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 #1, C. Kern, from 20 August 2014.

We are grateful for the comments, corrections and suggestions of Referee #1.
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 1. Referring to the Specific Corrections

Comment (1)
Title: The word "continuous" in the title appears to imply that the mapping measurements are performed continuously. Usually, continuous measurements are ones that are performed without interruption for a long period of time, e.g. days or weeks. This is clearly not the case here. I assume the authors are referring to the lack of gaps in the mapping data, but perhaps this can be expressed in some other way, e.g. "two dimensional trace gas mapping" or "high resolution trace gas mapping"?

Answer to Comment (1)
We agree with the Referee, also his assumption on our intention is correct. We decide for changing the title to "A wide field-of-view imaging DOAS instrument for two dimensional trace gas mapping from aircraft".

Comment (2)
P3592, L13 – The spatial resolution depends on flight altitude. This should probably be mentioned here. Also see line 17, where a spatial resolution of 30m is given. This also depends on altitude, correct?

Answer to Comment (2)
Yes, that is correct. Across track spatial resolution always depends on flight altitude in this study. This information is included now in the text of the revised manuscript in both mentioned places.

Comment (3)
P3593, L7 – Here, perhaps you should more clearly explain that the reaction of NO with ozone is in steady state with the photolysis of NO_2 unless this system is perturbed e.g. by the input of NO from an industrial stack into the atmosphere.

Answer to Comment (3)
We have added the following sentence at the respective point in the text after the description of the formation and loss processes.
These processes are in steady state in an unperturbed situation, i.e. as long as no local NO pollution sources are present.

Comment (4)
L13 – It's probably better to state that tropospheric O_3 "impacts" air quality, since it does have a few positive impacts as well as the negative ones.

Answer to Comment (4)
Yes, we have changed the wording accordingly.

Comment (5)

P3594, L27 – You mention two previous applications of imaging systems on aircraft but fail to mention the application by General et al who flew a similar instrument (HAIDI) to characterize the plume of Mt. Etna. I believe that this instrument is more similar to the AirMAP than the other two that you do mention here. Please include a brief description of the HAIDI and a short comparison of the AirMAP and the HAIDI, as you do for the other two. Please be certain to include the relevant references for this too.

Answer to Comment (5)

It was simply not possible at the time to cite the above mentioned studies.
Our manuscript was submitted in December 2013, i.e. before the respective studies were published. During the review period, further studies became available. Of course we now include the publications of General et al. in the revised version of our paper, as well as a discussion of the HAIDI.
The following section is included in the introduction now.

Recently, the new imaging instrument HAIDI has been reported, which has been successfully applied to measurements of NO_2, SO_2, BrO and OClO from anthropogenic emissions, in Polar regions and within volcanic plumes (General et al., 2014, General et al., 2015). The HAIDI consists of three DOAS instruments, which point at different directions and are used either in whisk broom or push broom mode (Schowengerdt et al., 2007). Observations yield spatial trace gas distributions at ground resolutions below 100 m as well as information on the vertical distribution.

Schowengerdt, R. A.: Remote Sensing - Models and Methods for Image Processing, Academic Press, Burlington, doi:http://dx.doi.org/10.1016/B978-012369407-2/50006-1, http://www.sciencedirect.com/science/article/pii/1335 B9780123694072500061, 2007.

Comment (6)

P3595, L9 – Does this sentence still hold true when compared to HAIDI?

Answer to Comment (6)

This comment refers to the sentence ”The present study introduces the Airborne imaging DOAS instrument for Measurements of Atmospheric Pollution (AirMAP), which is well suited for trace gas mapping of comparably small scale emissions at fine spatial resolution and better spatial coverage as compared to previous studies.“

The HAIDI is a sophisticated instrument featuring a number of advantages, being a very flexible measurement system with three DOAS instruments, of which one is always used as a nadir whiskbroom scanner. Full 2-D spatial coverage within the swath is possible for the HAIDI instrument. However, this depends on the chosen measurement sequence and influential factors such as the flight altitude. If the flight altitude changes the measurement sequence would need to be adapted to avoid spatial gaps. Indeed, useful settings chosen for the HAIDI campaigns as published in the two manuscripts cited above feature some spatial gaps.
The advantage of AirMAP addressed by the sentence in question is its good spatial coverage as there are no spatial gaps along flight direction, irrespective of the situation. With fixed detector settings only the aspect ratio of the ground pixels and the swath width change for different aircraft altitudes and speeds allowing a flexible operation and flight planning.
This holds true as long as the exposure time is above the read out time of the storage area of the frame transfer CCD. This is the only requirement and it is well fulfilled in all our applications.
Nevertheless, the sentence in question is changed as follows, omitting the direct comparison to the alternative instruments.

