Egbers M, Lerot C., van Roozendael, ., Lambert J-C., Granville J., M., Koukouli M., Balis D., "Clouds as Scattering Layers: Improved Retrieval of GOME-2 Total Column Products", EUMETSAT Meteorological Satellite Conference, Bath, September 2009.

Fig. 1: Please add a panel showing the resulting tropospheric column (also the negative values!).
Fig. 2 below shows the GOME-2 tropospheric columns for 22 Feb. 2008, with negative values plotted in dark blue. Unfortunately, most part of the NH above 30ºN is covered by clouds on this day. Therefore, we have not included this plot in Fig. 1 in the paper. As

mentioned above, we did expand the discussion on the uncertainty in the stratospheric NO_2 distribution for 22 Feb. 2008 in Sect. 5.1.

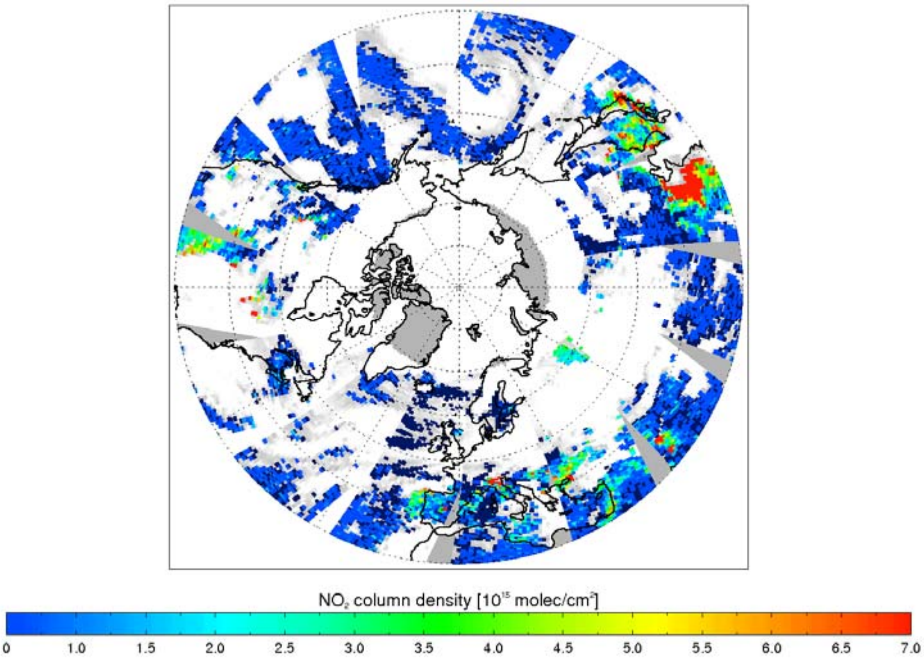

Fig. 2. Tropospheric NO_2 distribution from GOME-2 for 22 February 2008. Only measurements with a cloud radiance fraction < 50% are plotted. Measurements with negative tropospheric columns are plotted in dark blue.

Fig. 3: GOME for 2007 for Europe?
We have not changed the GOME/ERS-2 for Europe as explained above.

Fig. 4: Please modify the colorscale such that it is possible to recognize negative columns.
We have updated Fig. 3 and 4 in the paper, so that it is possible to recognize negative columns. Fig 3. below shows the new figure for East Asia. Since both regions are mostly polluted and the plots show yearly averages, the areas with negative columns are relatively small and scattered. As mentioned above, we do not think it is within the scope of this paper to include a detailed discussion on negative NO_2 columns.

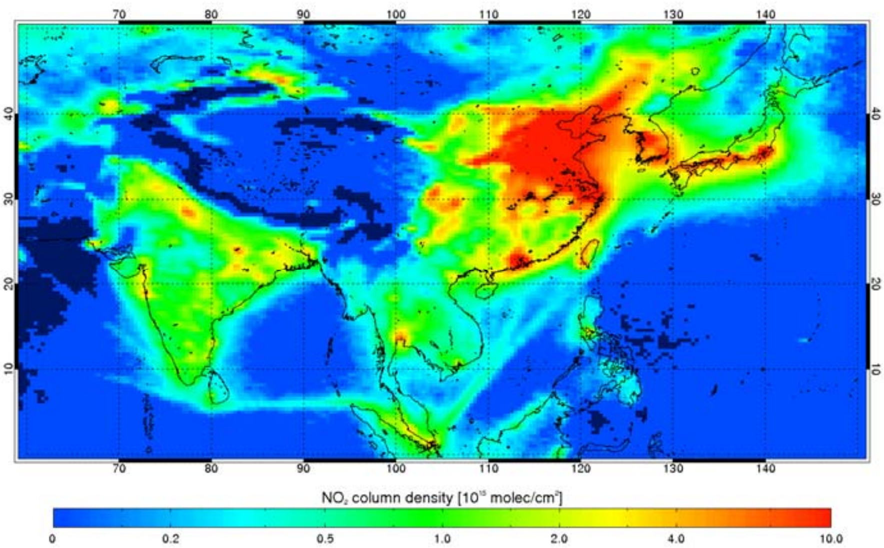

Fig. 3. Average tropospheric NO_2 columns over East Asia measured by GOME-2 for 2007-2009. Areas with negative tropospheric columns are plotted in dark blue.

Fig. 8: Please substitute or add 22 Feb 2008
Done. See comment on “P1639 L22” above

Figs. 10, 12, 14, 15: Linear regression is not appropriate for these scatterplots, as both x and y have uncertainties and the resulting slopes should not depend on the arbitrary choice of the variable used as x. So please use orthogonal regression; see http://www.atmos-chem-phys.net/8/5477/2008/acp-8-5477-2008.html
The statistics on the scatter plots have been updated using orthogonal regression as described in Cantrell [2008].

