Response to Reviewer #2, comments to “The CU 2-dimensional MAX-DOAS instrument - part 1: retrieval of NO_2 in 3 dimensions 1 and azimuth dependent OVOC ratios” by Ortega et al., 2014.

We greatly appreciate referee #2 for the evaluation of our paper and for the several detailed comments, we feel that helped to improve the paper. Please see below our answers.

Black: Referee’s comments
Blue: Author’s reply
Green: sentence added/modified in the manuscript

General Comments:

(1) In the abstract and introduction, the new instrument and 2D-MAX-DOAS are frequently praised as new, innovative technique implying somehow that the 2D-MAX-DOAS technique is invented here. Please go through the text and rephrase carefully. 2D-MAX-DOAS is a well-established technique used by many groups (NASA, Heidelberg, BIRA, Bremen, Washington and many others). Instead, the authors should make the point that they present a measurement strategy which exploits the time consuming 2D-MAX-DOAS very efficiently (i.e., performing profile retrievals in each azimuth direction would require complete elevation scans and thus much more time).

During the access review process Referee # 1 already had provided a detailed review with a similar point. In the revised manuscript we have added reference to Piters et al. (2013), and also added the names of all the institutions that are at present carrying out 2D MAX-DOAS measurements. We have conducted a thorough literature review and do not find any missing references. To our knowledge, no previous literature has documented the retrieval of trace gases in 3D from 2D MAX-DOAS. The emphasis on the retrieval method has been clear from the first sentence of the abstract. We have added emphasis about “a measurement strategy, which exploits the time consuming 2D-MAX-DOAS very efficiently (i.e., performing profile retrievals in each azimuth direction would require complete elevation scans and thus much more time).” in the introduction and conclusion section of the revised manuscript. The modified text in the introduction reads as:

“Traditional MAX-DOAS observations have been performed using one or two azimuth views, and have highlighted the need for studying inhomogeneities in the air mass close to the instruments (Brinksma et al., 2008). Thus far, the analysis of MAX-DOAS measurements have been limited to at most four azimuth angles (AA). For example, Wagner et al. (2011) used three telescopes to measure at three fixed azimuth views and several EA simultaneously to estimate HCHO, and NO_2. Recently, Sinreich et al. (2013) introduced a parameterization approach to interpret measurements of near surface concentrations of NO_2 in three azimuth directions, and demonstrated the validity of this approach by comparing with two long-path DOAS instruments facing in opposite direction. Wang et al. (2014) presented measurements under four azimuth viewing angles and with a fixed (low) EA to retrieve surface mixing ratios of several trace gases. 2D-MAX-DOAS is a rapidly emerging technique that points to any azimuth angle (AA); the data interpretation is complex. Piters et al. (2012) describes several 2D-MAX-DOAS instruments from BIRA, University of Bremen, University of Heidelberg, Washington State University, and NASA. Thus far, only direct sun irradiance measurements with a 2D telescope have been used to obtain columns of NO_2 without sophisticated radiative transfer calculations (Herman et al., 2009). A retrieval strategy to measure 3D distributions of gases independent of solar position is complex, and currently missing in the literature to the best of our knowledge. Our retrieval strategy exploits 2D-MAX-DOAS measurements very efficiently, i.e., a full profile retrieval is conducted for a subset (here one) azimuth direction, and used to assess azimuth dependencies of near-surface VMR using a parameterization approach that builds on Sinreich et al. (2013)."

(2) There is no doubt that the authors built a very good instrument, but if the focus is so much on the new instrument (even the title states it) the authors have to point out what the new, innovative aspect is. In principle, no component of the instrument is new, but they are combined and arranged in a different way. For example, Heidelberg and Bremen are doing 2D-MAX-DOAS, Heidelberg even with a rotating prism; BIRA, NASA and Washington can perform direct sun measurements, Washington even with an integrating sphere. I see insufficient references to these instruments. My personal preference was to include a section describing other existing instruments and their capabilities (with references). Then, the authors could highlight advantages of their instrument (maybe stating that they combined the best aspects of existing technologies, which is true to my opinion).

See our above response. We believe our citations of the literature are complete in the revised manuscript. There is no previous publications that would present 2D measurements of trace gases to our knowledge. If Referee #2 knows of any peer reviewed literature that we may have missed, we would be happy to include it.