The present study introduces the push broom Airborne imaging DOAS instrument for Measurements of Atmospheric Pollution (AirMAP), which is well suited for trace gas mapping of comparably small scale emissions at fine spatial resolution. The full spatial coverage within the given swath is independent of flight altitude, aircraft speed and measurement sequence.

Comment (7)
P3596, L26 – Please be more specific with regard to the fiber diameter. Is the core-to-core separation the distance from the center of one fiber to the center of the next, or is this the distance that the cladding in between two adjacent fibers takes up? Both dimensions are important. The distance from the center of one fiber to the center of the next combined with the focal length of the imaging optics effectively gives the spatial resolution, independent of flight altitude, correct?

Answer to Comment (7)
The core-to-core separation given in the text is indeed regarded as the distance from the center of one fibre to the center of the next fibre. And it is true that this dimension determines the limit of the spatial resolution across flight direction. The spatial resolution in terms of the viewing angle is independent of flight altitude, while the spatial resolution in terms of actual distances on ground is still flight altitude dependent. The respective sentence is changed in the following way.

The observed ground scene is imaged onto the entrance of a light guide consisting of 38 sorted single glass fibres, which are vertically aligned in the same sequence at either end with a center-to-center separation of 220 µm, i.e. the distance from the center of one fibre to the center of the next. The dimension of the fibres without cladding is 193 µm. The distance from one fibre to the next determines the limit of the spatial resolution in across flight direction in terms of viewing angle.

Comment (8)
P3597,L7 – Please give a reference for pushbroom and whiskbroom imaging. I believe that your reference to Lohberger et al e.g. describes this.

Answer to Comment (8)
We include the reference to online publication Schowengerdt et al, 2007 where the terms are fully explained.

Schowengerdt, R. A.: Remote Sensing - Models and Methods for Image Processing, Academic Press, Burlington, doi:http://dx.doi.org/10.1016/B978-012369407-2/50006-1, http://www.sciencedirect.com/science/article/pii/1335 B9780123694072500061, 2007.

Comment (9)
3598, L21 – This sentence explains several different concepts, so I'm not really sure what the "latter" refers to here. Also not sure what the "basis for mapping purposes" means. Please be more specific.

Answer to Comment (9)
We have rephrased the sentence in order to clarify the meaning.

The along track IFOV projected onto the ground is usually smaller than the travelled distance during the exposure time and is therefore neglected in the trace gas maps. The length of displayed ground pixels in flight direction is determined here by the travelled distance during one exposure, also taking into account the aircraft attitude at the start and end of an exposure.

Comment (10)
L23 – Change to "a ground pixel size of 30m along track is achieved". Again, this must depend on flight altitude, correct?

Answer to Comment (10)
We have changed the word order as suggested. According to our better description now in the part above, the travelled distance determines the displayed ground pixel length in flight direction in the trace gas maps. While the IFOV projected to the ground would be flight altitude dependent, the travelled distance is not.

Comment (11)
P3602, L20 – You show that the spectral resolution is slightly degraded for pixels close to the edge of the sensor. You write that the "varying image quality along the spatial axis" is responsible. I assume you mean that aberrations of the object lens become significant for rays ending close to the detector edge. Perhaps clarify this sentence. Also, doesn't this mean that the spatial resolution is also degraded (in cross-track direction) for those pixels? Is that an issue at all? Perhaps the pixels actually begin to overlap slightly? I don't think it's mentioned in the text.

Answer to Comment (11)
The image aberrations also affect the spatial resolution towards the detector edge. It is mainly the spectrometer that causes optical aberrations in the image on the CCD chip. These aberrations become especially well visible in the outermost viewing directions. The utilized CCD chip is fairly small (8 x mm²), and therefore the degradation in spatial direction is not too large and the individual spatial directions remain well separated.
We change the wording of the sentence and add a comment on the degradation of the spatial resolution.

Due to image aberrations for off-axis object points, the SRF has a different shape and width for the different viewing angles. In addition, the image quality along the spatial axis is slightly degraded towards the detector edges. However, the individual spatial directions remain well separated.

Comment (12)
P3603, L3 – You mention that the SRF shape and width do not vary with wavelength. This is indeed an important characteristic, but it is seldom perfectly fulfilled. It is quite common for spectrometers to show some degree of variation in shape at least. There are various ways of dealing with this – from simply ignoring minor variations to using a variable SRF for cross-section convolution. In any case, it might be useful to include a little more information on the tests that were conducted to ensure there was no significant variation in the SRF, and perhaps state that the SRF was 'reasonably constant' or that 'no variations were detected', rather than discounting this effect altogether.

