AMT-2016-366: Responses to Anonymous Reviewer 1

Stephen Broccardo

June 20, 2017

Thank you for the positive review and comments.

The title of the paper has been changed to reflect the fact that the results are specifically from the Highveld.

In response to the specific comments:

Page 2, Line 32: *The slant density is the integral of the path length times the number density of that absorber, not the concentration (which describes the number density absorber as a fraction of the total air density).*

Response and action

Agreed. Edited p2. line 32 and p3. line 4: changed the word "concentration" to "molecular number density".

Page 3, Line 4: "to a first approximation, is slanted" is a bit confusing. Do you mean because of geometry?

Response and action

Yes, a geometric first approximation is what is meant. Added the word "geometric" to the sentence.

Page 3, Line 12: *The analysis of the NO_2 slant column is skimmed over without really any detail. I realize there is another paper describing this process, but could you say a few words about what other absorbers and parameters are fit, as well as individual fitting uncertainties from noise or systematic uncertainties? Also, not much info on instrument. What is SNR, are these from spectra that have been co-added spatially, what is the size of the CCD array (pixels), spectral resolution, spectral sampling etc? What is used for a reference spectrum?*

Page 4, Line 2: *Can you expand just briefly on why a photolytic converter is desirable? Also, why do you present NOy and not NO_2?*

Response and Action

In response to these two questions, the description of the measurements has been expanded. The paragraphs now read:

A DOAS instrument based on an Acton 300i imaging spectrograph employing a pushbroom viewing geometry, where each line of pixels across the instrument's swath is captured simulaneously on an Andor DU-420BU CCD, was fitted into the Aerocommander 690A. This CCD has 255 pixels in the across-track dimension and 1024 pixels in the spectral direction. The temperature of the spectrograph was kept stable at 30°C using a thermostatic heater in an insulated box, and the CCD temperature was set at -20°C using its own in-built thermo-electric cooler. Eight spectra were co-added into 32 across-track pixels, each with an across-track footprint of approximately 70m, assuming a flight altitude of 4500m above the ground. This was done in order to make optimum use of the optical resolution of the instrument. Along-track resolution is determined by the aircraft speed and the integration time on the instrument, which was adjusted automatically in-flight to avoid saturation of the CCD, and is generally about 100m (Heue et al, 2008). In the present study only the nadir pixel of the iDOAS is used.

Slant-column densities were retrieved using the WinDOAS software package. Absorption cross-sections for NO_2 (Vandaele et al, 1998), ozone (Burrows et al, 1999), water vapour (Rothman et al, 1998), O_4 (Greenblatt et al, 1990) were fitted across a spectral range of 432nm to 464nm. The Ring effect was accounted for using a appropriate cross-section calculated using the DOASIS software (Kraus, 2006). A reference spectrum was chosen from an appropriate location along the flight track far from known sources implying that slant-column densities from WinDOAS are in fact differential slant column densites. Satellite retrievals use a similar technique, using a measurement over remote ocean areas as an approximation of zero-NO_2. We adjust our slant-column densities using an offset in order to bring the vertical column densities from the iDOAS into line with the appropriate satellite measurement (either OMI or SCIAMACHY) in background areas of our flight track.

In addition to the imaging DOAS (iDOAS) instrument, the aircraft carried a Particle Measurement Systems Passive Cavity Aerosol Spectrometer Probe 100X (PCASP), operated with the pre-heater switched on; and a Thermo Scientific 42i chemiluminescence instrument with a molybdenum converter in the cabin, plumbed into the aircraft's scientific-air inlet in order to measure in-situ NO_y. In such instruments the converter converts NO_2 to NO, which is then measured by chemiluminescence; however a molybedenum converter also converts other nitrogen species. This can be avoided using a photolytic converter, however an instrument with a photolytic converter to measure NO_2 was not within the project's budget. The aircraft is fitted with a Rosemount ambient temperature sensor, and a separate pitot-static system for measurement and logging of static and dynamic pressure. The humidity sensor fitted to the aircraft did not function during this campaign. The aircraft's data aquisition system also logged parameters from a GPS (Global Positioning System) receiver.

Section 2: *Subheadings would increase the readability of this section. For example: "iDOAS NO_2", "In situ measurements", "Satellite observations" etc.*

Response and action

Subheadings have been added, and some of the paragraphs re-arranged to be under the relevant heading.

Page 4, Line 16: *This only the best case at nadir. The sides of the OMI swath are much larger.*

Response and action

The broadening of the OMI ground-pixel size towards the edges of the swath has been clarified, with the addition of the sentence "OMI pixels broaden in the across track direction as the viewing angle moves away from nadir".

Page 4, Line 17: *Can you give uncertainties in satellite VCD's? These can be quite large.*

Response

Uncertainties in the satellite VCD's are provided in the data files from the satellite retrievals, and could be shown. However, the figures in the present paper are already quite busy, and the focus is on AMF uncertainties in the iDOAS and satellite measurements (which, as shown in the response to reviewer 3's comments, we feel may be more uncertain than previously thought) and variability within each satellite pixel. We feel that trying to show too much will detract from this focus.