(3) As a comment: There is a lack of independent ground-based measurements (e.g., in situ) to validate the results and the presented approach. This is a pity since Mainz (as any other larger German city) is performing routinely in situ air quality measurements. There are 6 stations in Mainz which measure at least NO2 (see link below, unfortunately only in German). Normally, there are some short-term results available online, but these people are often helpful if asked for data. http://www.luft-rlp.de/aktuell/stationswerte/stickstoffdioxid/index.php
This data would have been well-suited to validate range-resolved NO2 in different azimuth directions (as shown in Fig. 9)

We have responded to a similar comment in detail as part of our response to Reviewer#1. The revised Section 4.6 now discusses a comparison of the parameterization approach with optimal estimation. This section has been further expanded in response to the comments from all three reviewers. In particular, we have added a new sub-section 4.6.1 that compares the near surface NO_2 VMR from the parameterization approach with that derived from optimal estimation. Also we have revised Section 4.6.2 to make our rationale transparent for why a comparison with the network in-situ sensors has not been attempted in light of the more fundamental sampling problem of inhomogeneous distributions. We have also added reference to Remmers et al. 2015, manuscript in preparation, which presents a comparison of NO_2 dSCDs from different 2D-MAX-DOAS instruments during MAD-CAT. We refer the Reviewer to check the added text in our response to Reviewer #1.

(4) There is something strange with the fit settings (Tab. 3 and 4). For example, Tab. 4 states that CHOCHO and NO2 fits in the range of 434-460 nm are using cross sections of BrO, HCHO (UV absorbers), but not CHOCHO itself which would be number 6 according to Tab. 3.
In addition, if the authors include two ozone cross-sections at different temperatures in the CHOCHO fit in the Visible, please shortly explain why this is necessary.

Thanks for pointing this out. This was an edit error in table 4. We have corrected table 4 accordingly.

(5) As being based on Sinreich et al. (2013) the authors should include a brief section summarizing this study and the basic idea and findings of the parameterization approach. In this context, please clarify which preconditions exist for the parameterization approach. Sinreich et al. (2013) state that collapsing of slant columns is a necessary condition. Was this the case for the MAD-CAT campaign? Please provide evidence. If yes, is your approach limited to conditions of very high aerosol scenarios? Please clarify and give recommendations, under which conditions your approach is applicable.

In section 3.2.3 we have added the short description below summarizing the basic idea of the parameterization approach presented in Sinreich et al. (2013). The text reads as:

“We apply the parameterization approach introduced in Sinreich et al. (2013) to the AA scan to obtain the horizontal near-surface mixing ratios of NO_2. Shortly, Sinreich et al. (2013) pointed out that dSCD obtained from MAX-DOAS in a single and low EA can be converted into near surface box average mixing ratios by means of parameterization of the RTM. In the following, we use terminology consistent with that of Sinreich et al. (2013), however we refer to MLH instead of planetary boundary layer (PBL) to avoid confusion with the meteorological term. Briefly, the differential light path of the EA with respect to the reference is determined by means of O_4 dSCD weighted by a correction factor. The correction factor, f_c, accounts for the different O_4 and NO_2 vertical profile shapes and the difference in absorption wavelengths. Sinreich et al.

(2013) showed that this method does not depend on the actual aerosol load, and only weakly depends on the aerosol layer height, if sufficient aerosol is present. This was assessed in Sinreich et al. by a 'collapsing' of the O_4 dSCDs in a set of two low EA, which is however not a pre-requisite for this approach to work for very low EA (see Supplment text in Sinreich et al., 2013; Wang et al., 2014). An important pre-requisite knowledge to calculate f_c is knowledge about the trace gas MLH. As long as this height is higher than 500 m, the method is insensitive to the actual value of the trace gas layer height. We apply this method to multiple wavelengths to average NO_2 over different horizontal distances. At each wavelength the near-surface volume mixing ratio of NO_2 can be evaluated using the following equation (Sinreich et al., 2013):"

(6) The authors used a profile retrieval to obtain the PBL from measurements in different elevation angles (but only 1 azimuth direction) and then the PBL from this for the parameterization approach, i.e. for retrieving trace gas VMR in different azimuthal directions. This requires a constant PBL in all directions (and all ranges), which would require a flat terrain. Is this the case for the measurement site or are changes of the PBL or the terrain taken into account?

As described in the text above we use terminology consistent with that of Sinreich et al. (2013), however we refer to the NO_2 mixing layer height (MLH_{NO2}) instead of planetary boundary layer (PBL) to avoid confusion with the meteorological term.

The Reviewer is correct, we use the standard azimuth angle to estimate the MLH_{NO2}, which then is used to calculate the correction factors needed to retrieve azimuth distribution of NO_2. As stated in section 4.1.2 the terrain effect may play a role, especially at longer wavelengths. In order to avoid complexity in the manipulation of the MLH_{NO2} we used the time resolved average MLH_{NO2} obtained with the three wavelengths, and we use the standard deviation as a proxy of the mixing height variability to conduct a sensitivity study and assess the error in the calculation of the correction factor (as stated in section 4.1.2). This implicitly includes terrain effects. We agree that azimuth directionality can be taken into account, however during our setup only a single azimuth view was used to retrieve vertical profiles of NO_2. Hence, in the conclusions we have suggested that more azimuth views could help to estimate PBL variability and decrease even more the error in the correction factors. The sentence added in the conclusion reads as follow:

"For future 2D-MAX-DOAS measurements we recommend the use of more than one azimuth view to estimate the MLH_{NO2} in several directions, and account for possible terrain effects."