Answer to Comment (12)
Yes, it is seldom perfectly true that the SRF shape and width do not vary with wavelength. That is the reason why the point is mentioned, but we did not say that this is generally an issue. Again, the relatively small size of the CCD chip prevents the instrument from seeing the increasing problems at further distances from the optical axis. We have performed laboratory measurements of one HgCd emission line. The emission line was moved in steps over the CCD image by turning the grating assuming that optical aberrations dominate over image differences caused by the grating position. When overlaying two measurements, one with the line at the left spectral edge and one at the right edge, no variations in the line shape were detected. We have added two sentences to the respective paragraph in the text.

Most importantly, the SRF shape and width for each individual viewing direction are reasonably constant along the wavelength axis (not shown). Such a dependency may in general occur and would become more important for larger CCD chips. However, no variations in the SRF spectral shape were detected in laboratory tests with the AirMAP. This stability is required for accurate and consistent trace gas retrieval results.

Comment (13)
P3604, L7 – Can you very briefly explain why a "constant intensity offset" is fitted? I assume this means that a constant intensity Ic is subtracted from (or added to) the measurement before the linear fit is used to derive column amounts? Ic is then varied in the non-linear fit? Is Ic meant to describe the contribution of stray light in the spectrometer? If so, is the assumption that this stray light is constant across the detector a valid one?

Answer to Comment (13)
Stray light is a common effect leading to an intensity offset Ic across the CCD chip. In our case, where the logarithm of the ratio between actual intensity and reference intensity (i.e. ln I/I0) is modelled by the DOAS retrieval, an offset Ic on the actual intensity I causes (as long as the offset is small with respect to the reference intensity I0) an additional term in the optical density. This additional term is the ratio between the offset and the actual intensity Ic/I. Ic is found by the non-linear fitting routine while the inverted intensity 1/I is used from the actual measurement.
As this effect has the spectral shape of the inverted intensity, it is similar to other common effects such as the Ring effect, polarization effect and others. Therefore, it is not easily possible to separate this as the stray light effect alone.

In our case, the assumption of a constant intensity offset by stray light or other effects is an assumption, although this is not generally the case. In the process of finding the best fitting parameters, a test is performed assuming an intensity offset which is linearly changing over the CCD chip. This will include an additional term and fitting parameter in the fitting routine. As such an extension of the number of fitting parameters does not significantly improve the fit result in terms of RMS, only one term is used for the correction of an intensity offset.

Comment (14)
L19 – Significant spatial variations along flight direction are described here, but fig 7 shows a time series. Perhaps it is better to speak of temporal variations here, although I understand that they correspond to a spatial variation. Also, some of this variation is clearly noise. It might be a good idea to mention the magnitude of the noise, and then say that the temporal variations in the data are clearly larger than the noise alone.

Answer to Comment (14)
Surely, the reviewer refers to Fig. 6. We had hoped that by including the precise explanations, i.e. that the time axis shows spatial variations due to the moving along flight direction, the situation would be clear. Using the term "temporal variations" could wrongly lead the reader to think we would actually observe changes in the NO_2 over time, which we don't. Therefore we prefer to speak then of "variations" alone instead of "temporal variations", but simply omit the word "spatial" to hopefully avoid confusion. The sentence now reads:

With an exposure time of 0.5 s this high resolution time series shows strong variations along flight direction, i.e. along the time axis, as well as considerable spatial variation across flight direction, visible through the differences between the three viewing directions.

It is correct that some of the variation is due to noise. We add the following sentences to clarify.

Some part of the variation is due to noise, which is in the range of a few times 10^{15} molecules/cm^2 for the slant columns as discussed below. The observed variations along and across flight direction are clearly larger than the noise alone.

Comment (15)
P3606, L3 – Here and elsewhere the "central flight pattern" is mentioned. It did not become clear to me what part of the flight track this actually refers to. It seems to me that all the plots of the flight track over the power plant show the same area. In this case, maybe it is better to simply refer to the "flight track above the power plant area".

Answer to Comment (15)
We have included the explanation in the first instance where the central flight pattern is mentioned (Section 4.1). Then the usage of the term is retained in the other instances.

Typical flight speed during the central flight pattern (i.e. the flight track above the power plant area) is around 60 m/s.

Comment (16)

P3606, L9 – You mention that the NO2 slant column amounts do not exhibit stripy features and are generally consistent for different viewing angles. You state that these features would appear if there were viewing angle dependencies of the slant column results. In the next section, you then show that the AMF depends on the viewing angle, and use a geometrical approximation to correct for this effect. These two statements appear to be in conflict with one another. Shouldn't the slant columns exhibit enhanced values towards the sensor edges when compared to the nadir observations? Granted, this would only be visible in areas in which significant amounts of NO2 were actually located beneath the aircraft, particularly above the power plant plume. I'm a little bit confused by this finding.

