We thank both reviewers for helpful comments that have led to improvements in the manuscript. Our responses are given below in boldface. We have also boldfaced parts of the referee's comments to highlight certain points. We have added new sections and figures to in the main text and the Appendix to address detailed points raised by the reviews. Finally, we have added numbers to the comments from Referee #1 for convenience.

Anonymous Referee #1
Received and published: 20 March 2014

In this manuscript, data on free tropospheric NO2 is retrieved from OMI observations using data taken at different cloud conditions. The retrievals are validated by comparison with data from airborne in-situ observations and reasonable agreement is found. Using a long time series (3 years), a coarse climatology of upper tropospheric NO2 is created, showing interesting seasonality in its geographic distribution. As verification, the climatology is compared to results from the GMI model. As a side product, an estimate of the stratospheric column is derived which is compared to the operational OMI stratospheric NO2 product and the GMI model atmosphere.

The paper is **clearly structured, well written** and reports on a novel satellite data product, free tropospheric NO2 amounts. **The technique used has** to my knowledge **never before been applied to NO2** and the results are **interesting** as very little is known about spatial distribution and seasonality of NO2 in the upper troposphere. The approach taken and the methods used are **sound** and **nicely described**, and a thorough discussion of uncertainties and results is provided. There are however several points which I think need to be improved or re-considered in the manuscript, and I therefore recommend publication in ACP only after my comments listed below have been taken into account.

> Thank you very much for your positive comments.

Major Comments

1) A geometric AMF is used for computation of the NO2 vertical columns over clouds. While this is probably a very good approximation above a cloud, it is not a good approximation for NO2 within a cloud. As the cloud top pressure from OMCLDO2 and OMCLDRR give the cloud optical centroid pressure, there always is a contribution from NO2 within the cloud which will be seen with another AMF. This is further complicated by the sampling issue discussed below. I think this needs to be discussed.

Since the comment 1 of the Referee #2 points out the exactly same point, we respond to the both comments here together.

Comment 1 of Referee #2) The main issue I have with the method by the authors concerns their use of a simple geometric air mass factor (AMF) for converting the slant columns into the above-cloud vertical columns that are at the basis of their method (Eq. (6)). Using a geometric AMF may be a reasonable choice for retrievals of stratospheric NO2 columns, but it will lead to considerable errors for above-cloud retrievals, because the sensitivity to NO2 within and also above the cloud is strongly enhanced by the bright cloud. This is clearly indicated in the radiative transfer studies shown in e.g. Hild et al. [2002], Eskes and Boersma [2003], and Boersma et al. [2005]. The authors should therefore revisit their geometrical AMFs and replace these by more realistic AMFs that take into account the increased sensitivity above the effective cloud pressure level (and still discard the NO2 below as is done in the geometric AMF). See also the study by Beirle et al. [2006]. The more realistic AMFs will certainly be higher than the simple geometric AMFs used here, and their use will improve the agreement between the OMI-derived and GMI modelled mixing ratios, and between OMI and INTEX-B.

> We have added a new section (Sect. 3.2) that discusses effects of geometric and "cloudy" AMF obtained using certain assumptions about cloud optical depth profiles, etc.

> **3.2 Comparison of NO2 VMRs derived using geometric and near-Lambertian AMFs in complex (realistic) cloudy conditions**
>
> **In this subsection, we attempt to assess potential errors in our approach owing to various AMF assumptions. To do this, we first simulate OMI cloud and slant column measurements in realistic cloudy conditions using the LInearized Discrete Ordinate Radiative Transfer (LIDORT) model (Spurr et al., 2001). For these simulations, we use the C1 cloud model (Diermendjian, 1969) and all calculations are performed at 440 nm. Similar calculations were performed at shorter UV wavelengths by Ziemke et al. (2009).**
>
> **We then retrieve VMRs based on the geometric AMF assumption. Previous radiative transfer studies have shown that there is enhanced scattering and absorption (e.g., of NO2) within and above bright clouds (Hild et al., 2002; Eskes and Boersma, 2003; Boersma et al., 2005; Beirle et al, 2006; Beirle et al., 2009; Ziemke et al., 2009). A near-Lambertian (i.e., scattering cloud with high optical depth uniformly distributed over a thin layer) may also be a**

reasonable AMF formulation to use in a cloud-slicing approach. We therefore also examine the use of such an AMF for determining free-tropopsheric NO2 mixing ratios.