In addition, parameterization approaches are normally well-suited for box-like profiles. While this is likely at the (polluted) measurement location, on the other hand, the authors use a profile retrieval with an exponentially decreasing apriori profile (and the retrieved profiles in Fig. 6 are also not box shaped) and a factor of 1/2e to determine the boundary layer height which is then used in the parameterization approach for other azimuth directions (and I guess the conversion from VMR to VC in Sect. 4.5 using the PBL is again based on a box-profile assumption?). This is a contradiction.

This is not necessarily a contradiction, as our definition of NO_2 mixing height is based on the actual retrieved NO_2 profile. We have conducted additional sensitivity studies to estimate the mixing height under an assumption of a box-profile shape, and find that the sensitivity in the effect on the correction factor, and thus the near surface VMR is small (< 10%).

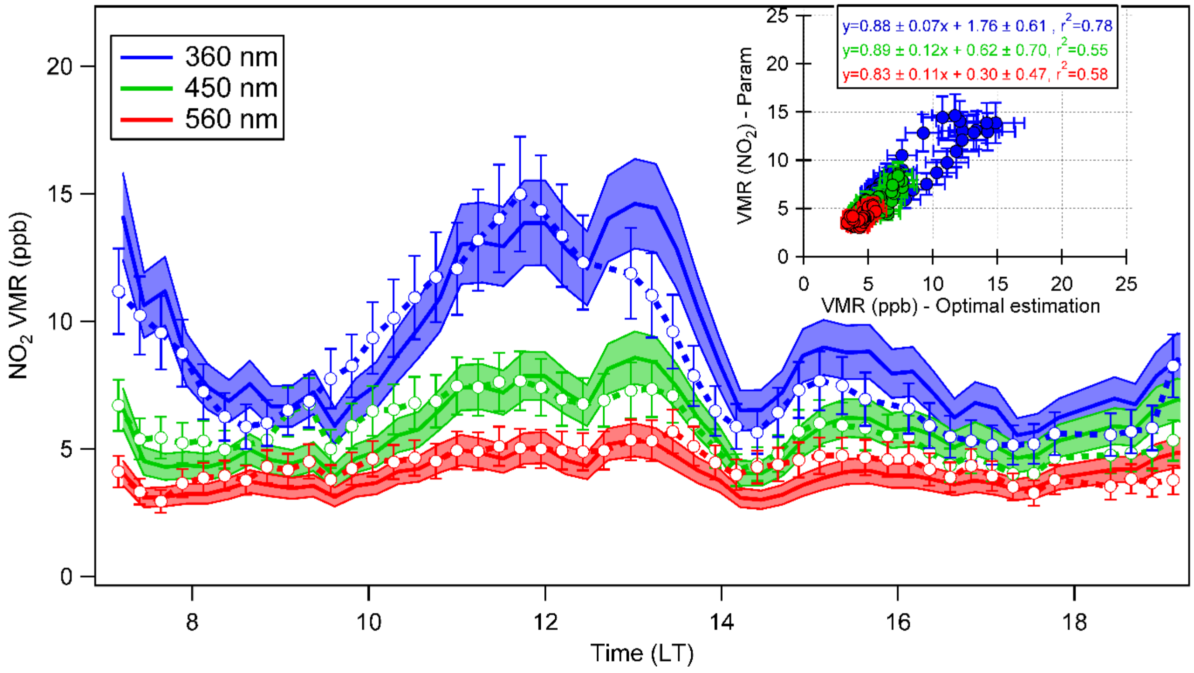
(7) Figure 8 shows that the main azimuthal viewing direction is very close to Frankfurt Airport which is only about 20 km away (i.e. within the effective radius) and one of Europe's main hubs. Thus, enhanced uplifted layers of NO2 (and aerosols) are likely in this direction in which the profile retrieval is performed. The MAX-DOAS profile retrieval has normally high sensitivity only close to the ground, while the retrieved profile in higher altitudes is determined mostly by the apriori profile (which is an exponential decrease in the present study). Thus, the authors should elaborate sensitivity tests for uplifted NO2 and possible impacts on their retrieval.

Thanks for pointing out this interesting idea. In fact that would make an interesting study. However in the current manuscript we make no attempt to retrieve NO_2 in the upper layers. Surprisingly however, in earlier work with airborne MAX-DOAS mapping NO_2 VCDs over the Denver International Airport we did not find significant enhancements of NO_2 VCDs above the airport compared to its (mostly rural) surroundings. Aircraft are important sources of NOx primarily in the corridors used by aircraft during take-off. Studies that exploit the sensitivity towards layers aloft with MAX-DOAS are still in its infancy, and deserve further attention. However, for the present setup the NOx in the boundary layer will need to be decoupled from NOx aloft. We consider this topic is beyond the scope of this (quite complex) method paper.