Answer to Comment (16)

Yes, it is true that the slant columns should exhibit enhanced values towards the sensor edges. This general dependency is accounted for in the vertical columns by the viewing angle correction of the AMF. In the slant columns of our observations, this dependency is not or only hardly visible due to several reasons. First of all, the effect is rather small in our LOS range. The effect is much larger for instruments such as OMI (Ozone Monitoring Instrument) with LOS that exceed 50°. Geometrically, the light path length between nadir and the outermost LOS only differs by less than 5% in our case. In addition, the stratospheric NO_2, for which this U-shape dependency is prominent, is effectively removed in our results by the use of the reference intensity. Also we did not fly over an extended homogeneous field of enhanced NO_2, which would lead to a more prominently visible dependency as well. Some part of the dependency may also be covered by noise influence.

When we state that our instrument does not show viewing angle dependencies, we were thinking of irregular dependencies, i.e. non-uniformities in the slant column depending on viewing angle in contrast to the smooth variation described above. The "stripy features" refer to potential irregular effects in the sense that certain viewing directions have the tendency to always show larger values than others. In fact, due to the varying slit function (SRF), the 35 viewing directions of our instrument could be regarded as 35 individual instruments. For individual instruments, small biases or offsets in the retrieval results could have occurred, e.g., in case SRF variations and spectral calibration variations between the different viewing directions are not taken sufficiently into account. Such dependencies are not present in the AirMAP results at the given precision.

We hope to clarify the statement by specifying the mentioned features as "irregular" viewing angle dependencies and by describing the regular dependency here as well. In addition, we explain what we mean by "stripy features".

For the analysis of the AirMAP observations no post-processing in the form of destriping has been applied. Such a procedure would be necessary in case of irregular viewing angle dependencies in the trace gas results, i.e. non-uniformities in the slant column values (Dobber et al., 2008; Popp et al., 2012). The LOS correction applied in the next section takes into account only the smooth viewing angle dependency due to a slightly longer light path for observations at the sensor edges. The single viewing directions of AirMAP yield consistent results, which do not exhibit stripy features, i.e. in the sense that certain viewing directions would have the tendency to always show larger values than others, neither in the instrument calibration parameters nor in the retrieved NO_2 column amount.

Comment (17)

*P3607, equations – These equations and SCIATRAN model results are valid when observing trace gas distributions on spatial scales that are larger than the lateral distance that light travels on its way from the aircraft altitude to the ground and back up again to the instrument. In the worst case, this length scale L = H * (tan(Θ) + tan(Θi)). For e.g. a solar zenith angle of 30 degrees and a viewing angle of 20 degrees, this length scale L approaches the flight altitude H. If flying at 1km altitude, this* *means that in this admittedly worst case, the radiation passed through the atmosphere about 1km away on its way down to the Earth. If measuring a plume of smaller dimensions at considerable altitude, the radiation might miss the plume on its way down, only passing through it on its way up after having been scattered on the ground. These 3 dimensional effects obviously can affect the AMF in some cases, particularly when observing localized plumes. It will likely be quite difficult to quantitatively deal with these 3 dimensional effects in this study. Also, the stack plume is likely quite close to the ground and so the issue might be negligible. However, I feel that the issue is at least worth mentioning and it could be worth conducting a back-of-the-envelope calculation to determine if it is relevant here or not, thus also alerting others that in the case of high altitude plumes measured under high solar zenith angles, this can be problematic.*

Answer to Comment (17)

We agree that in general, the described 3-dimensional effects are very interesting and might be important. It is surely true that dealing explicitly with these effects is out of scope for the current study.

As described in the comment, the 3-dimensional effects can lead to a different (smaller) AMF when the light beam passes through the trace gas plume only on one way (downwards or upwards) due to the horizontal localisation of the plume. In addition, the 3-D effects may lead to a displacement of the trace gas assignment to the ground pixel, as the plume is not only (or not at all) situated above the respective ground pixel but somewhere along the light path and will therefore be assigned to a different horizontal location.

However, this displacement is smaller than the maximum length scale L.

The observed NO_2 in a viewing direction Θ_i is assigned to the projected ground pixel in direction of Θ_i and not to the location of the aircraft. Therefore, the displacement D is considerably smaller than the length scale L. For the respective worst case, either (a) $D = H * \tan(\Theta)$ for a localised plume at flight altitude which is displaced from the projected ground pixel away from the aircraft, or (b) $D = H * \tan(\Theta_i)$ for an elevated plume right below the aircraft. For case (a), the light beam passes through the plume on its way to the ground. For case (b), the light beam passes through the plume on the way from the ground to the aircraft. Maximum displacements may still range in the order of the flight altitude. However, as described below, the worst case is not fulfilled in our situation.

