AMT-2016-366: Responses to Anonymous Reviewer 3

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June 20, 2017

Thank you for your review and comments, which have added substantial value to the paper.

Reviewer comment 1: *Four research flights are analysed and discussed in quite some detail, but the interpretation of differences between airborne and satellite retrievals could go more into depth. Based on the present manuscript, the reader might get the impression that systematic differences in tropospheric NO_2 columns between satellite and iDOAS can be explained solely (or largely) by horizontal variability in the tropospheric NO_2 columns on a scale that is smaller than the typical size of satellite pixels. What would be particularly relevant is to investigate further the potential impact of profile shape assumptions for NO_2 and aerosols in explaining the difference between satellite and airborne measurements over the most polluted regions. Close to major point sources one may expect not only to find locally quite extreme tropospheric NO_2 column abundances, but at the same locations also the NO_2 profile shape may deviate considerably from other places further away from the main sources. In this context, it may be relevant to distinguish explicitely four profiles: the true profile at the spatial resolution of the aircraft measurements (P true air), the profile used in the airborne retrieval (P prior air), the true profile at the resolution of the satellite measurements (P true sat) and the profile used in the satellite retrieval (P prior sat). Differences in tropospheric NO_2 column retrievals (space-borne vs airborne) cannot be interpreted without taking into account these four profile shapes in the discussion. How much do the authors think P rue air can deviate from P prior air close to the main sources (same for P true sat and P prior sat). Furthermore the AMF is not only affected by the (different) profile shapes, but also by the block-AMFs, and these are not identical for the satellite and the airborne point of view. This should be taken into account as well.*

Despite the length of this comment, I would suggest to add just one or two paragraphs addresssing this point and providing some first order estimates. It would for instance be enlightening to the reader if the impact of making *wrong profile shape assumptions is worked out for one hypothetical scenario. For instance (it is up to the authors to deviate from this concrete suggestion): scale height for P true air is 0.2km (e.g. close to strong isolated source); scale height for P true sat is 0.4km (averaged over a larger region the true profile is less dominated by the local source); scale heigh for P prior air is 0.6km (this number is used in the present study); scale height for P prior sat is taken from the profile used in DOMINOv2 product over this region. Block AMFs should be applied for a representative SZA and surface reflectance. When combined, this information should provide the reader with a first order quantitative estimate of local AMF fluctuations near a strong plume: to what extent can this explain the discrepancy between the satellite and airborne retrieval? Or perhaps it is concluded that - when taking this effect into account - the observed discrepancy increases even further.*

Response

This is a good idea. We have developed a further suite of vertical profile scenarios, based on Scenarios 11 and 12. These scenarios have a variety of scale-heights from 0.2km to 1.4km.

Like in the previously presented model scenarios, AMFs were calculated for each profile shape with all the permutations of SSA set at 0.82, 0.90, and 0.98; and surface albedo set at 0.02, 0.05, 0.08, and 0.11. All these profiles, like their parent profiles of scenario 11 and 12, have the surface elevation set to 1400m. In order to address the reviewer's next comment pertaining to the use of a fixed AOT, calculations were repeated with the AOT set to 0.1, 0.3 and 0.5.

What is remarkable from Figure 1 in this response, is that the trend in AMF with decreasing scale-height is negative for scenarios without an elevated layer, and positive for scenarios with such a layer. Such layers have been observed in this and other measurement campaigns. This result implies that close to a surface NO_2 source, such as the city of Johannesburg, the error from incorrect choice of *a-priori* vertical NO_2 profile cannot be determined without an actual profile measurement.