(8) How does the retrieved profile (from optimal estimation) in the main azimuthal direction look in comparison to the VMRNO2 calculated from Eq. 5 with fc from Eq. 6 (where the PBL from profiling is needed for) in the same azimuthal direction? Are the results consistent? Please give an example plot of this and a short comment (I guess one would expect that VMR_NO2 from Eq. 5 and 6 is some average of the profile in the lowest layers?).

We provide a plot comparing the near surface VMR from OE and the parameterization approach for NO_2 in the standard azimuth view as part of Fig. S5. We have further added the following text to the revised manuscript as part of the new subsection 4.6.1: Comparison of near-surface NO_2 VMR:

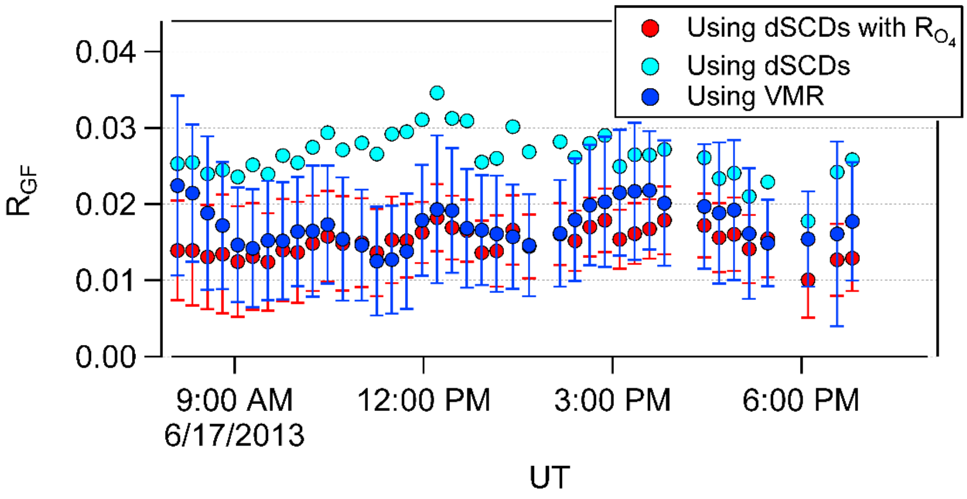
“The multi-wavelength NO_2 near-surface VMR retrieved with the parameterization approach are compared with those derived from OE for the standard azimuth view in Figure S5. Note that the two approaches are independent, and do in particular not use identical NO_2 dSCD. As pointed out in sections 3.2.3 and 3.3 the NO_2 derived with the parameterization approach uses only data from the lowest EA, and the mixing ratio is characteristic only over the differential effective path length and height in the lower portion of the MLH. By contrast, the OE uses NO_2 dSCDs from a full set of EA. In reality, the comparison of MLH_{NO2} in Section 4.1.2 shows that there is some variability in NO_2 with altitude. The near surface VMR is subject to smoothness and assumptions about a priori profile shape/magnitude and error covariance matrices, and thus differences in Fig. S5 may represent actual gradients in the chemical state of the atmosphere, and should not be interpreted exclusively as an ‘error’ of the retrieval strategy’. In order to account for the different horizontal range sensitivity we have averaged the NO_2 profiles from figure 6 to a similar representative height of the parameterization approach for each wavelength. In general, both methods capture the NO_2 variability within the spatial scale for each wavelength. The slopes of a linear regression (parameterization versus OE) varied from 0.83-0.88 and intercepts from 0.11-1.76 ppbv NO_2 (see insert on Figure S5). There is further a general good correlation, with the R^2 ranges between 0.55-0.78, for each wavelength considering the difference in sensitivity of air masses probed by the two methods.”



“Figure S5. Multi-wavelength comparison of NO_2 time series using the parameterization approach (continuous lines) and optimal estimation (open circles). In order to account for the different range sensitivity we have averaged the NO_2 profiles from figure 6 to a similar representative height of the parameterization approach for each wavelength. In this case we have used a height of 0.2, 0.6, and 1.0 Km for the 360, 450, and 560nm. The insert shows the linear correlation for each wavelength."

(9) The trace gas ratios (CHOCHO-to-HCHO etc., Sect. 3.4 and 4.4) are not calculated from the onion peeling approach, but directly from slant columns. Differences in the path length in the UV (HCHO) and Visible (CHOCHO) are accounted for using a correction factor determined by O4 slant columns in the Visible and UV, which I think is valid (R_O4 in Eq. 9). However, parts of the CHOCHO information come from a distance from where no HCHO information comes from. For example, it can happen that a source of CHOCHO and HCHO is in a larger distance and can be seen only in the Visible (this is even more likely in an urban, inhomogeneous environment like the Frankfurt-Mainz area). Then, the CHOCHO slant column would be enhanced while the HCHO slant column is not enhanced and the simple conversion based on R_O4 does not take care of this. Thus, I think using range-resolved results from the onion-peeling is the much better way, especially since this technique is demonstrated in the previous section (of course the ratio of CHOCHO to HCHO is impossible then, but for the ratios with NO2 this is possible).

