## Author reply Referee #1

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-329, 2016.

We would like to thank referee #1 for his useful comments and detailed questions.

<u>Legend:</u>

- **Referee comment (bold)**

*Author's answer (italics)*

Modified manuscript text (blue)

The paper describes NO2 measurements performed over Bucharest during the AROMAT campaign in September 2014. Nitrogen dioxide was observed with the Airborne imaging DOAS instrument AirMAP from the University Bremen. The focus of the paper is on the data retrieval and a short interpretation of the observed tropospheric column densities. To large parts it is interesting to read especially the albedo correction is very a nice idea. In some parts however the paper might be shortened a bit to stress out the main focus (AMF calculations with the highly varying albedo and the interpretation of the data). Therefore I suggest:

- **Shifting the stratospheric correction (4.1.2) to the appendix. After half a page the authors state that the corrections discussed above were neglected here. From Table 1 one can easily calculate that the difference in the stratospheric AMFs is close 0.2 for flight 2 and around 0.05 for flight 1. Moreover parts of this section focus on the satellite retrieval and are not directly related to the airborne instrument, though used for the correction in general.**

*The stratospheric correction was not neglected in the analysis. The respective sentence reads: "As the measurements shown in this study were performed around noon, the diurnal variation in $SCD^{strat}$ are very small and could thus be neglected." We intended to use the subjunctive and did not state that we neglected the stratospheric correction. We acknowledge the ambiguity of that sentence and now changed it, see below. Since we have used the stratospheric correction in our retrieval, we prefer to keep this section in the main part of the manuscript.*

As the measurements shown in this study were performed around noon, the diurnal variations in $SCD^{strat}$ are very small and stratospheric correction is of minor importance.

*Considering the comment of Referee #2 we have also added the following two sentences to the text.*

For the two flights on 2014-09-08 and 2014-09-09 the stratospheric VCD was estimated to be around 3 x $10^{15}$ molec $cm^{-2}$.The maximum change in the stratospheric SCD with respect to the reference spectrum, $\Delta SCD^{strat}$, was 1.5 x $10^{14}$ molec $cm^{-2}$ and 3 x $10^{14}$ molec $cm^{-2}$, respectively.

- **Shortening the description of the aerosol profile chosen and the other AMF parameters. I am not sure whether it suitable to use an aerosol profile from August 2015 for measurements in September 2014. But if a typical profile is assumed it might be used.**

*In order to improve the readability of that section we have now put the information about the profiles of aerosols (and $NO_2$) into separate subsections.*

*To answer the comment about the suitability of the used aerosol profile: Climatological data of the AOD in Bucharest is available on the AERONET website (https://aeronet.gsfc.nasa.gov/new_web/V2/climo_new/Bucharest_Inoe_500.html#2014). For September 2014, the monthly average of the AOD at 450nm is 0.26 +-0.1 (mean+-std). The used profile 31a (AOD=0.22) is the closest match of the experimental profiles to the climatological AOD value and is within the standard deviation of the monthly average. We thus assume that the used profile can be regarded as a typical profile for this location and season although the real aerosol profile during the measurements can of course be different. We have now added this information in the text.*

For the analysis of both flights in Sect. 7, we have used the profile FUBISS 31a. The AOD of this profile is the closest match to the monthly average AOD, available from AERONET measurements, having a value of $AOD_{450} = 0.26\pm0.1$, (mean±stdev) (https://aeronet.gsfc.nasa.gov/new_web/V2/climo_new/Bucharest_Inoe_500.html#2014).

- **For the albedo correction two reference pixels from the MOD09A1 database, which has a resolution of 0.1 degree, are chosen. Why is the reference taken over the city? According to the map the intensity varies by a factor of 2 with high small scale variations. An area with less variability might be a better choice?**

*In general we would agree that an area with less variability would be a better choice. However, practical limitations make it almost impossible to find such an area.*

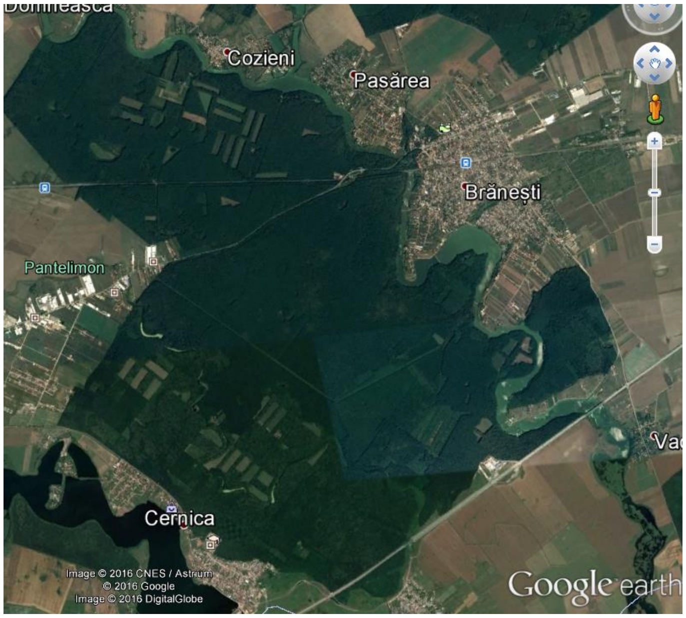
*1. The scaling factor ($I_{modRef}$ / $I_{measRef}$) is determined for each viewing direction separately because the individual fibres have a non-uniform transmission. In order to discriminate the influence of outliers, such as high intensities caused by sun-glint or direct reflection it is desirable to average over many measurements. To determine the surface reflectance, the whole swath (>3km) must thus cover a homogeneous surface over a long distance along-track.*
*Possible candidates for surfaces with a homogeneous surface reflectance are (a) the lake in the north-west and (b) the forest in the north-east of the covered area. In the first attempts of the method development we tried to use these areas together with reflectance data from the USGS spectral library (https://speclab.cr.usgs.gov/spectral-lib.html) and the ASTER spectral library*