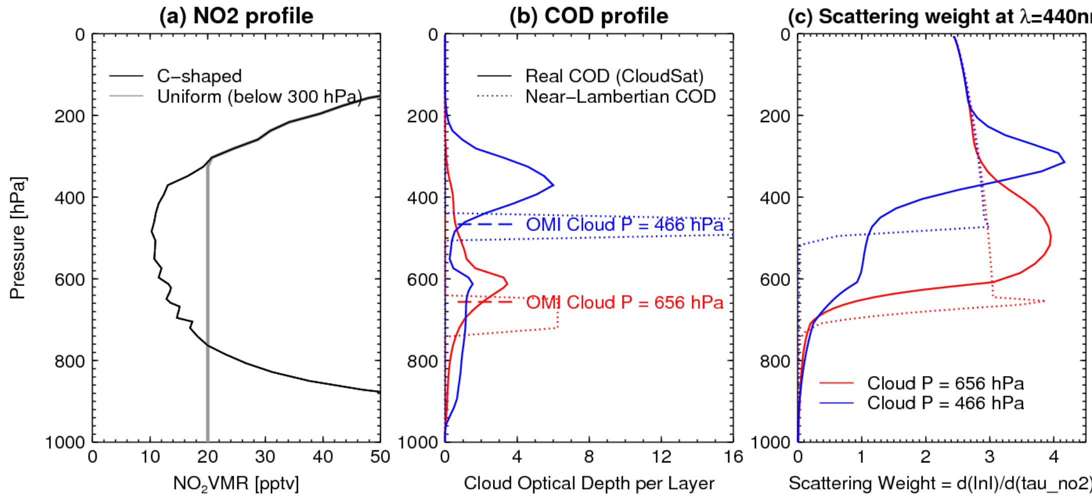

Fig. 2. Experimental settings to simulate OMI above-cloud NO2 VCD observations: (a) NO2 profiles used in the AMF calculations, (b) cloud optical depth (COD) profiles used in the radiative transfer calculations, and (c) scattering weight profiles from the radiative transfer calculations corresponding to COD profiles in (b). See text for more details.

In order to accurately simulate OMI measurements, we first need a realistic NO2 profile. Here, we use a C-shaped profile generated by the GMI model in polluted conditions shown in Fig. 2a. We also need to use realistic cloud optical depth (COD) profiles. A combination of CloudSat/MODIS data (i.e., the CloudSat 2B-TAU product) provides a source of such data (CloudSat, 2008). Examples of COD profiles are shown in Figure 2b (solid lines). The red solid line in Fig. 2b shows a Gaussian-like COD profile where the reported collocated OMI cloud optical centroid pressure (OCP) was 656 hPa. The blue line shows another example of a multi-layer vertically-extended cloud. These profiles are from a tropical deep convective complex and were also used in the study of Vasilkov et al. (2008). Figure 2c shows the corresponding scattering weights (solid lines) for these cloud profiles. For both cases there is enhanced weighting in the top portion of the cloud with decreasing weights in the bottom portions. The calculations were performed at SZA=46° at nadir.

Without a priori knowledge of the COD profile (which is the case in general) and with only a single retrieved OMI cloud OCP value for each observation, we must make assumptions in order to compute scattering weights. For example, we may assume that the COD profile is uniform and optically thick (total COD=25) within a thin layer (1\,km geometrical thickness). The dotted lines in Figure 2b show such clouds that would produce the observed OMI cloud OCP. The scattering weights corresponding to these uniform profiles are shown in Fig. 2c (dotted lines). Although the scattering weights from the uniform clouds show slightly enhanced scattering above the cloud OCP (including both the very top portion of the cloud as well as above the physical cloud top), they do not reproduce the shape of the scattering weights from the CloudSat optical depth profiles.