We partially agree. As mentioned in the text the ratios calculated within the same window do not carry high uncertainty, however the CHOCHO-to-HCHO ratio are subject to higher uncertainty because of the difference in wavelength. We believe this is partially corrected with the R_{O4} factor, which accounts for the different path length. In order to assess the reliability of the R_{GF} calculated with the dSCD we have retrieved vertical profiles of CHOCHO and HCHO for the standard azimuth view using OE. The near surface VMR of CHOCHO and HCHO are used to calculate $R_{GF\text{-}OE}$ and compared with the R_{GF} evaluated earlier. The time series of both methods show no significant difference and the values are always between error bars (see figure below) only if R_{O4} is accounted for in the calculation. If the R_{O4} factor is not used RGF is overestimated by up to 45% based on the ratio of dSCDs.



We have modified part of the description of the ratio calculation (section 3.4) to make it more explicit:

“The azimuthal distribution of the trace gas ratios in the boundary layer directly uses the respective trace gas dSCDs. R_{FN} and R_{GN} are calculated with the dSCD obtained in the same wavelength window to assure the optical path lengths are directly comparable. However, R_{GF} needs special attention since HCHO and CHOCHO retrievals employ different wavelengths during the DOAS analysis (Table 3), and thus reflect different optical paths.”

Another important advantage which arises from using dSCD in the azimuth scan is that no complex and laborious RTM is necessary, resulting in a fast retrieval to determine near-real time air mass chemistry.”

We also have added a short description at the end of section 4.4 to describe the error of the trace gas ratios:

“The error on the ratios, calculated with the DOAS fit error propagation of the dSCDs, show errors lower than 5%, 10%, and 20% for R_{FN}, R_{GN}, and R_{GF} respectively.“

In addition: Despite that the site of study is an urban area in Mainz, some biogenic contribution of VOC could be expected at least in some of the azimuth directions due to major growth of plants corresponding to the warm season and then major isoprene emissions. Could the authors comment on this?

HCHO and CHOCHO are two intermediate species produced in the oxidation of biogenic and anthropogenic VOC. As stated in sections 3.4 and 4.4 the ratio of CHOCHO to HCHO (R_{GF}) has been proposed as a metric to identify VOC emission types. In our study, the only available variable to pinpoint possible major contribution of biogenic VOC is the R_{GF}. Satellite studies reported that for R_{GF} lower than 0.04 the air mass is associated with anthropogenic VOCs emission sources while indices above 0.04 might relate to biogenic emission sources. As pointed out in section 4.4 and Fig. 10 the R_{GF} values are always lower than 0.02. Even when there is an increase of HCHO and CHOCHO in the South direction in the late afternoon (see HCHO to NO_2 (R_{FN}) and CHOCHO to NO_2 (R_{FN}) on Fig 10) the value of R_{GF} remains low, which indicates the dominance of anthropogenic influences in Mainz based on our current understanding of R_{GF}. As pointed out in section 4.4 the absolute values of the R_{GF} differ between satellite and in-situ observations, and the interpretation of these ratio warrants further investigation.

(10) As MAD-CAT is an intercomparison campaign and many international groups gathered for several weeks, do you know if there will be further results/publications (if yes, the authors should announce this as being the first publication)?

Upcoming publication/results have been introduced accordingly along the manuscript.

Specific comments:

P. 11655, L. 1-3: 2D-MAX-DOAS is well-known (rephrase or delete the sentence)

As mentioned before we agree that 2D-MAX-DOAS measurements have already been performed before, but there is a severe shortage of publications on the topic given the amount of work several groups have spent developing instruments. The sentence now reads:

"3D distributions measurements with 2D-MAX-DOAS provides an innovative, regional perspective about trace gases, their spatial and temporal concentration gradients, and maximizes information to compare near-surface observations with atmospheric models and satellites."

P. 11656, L. 4-6 "The development … currently missing". Again one of the sentences that have to be rephrased. You may want to include for example Piters et al. (2012) for MAX-DOAS instruments already measuring in (more than 4) different azimuthal directions.
Piters et al.: The Cabauw Intercomparison campaign for Nitrogen Dioxide measuring Instruments (CINDI): design, execution, and early results, AMT, 2012.