The 3-D effect has most influence at large SZA, if in addition the sun, aircraft and viewing direction are in one plane, i.e. if the sun is exactly to the left or right of the aircraft (+/-90° azimuth angle with respect to flight direction) and for plumes which are strongly localised and are situated at large altitudes.

This supposed worst case is not fulfilled in the present study.

- The plume is located at lower altitudes than the aircraft. The stack height is 275 m, and also taking into account the plume rise due to the warm exhaust gases, the plume will not be localised in a thin layer right below the aircraft. The plume is initially located at altitudes somewhere half way between ground and aircraft.
- The plume spreads vertically within the strongly mixed boundary layer.
- The sun is not in the same plane as the aircraft and viewing direction for the plume overpasses.

It is also worth noting that the geometric approximation, which is only used for LOS correction in the study, is not exactly fulfilled. The box AMF (cf. Fig. 10) is smaller close to the ground which means that part of the light is scattered in the atmosphere before it reaches the ground. The direct light beams that travel from the sun to a point in the atmosphere below the aircraft and are then scattered into direction Θ_i, are less displaced from the aircraft on their way downwards as compared to those that reach the ground before being scattered or reflected up again. Therefore, the expected influence on the assigned horizontal location of the observed trace gas absorption is decreased as compared to the worst case. Multiple scattering in the real atmosphere (as opposed to the descriptive imagination of single straight light rays scattered only once) further reduces the influence of 3-D effects.

The stability of the atmosphere determines how the plume spreads in horizontal and vertical directions. In our case, the plume spreads similarly in both dimensions. As soon as the plume reaches altitudes around the flight altitude, the horizontal spread is also large so that 3-D effects become less important. In our case, the plume is located at lower altitudes before it obtains such a large extent. The plume is emitted at stack height (275 m) and then rises due to the warm exhaust gases for a few hundred meters. The plume already spreads during the rising.
A rough calculation is done, also taking into account a solar azimuth angle to the aircraft. Typical settings for our case study are chosen, except for the LOS, which is taken close to its maximum. The required horizontal plume extent PE is then calculated. PE is the extent that would lead geometric light beams to pass through the plume on both ways (downwards from the sun and upwards into the respective LOS to the instrument).

The following values have been used:
$SZA = 40°$, $LOS = 20°$, $\alpha_{azim} = 45°$, $Alt_{Plume}=600m$

The length scales L_{LOS} due to viewing angle, and L_{SZA} due to the SZA, are given by:

$L_{LOS}=Alt_{Plume}\cdot \tan(LOS)$ and $L_{SZA}=Alt_{Plume}\cdot \tan(SZA)$

The Plume Extent PE then reads:

$$PE = \left[\left(L_{LOS} + L_{SZA}\cdot \sin(\alpha_{azim})\right)^2 + \left(L_{SZA} * \cos(\alpha_{azim})\right)^2\right]^{\frac{1}{2}}$$

For the given situation, the plume would need to have a diameter of 675 m, (360 m across flight direction and 570 m along flight direction). Then, both light beams would pass through the plume. For the case used in our emission calculations, the plume has already broadened more than these distances, but close to the stack, the plume extent is of this order of magnitude or smaller. However, at the beginning the plume is at lower altitudes. Rising and horizontal spreading of the plume both happen at the same time and have opposing influences with respect to the 3-D effects.
In addition, the above calculation is for the outermost viewing directions and for light beams that actually travel right down to the ground and back. However, the effective plume altitude (altitude of plume above scattering altitude) is on average smaller than the real plume altitude, and the geometric approximation is not perfectly fulfilled.

It can be concluded, that for our case, the 3-D effects are not too important, but they may become relevant close to the stack for the outermost viewing directions. The main effect then would be an overestimation of the AMF. An overestimation of the AMF leads to a smaller vertical column of NO_2 than is actually present. For special cases, however, the 3-D effects can be very important and the relevant length scales should be inspected.

Regarding our manuscript, we added the following comment at the end of the respective Section (6.3).

It is worth noting, that neither SCIATRAN nor the geometric approximation are taking into account 3-dimensional effects. The importance of such 3-D effects increases for increasing SZA, increasing LOS angle and for a plume which is horizontally confined as well as situated at high altitudes directly below the aircraft. For such a case, an individual light beam may travel through the plume only once, e.g. either on the way from sun to the ground or from the ground to the instrument. In this case, the AMF would be overestimated and the resulting vertical column would be underestimated. In addition, the assignment of the measured trace gas amount to the ground pixel may be affected. Multiple scattering on the other hand reduces the influence of 3-D effects.
Close to the stack, 3-D effects might be present in our case, further away from the stack, however, the plume has already spread in horizontal and vertical directions, so that the influence of 3-D effects becomes less relevant.