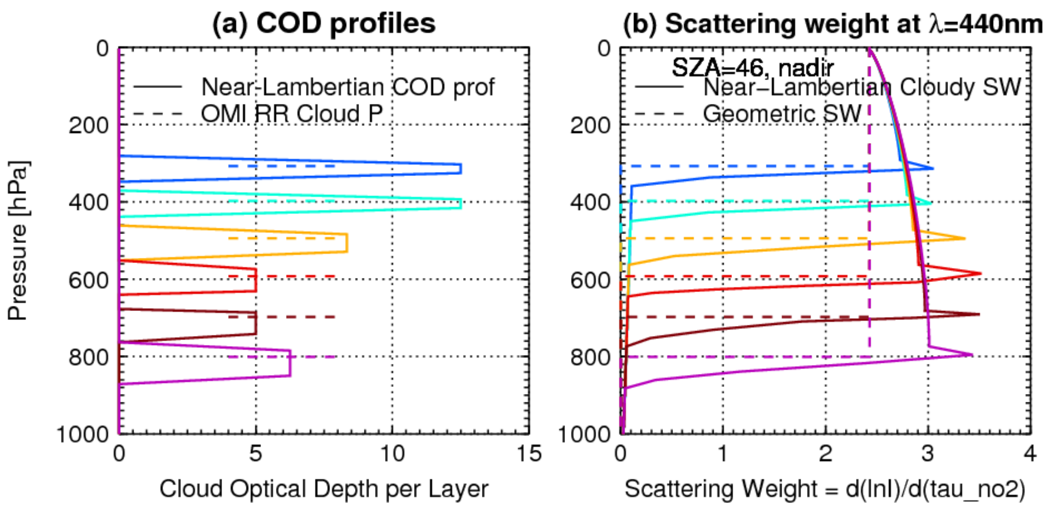

Fig. 3. (a) Near-Lambertian COD profiles (solid lines) that correspond to various cloud OCPs (dashed lines), (b) scattering weights calculated using near-Lambertian COD profiles (solid lines) accompanied by geometric weighting functions (dashed lines).

Figure 3a shows near-Lambertian COD profiles at different cloud OCPs. The corresponding scattering weights for these clouds are shown in Fig. 3b along with geometric weighting functions; the latter assumes uniform weighting related to the viewing geometry (i.e., sec(SZA)+sec(VZA), where SZA and VZA are the solar and viewing zenith angles, respectively) above the cloud OCP with zero weighting below. The overall shape of the scattering weights for near-Lambertia clouds does not vary much with cloud OCP; however the amount of enhanced scattering above and inside the cloud depends upon the cloud OCP.

We next compute (1) geometric AMFs and (2) near-Lambertian AMFs using our scattering weight calculations. The difference between above-cloud NO2 VCDs computed using geometric and near-Lambertian AMFs varies with the viewing geometry, cloud OCP, and a priori NO2 profile. Above-cloud NO2 VCDs from the geometric AMFs are larger than those from the near-Lambertian AMFs in most viewing geometries, except where the solar zenith angles are greater than ~70°. In moderate viewing geometries (SZA<70°), the differences are larger when the cloud OCP is greater (low clouds). The VCDs computed using the geometric AMFs are higher than with near-Lambertian AMFs by up to maxima of 5% (14%) for the C-shaped (uniform) NO2 profiles. For the remainder of this section and in appendices, we focus on results using near-Lambertian AMFs with the C-shaped NO2 profile.

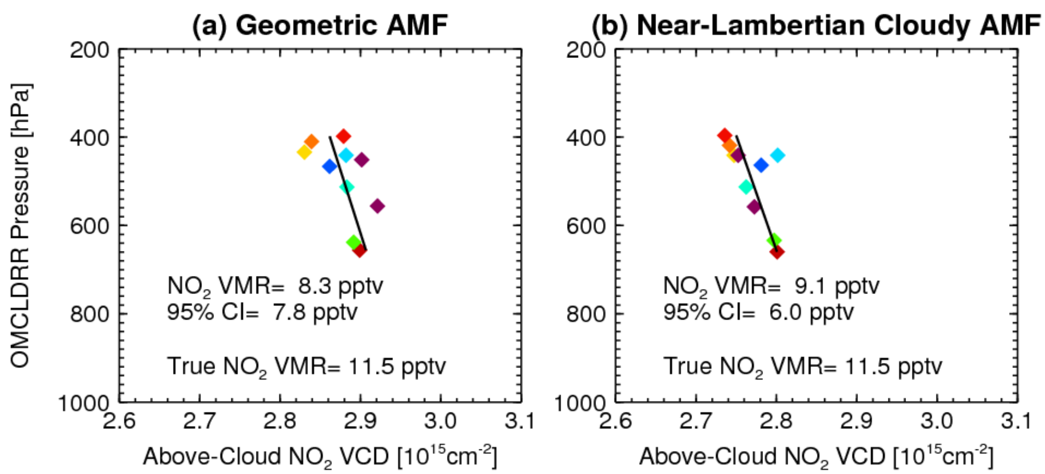

Fig. 4. NO2 VMRs derived from simulated OMI cloud OCPs and above-cloud NO2 VCDs using (a) geometric AMFs, and (b) near-Lambertian cloudy AMFs.

We next simulate SCDs for 10 different cloud optical depth profiles from CloudSat/MODIS using LIDORT at nadir and SZA=46° for the C-shaped NO2 profile in Fig.2a. Figure 4a and b shows the simulated above-cloud VCDs derived using geometric and near-Lambertian AMFs, respectively, versus the corresponding cloud OCPs. We then derive NO2 VMRs from the slopes for these two AMFs. The derived NO2 VMRs, 95% confidence interval, and the true NO2 VMR are presented.