We have added the reference, and modified the second paragraph in the introduction:

"MAX-DOAS instruments collect scattered sunlight at different elevation angles (EA) above the horizon. Spectra recorded at several EA between the horizon and the zenith view greatly enhanced photon paths and sensitivity inside the boundary layer, which can be used to derive vertical profile information (Hönninger et al., 2004). Traditional MAX-DOAS observations have been performed using one or two azimuth views, and have highlighted the need for studying inhomogeneities in the air mass close to the instruments (Brinksma et al., 2008). Thus far, the analysis of MAX-DOAS measurements have been limited to at most four azimuth angles (AA). For example, Wagner et al. (2011) used three telescopes to measure at three fixed azimuth views and several EA simultaneously to estimate HCHO, and NO_2. Recently, Sinreich et al. (2013) introduced a parameterization approach to interpret measurements of near surface concentrations of NO_2 in three azimuth directions, and demonstrated the validity of this approach by comparing with two long-path DOAS instruments facing in opposite direction. Wang et al. (2014) presented measurements under four azimuth viewing angles and with a fixed (low) EA to retrieve surface mixing ratios of several trace gases. 2D-MAX-DOAS is a rapidly emerging technique that points to any azimuth angle (AA); the data interpretation is complex. Piters et al. (2012) describes several 2D-MAX-DOAS instruments from BIRA, University of Bremen, University of Heidelberg, Washington State University, and NASA. Thus far, only direct sun irradiance measurements with a 2D telescope have been used to obtain columns of NO_2 without sophisticated radiative transfer calculations (Herman et al., 2009). A retrieval strategy to measure 3D distributions of gases independent of solar position is complex, and currently missing in the literature to the best of our knowledge. Our retrieval strategy exploits 2D-MAX-DOAS measurements very efficiently, i.e., a full profile retrieval is conducted for a subset (here one)

azimuth direction, and used to assess azimuth dependencies of near-surface VMR using a parameterization approach that builds on Sinreich et al. (2013).
"

P. 11658, L. 10 That means you are not using a fiber bundle? Does the mono-fiber preserve polarization?

The short and introductory description in section 2.1.1 has been modified as follow in order to make it clear:

"The photons are directed onto an f/4 2.54 cm lens via the same optical axis; the lens focuses the light into the mono-fiber which is coupled to the fiber bundle (see section 2.3)."

P. 11658, L. 12: "was designed to exhibit a low residual error". Please explain what is the special thing that reduces the residual error (since this addresses the spectrometers and not the telescope).

The low residual error is primarily the consequence of light-strong optics, large diameter fibers, mixing-fibers to assure homogeneous illumination of the spectrometers etc, as is described in detail in Coburn et al. (2010).

P. 11658 L. 12 (and figure 1): The light from the integrating sphere and the scattered light view ports are using the same lens, correct (which is not shown in Fig. 1, but would be located at the right edge of the Fig.1)?

Thanks for the observation. We have added a short description in Fig. 1 to make clear that the lens is on the right side of the prism/integrating sphere.

P. 11656, L. 21-23 and P. 11658, L. 20-22: The two modes are not special to the instrument which is able to point in any direction, I think. Moreover, these three modes have to do with the retrieval strategy that is presented in this study which should be pointed out more clearly.

The three modes are defined by software, and are thus characteristic of the way we operate our 2D-MAX-DOAS instrument. We have modified the revised manuscript to make it clear

"The different modes, defined by software, maximize sampling of the horizontal and vertical distribution of trace gases with a single instrument and with fast time resolution."

P. 11659, Sect. 2.1.3 I find the alignment procedure really interesting. For the direct sun measurement (I know, this is the topic of another paper), do you use the same alignment procedure, so is this accurate enough for doing direct sun DOAS (without tracking)?

Thanks for the interest on this. This in fact is a topic of the second part of the paper. Shortly, yes the same procedure is applied for direct sun measurements. However, with direct-sun observations it is less important to have accurate pointing than for mode 2 measurements (almucantar scans). As pointed out by Herman et al. (2009) the integrating sphere minimizes the effect of pointing errors with direct sun observations. The effect of the pointing inaccuracy, FOV, among others will be described in the second part.

P. 11660, L. 18. Do you care for effects like atmospheric refraction when using a distant object for calibration of the EA, i.e. effects that make the object appear at another angle as the geometric angle? How large do you estimate such effect?

We have made no further attempts to account for atmospheric refraction. Refraction may play an important role for extraterrestrial objects where the light path does not follow a straight line but bends due to the effect of the air refractive index making appear objects higher than their real position. On the other hand, for close objects as the hill used for the EA adjustment, refraction may not play a huge difference. Nevertheless, we have seen that for extraterrestrial objects close to the horizon the effect of refraction would be less than 0.2˚ (depending on T, and P, and wavelength) (Gisi et al., 2011). In section 2.1.3 we added the following text:

"We have made no further attempts to account for atmospheric refraction, which for extraterrestrial objects close to the horizon the effect of refraction would be less than 0.2˚ (depending on temperature, pressure, and wavelength) (Gisi et al., 2011)."

P. 11661, L. 10: I would put the information that you restrict to a case study already to the introduction. Furthermore, this raises again questions about possible upcoming publications/results.

In the revised manuscript we describe that results are restricted for the case study. Several upcoming publications of results of various instrument inter-comparisons (e.g., for NO_2, HONO, CHOCHO) have been introduced accordingly along the manuscript.

