Author Comment on „A wide field-of-view imaging DOAS instrument for continuous trace gas mapping from aircraft“ by A. Schönhardt et al.

Referring to the Referee Comment of Referee #2, from 23 April 2014.

We are grateful for the comments, corrections and suggestions of Referee #2.
In the following, we address all the points raised by the referee. Original comments are shown in black italics, our answers in black normal font and new text for the manuscript in blue.

Part 2. major points

Comment (1)
The plume emissions are performed for a distance of 6 km to the stack, which is probably good to estimate emission correctly. On the other hand this instruments offers the possibility to study the chemical processes on a small scale (30 × 30 m). For this purpose a more detailed discussion of the observation close to the stack would be of interest.

Answer to Comment (1)
Mapping the emission plume of point sources in detail is a strength of the AirMAP instrument. The current study aims at demonstrating this ability. We agree on showing more details on the plume overpasses closer to the stack and we discuss the observations. In total, there are five overpasses close in time at different distances from the stack. A new figure is included, and shall replace Figure 14 in the AMTD version. Figure 13 and 14 are now swapped in the revised manuscript. A more detailed discussion of the plume chemistry, however, shall not be included as we consider this not the right place to do so. That would be a paper of its own and should be placed in a journal other than AMT. An additional figure is included (Figure 15) which shows the integrated NO_2 amount across the plume with respect to the distance of the stack. The integrated NO_2 amount (line integral) takes into account the ground pixel length as well as the relative angle between the cross section and the direction of plume movement, i.e. the wind direction. Cross sections of the five overpasses are included in this figure, and data is based on the LOS35 evaluation. So in total, the integrated NO_2 amount from 175 cross sections is displayed.

New text included in Section 7:

The NO_2 plume is investigated during several overpasses.
In Fig. 14, five overpasses over the exhaust plume are displayed. They show the NO_2 measurements at different distances downwind of the power plant stack. The overpass furthest away has a distance of around 6 km from the stack and is used for an emission estimate.
Many details in the plume structure are resolved by the AirMAP measurements. At 09:40 UTC close to the stack, NO_2 amounts are still rather low, as NO_2 needs time to form from NO and ozone. Especially the two overpasses at 09:52 and 10:00 UTC show that the plume structure is strongly inhomogeneous, at 09:52 UTC the largest NO_2 amounts are not found in the lateral center but towards the southern edge of the plume, at 10:00 UTC an interruption of the plume in wind direction (across track), i.e. a discontinuity due to atmospheric turbulence, is observed.
Figure 15 shows the integrated NO_2 amount across the plume with respect to the distance of the stack. The integrated NO_2 amount (line integral) takes into account the relative angle between the cross section and the direction of plume movement, i.e. the wind direction, see discussion below. Cross sections of the five overpasses are included in this figure, and data is based on the LOS35 evaluation. In total, the results from 175 cross sections are shown.
The detailed maps of the plume and the integrated NO_2 amounts show that emission estimates from single cross sections would lead to fairly different results. NO_2 emission rates Q from the power plant point source are derived using the fifth cross section from the overpass furthest away from the stack.