Action

The following paragraphs and the figure have been added to the discussion on page 14:

It is instructive to evaluate the potential air-mass factor error that might be made by assuming an incorrect vertical profile of NO_2. Several more radiative-transfer modelling scenarios are introduced, based on scenarios 11 and 12, i.e. with an exponentially-decreasing profile, surface elevation set at 1400m, some profiles with an elevated layer of NO_2 and some without. The scale height of the profiles is varied from 1400m to 200m, and radiative

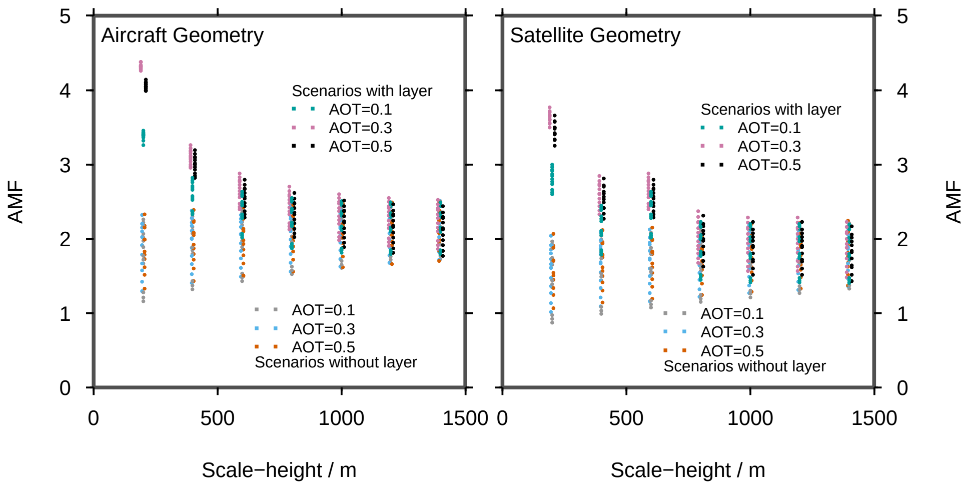

Figure 1: AMFs for scenarios of varying scale-height, for aircraft viewing geometry (left) and satellite viewing geometry (right). For each scenario of scale-height and AOT, variability in the AMF is due to variations in surface albedo and single-scattering albedo.

transfer calculations are done at a representative solar zenith angle of 55°. Once again air-mass factors for permutations of AOT of 0.1, 0.3, and 0.5, and SSA of 0.82, 0.90, and 0.98 are calculated. Results for aircraft- and satellite viewing geometry are presented in Fig. 8. It can be seen that the AMF increases for scenarios with an elevated NO_2 layer as the vertical profile scale-height is decreased. In contrast, the AMF for scenarios without such a layer decreases as the scale-height is reduced. In the satellite viewing geometry, the behaviour is slightly different, with a flattening off of the AMFs with scale-heights of 600m and 400m, compared to the aircraft viewing geometry. This behaviour can likely be explained by examination of the block-AMFs for the two cases, however such analysis is beyond the scope of the present study.

We might estimate the VCD error arising from AMF uncertainty for the iDOAS using two profiles: the true profile at the spatial scale of the instrument, P_{true} and the profile used in the AMF calculation P_{prior}, along with the associated AMFs: AMF_{true} and AMF_{prior}. If P_{prior} is an exponentially-decreasing profile with scale-height of 1000m either with- or without an elevated layer, AMF_{prior} will lie between approximately 1.6 and 2.6. Close to a surface source of NO_2 P_{true} might have a much smaller scale-height, for example 400m. In the case of a profile with an elevated layer, AMF_{true} should be between 2.5 and 3.2. Using the mid-points of the uncertainty ranges of AMF_{true} and AMF_{prior}, this will lead to a 26% overestimation of the VCD. In the case of P_{true} having no elevated layer, AMF_{true} will lie between approximately 1.2 and 2.3, leading to a 20% underestimation of the

VCD from the use of AMF_{prior}.

In the case of a satellite measurement, a representative profile for the satellite pixel is likely to have a larger scale-height, since more background areas will be included in the measurement along with the surface source and the discrepancy between AMF_{prior} and AMF_{true} will be less, but will behave in a similar manner to that described above. This highlights the need for an improved P_{prior} as the spatial resolution of the measurement improves.