*(http://speclib.jpl.nasa.gov/). However, using these areas and spectral libraries did not provide satisfying results.*

*The lake is not ideal because the water surface shows strong directional effects (cf. Fig 13 in paper) and thus the reflectance depends on the relative azimuth of the measurements. Furthermore its area is relatively small and does not allow averaging over many measurements.*

*The forest proved to be problematic for several reasons:*

- *The spectral databases contain spectra for different vegetation types, but the forest as a whole appears darker than individual leaves. This can be explained by the fact, that the tree canopy and the ground below absorb more radiation than an individual leave would. Using the reflectance values from the spectral libraries as $A_{Ref}$ thus resulted in too large surface reflectances.*
- *The vegetation in the forest area is not uniform. The woodland is interrupted by several forest glades (cf. figure below) having a different reflectance than the woodlands. When these forest glades are excluded from the surface reflectance reference area it cannot be assured that enough intensity measurements can be averaged.*




*2. We have chosen to use a global climatology to allow the application of the method in a consistent way for measurements in different regions and different times of the year. By using 0.1°x0.1° grid cells in the urban area a lot of AirMAP measurements (approx. 1300-2000 measurements per viewing direction) are averaged. This averaging makes the method more robust against outliers and directional effects of the surface. We have shown that the presented method gives consistent and satisfying results.*

*Another possibility would be to use satellite data on a daily basis. Reflectance data is available in relatively high spatial resolution from instruments such as MODIS (500m) or Landsat. However these* *datasets may also be useless if the overpasses on the days of the research flights are affected by clouds. Thus we have chosen to use a climatology, which is based on MODIS data.*

- **Airborne measurements provide high resolution and good data that might be used for many interesting scientific questions. The comparison to satellites is from my point of view not really an interesting question; we did that too often in the past. So I thank the authors that they did resisted this temptation.**

**Detailed comments:**

- **P3 l24 & p4 l1: The area of the city in km2 and the area covered by the flights are not necessary. The maps show that most of the city including some suburbs were covered.**

*Although this information may not be necessary for the interpretation of the data shown here, this information about the area covered and duration may be interesting for the reader. Thus we prefer to keep this information in the text.*

- **P4 l1: Flight 1 (2014-09-08) started in the north and ended in the south, on this day the wind blew from north east (figure 2). Is it possible that the aircrafts plume was observed? The influence might be small and can hardly be estimated or?**

*If the aircraft's plume was observed during the flight, the influence must be small. Otherwise these emissions should show up in the dSCD results, especially in the clean regions. Either the $NO_2$ signal is too small to be detected by AirMAP, or the plume was not within AirMAP's field of view (FOV).*

*In order to check if the aircraft's plume could have been transported into AirMAP's FOV, we can estimate if the sum of the half swath width(w) (in upwind direction, at aircraft plume altitude) and the travelled distance by an air parcel into direction of the next flight line ($d_{South}$) is larger than the separation of the flight tracks ($d_{Track}$)*

$$test = (d_{South} + w) - d_{Track}$$

*If $test > 0$, then the aircraft's plume from the previous track was in AirMAP's FOV.*

*If $test \leq 0$, then the aircraft's plume from the previous track was not in AirMAP's FOV.*

*To calculate the displacement of exhaust $NO_x$ in southern direction the following formula can be used:*

$$\mathrm{d_{South}} = \mathrm{time} \times \mathrm{windSpeed} \times \cos(\mathrm{windDir})$$

*On 2014-08-08, the wind was coming from 57°N with a speed of 1.1 $ms^{-1}$. The duration of one flight leg is about 10 minutes. Thus, the maximal time between adjacent measurement locations is 20 minutes. Plugging these values into the equation above yields:*

$$\begin{aligned} d_{South} &= 1200s \times 1.1\ ms^{-1} \times \cos(57°) \\ &= 719\ m \end{aligned}$$

*The half swath width of AirMAP can be calculated as follows:*

$$
\begin{aligned}
w &= H \times \tan\left(\frac{FOV_{across}}{2}\right) \\
&= H \times \tan\left(\frac{51.7°}{2}\right) \\
&= H \times 0.48
\end{aligned}
$$