P. 11661, L. 24 "1.7 mm optical mono-fiber" This is the outer diameter of the mono-fiber, correct? Otherwise, there is a discrepancy with the theoretical FOV = 2*arctan(d/2f) with d = 1.7 mm and f = 4*25.4 mm which would give a value of 0.95°.

Thanks for the observation. We were using a FOV from an earlier deployment. The revised manuscript has been updated with the correct theoretical FOV of 0.95˚.

P. 11662, Sect. 3.1 The authors should at least mention that the DOAS technique is based on the

Lambert Beer law. In addition, one could show the DOAS equation, so that also readers who are not familiar with DOAS can conclude which "reference spectrum" is meant (P. 11663, L. 5)

We adopt the reviewer suggestion to mention the Lambert Beer law in section 3.1. We think there is ample references that cite the DOAS equation among the references cited (e.g, Stutz and Platt, 2008).

P. 11663, L. 7 "… a Ring cross section is calculated from each reference spectrum…" This is performed as suggested by Wagner et al. (2009)? If yes, please give that reference here (or another reference describing the method you used).
Wagner et al.: "Three-dimensional simulation of the Ring effect…", AMT, 2009.

Thanks for the suggestion. We have included Wagner et al. (2009).

P. 11661, Sect. 2.3 In my opinion, the first paragraph of Sect. 2.3 belongs to the instrument description where I was missing information about the non-telescope-part of the instrument.

Thanks for the observation. We have moved text from lines 12-17, page 11661 into Section 2.1.

P. 11665, L. 16 How do you know if the minimum is local or global?

We have rephrased the revised manuscript as follows:

"The aerosol extinction convergence criteria are achieved when the residuals of the cost function in Eq. (1) fall into a valley under certain feasible set of extinction profiles calculated with the constrain parameters."

P. 11666, L. 9-11 This is somehow not a complete sentence…

We have re-phrased the sentence as follow:

"The $\boldsymbol{S_a}$ matrix was treated as a tuned parameter in order to avoid non-real oscillations in the retrieved profiles (Clémer et al., 2010; Baidar et al., 2013; Hendrick et al., 2013.). The diagonal elements of the $\boldsymbol{S_a}$ were set to account for large variations, up to 100%, of the initial a priori profiles. The nondiagonal in the $\boldsymbol{S_a}$ matrix were correlated through the altitudes (z) with a well-used exponential Gaussian decay function (Barret et al., 2003; Clémer et al., 2010):"

"In this equation z_i and z_j are the altitudes of the i-th and j-th grid layers respectively and γ is the correlation parameter, which was set to 0.3 km, similar to the inversion grid height. The wavelengths used to retrieve NO_2 vertical profiles are the same as for the aerosol extinction profiles (350 nm, 450 nm, and 560 nm)."

P. 1166, Sect. 3.2.3 Please include a brief description/summary of Sinreich et al. (2013) and the correction factors, including pre-conditions and limitations, if existent.

Please see our response of the general comment #5 above.

P. 11670, L. 19 "… points out that the aerosol load around Mainz is homogeneous". This is an interesting result and it is also surprising. Looking at a map of Mainz and its surrounding (radius of 20 km, which is the effective radius), one finds large inhomogeneity in population (expecting less pollution to the North-West and more pollution to the North and North-East). Isn't this also in contradiction to the azimuthal NO2 distribution derived later? Is there an explanation for the aerosol to be homogeneous?

We have modified the sentence to read: "The agreement between the two instruments could be coincidental, or indicate that the aerosol load around the city of Mainz is homogeneous."

As in others cities the NO_2 and aerosol might not be co-located due to different emission sources, transport, and life time. There is two possible reasons for why variations in aerosol optical thickness could be a more homogeneously distributed than NO_2 VCDs. First, the lifetime of aerosols is much longer than that of NO_2, which leads to a more homogeneous distribution. Second, most of the aerosol mass of accumulation mode sized particles (the most active size in terms of the number weighed optical properties) is from secondary sources, i.e., is formed in the atmosphere as opposed to emitted. However, we did not mean to limit this statement as narrowly, and much work remains to be done to assess azimuth distributions of AOD, and the spatial scales over which AOD varies in the atmosphere.

P. 11671 L. 10-14 Please consider to show a picture of the averaging kernel from a measurement from 17 June 2013 to demonstrate your comment.

An averaging kernel example is now shown on the modified Fig. 6.

P. 11671, L. 20 Where does the factor of 1/2e for determining the PBL height come from? Is it a commonly used factor, just arbitrarily chosen or from any literature (then please give a reference).

The 1/2e factor for determine the MLH_{NO2} is motivated by the fact that the NO_2 is distributed along this altitude (Fig 6) and may be a good height to calculate the correction factors. As mentioned along the manuscript we have conducted sensitivity studies to determine the effect of this variability, which has been counted on the uncertainty described on the new table 6.