Comment (18)

P3613, L15 and Figure 17 – This shows some variability in the NO2 emission rate as a function of distance from the stack .The variability in the plot is largely within the measurement error. However, the flight track shows that there were several additional transects of the power plant plume that were conducted closer in. Adding emission rates derived these to the figure could potentially show the expected increase with time beyond the measurement error, and this plot might then even be used to attempt to improve the constraints on the NOx emission rate (e.g.by assuming a constant O3 concentration and photolysis rate and fitting for an exponential approach to the steady state NO/NO2 ratio). Then one might not have to guess a value for r. Personally, I think that would be quite interesting and I wonder if you might consider it.

Answer to Comment (18)

We agree that information about the build-up of NO_2 along the plume can be achieved by including our measurements close to the stack. In Figure 14, the overpasses closer to the power plant are now included. See also Comment (1) of Referee #2.

In addition, a new figure (Figure 15) now shows the integrated NO_2 amount across the plume for all five overpasses. Results are taken from the LOS35 analysis, so 35 observations for each overpass are included, amounting to 175 data points. The build-up of NO_2 with increasing distance from the stack is clearly seen.

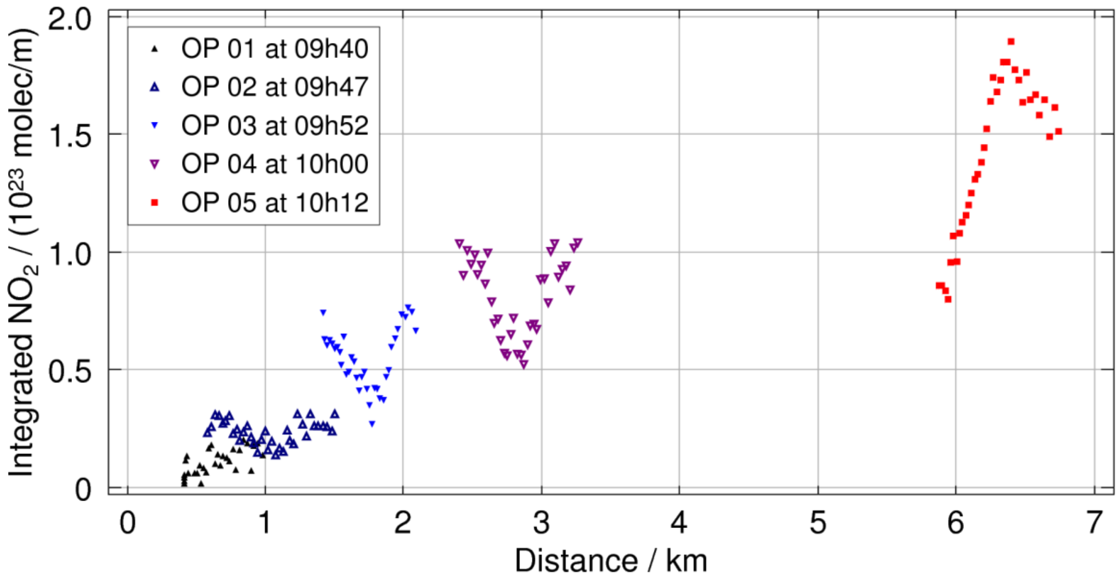

Figure 15 Integrated NO_2 amount from five individual overpasses (OP) at different times and distances from the stack. Results are taken from the LOS35 evaluation, therefore in total 175 cross sections through the plume are included in this diagram.

Considering the basic NO_x chemistry which influences the NO_2 content in the exhaust plume, one can estimate the behaviour of the NO_2 plume. The relevant chemical reactions are the following:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$
$$NO_2 + h\nu \rightarrow NO + O \qquad (2)$$

They lead to differential equations:

NO_2 formation : $\frac{d}{dt}[NO_2]_{(1)} = k_1 \cdot [NO] \cdot [O_3]$

NO_2 loss: $\frac{d}{dt}[NO_2]_{(2)} = -J \cdot [NO_2]$

In total: $\frac{d}{dt}[NO_2] = k_1 \cdot [NO] \cdot [O_3] - J \cdot [NO_2]$

$$= k_1 \cdot [NO_x] \cdot [O_3] - (k_1 \cdot [O_3] + J) \cdot [NO_2] \qquad (3)$$

(using the relation $[NO_x]=[NO]+[NO_2]$).