*, where $H$ is the vertical distance to the object of interest (the possible plume). Because the aircraft altitude was constant, the vertical distance to the plume is determined by the vertical movement of the air, i.e. the distance an air parcel has travelled into ground direction. Because the speed of the downward vertical transport is not known, it is simply set to the same value as the horizontal wind speed. Using this assumption, an air parcel may be transported into ground direction with:*

$$d_{vertical} = 1200s \times 1.1\ ms^{-1} = 1320m$$

*Setting $H$ to $d_{vertical}$ results in a half swath width of $w = 634\ m$..*

*The distance between adjacent tracks is about $d_{Track} = 1900m$.*

*Now these values can be plugged into the first equation to see whether the aircraft's plume could be observed.*

$$
\begin{aligned}
test &= (719m + 634m) - 1900m \\
&= 1353m - 1900m \\
&= -547m
\end{aligned}
$$

*This result is much smaller than 1 indicating that an observation of the aircraft's plume is rather unlikely because the locations of the outermost viewing direction and the exhaust $NO_x$ are separated by more than 500m.*

*The approach developed above has of course many uncertainties involved. First of all, the horizontal wind speed at flight altitude is probably larger than the measured wind speed on ground. On the other hand, the vertical wind speed is usually much lower than the horizontal wind speed. However, even if the air masses could have been transported into AirMAP's field of view, we do not see any influence on our measurements.*

- **P4 l2: include "around local noon" in the flight description.**

Both flights were performed around local noon under cloud free and sunny conditions with low wind speeds (< 1.5 $ms^{-1}$ )

- **P5 l18: The grating with 600 g/mm and 500 nm blaze wavelength was used. The other gratings (the Acton 300i allows up to 3) are not important in this context. The same grating was already used in Schönhardt et al. (2014).**

*The 300g/mm grating was also used during the campaign, but not for the flights presented here. In future publications we may show results, in which the 300g/mm grating was used for measurements in the UV spectral range. In order to inform the reader about the availability of that different instrumental setup we prefer to keep that information in the text.*

- **P5 l25: Does the size of the ground pixel change towards the edges of the swath? Later on it is mentioned for large roll angles of the plane but under normal flight conditions?**

*Image distortions, such as barrel or cushion distortion were not characterized in the lab. However, the manufacturer's specification states that the objective has a low distortion. Thus it is assumed that the instantaneous field of view of the viewing directions is constant in terms of solid angle. When the footprint is projected on the ground it becomes larger with increasing VZA. This increase affects the across-track width, as well as the along-track width of the ground footprint. The across-track width of a ground pixel can be described by the difference of the tangens between the VZA of the outer vertex ($VZA_{outer}$) and the VZA of the inner vertex ($VZA_{inner}$) of a viewing direction i.*

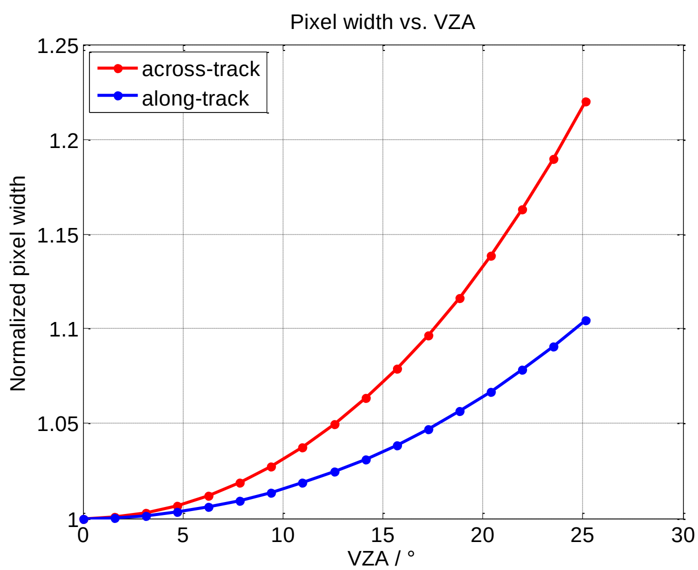
$$PixelWidth_{across} = H \times \tan\left(VZA_{i_{outer}}\right) - \tan\left(VZA_{i_{inner}}\right)$$

The instantaneous pixel width in along-track direction can be described by:

$$iPixelWidth_{along} = \frac{1}{\cos(VZA_i)} \times H \times 2 \times \tan\left(\frac{iFOV_{along}}{2}\right)$$

Where H is the altitude a.g.l. and $iFOV_{along}$ is the instantaneous field-of-view in along-track direction.

*The figure below shows the increase in pixel width with increasing VZA normalized to the central viewing direction and illustrates that the across-track pixel width increases by about 20% for the outer viewing directions during a level flight (not in turns). The along track pixel width increases by about 10%.*



- **P6 l3: There is an updated version of the solar atlas by Chance and Kurucz (2010), you might use instead.**

*Thank you for the suggestion. We will test the updated version of the solar atlas. However, we only use the solar atlas, convoluted with the slit function for the calibration of the spectra. Considering the spectral resolution of the instrument, the changes are expected to be very small.*

- **P7 l4: "multi measurements in one grid cell were averaged" how good do the multi measurements agree? At least for the vertical column that might be interesting to see.**

*Variability between multiple measurements in one grid cell can have several reasons. The measurements are affected by fitting noise, directional effects of the surface, differences in light path from geometry (sun and instrument), imperfect geolocation and of course temporal variability in $NO_2$.*

