Reply to RC1 by Emmanuel Dekemper

The authors would like to thank Emmanuel Dekemper 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 Emmanuel Dekemper 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.

The manuscript presents a new instrument for the ground based remote sensing of atmospheric NO2. The sensitivity to the target species is provided by the principle of the gas correlation spectroscopy (GCS), but the main objective of this instrument is to become a quantitative imager of 2D fields of NO2. This NO2 imaging capability introduces this new instrumental concept in the very small family of imaging instruments capable of retrieving quantitative information on UV-VIS species (SO2 cameras, AOTF-based NO2 camera, and I-DOAS instruments to my knowledge). As such, this manuscript bears a strong original content, and can be considered as the seminal paper for a new class of NO2 imaging instruments.

The paper is well structured: it first explains the principles of the GCS, then presents a prototype and some results obtained in the laboratory, followed by the analysis of data acquired during an outdoor campaign at a coal-firing power plant. The quality of the text is very good: descriptions are exhaustive, the English is of very good level, and most figures are clear. The reader will not only find all the needed details to fully grasp the instrument principle, and what it can do, but he will also discover a honest account of real-life conditions performance, which is very much appreciated.

In the following, I'm listing a number of questions/remarks on scientific aspects. This review ends with the few typos/technical issues that I could spot.

Specific comments/questions:

My comments follow the structure of the paper:

- Introduction: The first part of the section lacks a few references about the harmfulness of NO2. Also, I believe that NO2 exposure limits recommended by the WHO have recently been lowered for NO2. Please check this and update if needed. Also, it might be interesting to list the previous remote sensing instruments which have employed the GCS method. I know about the satellite instrument HALOE, but others, even ground based might have existed.

The section has been updated accordingly. The statement regarding the health risks of NO_2 exposure now has two references. The WHO recommendation for annual exposure was changed to the most recent value (2021) of 10 μg m^{-3}.

Three other applications of GCS methods were already listed where GCS is introduced in the introduction: Ward and Zwick (1975); Drummond et al. (1995); Wu et al. (2018). Each of the references deals with a different practical application of the method. A reference to the HALOE instrument (Baker et al. (1986)) was added.

- Section 2.1: In the beginning of the section it could be wise to simply remind that the model assumes that the pressure and temperature dependence of the cross-section is neglected. Also, I appreciate, in eq.(4) that the authors didn't overlook the temporal variability by applying an integration over time. But as no parameter is exhibiting any temporal dependence, I think it is ok to replace the integral by a product with the exposure time.

A sentence explaining that the pressure and temperature dependence of the absorption cross sections were ignored throughout section 2 was added:

"In the following, all absorption cross sections are considered constant, i.e. their slight dependence on pressure and temperature is neglected."

The specified integral can indeed be written as a product, please refer to eq. (13).

- Section 2.1, line 123 and below: You are rightfully stressing the delicate alignment of the two optical channels. Actually, as your processing algorithm contains a normalization of one image by the other, the consequence is that the derived quantity gets more sensitive to parameters which might not be as identical as you expect. For instance, the pixel response non-uniformity (PRNU) map of the two sensors could be quite different, and therefore not get properly cancelled by the normalization. Therefore I think it is good to acknowledge the potential differences across the two channels by adding a subscript in eq.(7)-(8) and further. In particular, I would add the subscript "c" to \eta and \mu in eq.(8). With this notation, the reader will be in a better position to appreciate the attempt of canceling the optical effects with the normalization performed in eq.(12).

The correction procedure used by the authors ("flat field correction", or FFC) is a well-established method of correcting non-homogeneities in images, which works based on dividing the measurement image by a reference image taken against a radiometrically uniform background. This procedure also accounts for fixed pattern noise in general, including PRNU by gain normalisation. The authors have clarified this by text, adding the sentence

"Even with entirely identical optical setups, the two camera sensors may have slightly different pixel response non-uniformity (PRNU) maps."

in order to clarify, that PRNU is corrected by the FFC.

The authors have decided to omit the proposed subscript c to η and μ. The reason is that the FFC is assumed to correct η and μ to the same values for both channels.

- Section 2.2, line 145: The assumed range of NO2 SCDs should be justified.

A sentence justifying the assumed range of NO_2 SCDs was added:

"The assumed range of NO_2 column densities is justified as follows: In order to measure column densities much lower than 10^{16} molec cm^{-2}, the exposure time would need to be increased significantly, resulting in poor temporal resolution. At the same time, even strong NO_2 pollutions in the atmosphere typically do not exceed 10^{18} molec cm^{-2}, assuming realistic viewing geometries."

- Section 2.2, line 161: From my own experience, relying on manufacturer data is risky. For quantitative measurements as the ones you intend to do, I would recommend to measure the detector parameters, and the optics transmittance by yourself, with the needed spectral resolution (which is very often very coarse in manufacturer's datasheet).