The differential equation is solved by the following exponential term

$$[NO_2](t) = \frac{k_1 \cdot [NO_x] \cdot [O_3]}{k_1 \cdot [O_3] + J} \cdot \left(1 - e^{-(k_1 \cdot [O_3] + J) \cdot t}\right) \qquad (4)$$

However, for this exponential behaviour, the assumption is necessary, that $[O_3]$ and $[NO_x]$ are constant. As a first approximation, NO_2 amount increases until a steady state amount $[NO_2]_{st.st.}$ is achieved, given by the prefactor in equation (4).

$$[NO_2]_{st.st.} = \frac{k_1 \cdot [NO_x] \cdot [O_3]}{k_1 \cdot [O_3] + J} = \frac{k_1 [NO]_{st.st.} [O_3]}{J} \qquad (5)$$

The NO_2 amount in close proximity of the stack is close to zero and rises until some kilometres from the stack the steady state amount is achieved. In between, only a lower limit of the emitted NO_x would be determined if NO_2 is measured and steady state is already assumed.

Using eq. 4 as approximation of the temporal build-up of NO_2 neglects the fact that the concentrations of the two species vary along the plume, as mixing with ambient air takes place (i.e. O_3 is variable) and, especially, the plume concentration is diluted (i.e. NO_2 is time dependent and cannot be assumed constant in the integration of eq. 3). Detailed information on these processes is not measured.

As no further trace gases within the plume were measured, no useful information about the O_3 and total NO_x contents and their development is available here. In addition, our measurements show that the plume behaviour is strongly inhomogeneous, so that a more sophisticated plume modelling would be needed. We consider the detailed chemical analysis of the exhaust plume to be out of the focus of this more technical paper.
For a reasonable estimate of the order of magnitude of the NO_x emission, the cross sections through the plume furthest away from the stack are used.

Comment (19)
P3617, L1 – I was a little bit confused about this section. First you derive VC and Int for both the motorway and the bright field. Then you seemingly subtract the VC derived for the field from the VC derived from the motorway. I thought this was done to correct for variability in the AMF caused by the enhanced albedo (which is at least similar for the two locations). You arrive at 3±2.2e15 molec/cm2. However, you then describe how much an additional correction for variations in the albedo would affect the values. Didn't you already correct for this effect by using a differential measurement, i.e. looking at the motorway relative to the bright field?

Answer to Comment (19)
The subtraction of the two VC in the discussion on P3618 was performed in order to discuss the NO_2 amount above background, i.e. the enhanced NO_2 above the motorway as compared to the bright field, assuming that the enhancement is caused by the traffic on the motorway.
The intensity is larger above the bright field than above the motorway, however, the two intensity values are equal within their error margins. Therefore, the larger NO_2 above the motorway cannot be explained by the large reflectivity from the road surface.

The estimation of the AMF influence on the NO_2 amount is addressed then (P3618 L13) because the absolute NO_2 column above the motorway is slightly overestimated. The albedo influence on the AMF has not explicitly been taken into account initially. A larger albedo enhances both contributions to the observed NO_2 column, the NO_2 from the motorway and the background amount, in the same way. Only the influence on the difference is discussed in comparison to the study by Pundt et al. who derive NO_2 emissions from a motorway.

Part 2. Referring to the Minor Corrections

Particularly on the first few pages, there are a large number of sentences that do not follow the basic structure of subject, predicate, object (S-P-O) but instead start with the object or a subordinate clause instead. While grammatically correct, overuse of this inverted sentence structure tends to make the material more difficult to comprehend. I recommend rewording some of these sentences to a more simple S-P-O sentence structure. Examples of these inverted sentences include:
Abstract first sentence: "For the purpose of trace gas measurements and pollution mapping, the AirMAP has been developed, characterized and successfully operated from aircraft"
might be changed to:
"The AirMAP has been developed, characterized and successfully operated from aircraft for the purpose of trace gas measurements and pollution mapping."
→We have changed the word order and additionally split the sentence into two.

Abstract second sentence
→ Done.

Abstract line 8 – "With a wide-angle…"
→ Done. Additionally split into two sentences.

Abstract line 14 – "From a maximum.."
→ As an exception we wish to keep the word order in this case as we feel that its meaning is better understandable this way.

Abstract line 18 – "For accurate spatial…"
→ Done.

In addition, word order in the following sentences has been changed:
P3593 L14, L16

P3592,L14 – Replace "single" with "individual" in "… by 35 individual fibers…"
→ Done.

L22 – use simple past: "AirMAP was operated on the…"
→ Done.

L24 – "AirMAP clearly DETECTED the emission plume downwind OF the …"
→ Done.