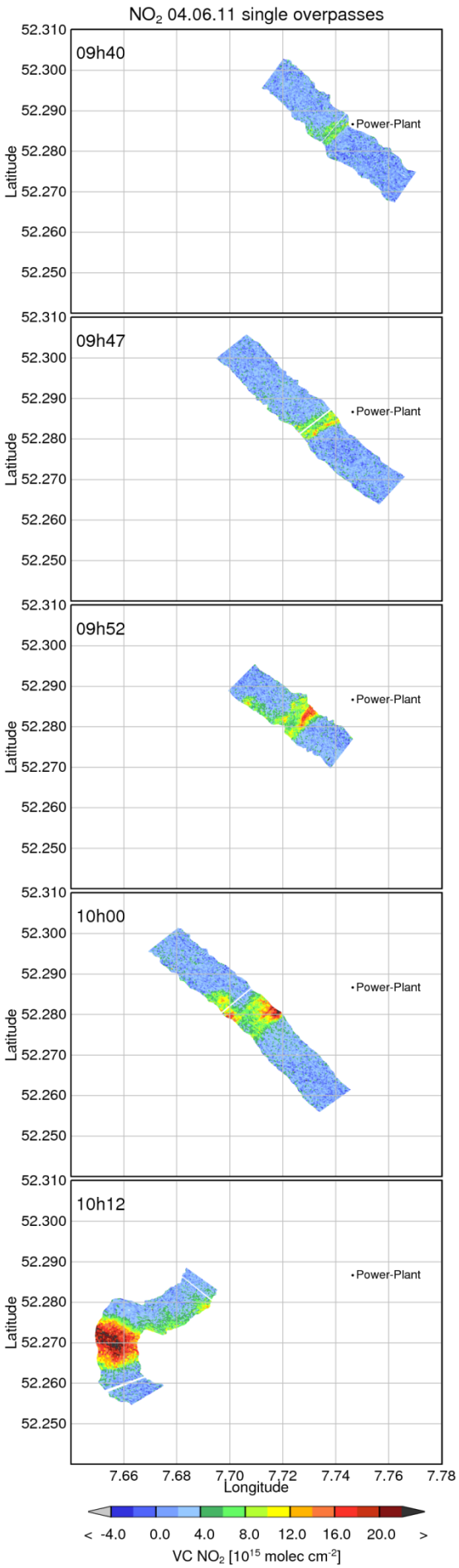
NO2 04.06.11 single overpasses
09h40
09h47
09h52
10h00
10h12
Power-Plant
Latitude
52.310
52.300
52.290
52.280
52.270
52.260
52.250
7.66
7.68
7.70
7.72
7.74
7.76
7.78
Longitude
< -4.0
0.0
4.0
8.0
12.0
16.0
20.0
>
VC NO2 [10^15 molec cm^-2]

Figure 13 Overpasses at five different times between 09:40 and 10:12 UTC over the NO_2 plume. The latest overpass at 10:12 UTC has a distance of around 6 km from the power plant and is used for the emission flux calculation.

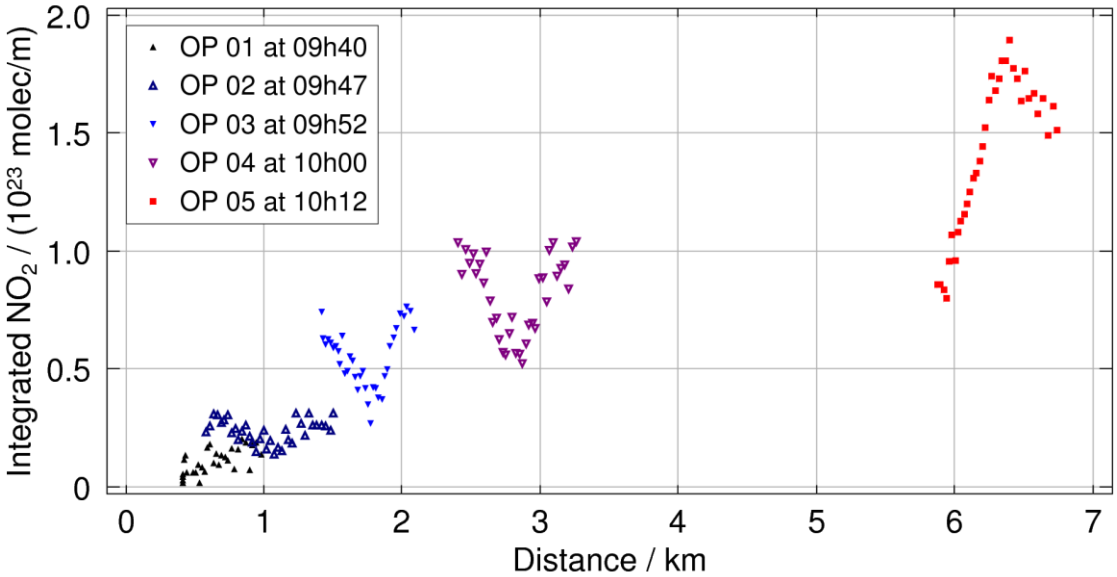

Figure 15 Integrated NO_2 amount from five individual overpasses (OP) at different times and distances from the stack. Results are taken from the LOS35 evaluation, therefore in total 175 cross sections through the plume are included in this diagram.