The authors agree in principle. However, the measurement method relies on narrowband spectral variations. Broadband transmissions, like that of the camera lens or the quantum efficiency mainly affect the SNR, but hardly the overall instrument response. Other instrument specific quantities (like the transmission spectrum of the bandpass filter) were validated by laboratory measurements and were found to agree with the vendor's information.

- Section 2.3: This section seems to be an attempt to get rid of the spectral integrals inherent to the method. In my opinion, the price to pay to achieve an analytical, integral-free expression for the effective optical thickness is too high. The reader can also be disturbed as the objective of this attempt is not fully clear. I would recommend to remove this section, in favor of stressing again that the measurements are not spectrally-resolved, and therefore a direct access to the target air mass SCD is not possible.

The purpose of this section is to present a purely analytical expression of the instrument response in a GCS-based measurement for arbitrary target gasses. The section is not highly relevant for the GCS-based NO_2 camera. However, for new GCS-based remote sensing methods it could be used for quick assessment of the instrument's capabilities because no knowledge of the spectral structure of the absorption cross section is required.

The authors have decided to move this section to the appendix (see Appendix A).

- Section 3, line 258: I was a bit surprised by the reported dark signals. It is quite strange to report values at temperatures well above the normal working conditions (e.g. 20°C). Also, the dark signal precision (24+/-9) is quite poor, much worse than the shot noise. This questions the accuracy of the dark current removal. But as the exposure times are orders of magnitude smaller than the second, I guess it is not really an issue. However, the text does not address the characterization of the ADC gain, which might be pixel-dependent, and detector-dependent. Perhaps providing the steps that you apply during the "L1" processing for the data would help the reader to understand exactly what is done.

The dark signal is listed at a temperature of 50 °C, because this is the average operating temperature of the two camera modules. This significant warm-up is due to the small form factor of the modules. The original sentence

"A thermal dark signal of (24 ± 9) e$^-$ s^{-1} at a sensor temperature of 50 °C and a doubling temperature of (6.1 ± 0.1) °C were found"

was changed to

"A thermal dark signal of (24 ± 9) e$^-$ s^{-1} at a sensor temperature of 50 °C, which is approximately the average operating temperature of the camera modules due to their small form factor, and a doubling temperature of (6.1 ± 0.1) °C were found".

Overall, the information on dark signal is included for the purpose of completeness and supposed to tell the reader, that a dark signal correction is obsolete. This is also stressed in the short example

"(. . .) the contribution of the dark signal to the total measured camera signal is negligibly small (e.g. below 0.05 % for an exposure time of 30 ms and a sensor saturation of 50 %)".

The gain of the camera modules is set to a fixed value. As explained earlier, non-uniformities of the ADC gain are assumed to be accounted for by the flat field correction.

An explanation of the "L1" processing of the data was added by the authors:

"From a technical perspective the retrieval of the camera data follows the typical pattern of digital imaging: Inside the camera modules, the incoming photons detach electrons from the semiconductor material of the camera chip (characterized by η). That charge is digitized (characterized by the fixed ADC gain K in units of [e$^-$ ph^{-1}]) and saved as 16-Bit grayscale image files.".

- Section 4.1, eq.(25): This equation is not correct, as the ratio of the J-terms, followed by the log does not remove the spectral integrals contained in each J-term. At best, you obtain a biased estimate of the column. The importance of this bias can be assessed with a numerical experiment. Negligible or not, this aspect should be addressed.

This point was rightfully raised. Indeed, the term in eq. (25) is only an approximation of the column inside the gas cell of the instrument. A numerical experiment was run to assess the quality of this approximation. A new figure:
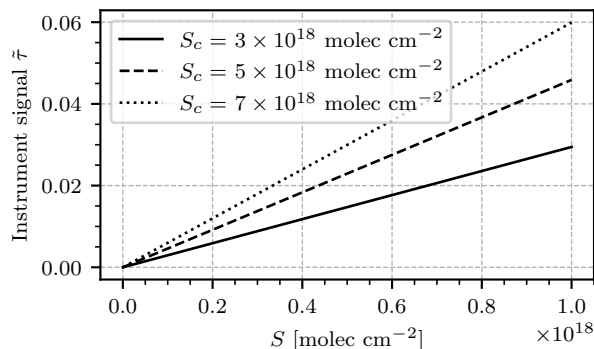
and an explanatory paragraph

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 $\overline{\sigma} \approx 5.1 \cdot 10^{-19}$ cm^2 molec^{-1} is the absorption cross section of 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 $S_c = 4 \cdot 10^{18}$ 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 $2 \cdot 10^{17}$ molec cm^{-2}.

was added.