The errors in derived NO2 VMRs are similar for both AMF assumptions;

errors are in the range 20-30% with a somewhat higher error and larger confidence interval for the geometric AMF assumption. The two points deviating from the others in the near-Lambertian AMF scenario result from multi-layer clouds.

In the remainder of this paper, we show results based on the geometric AMF. We show sample results derived with near-Lambertian AMFs in Append. D2. In brief, the results derived using both AMFs display similar spatial and seasonal variability, although the NO2 VMR magnitudes are somewhat smaller using the near-Lambertian AMFs.

Appendix D2 Near-Lambertian AMF sample results

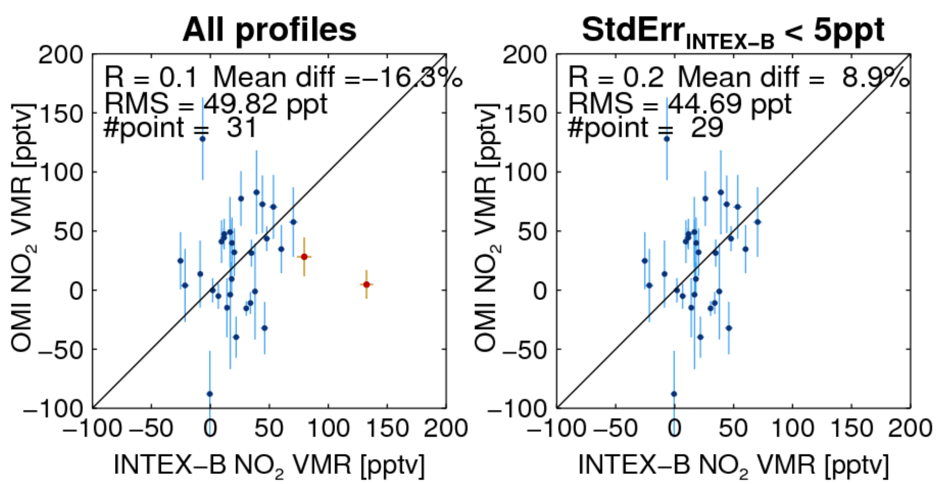

Fig. D3. Similar to Fig. 6 but using near-Lambertian cloudy AMF.

Here, we show results obtained using near-Lambertian cloudy AMFs with the OMCLDRR cloud OCP values. Similar to Fig. 6, Fig. D3 shows a scattergram of INTEX-B and OMI cloud slicing NO2 VMRs. The left panel shows all available matchups between INTEX-B and OMI, and the right panel shows matchups where the standard error of the mean of INTEX-B measurements < 5 pptv. The mean difference between INTEX-B and OMI NO2 VMRs is smaller when using near-Lambertian AMF as compared with the geometric AMF. However, the RMS difference between INTEX-B and OMI NO2 VMRs is greater with near-Lambertian AMFs.

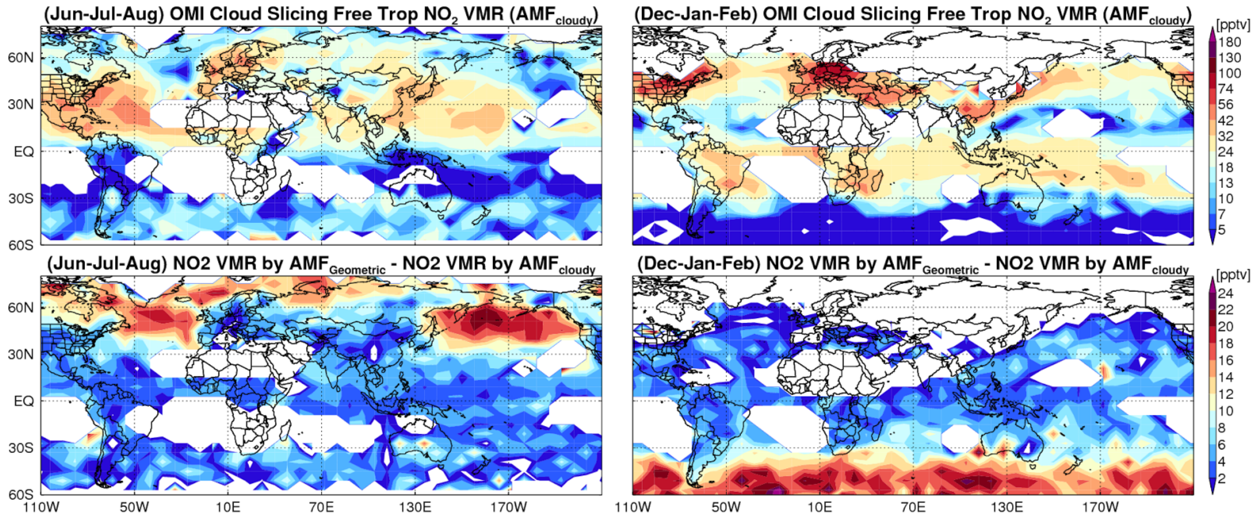

Fig. D4. For Jun.-Aug. (left) and Dec.-Feb. (right) averaged over 2005-2007, Top: Global maps of NO2 VMR calculated using near-Lambertian cloudy AMFs; Bottom: Difference in NO2 VMRs computed using geometric and near-Lambertian AMFs.