Reviewer comment 2: *Although aerosols are not entirely neglected in this study, they receive little attention considering the fact that for all four flights - each covering distances of hundreds of kilometers - just one fixed value is assumed for the AOT. It is quite remarkable that the uncertainty range of the AMF is derived using a look-up table that does include variability of the single-scattering albedo, but not of the AOT. Over a region where the variability in NO_2 is so large, it is almost unthinkable that the AOT can be approximated with a single value. To some extent the same argumentation as given above (in the vicinity of a strong pollution source the NO_2 profile shapes may show considerable spatial variability) can be given here as well: in the same region the AOT may show a substantial variability (although probably less extreme than for NO_2). In my opinion this point should at least be mentioned. It would be even better to find satellite AOT data (e.g. from MODIS) for the days of the research flights to provide more insight into the relevant parameter*

Agreed. The approach taken to constrain AMF uncertainty arising from profile shape, SSA, surface albedo, surface elevation and SZA in this paper, by calculating all the permutations of these parameters, is extended to the AOT. The above parameters are further permuted with AOT's of 0.1, 0.3 and 0.5. 2-dimensional plots of AMF vs SZA (which could be thought of as slices of the discussion paper's Figure 5) are shown below for each surface albedo, with the original modelling highlighted in orange, and the additional permutations with the and lower and higher AOT in grey and blue-green respectively.

The increase in the range of AMF uncertainty derived from the present approach of modelling all permutations, as a result of the extra two AOT's used is not as large as might be anticipated. Nevertheless, the new values of minimum and maximum AMF will be used and figures, tables and discussion in the manuscript will be updated. In addition, a mistake in scenario 12, where the incorrect vertical profile of NO_2 was used, has been corrected.

Reviewer comment 3: *In the manuscript the discrepancies found between iDOAS and OMI (SCIAMACHY) are not compared to results from other validation studies, e.g. where OMI retrievals are compared to MAX-DOAS observations. In the last years many of such studies were doen, with MAX-*