Page 4, Line 20: *I got confused here as on initial reading it sounded like the TM4NO2A was OMI data but with SCIAMACHY stratospheric slant columns as strat columns had just been mentioned.*

Action

The sentence has been changed to: "The TM4NO2A product is a product using slant column measurements from the SCIAMACHY satellite instrument and a similar scheme using model profiles and stratospheric columns from the TM4 model."

Page 4, Line 26: *Suggest mentioning swath width here and how many across track pixels there are here.*

Action

This has been added to the improved description of the DOAS measurements earlier in the section.

Page 5, Line 12: *Why do you average to 1.2 km? If purpose is to examine intra-pixel variability, how much cross-track and along-track information are you losing? Is this done to reduce error from noise?*

Response

This is in fact a time-based average (10 second moving average), which at the flight speed works out to approximately 1.2km. The iDOAS has 32 across-track pixels, we use only pixel 15 (counting from 0) in this paper.

Along-track averaging is used to reduce noise, for example an earlier version of Figure 7, without the averaging, and using a fixed AMF rather than attempting to quantify iDOAS AMF uncertainty is shown below.

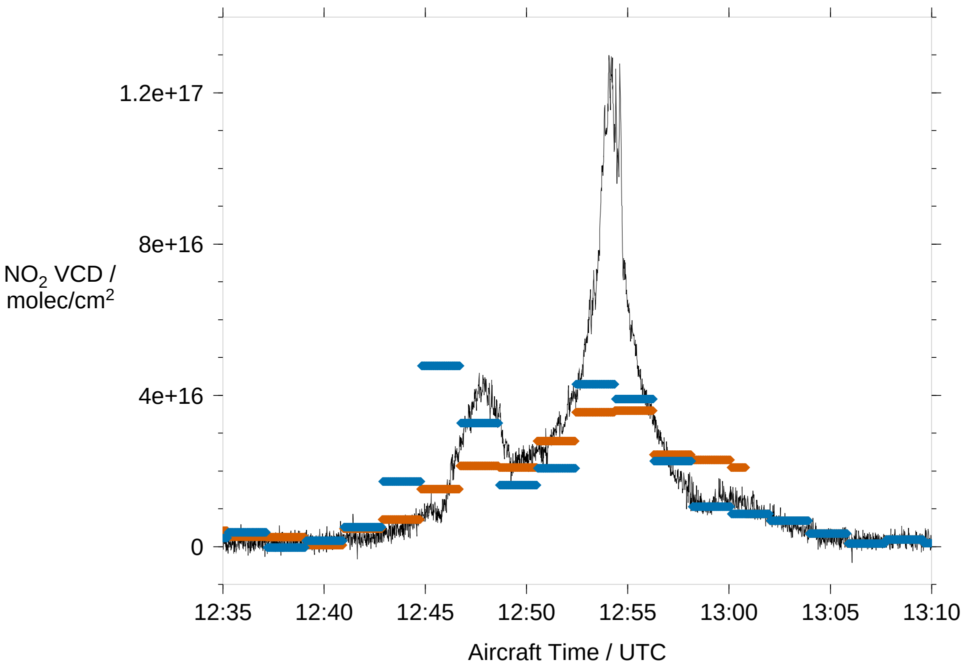

Figure 1: Airborne iDOAS measurements at full resolution (i.e. approx. 80m) on 9 August 2007 compared with OMI DOMINO V2 at aircraft nadir (orange) and one pixel upwind of (blue). A fixed AMF of 1.6 is used for the aircraft measurements in this figure.

Action

In the section on comparing DOAS measurements to the satellite (p5, line 9, which has now been moved under the appropriate sub-heading) the following has been added: "…the first is to average 80m-resolution nadir iDOAS measurements using a ten-second moving average in order to smooth out fine-spatial-scale variations and make a comparison with the much larger satellite pixels." and "The second approach is to calculate the mean and standard deviation of all nadir iDOAS measurements ..."

Page 6, Line 2: *Are there only 8 profiles total and what are locations? Maybe mention here to put in context. This intro to the section is a bit confusing as it presents the conclusion all of a sudden without referencing the data/figures. Maybe add an introductory sentence to ease into the analysis.*

Response and action

Yes there are only 8 profiles. An introductory sentence has been added: "During each of the flights, a vertical profile measurement was performed before and after the satellite-tracking portion of the flight."

Page 6: *Is the representation of some profiles as exponential valid in this region? Do you have any surface observations, or model profiles to check against? I realize this is done for contraining the error more than anything, so probably doesn't make a big difference, but I'm just curious.*

Response

We don't have any model profiles that we feel will be helpful. The state of modelling in South Africa is poor, mostly relevant to regulatory compliance. There are some other unpublished vertical profiles from other campaigns, but we chose to use only those from the 2007 iDOAS campaign. Near to surface sources, and at the spatial scale of the iDOAS, an exponential profile seems more appropriate. This is difficult to confirm, even with an aircraft profile measurement, since aircraft will typically climb and descend in a racecourse pattern, with straight portions of two minutes' flying between 180° turns, amounting to almost 10km of flying. Perhaps an ex-military pilot would be willing to fly more aggressively to make the horizontal extent of vertical profile measurements smaller. This would also require greater coordination with air-traffic control, who might find this sort of maneuvering unusual. A vertical profile climatology would be useful over the Highveld and in fact most regions of the world.