Comment (2)

In section 7.4 the conversion of NO to NO2 is described as limitation of the flux estimate, is it possible that this "limitation" might be used to gain additional information about the mixing in of O3 and the conversion mechanism as in Louban et al., 2009 (DOI 10.1007/s00445-008-0262-6) for BrO?

Answer to Comment (2)

The study by Louban et al. shows excellent imaging DOAS measurements of BrO and SO_2 in a volcanic plume taken from ground. Louban et al. estimate the order of magnitude for the Br/BrO ratio by using the relevant reaction constants and assuming that O_3 is readily available. A value for the concentration of O_3 needs to be assumed in order to derive a number for Br/BrO here. Of course we can do this in our case as well. However, there is not more information in that procedure than in estimating a plausible ratio r. The actual O_3 concentration is not measured so a similar uncertainty results from this approach also. An estimation of r resulting from assumptions on O_3 concentration as well as the reaction constants is given below.

In addition, Louban et al measure SO_2, which is chemically stable along the plume, and they determine BrO/SO_2 ratios. This way, they can eliminate the effect of dilution along the plume and then analyse the ongoing bromine chemistry. In our case, we do not have an additional stable species in the plume that we measure. That means a similar mechanism can unfortunately not be applied here.

Nevertheless, it is true that more information is contained in our measurements than what we actually use. We did not plan to include a chemical analysis of the plume in this AMT paper. However, while a detailed analysis is outside the focus of this more technical paper, we include an investigation of the NO_2 variation along the plume, using also measurements closer to the stack, see also answers to Comment (1) here, as well as to Comment (18) of Reviewer #1.

Estimation of r from reaction rates and ozone

As an alternative to the procedure in our manuscript, which is assuming a reasonable value for the ratio $r = [NO]/[NO_2]$, one can also use reasonable estimates and documented values for the quantities entering equation (4) and (5), i.e. numbers for $k_1, [O_3]$ and J would be needed then instead of the ratio r.
The following values apply:

Reaction rate $k_1 = 1.9 \cdot 10^{-4} \frac{cm^3}{molec \cdot s}$ (Sander et al., 2011) and

photolysis frequency $J = 0.007\ s^{-1}$ (Koepke et al., 2010) for $SZA \approx 40°$

Tropospheric O_3 is a strongly varying quantity and was not measured within the plume. In summer, ambient O_3 in rural areas is around $7.5 \cdot 10^{11}$ molec/cm^3 , i.e. 30 ppbv, while in urban areas values between 40-70 ppbv are more typical and the concentration can still be substantially larger.

Ozone values between 40 and 70 ppb correspond to values of r between 0.37 and 0.21, respectively. The value 0.25 for r used in our study lies within this typical range.

References:

Sander, S. P., J. Abbatt, J. R. Barker, J. B. Burkholder, R. R. Friedl, D. M. Golden, R. E. Huie, C. E. Kolb, M. J. Kurylo, G. K. Moortgat, V. L. Orkin and P. H. Wine: "Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 17", JPL Publication 10-6, Jet Propulsion Laboratory, Pasadena, 2011, http://jpldataeval.jpl.nasa.gov.

Koepke, P., Garhammer, M., Hess, M., and Roeth, E.-P.: NO_2 photolysis frequencies in street canyons, Atmos. Chem. Phys., 10, 7457-7466, doi:10.5194/acp-10-7457-2010, 2010.

