Reply to RC2 by Referee #2

The authors would like to thank Referee #2 for the thorough review of their manuscript and the creative and helpful improvements. Below we reply to the raised issues one by one. Comments by referee #2 are printed in black, and the authors' replies in blue.

Please note, that during the review process, a new Figure and a new equation (Fig. 5 and eq. (15)) were added to the manuscript. All references to Figures or equations within this reply (except within direct quotes of sentences added to the revised version of the manuscript) refer to the **old** numbering, i.e. the one that the referees had seen when writing their reviews.

General evaluation

This manuscript introduces a novel measurement approach for the fast imaging of strong NO2 emission sources, such as stack plumes observed in power plants. The proposed NO2 camera is an application of the gas correlation spectroscopy, that was successfully used in the past e.g. for CO measurements in the infrared spectral range. The method is here extended to NO2 measurements in the visible spectral range. The study is largely exploratory in scope and concentrates on (1) a theoretical analysis of the measurement principle including estimates of the expected performances, (2) the verification of model predictions using a proof-of-concept instrument demonstrating the feasibility of the technique, (3) results from first measurements in the field at a large German power plant, and (4) comparisons with simultaneously recorded MAX-DOAS measurements. The proposed approach is very attractive since it is simple in concept and potentially inexpensive, which opens possibilities for future deployment at larger scale. In its current state, however, the system remains very experimental and a number of technical difficulties are still to be solved before such a camera can be ready for routine measurements in the field. Nevertheless I found the manuscript very interesting. The simple theoretical model is convincing and addresses several aspects of the measurements performances, such as sensitivity, selectivity, detection limit, etc. Model estimates are found to be in good agreement with actual measurements performed using the proof of concept instrument, which validates the approach. Also first measurements in the field show convincing results, for a stack plume of moderate strength. It also illustrates the main technical limitations of the current instrumental design. It also provides an interesting discussion on emission flux estimates performed using the camera, which arguably represents a promising application for future developments/applications. The last chapter on the comparison with MAX-DOAS measurements is however disappointing and somehow confusing. The authors struggle in a lengthy discussion to explain the potential reasons for a lack of agreement between both techniques, which result from a suboptimal operation of the DOAS system, a lack of time synchronisation and also calibration issues. In its current state, this comparison does not bring much to the study. I therefore strongly recommend to remove it and concentrate on an optimization of the experiment for a future publication. This reservation being made, I found the manuscript innovative, well written and definitely suitable for publication in AMT.

Detailed comments

Pg. 1, abstract: the first two sentences of the abstract could be omitted from the abstract. Such general information is generally provided in the introduction of the paper.

The two first sentences of the abstract have been removed.

Pg. 2, l. 54: I find the formulation “immanent asynchrony of the push-broom scheme” a little bit obscure in the present context. I suppose you mean that because of the need to scan in one spatial dimension, the information is recorded sequentially, which can lead to image deformation effects. Please confirm or clarify.

The interpretation of Referee #2 is correct. “immanent asynchrony of the push-broom scheme” means that when recording an image by scanning row by row, the individual rows composing the final image are asynchronous relative to each other.

The authors have changed the specified paragraph to:

Although modern hyperspectral cameras can reach adequate spatio-temporal resolution, some problems remain. Methods that rely on a push-broom scheme suffer from time delays between the rows (or columns) of the recorded images. Furthermore, spectrally resolving instruments are usually expensive and bulky.

Pg. 3, l. 74: the instrument concept requires that the NO2 contained in one of the cells remains stable during measurements. Doesn't this requirement imply that the cell temperature must be stabilized to maintain the NO2/N2O4 ratio at a constant value? This question should maybe be addressed in the section dealing with instrument model calculations and uncertainties of the method.

This is a valid point and was considered by the authors. When setting up the experiments, the instrument was left running for a while (10 – 30 minutes) so that thermal equilibrium could settle inside the instrument case.

Remaining variations of S_c principally have an influence on the measurement, because the instrument calibration depends on S_c. However, the correct calibration factor $k^{-1}(S_c)$ can be derived from S_c (see Fig. 4 for context), which again can be computed from each camera image, provided that an off-plume region with $S = 0$ is contained in it. For context, see eq. (25).