P. 11674, Sect. 4.2.2 In the error discussion you should include changes of the PBL due to the terrain.

In our response to reviewer#1 we have conducted a sensitivity study on the effect of different definitions of MLH on the correction factor used for VMR conversion. For details see our response to reviewer#1, and the revised manuscript section 4.1.2.

P. 11676, Sect. 4.4 In contrast to the onion-peeling approach which is performed only for the case study, the trace gas ratios were calculated for the whole campaign and your discussion is based on this? Maybe I missed that, but please make this clear.

Thanks for the observation. We have modified section 4.4 to make clear that results of the trace gas ratio are for the case study, with potential to apply it for the whole campaign. We decided to present results just of the case study to relate them with the NO_2 results.

P. 11678, L. 21-23: Please explain how you calculated the NO2 VMR into VC using the PBL. If you perform this transformation assuming a box-profile, this is again in contradiction to the apriori profile used in the profile retrieval (and to the retrieved profiles in Fig. 6).

As mentioned in section 4.5 the NO_2 VCD in the azimuth sectors are calculated using average VMR and the PBL estimated with retrieved NO_2 profiles. As pointed out previously this is not necessarily a contradiction, as our definition of the PBL is based on the actually retrieved NO_2 profile. The point we make in comparing NO_2 VCD with OMI measurements (Fig. 11, Section 4.5) is that the NO_2 VCD obtained from azimuth distributions of NO_2 measured at 560nm wavelength represents an area that is more directly comparable to the area probed by the OMI pixel (top panel in Fig. 11). In order to see possible differences in the VCD assuming box-type profile we have performed an additional sensitivity exercise. In this exercise the NO_2 VCD in the azimuth standard view is calculated in two different ways: (1) using the NO2 VMR from the parameterization approach assuming a box-profile and the PBL estimated with the 1/2e decrease in NO_2 and (2) using the NO_2 vertical profile integrated on Fig. 6. As expected, the NO_2 VCD retrieved from 1 is overestimated by a factor of 15-20%. In this context, we have added an explanation of this possible overestimation in section 4.5:

"Note that even though the error of the OMI based NO_2 are quite large due to the single overpass the overestimation of the MAX-DOAS obtained in the UV is still significant. Further research should be done to investigate the azimuth distribution and wavelength dependence with long time series. It is interesting to note that the assumption made to calculate the azimuth NO_2 VCD using the azimuthal NO_2 VMR would be overestimated based on the NO_2 vertical profiles (Fig.6), which are not box profile type. To establish the possible overestimation we performed a sensitivity study where the NO_2 VCD in the standard azimuth view was calculated integrating the NO_2 vertical profiles and compare them with the assumption of using the MLH_{NO2} estimated previously. As expected, the NO_2 VCD is overestimated by a factor of 15-20% if we use the assumption of the MLH_{NO2}. In this context, the agreement of the NO_2 VCD at 560 nm would be even better and the overestimation of the 350, and 450 nm NO_2 VCD would be in the order of 35% and 25% respectively."

P. 11681, L. 10-11 "We present the first fast 2D-MAX-DOAS measurements…" Again, please be more careful and rephrase (e.g., find other fast 2D-MAX-DOAS instruments in Piters et al. 2012).

In the revised manuscript we have removed the word "first", although there is little detail given in Piters et al. to assess the 'speed' of other 2D MAX-DOAS instrument. The sentence reads as follow:

"The time resolution of our fast 2D-MAX-DOAS measurements (here 14 min) for a complete 360˚ azimuth view is deemed sufficient to document changes in the atmospheric distributions of NO_2, HCHO, CHOCHO that occur on 30 min or longer timescales. A retrieval strategy is presented that exploits the time consuming 2D-MAX-DOAS technique very efficiently. For future 2D-MAX-DOAS measurements we recommend the use of more than one azimuth view to estimate the MLH_{NO2} in several directions, and account for possible terrain effects."

Figure 1: The authors state that the fiber does not move, what is clear for the elevation due to the prism. But when changing the azimuth, does the fiber move? Is there an advantage of not moving the fiber (other than mechanical stress) because when taking a measurement the fiber does not move anyways?

We have modified the caption of Fig. 1 to make clear that the fiber does not move in any of the measurement modes. See our response to reviewer#1.

Figure 2: Why is there shown an example from a previous campaign? I would prefer an example from the campaign the paper is about.

The MAD-CAT campaign was not our first deployment of this instrument, and the most thorough characterization was done as part of a previous setup that is however identical to the one deployed during MAD-CAT. Given the method focus of this paper, we believe this section is well-suited for the presentation of the alignment procedure, and would like to keep the azimuth alignment as it currently is.

Figure 3: Why showing fit examples from 06 July 2013? All the study is about 17 June 2013, so please provide fit examples from this day.

Thanks for the suggestion. We have changed the fit examples from 17 June, 2013 in the revised manuscript.

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