Comment (3)
Although the authors demonstrate very well the instruments ability to detect a low level NO2 enhancement above a motorway the uncertainty seems quite high (section 9). Therefore I am not sure how useful the comparison to ground based observation really is, especially as these were not performed simultaneously at the same road. Hence the emission should only be compared with respect to the order of magnitude for a medium sized highway (55 000 cars/day). The authors might emphasis this difference more clearly.

Answer to Comment (3)
The comment is reasonable since the uncertainty on the low level NO_2 is fairly large. However, the NO_2 enhancement above the motorway is significant and there are hardly any direct observations of NO_2 from car traffic by DOAS measurements. Therefore, we want to set our observations into perspective by comparing to the other study. This is consequently not meant as a validating comparison, and we accept the recommendation to emphasise the limitation more clearly.

We add the following sentence at the beginning of the comparison:

This study is used in order to compare the order of magnitude of NO_2 column amounts for a medium-sized highway.

Part 3. smaller changes and technical points

• p 3596 l 15-21: I am not sure if Wang et al., 2005 is a proper reference for the description of the spectrograph, maybe Wang et al., 2006 or Bruns 2004 (PhDthesis) are better references. I was a bit confused about the small wavelength interval detected by the instrument. Is it caused by the smaller detector size compared to Wang et al., 2006? In Wang et al., 2006 a wavelength range of ~140nm is given while here the total range is only 41 nm.

The spectrograph is not explained in detail in the papers by Wang et al. (2005), the comment was meant as additional information, that it is actually the same spectrograph as was used by the cited studies. We include the citation of Bruns (2004) instead. And yes, the wavelength range is smaller as in Wang et al. (2005) and Bruns (2004) because the described frame transfer CCD has a smaller chip.

• p 3596 f l 26 - 2: The instrument includes a 200 µm fibre to illuminate a 100 µm entrance slit. It might be an idea to use a 100 µm fibre and omit the entrance slit. The risk of illumination the slit with half of the fibre is quite high i.e. misalignment of the fibre by 100 µm and this might cause an unpleasant slit function.

This is true. However, the optical alignment was carefully optimised in the laboratory before the campaign. With our setup, the entrance slit width can be adapted to the respective situation. In other applications a smaller slit width was needed, and thus the flexibility of the instrument has its advantages.

• p 3597 l 15 -25: In this section the readout time and shift time are compared. Please include the typical exposure time here as well. In line 23 on the following page it is said to be 0.5 sec. How is it determined, by the intensity of the previous measurement or is it fixed? Is there any risk that the 0.1 sec for readout are too long for the illumination of next spectra, e.g. when flying over bright clouds or snow. Thereby the spectra would be oversaturated.

The exposure time is included at this location in the text now.
The exposure time is kept fixed at 0.5 s. It was chosen as a relatively short time, which allows for small pixel sizes in flight direction and is short enough to usually avoid saturation effects. In case longer integration time is desired, co-adding may be performed during post-flight data analysis. If spectra would be oversaturated, they would be omitted from the results but this is not relevant for the clear-sky data shown and discussed in the manuscript.

• p 3903 l 20: I guess the authors used their "own" SCIAMACHY data available at http://www.iup.uni-bremen.de/sciamachy/ please add the respective reference or link to the data. Even though they were provided by one of the co-authors.

The reference to Richter et al. (2005), as well as the website http://www.iup.uni-bremen.de/doas/data_products.htm are included now.

Richter, A., Burrows, J. P., Nüß, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, doi:10.1038/nature04092, 2005.

• p 3604 l 20: Thank you for following my suggestion to extend the time series in figure 6. Please update the start and end times in the text as well.

Done.

• *p 3604 l 27: Please mention the time of the fit shown in figure 7 in the text as well.*

Done.