- Section 4.2.1: The vertical gradient that you observe like in figure 12 (a) is intriguing. You argue that it could come from the inhomogeneity of the reference measurements, but this does not really hold as the term J_ref(i,j)/J_c,ref(i,j) largely cancels broad variations of the sky signal across the FOV. While you have assessed the false signal induced by H2O, and O4, you haven't addressed the impact of not modelling the Rayleigh and aerosol extinctions. As the spectral integration is not removed in eq.(12), it is not excluded that the extinction by the Rayleigh scattering, given its wavelength-dependence (15% decrease over the filter bandwidth) is not completely canceled in the normalization. Given that the air mass increases as the pointing elevation decreases, this could perhaps explain the vertical gradient you are observing...

The relevance of such broadband extinction can be assessed as follows:

Rayleigh scattering is known to have a λ^{-4} wavelength dependency. This would be the dominant modulation of the radiance spectrum if there were no further broadband extinction due to aerosols ("blue sky"). Realistically, the modulation due to broadband extinction can be assumed to modulate the radiance spectrum with some factor between λ^{-4} and λ^{-0}. The influence of broadband extinction was estimated by modelling the two most extreme cases, i.e., computing the calibration curve of the instrument (like in Fig. 4) for (i) an unweighted radiance spectrum and (ii) a radiance spectrum weighted with λ^{-4}.

This study has shown that the contribution of the broadband extinction affects the instrument response by less than 1 % and is thus considered negligible.

Two explanatory paragraphs:

Furthermore, eq. (12) points towards a crucial benefit of the proposed measurement principle. While other correlation methods for remote sensing typically operate with two channels in different spectral domains (e.g. an on- and an off band channel in filter spectroscopy based SO_2 cameras), the spectral domain of the two channels is identical in GCS. Additionally, that domain is typically restricted to a few dozen nanometers using a bandpass filter. This makes the instrument insensitive to broadband extinction, i.e. by Rayleigh scattering or due to aerosols, given that their extinction coefficients vary only very slightly throughout the spectral domain the instrument operates in. The instrument response to broadband extinction is examined numerically in sec. 2.2.

and

In addition to water vapour and O_4 the modelled instrument response to broadband extinction was investigated. Rayleigh scattering has a wavelength dependence of λ^{-4}, while extinction due to larger particles shows weaker wavelength dependence. It was verified in a numerical experiment, that the instrument response curves displayed in Fig. 4 vary by less than 0.5 % when the assumed irradiance spectra are scaled by λ^{-4} and $\lambda^0 = 1$ respectively. This demonstrates that the NO_2 camera is practically insensitive to broadband extinction.

The origin of these vertical gradients remains an open and important question, however, the background fitting method we propose seems to be able to correct them efficiently enough.

- Section 4.2.1: You are reporting a column of 2.72x10^18 in the cell, whereas it was explained that the best performance is achieved with a cell of 4x10^18. I also had understood that this was the value used in the

prototype. Why such a deviation with respect to your ideal case? Is it caused by using eq.(25)? Why didn't you estimate this value with the DOAS instrument?

A strong deviation of the column inside the cell was observed on multiple field trips, whenever the instrument was exposed to an environment that was usually much colder and/or brighter than the laboratory, where the prototype was built. The loss of NO_2 in the gas cell could be explained by the $NO_2 \leftrightarrow N_2O_4$ equilibrium. Additionally, the higher the irradiance, to which the gas cell is exposed, the higher the photolysis rate of the contained NO_2. It was verified after the field trip, that the NO_2 column inside the instrument's gas cell had recovered after some time in the laboratory. It was once planned to deliberately "overfill" the gas cell, so that it reaches the ideal column density when brought outside. However, this turned out to be very hard to achieve with the available NO_2 reservoirs, from which the gas cell was filled.

Measuring the column inside the cell requires complete disassembly of the instrument, which was considered unnecessary and too risky in the field, given that the column inside the cell can be computed with sufficient accuracy from the measurement images themselves. Furthermore, the high optical depth of NO_2 in the cell (on the order of unity) is not suitable for applying the DOAS technique in the blue spectral range.

- Section 4.2.1, figure 14: It is very difficult for the reader to clearly distinguish the plume from the background. Please consider a color scale which would help the reader to observe the abundance of NO2 in the plume.

A colormap was chosen to make the plume more pronounced. For consistency, the same colormap was applied to the corresponding figures of sec. 4.2.

- Section 4.2.2, 2nd paragraph: This paragraph spans 41 lines, and contains a very technical discussion. Please consider splitting in more paragraphs, or even better, divide in subsections. A subsection dedicated to your analysis of the influence of the choice of reference area would make sense for instance.

This section was divided by introducing a new subsection named "4.2.3 Uncertainty analysis".