P3593, L6 – "NO2, is an important trace gas in the…"
→ Done.

L18 – "…NO2 lifetime, THE spatial and…"
→ Done.

P3595, L6 "… by an industrial consortium, AND first data…"
→ We have split the sentence instead.

P3596, L12 – Here in particular, I recommend switching to S-P-O sentence structure : "The NO2 column amount below the aircraft was observed during multiple overpasses over the power plant exhaust plume."
→ Done.

L18 – remove "currently" here. If you'd like to talk about other wavelengths, perhaps do so in the outlook.
→ Done.

L27 "… guide allows both optical imaging and flexible…"
→ Done.

P3597, L24 – "of the detector WAS performed…"
→ Done.

L27 – What are "additional instrumentation parts"? Do you mean "the rest of the AirMAP setup"?
→ We have changed this to "The following instrumental parts of AirMAP" as the mentioned parts are not "the rest of AirMAP" but those parts described in the next sentence.

P3599, L 10 – Recommend removing "adequately". This is implied by the rest of the sentence.
→ Done.

L12 – Is "AHRS" a standard acronym. I wasn't familiar with it, and I don't think it's introduced.
→ Yes, it is already introduced in an earlier section. See on P3595 L23 in the submitted version (Section 2, 3rd sentence).

P3600, L4 – Perhaps choose a different symbol for longitude? λ is already used elsewhere for wavelength.
→ λ is used for wavelength in a place where it is not necessarily needed (Eq. 6, and P3606 L26). Therefore, we decide to retain λ as symbol for longitude and avoid using it for wavelength.

P3601, L10 – Recommend replacing "right" with "correct"
→ Done.

P3602, L7 – Remove "far".
→ Done.

P3604, L6 – Replace "takes care of" with "accounts for"
→ Done.

L19 – Replace "strong" with "significant"
→ (probably means L24) Done.

P3605, L16 – This sentence is confusing, perhaps try: "The relative occurrence of a given NO2 column density is plotted in a histogram with a bin width of 0.5e15 molec/cm2.
→ Done.

P3606, L1 – "NO2 enhancements are CLEARLY above the detection limit"
→ Done.

P3607, L9 – "… 2.6 DIRECTLY below the aircraft…"
→ Done.

P3610, L7 – "… during a DESCENT INTO a regional airport…"
→ Done.

P3612, L26 – I believe the ratio of reaction rates for the NO – NO2 reactions is typically called the "Leighton Ratio". Perhaps introduce this term here? It might be useful for literature searches.
→ Done.

P3613, L16 – replace 'assured' with 'assumed'.
→ Done.

P3616, L9 – 30 m uncertainty for geolocation assuming what flight altitude?
→ Done.

L10 - replace "entering" with "that enter in"
→ Done.

L13 – replace "pieces" with "segments"
→ Done.

P3617, L28 – insert "with" after "meters, "
→ Done.

P3620, L5 – replace "have been" with "were"
→ Done.

L24 – Perhaps also mention OClO?
→ Done.

P3621, L1 – "focusing" is misspelled.
→ No, as "focussing" and "focusing" are both correct. We have changed it nevertheless.

Fig 3 caption – "… shows the HIGH REFLECTIVITY OF the motorway…"
→ Done.

Fig 5 – I can't distinguish between the solid and dotted lines in my copy
→ Neither in ours.. The line style of the dotted lines has been changed.

Fig 6 caption – Replace "strong" with "significant" or "considerable"
→ Done.

Fig 12 – It does appear that one part of the track sticks out a bit – in particular, the flight segment that was conducted approximately in north-south direction seems to give slightly lower values for the background than the rest of the data. Can this be easily explained?
→ It is true that the respective flight segment yields slightly smaller NO_2 amounts. We have considered several influencing factors which tend to result in lower NO_2 columns.
The reference intensity I0 contains some stratospheric NO_2 which cancels as long as I and I0 are taken within a short time interval. The north-south flight segment was recorded some time later than the other flight tracks above the power plant area. In addition, flight altitude was lower during the overpass over the power plant area so that part of the boundary layer NO_2 is missed by the measurement. The boundary layer height itself also changed during the survey. Instrumental effects may also play a role, however, measurement quality was stable during the flight.

We include the following passage in the manuscript.

One flight segment in north-south direction shows lower NO_2 amounts than the other flight tracks. The respective segment was flown latest in the pattern and at lower altitude. Therefore the track is rather narrow. The later time means that the SZA and the influence of stratospheric NO_2 have changed. This effect is noticeable but too small to entirely explain the observed change in the background NO_2. The lower flight altitude might cause that some NO_2 is missed by the measurement. Probably, additional effects influence the measurements in this flight track, which is however not further used in this study.