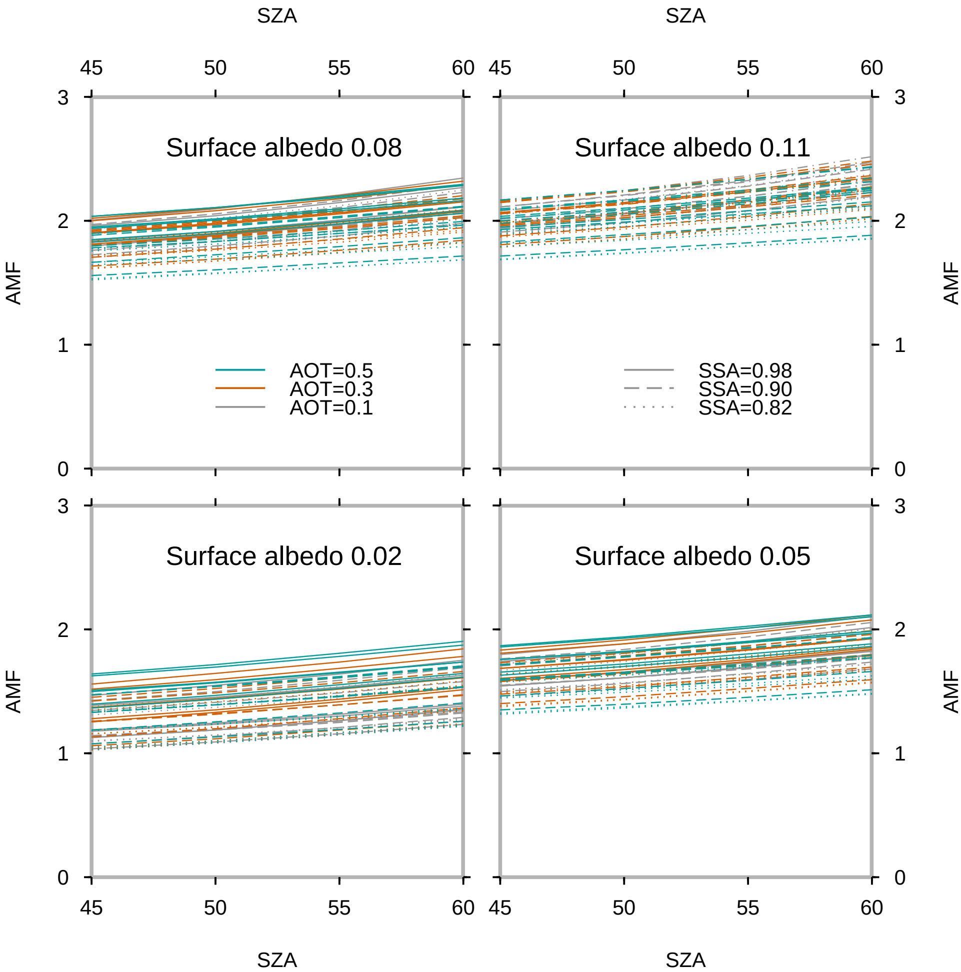

Figure 2: AMF versus SZA plots at different surface albedo's for scenarios with the surface elevation at sea level. AOT=0.3 (as in the discussion manuscript) is plotted in orange, and the additional scenarios of AOT=0.1 and 0.5 are plotted in grey.

DOAS instruments either in rural or in urban regions. It would be valuable to link the findings of this study to findings in such inter-comparisons.

Response and Action

The following paragraph has been added:

Comparison studies of ground-based multi-axis DOAS (MAX-DOAS) instruments with satellite measurements have given mixed results. Some studies (Irie et al, 2008; Hains et al 2010) showing MAX-DOAS results consistently lower than OMI. Kanaya et al (2014) shows DOMINOv2 biases of up to 50% lower than the MAX-DOAS, although the bias improves when only

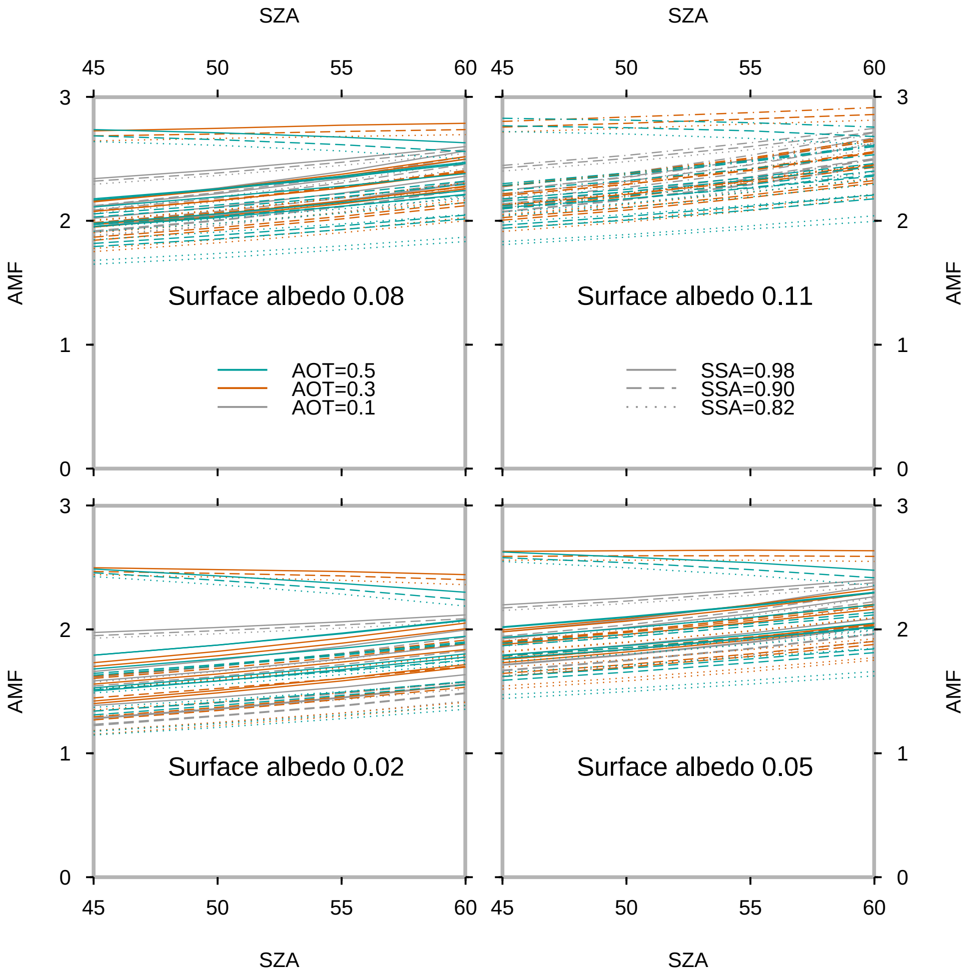

Figure 3: AMF versus SZA plots at different surface albedo's for scenarios with the surface elevation at 1400m above sea level. AOT=0.3 (as in the discussion manuscript) is plotted in orange, and the additional scenarios of AOT=0.1 and 0.5 are plotted in grey and blue-green respectively.