- Section 4.2.5: The comparison with the MAX-DOAS measurements delivers results which are a bit deceiving, but it also seems like you attempted to compare data of very different temporal resolution. Can't you revisit your comparison in order to make sure that you have the highest temporal consistency? In other words, one would expect to have used a different set of camera images for comparison with each MAX-DOAS point. Working with a global average doesn't put you in the best conditions... In addition, you report an average plume signal of 1x10^16, which is below your detection limit. This is striking for the reader, and would deserve a little explanation.

Referee #2 has strongly urged to remove this section entirely, which the authors have agreed to. The reason a global average was used is that the plume signal is extremely weak compared to the detection limit of the instrument. Using a global average allows to just barely identify the plume. The approach suggested by Emmanuel Dekemper was originally attempted by the authors but delivered useless results.

Technical comments

- General comment about significant decimal: Everywhere in the text, values are reported with too many decimals compared to the uncertainty of the measurements. This starts already in the abstract, but affects the manuscript globally. This particularly concerns the columns in molec./cm^2, and also the mass fluxes.

The authors agree, that in the paper some results are given with too many decimal places. Those decimals have been reduced to a sensible number of digits, i.e. the instrument's spatial resolution is now given as 0.9 m × 0.9 m instead of 0.92 m × 0.92 m

This change was not applied to intermediate results, e.g. the contents of (and references to) Table 1 and Table 2, which are used for further computation and should remain untouched.

- Figures 5, 6, 7, 20: those figures have issues with labels or annotations.

This issue was fixed.

- L4: gasses -> gases

This was changed as suggested.

- L17: "momentary" ?

This word was considered obsolete and removed without replacement.

- L19: "...is highly portable and cost efficient at building..." -> "...is highly portable for building..."

This was changed as suggested.

- L28: add a comma after "time"

The comma was added as suggested.

- Figure 1: Are you sure that (a) is not the filled cell, and (b) the empty one? This is what I would have assumed based on the brownish color of cell (a)...

This observation is correct, and the caption of the figure was changed accordingly.

- L64: "Lamber-Beer" -> "Lambert-Beer"

This was changed as suggested.

- I found the unit "phe" quite uncommon. Why not using "e-" for representing photo-electrons?

The units were changed as suggested.

- L118: I would not speak of "spectral channels" when referring to the two optical channels of the NO2 camera.

The authors prefer to leave this unchanged. They acknowledge, that "spectral channel" may be an unconventional use of the word, however it bears a strong analogy to DOAS terminology. In DOAS, the SCD of the target trace gas is extracted from the information contained in the many spectral channels, by application of the Lambert-Beer law. We compute an SCD from two pieces of information, namely J and J_c (or four pieces, if the reference signals are considered), also by application of the Lambert-Beer law. What we call "spectral channel" is therefore the analogue to the "spectral channel" in DOAS.

The sentence:

"We refer to the two measurements $J_{(i,j)}$ and $J_{c,(i,j)}$ as spectral channels in analogy to the widely used DOAS terminology."

was added to make the intention of the authors clearer.

- L119: "... functions as a measure ..." -> I think it is more correct to say that it is a quantity exhibiting a monotonic sensitivity to S.

The authors prefer to leave this unchanged, given that "functions as a measure" portrays the purpose of computing $\tilde{\tau}$ more clearly to the reader.

- L143: add a comma after "For this"

The comma was added as suggested.

- L147: In don't think that t_exp has been defined before

t_{exp} is now defined under eq. (4), where it first appears.

- L151: double "of"

The obsolete "of" was removed.

- L365: remove the comma after "showed"

The obsolete comma was removed.

- L384: "two cameras inside of the instrument" -> "two cameras inside the instrument"

This was changed as suggested.

- L386: "These displacements manifest as strong..." -> "These displacements manifest themselves as strong..."

This was changed as suggested.

- L392: you introduce the subscript "i" for your columns, whereas I guess that everywhere before, "i" was a row index (like in matrix algebra convention)...

The row indices have been changed from i to j, which would be the standard matrix algebra convention for a column index.

- L619: this value of 1/2 FPS is new. I had 1/12 in mind from the paper. Please check

In l. 15, the paper says 1/12 FPS, but in l. 619, the paper says indeed 1/2 FPS. The reason is, that the single images of the GKM measurement were recorded at a frame rate of 1/2 FPS, but in order to evaluate them, six consecutive images were averaged. This reduces the effective framerate to 1/12 FPS. However, the instrument is in principle able to record with frame rates higher than 1/12 FPS (again, here it was demonstrated, that 1/2 FPS is technically possible). An explanatory sentence

"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"

was added.

- L623: Split the paragraph after "expected."

This was changed as suggested.

- L630: Split the paragraph after "18.2%."

This was changed as suggested.