Similar to the first row of Fig. 7, the first row of Fig. D4 shows global maps of the free tropospheric NO2 climatology obtained with near-Lambertian AMFs. The second row of Fig. D4 shows the difference in NO2 computed using geometric and near-Lambertian AMFs. NO2 VMRs computed using near-Lambertian AMFs show similar spatial patterns and seasonality as compared with that computed using geometric AMFs; for example, both climatologies show high NO2 VMRs near major urban areas and the outflow regions and high NO2 in tropical regions affected by lightning. Overall, NO2 VMRs from near-Lambertian AMFs have lower magnitudes as compared with geometric AMF results. These VMR differences are highest in high-latitude oceanic areas during summer. This might result from the combination of cloud pressure and a priori NO2 profile used in near-Lambertian AMF formulation. In these regions, clouds form at very high pressure levels (low altitudes) as shown in the fifth row of Fig. C1, where geometric and near-Lambertian AMFs behave differently as explained in Sect. 3.2. Moreover, there is no ground-based NOx source, which makes the actual NO2 profile different from the C-shaped NO2 profile used in the near-Lambertian AMF calculations.

Owing to the relevance of Beirle et al. (2006) to this study, we have added a new sentence that refers to this paper in the Introduction:

“Beirle et al. (2006) also utilized GOME measurements in combination with US National Lightning Detection Network (NLDN) data to estimate lightning-produced NOx over the Gulf of Mexico.”

2) As pointed out in the manuscript, cloudy scenes differ from clear sky scenes in many respects as they are representative of other meteorological situations, photochemical regimes, transport patterns (frontal systems) and vertical NO2 distributions. Other satellite studies using cloudy data have highlighted the occurrence of transport events in cloudy situations (e.g. Stohl et al., 2003 or very recently Zien et al., 2013) as well as lightning (e.g. Beirle et al., 2009, Boersma et al., 2005) which will have important impacts on the statistical sampling of the free troposphere using cloud slicing. I’d suggest to discuss all these effects in a dedicated section in a qualitative way and to indicate the direction and size of the various effects that you expect. Some of the information is already present in the manuscript but should be collected and discussed in a more consistent way.

> Yes, cloudy scenes are indeed different from clear scenes regarding tropospheric NO2, in terms of meteorological situations and chemical conditions. Having a separate section describing these effects will make the paper easier to understand. Thank you very much for pointing this out.

In the revised manuscript, we have added a new section (4.2.1) to address these points.

4.2.1. Potential issues related to satellite sampling in cloudy conditions

Our derived climatology is representative of NO2 VMRs in highly cloudy conditions with significant cloud pressure variability as explained in Sect. 3.1. Consequently, NO2 VMRs are not obtained where clouds rarely form (e.g., Sahara) or where cloud pressure variability is small (e.g., oceanic areas with persistent low clouds due to subsidence, such as off the western coasts of South America and southern Africa). Therefore, it is important to interpret our results in the context of the observing conditions. In addition, when comparing cloud-slicing results with those from models, it is important to appropriately sample the model to reflect the observing conditions.

Here, we describe the potential differences between NO2 VMRs in cloudy

and all-sky conditions due to chemistry and transport. One important feature in cloudy conditions is lightning NOx production; it generally increases NO2 concentrations as compared with clear-skies. This is especially important in the tropics. In middle to high latitudes, the cloud-slicing NO2 VMRs are also derived in frontal storms, where uplift of boundary layer pollution and subsequent long-range transport frequently occurs (e.g., in the so-called warm conveyor belt) (Stohl et al., 2003; Zien et al., 2013). This may also increase cloud-slicing NO2 VMRs as compared with clear-sky conditions. In addition, NOx chemistry will be different in highly cloudy conditions as compared with clear-skies. NO2 photolysis rates may be increased above or within bright clouds, but decreased below them.

Comparison of NO2 VMRs from GMI in cloudy and all-sky conditions may provide an estimate of potential sampling biases. In general, the GMI cloudy NO2 VMRs are higher than those in all-sky conditions over urban regions (see Fig. C2 in Appendix C for GMI all-sky conditions). Therefore, in Sect. 4.2, for all comparisons we sample GMI in highly cloudy conditions (cloud optical depth > 10) and consider the potential sampling biases in the interpretation of our derived climatology.