remote surface sites are considered. This is attributed to both horizontal inhomogeneity within the OMI pixels and the inability of OMI to observe NO_2 close to the surface.

Reviewer comment 4: *On section 2: please provide some more details on the iDOAS observations. For instance: the field of view, number of pixels in across-track direction.*

Reviewer comment 5: *I am missing a formula that describes how VCD's are derived precisely from the (differential) slant column measurements. In my opinion, this should be described in more detail, although it has already been described elsewhere in full detail.*

Response

These two comments are similar to comments made by Reviewer 1. Details of the iDOAS and the retrieval have been expanded.

Reviewer comment 6: *The statements in Sect 3 are quite general. The words "usually" (p.3,l.3) and "frequently" (p.6,l.8) suggest a large number of profiles that are measured. However, these are not shown. Furthermore it is not clear if the profiles that are measured are representative for the plume or for more remote regions (see also the first comment).*

Response

It is true that these words express more confidence than what is warranted by the limited number of profiles measured during this campaign. The confidence that the authors feel is not from the profiles measured at the start and end of each iDOAS-measurement flight leg, which frequently were in background conditions; but rather from the literature on stable discontinuities over the Highveld, which is based on an analysis of long-term observations, and on how in the literature, elevated trace-gas and aerosol layers are frequently associated with these stable discontinuities, an observation corroborated by our own measurements. The impact of the presence or absence of such layers on the AMF has emerged as a finding of the present study, discussed above in the response to the reviewer's first comment.

Action

The language has been changed to reflect the above discussion.

Reviewer comment 7 *Figure 5 could be better readable if a grid was plotted on the left and right side of each cube. Furthermore it could be beneficial to use colours instead of different line styles and to provide a legend.*

Response

This was attempted, however the increased clutter in the diagram made it even more difficult to read.

Action

The 3-d figure will be replaced with the conventional plots shown in response to point 2 above. A 3-d figure will be retained to illustrate the principle of a minimum- and maximum-AMF plane.

Reviewer comment 8: *P.15, l24-25: "... indicating that ... 9 Aug". The terminology 'aged' versus 'young' might cause confusion, as some readers might wrongly think of 'photochemical aging'. It might be that what is here called an 'aged plume' is actually a region where the NO_2 profile is less shallow than for a 'young plume', and more in line with the prior NO_2 profile shape used for the OMI and/or iDOAS retrievals (see also the first* *comment). If this is the case, then one cannot say that OMI would be limited in its ability to capture the higher NO_2 gradient in the young plume because it is 'young'; for instance it could be more appropriate to say that the AMF derived using the prior profile shape used in the DOMINO product better matches the profile shape of an aged plume than the profile of a young plume. Please comment on this.*

Response

Indeed, the terminology may cause confusion. What is implied by an “aged” plume is one that is more dispersed in both the vertical and horizontal directions. The shallower horizontal gradients in a more dispersed, “aged”, plume are one reason why OMI might be better able to observe a more representative VCD, since the horizontal distribution of NO_2 across the OMI pixel is more homogeneous. This is what was meant in the discussion paper.

As the reviewer points out, the vertical dispersion of NO_2 in an “aged” plume could mean that the actual profile shape is closer to the *a-priori* profile used to calculate the satellite AMF. In addition, as shown in the figure above, potential errors in the AMF are less for profiles with a larger scale-height, and the divergence in the sign of the error for scenarios with- and without elevated layer found at low scale-heights disappears. This implies that AMF uncertainty will be smaller further downwind.

Action

The above discussion has been added, and the terminology has been changed.