*A possibility to investigate how good the averaged measurements agree is to look at the standard deviation of the measurements per grid cell. A map of the standard deviation within the grid cells is shown in Figure 1 (below) for the results from the flight on 2014-09-08. Note that the range of the color scale was reduced by a factor of 10 as compared to the $NO_2$ VCD map in the paper. Figure 2 (below) shows the number of measurements that were averaged in each grid cell. Grid cells that only contain measurements of one flight line generally average 2 measurements (along track). Grid cells in the regions of overlapping swaths from adjacent flight tracks contain about twice as many measurements. The standard deviation of the $NO_2$VCD in the regions of non-overlapping swaths (i.e. where N=2 or N=3) is about $1.7 \times 10^{15}$ molec/cm$^2$. In areas of overlapping swaths (N>=4), the standard deviation is $2.0 \times 10^{15}$molec/cm$^2$. The value of the standard deviation of the whole dataset is $1.6 \times 10^{15}$ molec/cm$^2$. In the case of grid cells corresponding to one flight line, these values can be explained to a large extent by the fit error of the DOAS analysis. When taking the mean of the quotient of the dSCD fit error of $NO_2$ and the corresponding AMF the result is:*

$$\frac{1}{n} \times \sum \frac{fiterror_{NO_2}}{AMF} = 1.44 \times 10^{15} molec\ cm^{-2}$$

*This value is a bit smaller than the value of the standard deviation for the non-overlapping swath regions. However, some natural variability of the $NO_2$ field also contributes to the variability of one grid cell. The standard deviation in the regions of overlapping swaths is larger, which is probably caused by natural variability of $NO_2$ between successive overpasses and by the fact that the ground spot is observed "from the other side" (from North to South, then South to North). This means that the air mass over which the measurement integrates is not the same.*

*Computing standard deviations for grid cells with only 2 to 4 samples may not be a good measure. An alternative approach is to compute absolute differences between multiple overpasses. For this we have separated the measurements according to the heading of the aircraft. Two data grids were produced,. One with $NO_2$ VCDs when the aircraft was heading to the East (yaw=90°+-5°), and one with $NO_2$VCDs when the aircraft was heading to the West (yaw=270°+-5°). This approach does not consider variability of measurements in one grid cell that were averaged along track, but the standard deviation can be computed from a much larger sample size. A histogram of absolute differences (East-West) is shown in*

Figure 3*(below). From almost 12000 compared grid cells, we find a mean close to zero and a standard deviation of 2.9 x $10^{15}$molec $cm^{-2}$.*