3) An estimate for zonal stratospheric NO2 columns is derived and compared to the operational product. The good agreement between the two independent estimates is taken as closure validation of the cloud slicing technique. While this looks good at first sight, I think that the two estimates are neither independent, nor does the agreement tell much about the quality of the cloud slicing product. The reason is that for a zonal average, even taking all OMI NO2 slant columns and applying a stratospheric AMF without any correction will lead to reasonable results. In the operational product, regions with known pollution are excluded, making the estimate better. In the cloud slicing product, only cloudy scenes are used, removing most of the BL pollution NO2, which again should result in a good estimate of the stratospheric NO2 without further processing.

The extrapolation to tropopause pressure (Fig. 1d) will remove the free tropospheric component from the above cloud total columns which at an estimated 30 ppt adds up to about 2E14 molec cm-2. This relatively small correction (which is the cloud slicing component of the stratospheric values shown) is of the same order as the differences between the two OMI stratospheric NO2 columns shown in Fig. 7. Thus the only conclusion I can draw from this comparison is that the free tropospheric columns derived with the cloud slicing method are so small, that they do not matter much for the stratospheric column. I therefore think that the whole discussion of the stratospheric columns needs to be revised (for example by showing the stratospheric estimate using all cloud slicing data but without extrapolation to tropopause height) or completely removed.

> We have deleted Sect. 4.4.

4) The known bias in the OMI NO2 slant columns is referred to in many places throughout the manuscript and used as explanation for higher than expected free tropospheric values. However, to my knowledge, the bias in the current OMI NO2 product is not a relative error but rather an absolute offset on the slant columns. As in the cloud slicing method the slope of a set of measurements at different cloud pressure is analysed, such an offset will not contribute significantly to the uncertainty at constant AMF. I therefore do not agree with the repeated statements explaining biases by the OMI SC problems and think they should be removed.

Since the comment 2 of the Referee #2 points out the exactly same point, we respond to the both comments here together.

Comment 2 of Referee #2) Rather than just citing a relative bias in the OMI NO2 slant columns, I propose to also quote the absolute bias in the vertical columns. Various studies (e.g. Belmonte-Rivas et al. [2014]; Krotkov [2012], Boersma et al. [2014]) suggest that the bias in the OMNO2A vertical (stratospheric) columns is rather constant over an orbit after converting the slant to vertical columns.

> Based on recent results not yet published, we cannot completely rule out the possibility of errors in our cloud slicing VMRs resulting from biases in the current version of OMI NO2 SCDs. The SCD bias looks almost additive in the VCD, but we cannot be sure that the bias is a purely additive offset. Thus we maintain the statements explaining the biases in the OMI SCDs in Sect. 2.1.2. However, we have deleted subsequent statements on the potential effects of these biases on our VMR results, such as "therefore our estimates from cloud slicing will be biased by the same amount". In addition, regarding comments by Referee #2, we have included the suggested references and quoted the bias in terms of VCD.

The corresponding part now reads:

> **"There is evidence that NO2 SCDs are positively biased (Krotkov et al., 2013; Boersma et al., 2014) which may lead to a high bias in NO2 VCD of 4-5 x 10^{14} (Boersma et al., 2014; Belmonte et al., 2014). The effect of this bias**

on our results is not yet clear. We plan to reprocess the OMI data when a new version of OMI SCDs is released."

5) The comparison between model and OMI free tropospheric NO2 VMR sounds OK in the text but looking at the figures, I hardly see any similarity. Both the spatial pattern and the absolute values are very different, and all the lightning signatures shown in the lower panels of Fig. 4 are clearly not reproduced in the OMI data. I think these discrepancies should become clearer in the text. It might also be worthwhile to mention the impact such differences in vertical distribution might have on tropospheric AMFs.

> Thank you very much for making a good point about the differences between OMI and GMI NO2 VMRs.

We have added the following phrase in Sect 4.2:

"**We note that the magnitudes of NO2 VMRs from GMI are generally lower than those from OMI NO2 cloud slicing. Beside the differences in magnitudes, the OMI VMR maps show some notable differences with respect to GMI, while the OMI and GMI tropospheric column maps in Appendix C look very similar."**

We have also added the sentences discussing the differences between OMI and GMI NO2 VMRs.

in Sect 4.2.2 Anthropogenic contributions:

"It is well known that boundary layer NO2 VMRs and thus tropospheric NO2 columns are higher in winter due to longer lifetimes. Our cloud slicing results show that seasonality of the OMI free tropospheric VMRs is similar to that in the boundary layer VMRs. However, this seasonality is not as apparent in the GMI model."