• *p 3606 l 16: Just a comment: The power stations discussed in Heue et al., 2008 have about 5 times the electrical output (~4100MW) compared to Ibbenbüren (~800 MW), this corresponds quite well with the ratios of the SCs. Also for the power station in Monticello (Texas,USA) (~2000MW) studied by Melamed et al., (2003) vertical column densities up to 8 × 1016 molec/cm2 were observed.*

We now include the comparison to the other studies more explicitly and mention the relevant numbers as well as the fuel type.

This is much smaller than values measured by Heue et al., (2008) above the huge South African Highveld power plants. In comparison, they observe slant columns up to 1.1×10^{17} molec/cm² for the coal and syngas fired Majuba power station (around 4,100 MW nominal capacity), and Melamed et al. (2003) observe vertical columns of up to 8×10^{16} molec/cm² above the lignite fired Monticello power station (Texas, USA, around 2,000 MW nominal capacity).

• *p 3607 eq. 7: Is this approximation really faster than doing simulation for nine viewing directions? (not including the roll angle of the aircraft) The influence will be small so there is no need to redo all the calculations. Aerosols are not yet included in the calculations, do the O4 images show any features comparable to the intensity or the NO2 images?*

The wavelength range of the spectrometer setup during the campaign was too small to perform an O_4 fit. A later instrument update now allows a larger fitting window, so that O_4 and NO_2 can be measured simultaneously. We could have done a simulation for all viewing directions also. However, there are much more than nine directions, especially due to the aircraft movement (esp. roll angle). Interpolation between some fixed calculated values would be necessary in any case.

• *p 3608 l 20-27: This section is slightly confusing. If I understand it correct you have two different effects: On the one hand NO is converted to NO2, thereby the NO2 SC increase on the other hand, the plume broadens thereby the SC decrease if you are further away from the source. However what really matters is the the integrated VC along the flight, and according to figure 17 it increases with increasing distance to the stack.*

Two effects occur along the plume – the total NO_2 content increases due to chemical conversion and dilution takes place due to plume broadening. However, the broadening is mentioned mainly as a description of the plume structure. The increase due to chemical conversion dominates over the dilution effect so that a decrease in SC or VC is not directly observed. A strong increase of the integrated VC (as well as individual VCs) is observed.

We have changed the last sentence to read:
In addition, the plume broadens while it is transported away from the stack, and the integrated VC across the plume increases with distance from the stack. This is shown in more detail in the following section.

• *p 3610 l 11 f: If the mixing layer height was 1300m and the flight altitude was 1100 m, the aircraft might have been flying through the plume. Was any in situ NOx instruments aboard? I am not sure if the geometric height of the stack is sufficient here, often the plume rises vertically directly at the beginning. Is this considered in the Gaussian dispersion?*

NO_x was unfortunately not measured in situ. The plume rise is certainly significant and was taken into account in order to estimate the vertical location of the plume at the investigated overpass at 6km distance. The spread was again needed to determine the wind speed and direction. At that distance the plume is already strongly spread over several hundred meters in the vertical direction. However, less than 5% are estimated to possibly be above the aircraft. Consequently, a minor part of the plume might be missed, but this is considered negligible at this point.

Part 4. references

• *Pundt, I., ... Losch, J., ..., doi 10.1016/j.atmosenv.2004.07.035, 2005. 3617, 3618*
change Losch to Lösch

Done.

Part 5. figures

• *please zoom in a bit more on figure 12? (comparable to figure 14) Because it is difficult to find any differences between LOS 9 and LOS 35. It is the improvement in the resolution, what the readers are interested in.*

We include a zoom in of each figure in addition to the overview figures, which we wish to keep.

• *Is figure 13 necessary?*

Maybe the figure is not necessary, but we think it might be helpful for the reader and prefer to keep it.

• *What is the resolution of figure 14 - LOS 35?*

Yes, we include the information in the figure now.

• *figure 18 is it useful to change the colour scale for the NO2 plot to a maximum close to 1 × 1015 molec/cm2? So the weak signals become more visible, but also the noise.*

Done. (Now Figure 19).