Figure 2 and 3: *Can you specify surface altitude here or show on figure? Is it the bottom of the y-axis?*

Response and action

Surface elevation at Richards Bay is sea level, and at Nelspruit the aircraft landed to refuel, so the profile is measured down to the surface. This information has been added to the captions and the text.

Figure 4 and relevant text: *What do you do for NO_2 profile in stratosphere in the model? Does NO_2 in the stratosphere contribute to the AMF or do you assume it cancels perfectly with the reference spectrum?*

Response

We assume that the change in stratospheric AMF during the course of a flight is small, and hence we assume that the stratospheric column cancels with the reference spectrum.

Section 5: *Could increase readability with subsection headings here.*

Subheadings have been added to separate the descriptions of flight on different days.

Page 12, Line 11: *Clearly the AMF changes drastically with the surface albedo. You are using your calculated AMF values to set bounds on the AMF error. How is the uncertainty in the AMF from uncertainty in the albedo determined (it's going to be high, with such a low resolution OMLER product)? Why not use MODIS albedo, or MODIS BRDF for an even better represention of the surface for high resolution observations from the aircraft?*

Response

Certainly, using a higher resolution albedo or BRDF would improve our calculation of the AMF, since surface properties have a large influence on the AMF. This would be the logical next step in sophistication of the radiative transfer modelling. It would appear that the uncertainty in the AMF from the profile shape is more important, and more difficult to quantify.

Page 15, Line 15: *I found this confusing. What is your reference? Do you have remote ocean measurements from the aircraft?*

Response

Reference spectra for our retrieval are chosen from background regions of the flight. Since background regions over land will have a higher column density than remote maritime regions measured by the satellites, we assume that the satellites' measurement of background regions over land are "correct" and we shift the aircraft measurements to match the satellite. This seems reasonable, since background regions have shallow horizontal gradients and the errors in the satellite measurements we describe will be small. This procedure does highlight the problem with using one nadir-viewing scattered-light instrument to validate another. Ideally some other independent measure of vertical column density, with less dependence on *a-priori* profile shape should be used as a reference for both the aircraft and satellite measurements.

Page 14, Line 14: *Could the same effect be achieved by putting the iDOAS on an aircraft that flies higher?*

Yes, a wider swath could be achieved by flying higher.

Page 14, 19: *"Underestimates" the peak only. What it's actually probably doing is just averaging out everything in the field of view (so you could equally say "overestimates the background").*

The regression line is fitted through iDOAS data that is averaged (along track) to the resolution of OMI, hence we are not comparing the peak iDOAS to OMI but rather a 1-dimensional spatial average.

Page 13, Line 5: *Not sure why you have to use two fitting schemes. Does that tell us anything?*

Response

The two fitting schemes attempt to demonstrate that the background measurements by the iDOAS and satellites are a better match than measurements close to sources. However, the reviewr makes a good point: it doesn't really tell us much.

Action

The inset figure has been removed.

Page 15: *You make a few comments about plume age, source etc. I noticed you used HYSPLIT earlier in the paper. Can it tell you anything about these specific cases?*

In cases where HYSPLIT proved to be useful for plume age estimates, this has been added to the discussion.

Page 16, Line 19: *Can you remind us of SCIAMACHY overpass time here? Also Figure 13 caption reads a bit like the times are for the satellite observations (I'm guessing it didn't take 50 minutes to fly over the region!)*

SCIAMACHY overpass times are shown in Figure 13 in each SCIAMACHY pixel. The approximate overpass time has been added to the caption.

Page 17, Line 6: *Not sure you can draw any conclusions about SCIAMACHY vs OMI at all here. There is a very limited amount of SCIAMACHY data at high NO_2 values. Obviously your slopes are very different on different days with OMI as well.*

True. We have edited the text appropriately to indicate that no real conclusion can be drawn.

Fig 6 and similar: *Can you specify that these are 1.2 km averages in caption or in text (which I'm assuming they must be?)*

Yes these are 1.2km averages. This can be added to the caption.

Figure 6: *I can't tell which is sub-aircraft pixel as it looks like flight was right down the border of two cross-track positions. Can you clarify this in*

text?

Yes, the flight track does cross from one OMI-row to the next. This has been clarified in the description of the figure.

Figure 7 and similar, *and relevant text: The average isn't technically over the "area" of the OMI pixel, which might have a 13x24 km2 size. Clarify.*

Agreed, it is the average of the full-resolution iDOAS measurements (from the nadir iDOAS pixel only) over the length of flight track within the OMI pixel. The captions and text have been amended to use the term "line-average".

Figure 7 and similar: *Specify colors for elevation/albedo subplot.*

A description of the colours has been added to the caption.

Figure 8: *Specify which are OMI and which are SCIAMACHY observations (maybe in legend?)*

Yes, specifying which are OMI and SCIAMACHY is a good idea. This has been done.