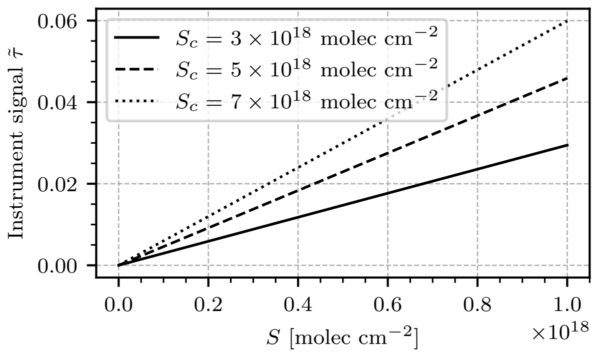
Following up to a comment in RC1, an additional explanatory paragraph regarding this procedure was placed in sec. 2.2:

During measurements, S_c must be determined so that $k^{-1}(S_c)$ can be computed. For this purpose, S_c could be directly measured using a second instrumental setup, such as a DOAS instrument. However, in many measuring scenarios it is more practical to determine S_c on the basis of the acquired images alone. For this purpose, an off-plume region of the imaged scene, where $S = 0$ is assumed, is used, and S_c is approximated by

$$S_c = \ln\left(J/J_c\right)/\overline{\sigma}$$

where $\overline{\sigma} \approx 5.1 \cdot 10^{-19}\ \mathrm{cm^2\ molec^{-1}}$ is the absorption cross section of $\mathrm{NO_2}$, averaged over the spectral range from 430 to 445 nm. The validity of this approximation was verified numerically, as displayed in Fig. 5. For a cell column density of $S_c = 4 \cdot 10^{18}\ \mathrm{molec\ cm^{-2}}$ (this value will be reasoned in the following paragraph), the proposed approximation underestimates the true value of S_c by less than $2 \cdot 10^{17}\ \mathrm{molec\ cm^{-2}}$.

along with a new Figure:



This should make it clear to the reader, that variations of S_c can be accounted for with sufficient accuracy.

Pg. 4, l. 93-96: are the units of radiances and irradiances important for this particular application? Certainly not for the measurement itself which is based on intensity ratios. But maybe this information is needed for the instrument model calculations. Please clarify whether absolute radiance values are used in this study.

Absolute radiances are used in the model calculations. In eq. (5), the radiance spectrum of the light source L_0 is a radiance spectrum in units W nm^{-1} m^{-2} sr^{-1}. L_0 is also a spectrum of absolute radiance values. The reason for that is that for the determination of the SNR, e.g. in eq. (16) and (17), terms arise that no longer only depend on signal ratios, but absolute camera signals (which depend on absolute incoming radiances). For example, increasing the incoming radiance by a factor of 100 increases the SNR by a factor of 10.

Pg. 4, l. 100: I suppose that the wavelength dependence of the quantum efficiency indicated here is a property of the silicium-based detectors used for the measurements, which explains the limited spectral range (UV-Vis-NIR). Note that the use of the sun as a light source also limits the applicable spectral range.

The authors agree, and the sentence

"The wavelength dependence of η typically restricts the integration to the near ultra violet (UV), the visible, and near infrared regions of the electromagnetic spectrum."

was changed to

"The wavelength dependence of η and the spectrum of the light source (typically scattered sunlight) usually restrict the integration to the near ultra violet (UV), the visible, and near infrared regions of the electromagnetic spectrum."

to reflect the Referee #2's comment.

Pg. 13, l. 269: the fact that the adjustment of the alignment of the two cameras is scene-dependent represents a major limitation for operation in the field. Can you further develop the reason why this is the case? I understood from the last sentence of the conclusions that the use of another instrumental design could solve this issue. It would be nice to introduce this possibility with a bit more details in the main part of the manuscript.

When the optical axes of the two cameras are not aligned, they will diverge significantly at long distances. For example, if the optical axes are shifted by just $0.1°$ ($0°$ would be perfectly parallel axes), an object in 2 km distance would appear shifted by $2\ \text{km} \cdot \sin(0.1°) \approx 3.5\ \text{m}$. Since eq. (12) computes the logarithmic ratio of the camera images, shifted structures are a source of strong false signals.