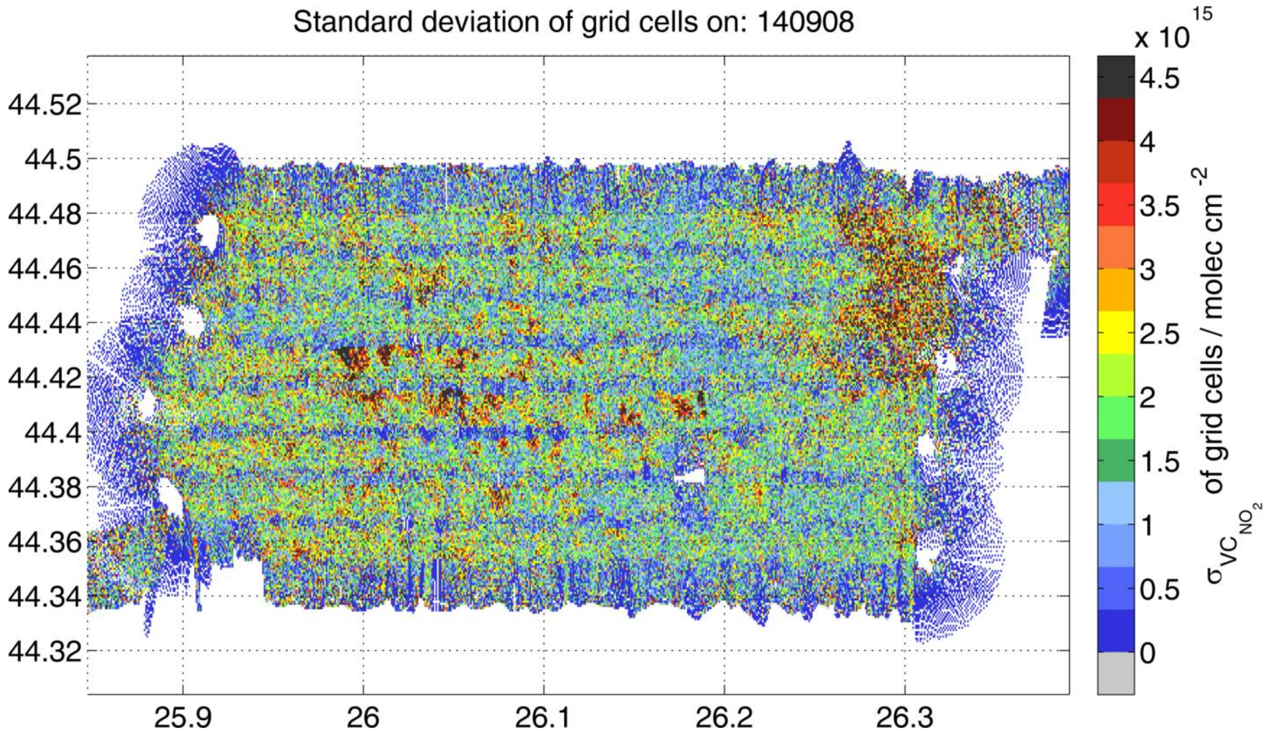



**Figure 1: Standard deviation (σ) of the NO2 VCD average per grid cell. Data is taken from the flight on 2014-09-08.**

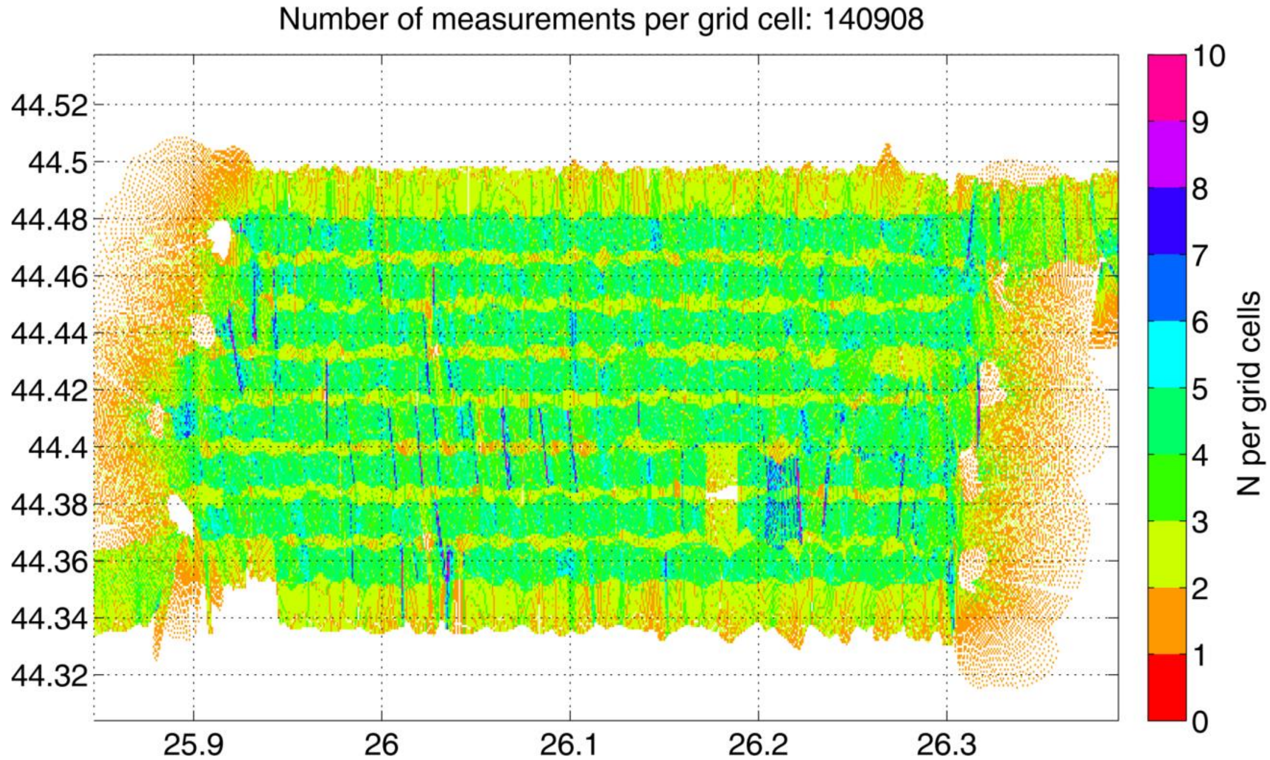



**Figure 2: Number of measurement averaged per grid cell. Data is taken from the flight on 2014-09-08.**

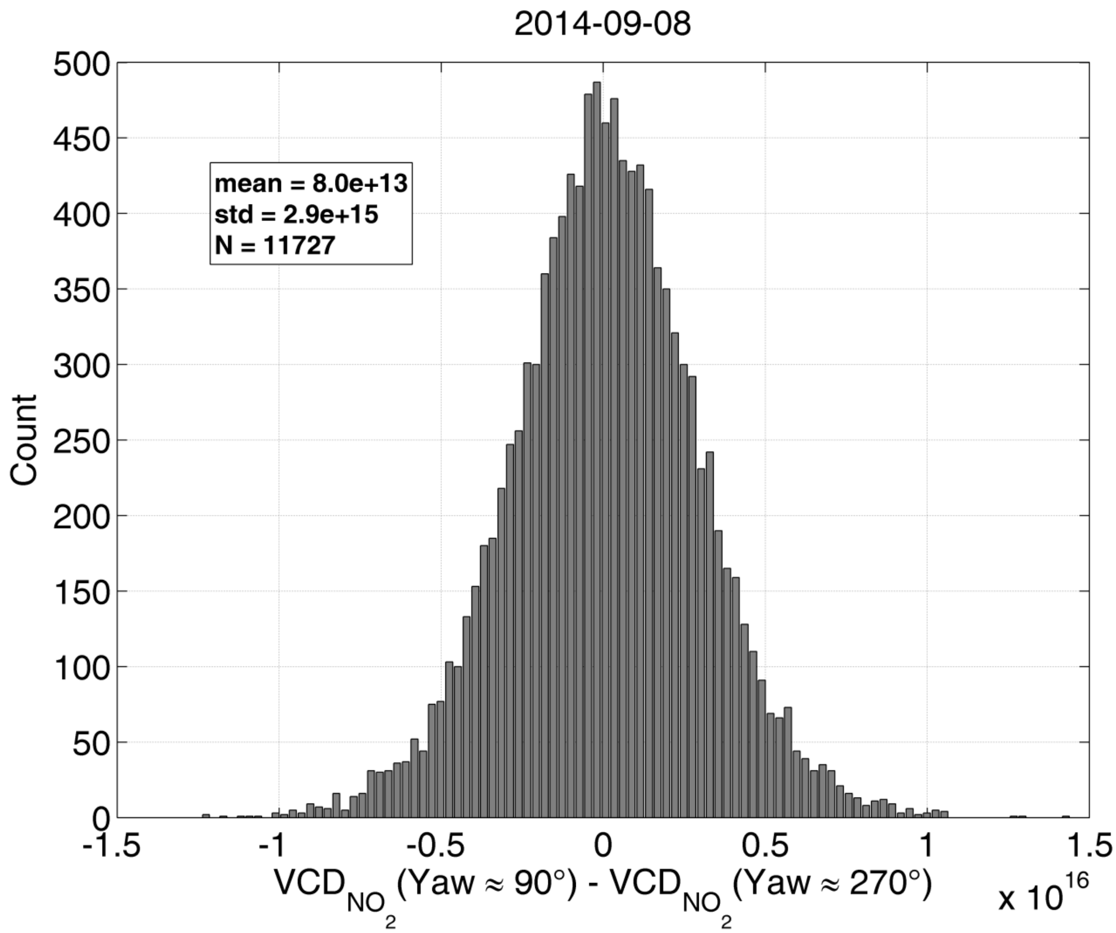



**Figure 3: Histogram of absolute differences between co-located $NO_2$ VCD from different overpasses during the same flight.**

- **P10 l5: Please replace "normal to the surface" by "vertical", the local surface might differ from the horizontal e.g. mountains.**

The change in light path length, as compared to a vertical path, is usually expressed in the form of an air mass factor (AMF), which is defined as the ratio of slant and vertical column densities:

- **P10 l13: The sentence beginning with "The dSCD and the AMF. . ." is not clear, please clarify.**

In these equations the terms dSCD and AMF refer to the trace gas amount fitted from a single spectrum and its corresponding air mass factor.

- **P13 figures 4 and 5: The BoxAMF for 440 nm are shown here and in section 5 the wavelength of 437.5 nm is mentioned (P17 l6), please use consistent data. Even though the difference in the AMF due to this wavelength change might be small,**

*We have computed new BAMF at the center wavelength of the fit window (437.5 nm). The new computations also used a finer altitude grid. Figures 4 & 5 were updated accordingly.*

- **Figure 5: Is it useful to add a vertical line at 0.04 and 0.1 for a better comparison to Figure 4?**

*Figure 5 was updated to show the AMF at 437.5 nm. Additionally the suggested vertical lines at 0.04 and 0.1 were added to support the reader.*

- **P14 Figure 6: Is the figure necessary? In this figure it looks if the VZA is mainly in flight direction.**