in Sect 4.2.3 Lightning contributions:

"While the locations of these apparent lightning-enhancements of NO2 over land are similar in summer in both GMI and OMI data sets, there are a few key differences to note: (1) the seasonality of the NO2 enhancements over tropical oceans shown in OMI data is not as apparent in the GMI output; in the OMI climatology, the enhancement in oceanic NO2 VMRs is present in

summer, while GMI shows less seasonal variability; (2) There is a stronger land/ocean contrast in GMI lightning-generated NO2 contribution than is seen in the OMI NO2 VMR climatology in regions where lightning may be playing a dominant role.”

in Sect. 5 Conclusions:

“However, some differences, particularly with respect to the seasonality of lightning-generated NO2 in the tropics and anthropogenic NO2 in the extra-tropics, are noted.”

Minor Comments

(a) p 1561 l9: It is contributes => It contributes
(b) p 1566 l27: I do not see why equation 4 is based on any assumptions on NO2 – this is about the cloud scene pressure.
(c) p 1569, l1: why is the lightning contribution derived using all scenes? Doesn’t this create a very different sampling than the satellite data?
(d) p 1569 and elsewhere: I’d prefer a small p for pressure
(e) p 1577 l12: in the both => in both
(f) p 1581 l29: over the North America => over North America

> Thank you for your very careful reading! We have fixed the manuscript as suggested in (a), (d), (e), and (f). See responses to (b) and (c) below.

(b) p 1566 l27: I do not see why equation 4 is based on any assumptions on NO2 – this is about the cloud scene pressure.

> We have re-written the sentences, which now reads:

“The derived NO2 VCD in a cloudy pixel can be interpreted as the total column from Pscene to the top of-the-atmosphere (i.e., the total column above Pscene), assuming that the NO2 profile is vertically uniform between Pterrain and Pc (Joiner et al., 2009).”

(c) p 1569, l1: why is the lightning contribution derived using all scenes? Doesn’t this create a very different sampling than the satellite data?

> Thank you pointing that out. We now show the GMI lightning contribution to free tropospheric NO2 screened for cloud optical depths > 10 in order to keep the sampling criteria relatively consistent. We note that the sampling criteria does not produce much differences in model results.

Anonymous Referee #2

The manuscript by S. Choi et al. presents a method to estimate NO2 concentrations in the troposphere above clouds from OMI satellite measurements. Tropospheric NO2 detection above and within clouds from satellite has been attempted before (e.g. Boersma et al. [2005]), but Choi et al. extend the approach to the global scale and also focus on polluted areas and all 4 seasons. The central idea is to compare "nearby" tropospheric columns that have been retrieved under situations of similar cloudiness, but with different cloud heights ("cloud slicing"). Assuming that the free tropospheric NO2 concentration is constant with altitude, the reduction of column with higher clouds can then be used to derive the free tropospheric, above-cloud NO2 concentration. This technique has been applied extensively for tropospheric ozone retrievals, but is **now being applied for the first time on tropospheric NO2**.

The authors **describe their method clearly**, and evaluate their method by comparing against independent aircraft measurements. That the validation does not prove to be an overwhelming success was to be expected, in view of the small difference signals, and the detection limit of the OMI retrievals. Nevertheless the above-cloud tropospheric NO2 climatology constructed by the authors appears to be a **compelling result**.

> Thank you very much for your positive comments.

Major comments

1. The main issue I have with the method by the authors concerns their use of a simple geometric air mass factor (AMF) for converting the slant columns into the above-cloud vertical columns that are at the basis of their method (Eq. (6)). Using a geometric AMF may be a reasonable choice for retrievals of stratospheric NO2 columns, but it will lead to considerable errors for above-cloud retrievals, because the sensitivity to NO2 within and also above the cloud is strongly enhanced by the bright cloud. This is clearly indicated in the radiative transfer studies shown in e.g. Hild et al. [2002], Eskes and Boersma [2003], and Boersma et al. [2005]. The authors should therefore revisit their geometrical AMFs and

replace these by more realistic AMFs that take into account the increased sensitivity above the effective cloud pressure level (and still discard the NO2 below as is done in the geometric AMF). See also the study by Beirle et al. [2006]. The more realistic AMFs will certainly be higher than the simple geometric AMFs used here, and their use will improve the agreement between the OMI-derived and GMI modelled mixing ratios, and between OMI and INTEX-B.

> Please see our response to comment 1 of the Anonymous Referee #1.