In theory, this problem could be solved once and for all by aligning the two axes perfectly and never touching the setup again. However, during transport of the instrument it is unavoidable, that the axes misalign slightly. Therefore, whenever the instrument arrives at the measurement site, the axes must be realigned.

Referee #2 has pointed out rightfully that the wording of this sentence is misleading and the corresponding sentence was changed from

"This adjustment is scene-dependent and of crucial importance in order to eliminate shifts in the FOVs of the two cameras"

to

"This adjustment is of crucial importance in order to eliminate shifts in the FOVs of the two cameras"

The alignment of the two cameras, only requires a few seconds using the thumb screws of the instrument and has not been regarded as a major limitation by the authors.

In l. 642, the idea of an instrument with a mutual optical setup for both channels and a beam splitter is briefly mentioned. Such an instrumental setup would have the potential to overcome shifts more easily, given that all light would be collected by a mutual lens.

Pg. 15, Table 1: Is there any particular reason why the uncertainty on the DOAS measurements of cell 2 so much larger than for other cells?

Please note that the uncertainty of cell 3 and cell 4 are larger than that of cell 2. The uncertainty is given by the uncertainty of the DOAS fit routine, but it may also vary from cell to cell, because the cells are of different size, have different glass thickness, etc.

Pg. 15, Fig. 9: this figure would gain being enlarged a little bit. Especially panel (a) is difficult to interpret.

The figure was enlarged as suggested.

Pg. 18, Fig. 12: again panels (a)(b) and (c) in this figure are very small and difficult to read. I suggest separating them from the two other panels and creating two separate figures. Since this figure shows the first illustration of an actual plume measurement with the camera, it deserves to be displayed in a more prominent way.

The panels (a), (b), and (c) of that Figure were enlarged but the authors decided to let them remain in a mutual Figure, given that panel (d) and (e) directly relate to them.

Pg. 19, l. 367: again these results demonstrate that the stability of the NO2 concentration in the reference cell is important, which suggests that an active stabilization of the temperature of the cell is needed (to constrain the NO2/N2O4) ratio.

This issue is resolved in the same manner as discussed in one of the previous points, regarding variations of S_c. When the measurement images are divided by the reference images taken against the blue sky (see eq. (12) for reference), and measurement image and reference image were recorded with different S_c, then a constant signal offset $\tilde{\tau}_0$ appears on the resulting signal image. This is corrected as explained in the paper and happens on an image-by-image basis. The variations that Referee #2 has mentioned here are fully corrected for.

Pg. 22, Fig. 22 and related discussion in pg. 23: the need to manually define the mask used to estimate the background 'out-of-plume' signal is also an important limiting factor for the technique. Do you see a possibility to overcome this difficulty either through an instrumental modification or by means of a more elaborated processing technique? If yes, it would be interesting to further discuss this question, maybe in a short section dedicated to perspectives for improvement of the technique.

This is a valid concern.

One solution would be to always use the "Full-FOV mask", see Fig. 16 (i). This eliminates a manual selection, but produces worse results. As Table 2 shows, the "Full-FOV mask" approach underestimates the NO2 SCD by approximately 25 %.

Another solution would be some form of automated plume detection. However, finding a general algorithm for such purposes is a very hard exercise. The authors would like to refrain from theorising much about this, but a short explanation was added to underline the importance of the plume mask to the whole evaluation:

"In the future, more elaborate methods for the separation of plume and background should be investigated. Generally, this would be achieved by image segmentation, for which a variety of methods exists. However, finding an ideal method that generalizes to other plume shapes and viewing geometries would require a study on its own."

Pg. 25, l. 465: at the end of the sentence, refer to section 4.2.4 where the question of the NO2/NOx ratio is explicitly analysed.

