

Atmos. Meas. Tech. Discuss., referee comment RC1  
<https://doi.org/10.5194/amt-2021-298-RC1>, 2021  
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## **Review of the GCS-based NO<sub>2</sub> camera by Kuhn et al.**

Emmanuel Dekemper (Referee)

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Referee comment on "The NO<sub>2</sub> camera based on gas correlation spectroscopy" by Leon Kuhn et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2021-298-RC1>, 2021

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### **General comments:**

The manuscript presents a new instrument for the ground based remote sensing of atmospheric NO<sub>2</sub>. 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 NO<sub>2</sub>. This NO<sub>2</sub> imaging capability introduces this new instrumental concept in the very small family of imaging instruments capable of retrieving quantitative information on UV-VIS species (SO<sub>2</sub> cameras, AOTF-based NO<sub>2</sub> 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 NO<sub>2</sub> 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 NO<sub>2</sub>. Also, I believe that NO<sub>2</sub> exposure limits recommended by the WHO have recently been lowered for NO<sub>2</sub>. 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.

- 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.

- 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).

- Section 2.2, line 145: The assumed range of NO<sub>2</sub> SCDs should be justified.

- 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).

- 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.

- 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.

- 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.

- 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 H<sub>2</sub>O, and O<sub>4</sub>, 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...

- Section 4.2.1: You are reporting a column of  $2.72 \times 10^{18}$  in the cell, whereas it was explained that the best performance is achieved with a cell of  $4 \times 10^{18}$ . 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?

- 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 NO<sub>2</sub> in the plume.

- 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.

- 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  $1 \times 10^{16}$ , which is below your detection limit. This is striking for the reader, and would deserve a little explanation.

## 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<sup>2</sup>, and also the mass fluxes.
- Figures 5, 6, 7, 20: those figures have issues with labels or annotations.
- L4: gasses -> gases
- L17: "momentary" ?
- L19: "...is highly portable and cost efficient at building..." -> "...is highly portable for building..."
- L28: add a comma after "time"
- 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)...
- L64: "Lamber-Beer" -> "Lambert-Beer"
- I found the unit "phe" quite uncommon. Why not using "e-" for representing photo-electrons?
- L118: I would not speak of "spectral channels" when referring to the two optical channels of the NO<sub>2</sub> camera.
- L119: "... functions as a measure ..." -> I think it is more correct to say that it is a quantity exhibiting a monotonic sensitivity to S.
- L143: add a comma after "For this"
- L147: In don't think that t<sub>exp</sub> has been defined before
- L151: double "of"
- L365: remove the comma after "showed"
- L384: "two cameras inside of the instrument" -> "two cameras inside the instrument"
- L386: "These displacements manifest as strong..." -> "These displacements manifest themselves as strong..."
- 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)...
- L619: this value of 1/2 FPS is new. I had 1/12 in mind from the paper. Please check
- L623: Split the paragraph after "expected."
- L630: Split the paragraph after "18.2%."