*We have updated the figure.*

- **• P18 l6 Add a point "g)" For each measurement the retrieved albedo was used for the AMF calculation.**

*Thank you for this suggestion. We have added the following sentence after point f).*
The above procedure yields scene specific surface reflectances for each measurement which are later used in the AMF calculations.

- **P19 Figure 11 and P20 Figure 12: these two figures both show a linear correlation and a histogram. It would be nice if the same scales for the two histograms were used.**

*Thank you for the suggestion. The histograms of Fig. 11 & Fig 12 now have the same scale.*

- **P21 Figure 13: The grey and green areas are slightly confusing, on the other hand they might be important. Does it look better or worse if you show the street lines only?**

*We have now plotted the map on a gray background to make the map more clear and less overloaded. Figure 13 was updated accordingly.*

- **P21 l1-5: For each measurement the individual retrieved albedo was used. This might be mentioned a bit earlier in the analysis description e.g. P18 l6 g) (above)**

*Done. See answer for comment P18 l6.*

- **P25 figure 17: The measured wind directions does not agree with the apparent distribution of the plume, in the emission estimates the later wind direction is considered. Specify in the discussion of the figure.**

*Thank you for this remark. We have now added the following sentence in the discussion of the figure:*
Comparing the apparent wind direction, as seen in the $NO_2$ distribution, to the data record of the meteorological station at Baneasa airport it is worth to note that there is good agreement on 2014-09-09, whereas a mismatch of the wind direction is observed on 2014-09-08.