Fig. 13: Please modify the range of the y-axis such that negative VCDs are also visible.
Negative tropopsheric values of GOME-2 have now been included in the comparison and the validation with MAXDOAS has been extended to March 2011. The figure below shows the updated version.

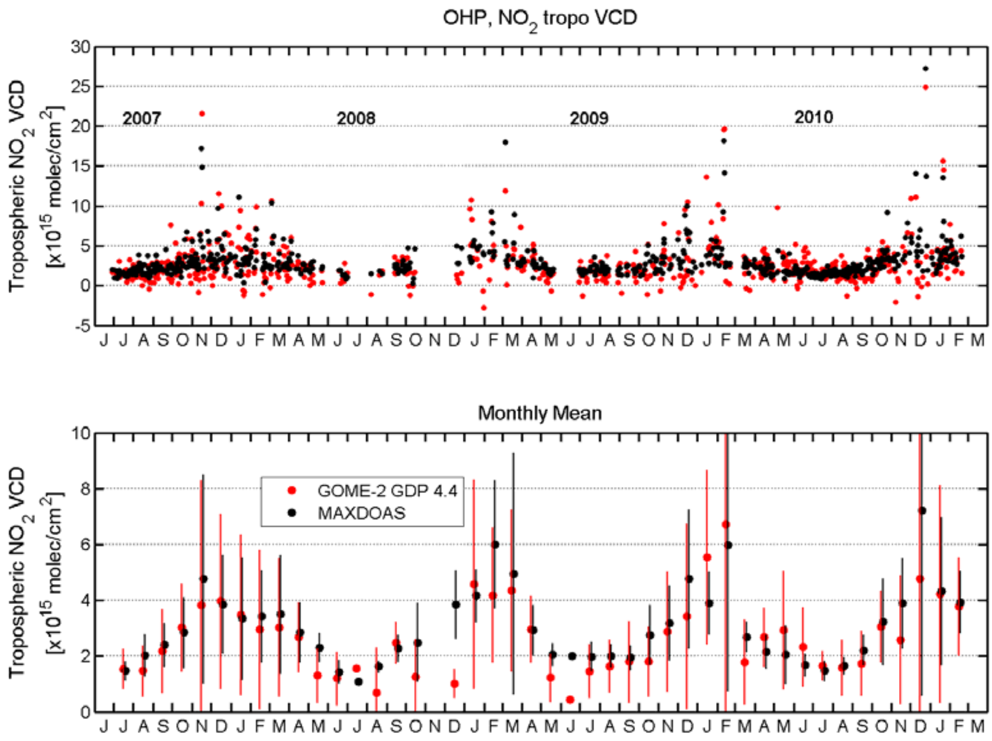

Fig. 4. Comparison of MAXDOAS and GOME-2 tropospheric NO_2 columns (mean value of all the pixels within 100 km around OHP, after cloud-free selection) from June 2007 to March 2011. In the first subplot daily values (only days with both successful measurements) are represented while the second subplot displays monthly averaged values and corresponding one sigma standard deviations.

Fig. 14: Add a similar scatterplot for the daily VCDs, as in Fig 13a.
Fig. 14 has been extended to March 2011 and a second scatter plot with the daily comparisons has been included (see figure below).

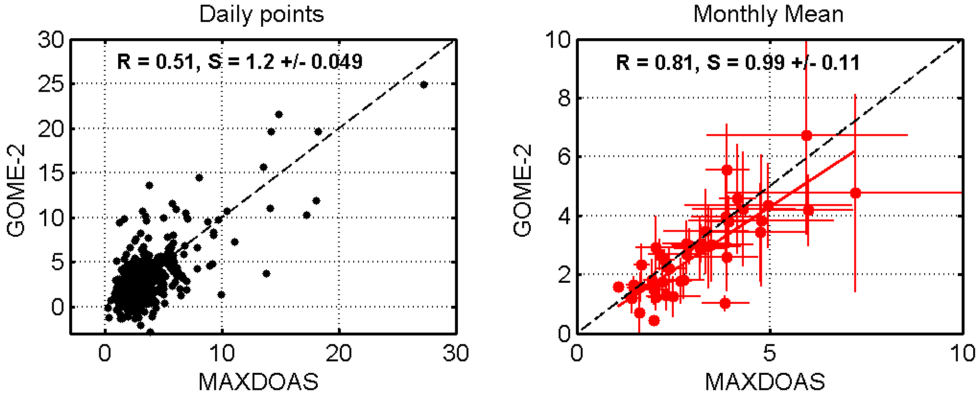

Fig. 5. Scatter plot of the daily and monthly averaged MAXDOAS and GOME-2 tropospheric NO_2 columns at OHP for the period displayed in Fig. 4. The correlation coefficient *R* and the slope *S* of the othogonal regression line are given as insert.

Please include a kind of outlook paragraph describing possible future improvements. In this context, also discuss the differences to the retrieval presented by Richter et al., AMTD, 2011, and explain how far the advances described there might be applied to the operational product as well.

We have added an outlook paragraph to Section 9 with planned future developments, including a discussion on the possible algorithm improvements described in Richter et al., 2011. In particular, the following improvements are planned to be applied to the operational product:

- use of an extended NO_2 DOAS fitting window (Richter et al., 2011).
- Implementation of a spike correction algorithm for the SAA in the level-0-to-1 processing (Richter et al., 2011).
- improvements in the stratospheric correction (see above).
- update of the a-priori (modelled) NO_2 profiles used for the AMF calculation.
- new surface albedo climatology (see above).
- improvements in the treatment of cloud and aerosols (see above).