A sentence was appended, referring to the specified section:

"The NO_2/NO_x *ratio of the plume is further investigated in sec. 4.2.5."*

Pg. 28, section 4.2.5: as already pointed out in my general comments, I strongly recommend to remove this section from the paper. My feeling is that it brings confusion and does not help consolidating the measurements obtained with the camera. I would suggest replacing it by a small section outlining the possible improvements that can be envisaged for the instrument and eventually the data evaluation.

The authors have decided to remove the specified section.

Furthermore, the final part of sec. 5 now includes a brief listing of further ideas for future improvements:

"In the future, the following improvements to the instrument should be implemented: Firstly, the optical setup inside the instrument can be further optimized. By including a beam splitter, the light for both sensor arrays could be collected from a mutual lens, thus eliminating the need to correct for differences in the otherwise two lenses as a potential error source, especially the cumbersome background fitting routine described in sec. 4.2.1. Additionally, there exist camera modules with much lower read-out time than the ones used in our prototype, increasing the overall photon budget available for measurements. Secondly, the instrument would benefit from thermal stabilization in order to maintain a more stable NO_2 *column inside its gas cell. This way, the evaluation procedure would rely less on successfully determining* S_c *(see sec. 2.2) and* $\tilde{\tau}_0$ *(see sec. 4.2.1) from an off-plume region of the camera images. Thirdly, when measuring* NO_2 *emissions from a strong source as in sec. 4.2, the evaluation routine could be made significantly less ambiguous by implementing an automated*

image segmentation algorithm to separate the plume and off-plume regions of the individual images."

Pg. 34, l. 1: in fact, if I understand correctly, the camera was operated at a reduced resolution of 1300 x 600 pixels (accounting for the windowing applied to reduce the read-out time).

Correct, the camera was operated on a reduced resolution of 1350×600 pixels (equalling an extent of 1245 m width and 551 m height, as seen in Fig. 14). A sentence was added a few paragraphs later, where the limitations in framerate and resolution specific to the GKM measurement are listed:

"In order to increase the SNR of this measurement and smooth the plume signal, sequences of six images were averaged over, reducing the effective frame rate to 1/12 FPS and the resolution to 1350×600 *pixels."*

Pg. 34, 623: '... a detection limit of about 2e16 molec/cm2 is expected...'. Here I would add that this was confirmed by measurements using the proof-of-concept instrument.

The sentence was changed from

"Furthermore, under realistic conditions, a detection limit of about $2 \cdot 10^{16}$ molec cm^{-2} *is expected."*

to

"Furthermore, under realistic conditions, a detection limit of about $2 \cdot 10^{16}$ molec cm^{-2} *is expected, which was later confirmed using the instrument prototype."*

Spelling, typos

Pg. 2, l. 44: remove 'either'

The word "either" was removed.

Pg. 6, l. 128: change 'In reality this latter condition need not be perfectly filled' by 'In reality this latter condition does not need to be perfectly filled'

This was changed as suggested, but the word "filled" was exchanged by "fulfilled".

Pg. 7, l. 151: 'of' is duplicated between 'choice' and 'particular'

The obsolete "of" was removed.

Pg. 10, Fig. 5: there seems to be a confusion of the 'S' and 'Sc' notations in this figure. To my understanding, the x-axis of panel (a) should be labelled as 'Sc' as well as the legend of panel (b). Please check and adjust as needed.

The Figures are labelled as intended by the authors. It also seems that the panels are already labelled as suggested by Referee #2: The x-axis of panel (a) is labelled as S_c, as well as the entries in the legend of panel (b).

Pg. 10, Fig. 6: the species of which cross-sections are dispˆlayed do not show up properly in the legend where the applied scaling factors are given.

This issue was fixed.

Pg. 13, l. 258: I suppose that the sensor temperature is fixed at -50°C, and not +50°C as indicated here.

The temperature is not fixed at all. The overall temperature of the camera sensors depends on the heat produced by the units themselves and the ambient temperature, to which the cameras are exposed. Due to their small form factor, the camera modules indeed heat up to a temperature of around +50 °C. However, exposure times are so short, that dark signal can be neglected, even at such high temperatures (see also our replies to RC1).

Pg. 22, Fig. 16, end of first line: replace ‘The left two’ by ‘the two left’

This was changed as suggested.