- **P25 l10 ff: The airborne measurements are very interesting by themselves. There is no need to compare to satellite data.**

*We are happy to hear that.*

- **P29 Table 6: just for completeness add the background correction here.**

*Table 6 was updated to include uncertainties related to the correction of the tropospheric background.*

- **P30 Table 7: Why is the uncertainty in the AMF calculation comparable, while MPIC uses a simple geometric approximation and the telescopes' elevation is 22 and the UGAL instruments points to the zenith and the AMF calculation is much more sophisticated. What is the typical resolution of these measurements for the local traffic and speed?**

*a. Uncertainties: The uncertainties stated were taken from the respective publications (Shaiganfar et al. 2011) for the MPIC retrieval and (Constantin et al. 2013) for the UGAL retrieval.*

*b. Resolution: The spatial resolution of the car measurements depends on the integration time used in the data acquisition and the speed of the car. Because the measurements were performed in an urban area, the speed is variable.*

*The statistics of the spatio-temporal resolution of the car measurements are summarized in the table below. The figure below the table shows the data from which these valued were computed.*

*In case of the MPIC instrument, which points to an elevation of 22°, the spatial resolution is coarser than the distance driven between consecutive measurements due to horizontal averaging. The magnitude of the spatial averaging depends on the height of the $NO_2$ profile. As already stated in Eq. 9 (manuscript), the horizontal averaging can be described by:*

$$dist_{ave} = \frac{Height_{NO_2}}{\tan(elevationAngle)}$$

*Assuming a box profile of 500 m, this results in a horizontal averaging of about 1.2 km, which has to be added to the distances driven by the MPIC car.*

| Parameter | MPIC | UGAL |
|---|---|---|
| **Temporal Resolution (mean) [s]** | 77 | 32 |
| **Temporal Resolution (median) [s]** | 62 | 32 |
| **Temporal Resolution (std) [s]** | 33 | 3 |
| **Spatial resolution (mean) [m]** | 423 | 285 |
| **Spatial resolution (median) [m]** | 348 | 276 |
| **Spatial resolution (std) [m]** | 447 | 218 |

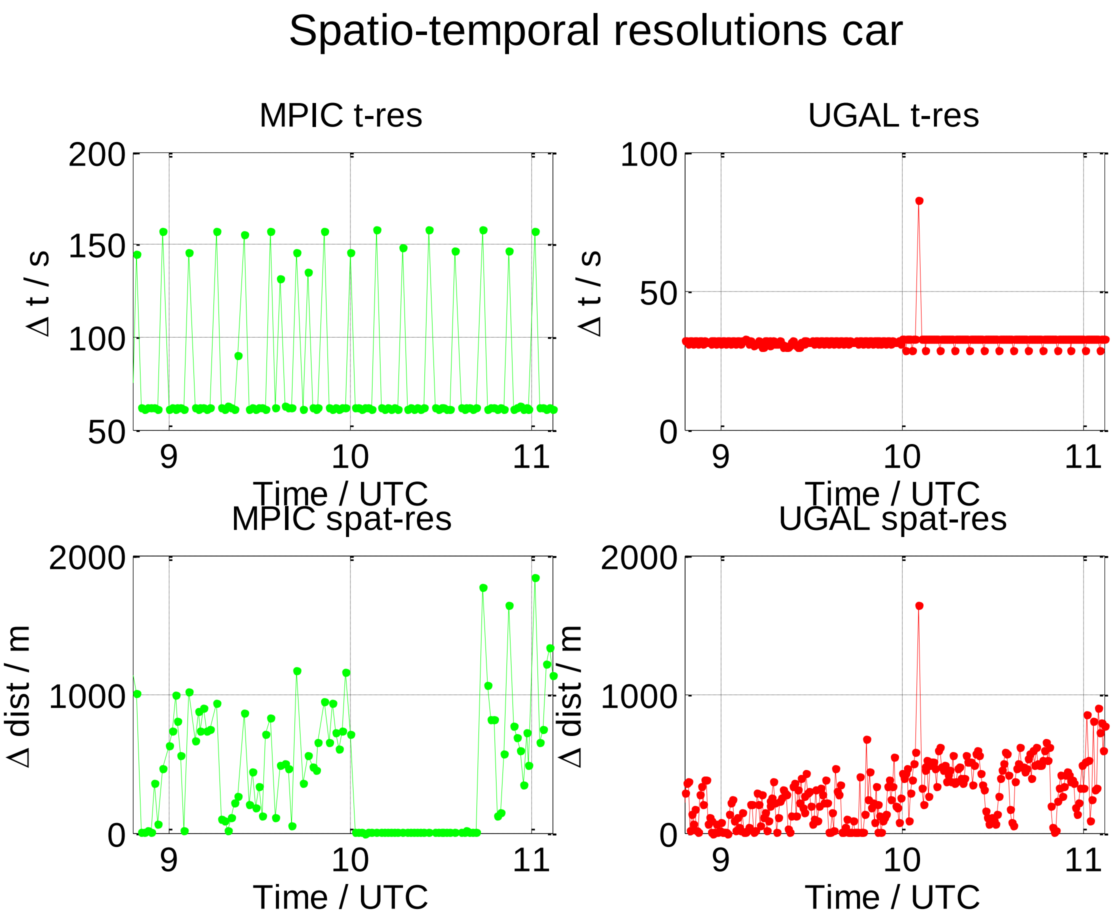



Figure 4: Upper panel: Temporal resolution of the car measurements (left: MPIC, right: UGAL). Lower panel: Spatial resolution i.e distance traveled by the car between the measurements

- **P31 l10: The mismatch caused by the forward direction of the MPIC instrument's telescope was not corrected for?**

*No, in the current approach the car's position was used for both instruments as co-location criterion. The comparison between airborne measurements and off-axis car-DOAS measurements may be improved in future analyses, taking into account the azimuth and the elevation angle of the measurements as well as the $NO_2$ profile.*