2. Rather than just citing a relative bias in the OMI NO2 slant columns, I propose to also quote the absolute bias in the vertical columns. Various studies (e.g. Belmonte-Rivas et al. [2014]; Krotkov [2012], Boersma et al. [2014]) suggest that the bias in the OMNO2A vertical (stratospheric) columns is rather constant over an orbit after converting the slant to vertical columns.

> Please see our response to comment 4 of the Anonymous Referee #1.

3. The assumption that the NO2 concentration does not change with altitude is generally defendable, but will lead to errors in case of lightning NOx production (and aircraft NOx). The 'profiles' shown in Fig. 6 of the manuscript show that the higher the cloud, the higher the inferred above-cloud NO2 concentrations. Such patterns have also been reported in the study by Boersma et al. [2005] from GOME, and high above-cloud NO2 has been observed from various aircraft campaigns near and even within thunderstorm clouds. This immediately shows that (1) a simple geometric AMF is inaccurate for such situations, and that the AMF should take into account the actual vertical sensitivity, and (2) AMFs should account for realistic a priori profile shapes that will be very different in lightning situations. The authors have all the means at hand with the GMI model and state-of-science radiative transfer codes.

> We have added a lengthy new subsection (Sect. 3.2) that addresses the bulk of this comment (e.g., errors due to the use of a simple geometric AMF and errors due to an incorrect profile shape assumption). Please see the response to comment 1 of Referee #1. Given the NO2 differences we see with GMI, particularly in areas influenced by lightning, and the result that the use of more sophisticated AMFs did not qualitatively change our conclusions, we do not feel at this time that using model-generated NO2 profiles will necessarily achieve a better result.

4. Section 4.2.2 should refer to the study by Boersma et al. [2005], as there are various parallels to be drawn. In that study, a considerable production of lightning NOx over the

tropical oceans was inferred from cloudy GOME measurements, with spatial patterns similar to those shown in the upper right panel of Figure 4. Similar to the GMI model here, the TM3 model used in their study also failed to reproduce a substantial source of lightning NOx production over the ocean, pointing at similar misrepresentations in the lightning parameterization in both models. Since these lightning parameterizations are still in use in many CTMs, it is important to point out that these are in need of improvement.

> Thank you very much for pointing out this relevant study for the discussion of lightning NOx. Indeed, it is a very important, interesting study in context of studying lightning NOx using satellite observations.

We have added the following sentence in the Introduction owing to the importance of Boersma et al. (2005) study:

> **"For example, Boersma et al. (2005) estimated the global lightning NOx production using GOME cloudy NO2 measurements."**

In order to highlight the connection from Boersma et al. (2005) to our study, we have added the following text to the revised manuscript:

> **"Boersma et al. (2005) have reported similar observations; they inferred a considerable amount of lightning-generated NO2 over tropical regions using cloudy GOME measurements with similar spatial patterns as shown in our cloud-slicing results. They also compared GOME-derived NO2 with that from the TM3 chemical transport model. Their study also showed some differences between observations and model simulations in cloudy conditions, presumably related to lightning parameterizations within chemical transport models."**

Minor comments

a) Please provide some quantitative estimates on the cloud pressure errors from the OMCLDRR and OMCLDO2 products.

> We have added the following sentences in Sect. 2.1.

> **"Acarreta et al. (2004) and Vasilkov et al. (2008) used radiative transfer**

calculations to estimate errors of OMI cloud optical centroid pressures. They estimate that errors should be in the range 50 hPa or less for a wide range of viewing condition and for moderate to high cloud effective fractions (or cloud optical thicknesses). Comparison of the two retrievals (OMCLDRR and OMCLDO2) has been used as a means to evaluate the retrieved cloud pressures after the launch of Aura OMI and may provide an upper limit on the errors (Sneep et al., 2008; Joiner et al., 2012). For effective cloud fractions > 0.75, the mean differences are 40 hPa (OMCLDO2 having higher pressures on average) over land and 25 hPa over ocean and standard deviations are approximately 63 hPa over both land and ocean (Joiner et al., 2012).”

b) P1565, L 22: ‘US/VIS wavelengths’ should be UV/Vis wavelengths.

> Thank you for your careful reading! We have fixed it.

c) P1579, section 4.4: to my opinion the results from the Belmonte-Rivas study should be cited here

> **Thank you very much for bringing to our attention to this new study, which is published in ACPD after we submitted this paper. We have referred to the Belmonte-Rivas study in Sect. 2.1 to provide more information on the potential bias in OMI NO2 SCDs. We deleted Section 4.4 which discusses stratospheric NO2 VCD as suggested by Referee #1.**

Overall, we have addressed the comments brought by the referees. During the process, we have modified the paper for a better flow according to the revision. For example, we have deleted the section explaining OMNO2B stratospheric column NO2, since we have deleted the stratospheric column derived from cloud slicing.