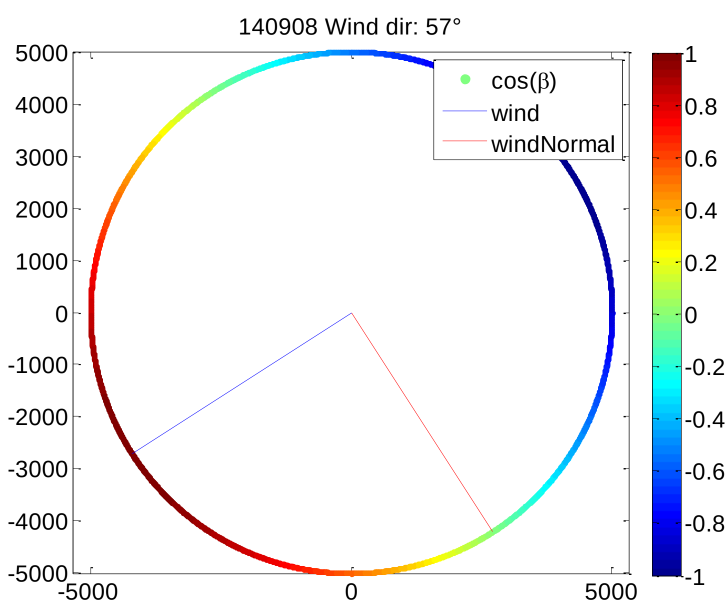
- **P33 l15 ff: The presented airborne measurements provide a much higher resolution than the OMI data used by Liu et al. (2016), moreover there are some "isolated" plumes like point source 9 in figure 15. If not for now but for the future it might be worth to estimate the NOx lifetime form the airborne measurements. Even the big Bucharest plume might be used, however here the assumption of a point source is no longer valid.**

*That would be an interesting research topic, indeed. However, for this paper such an analysis is out of scope.*

• **P33 l20 ff: This circular approach seems appropriate for a point source. However a city is rather an area source than a point source (p34 l13). Also emissions upwind of the centre are to be considered. In the current approach these emissions are counted at a distance of l and it is not important if the distance is upwind or downwind of the city. But in a Cartesian system with one axis along the wind direction, integrating perpendicular to the wind, this effect can be taken into account. As the authors already mentioned, in the circular approach with radii steps of 100 m the area increases between 0.06 km2 in the centre to 12.5 km2 between 9.9 and 10 km (p34 l13).**

*In our current approach emission upwind of the city center are considered as negative contribution to the total emission rate. We do not integrate the $NO_2$ over the area inside the circle, but along the circumference of the circle (line), see also P32 l13. The circle was only used to define the sampling points at different distances from the center. In that sense the method is similar to car-DOAS measurements when driving around the city on a circular road. The inflow of $NO_2$ rich air is accounted for in the term $cos(\beta)$, which is the angle between the normal of the wind direction and the azimuth of the line segment (P34 L1). This ensures that $NO_2$ transported into the encircled area becomes negative and has a negative contribution to the outflow. This is illustrated by the following figure which shows the term $cos(\beta)$ (color coded), the wind direction and the normal to the wind direction for the flight on 2014-09-08 at an example radius of 5km.*

The inflow of $NO_2$ enriched air is accounted for in the term cos(\beta), which is the angle between the normal of the wind direction and the azimuth of the line segment. This term ensures that $NO_2$ transported into the encircled area becomes negative and does not contribute to the emissions determined from within the circle. The term $\Delta s$ is the Euclidean distance between the sample locations.



**2 Typos and small corrections:**

- **P2 l3: "processes" instead of "precesses"**

*Fixed.*

- **P27 l2 and l5: the surface reflectance for Figure 4 is given with 5% or 0.04, one of the numbers is not correct. In the figure label it is 0.04.**

*Corrected.*

- **P33 l21: Is it really a Cartesian coordinate system. In a first approximation the geographical system on that small scale is Cartesian. To me the new one looks like a polar system.**

*We have used a Cartesian coordinate system. First, the grid of $NO_2$ VCDs was converted to Cartesian coordinates. Then the sampling angles $\phi$, were defined from 0° to 360° in steps of 0.1°. In the next step, the sampling locations were obtained by conversion from polar to Cartesian coordinates, using the respective radius R.*

$$x = \sin\left(\phi \cdot \frac{\pi}{180}\right) \cdot R$$
$$y = \cos\left(\phi \cdot \frac{\pi}{180}\right) \cdot R$$

**3 References:**

- **For some references the doi is not given, it is not mandatory but good practice.**
- **Constantin 2012**

*Unfortunately this publication does not have a DOI.*

- **, Heue (2005 and 2008),**
- **Lohberger 2004**
- **and Rozanov 2014.**

*Fixed.*

**please recheck the others as well**

- **For the two books Burrows, Borrell and Platt (2011) and Platt and Stutz (2008) there are brackets around the doi and doi is written in capital letters while in all other references it is not.**
- **Chance, K., and Kurucz, R. L., An improved high resolution solar reference spectrum for earth's atmosphere measurements in the ultraviolet, visible, and near infrared. J. Quant. Spect. Rad. Trans., 111, 1289–1295, doi: 10.1016/j.jqsrt.2010.01.036, 2010.**

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-329, 2016.