Atmospheric Measurement Techniques Discussions

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Review of the GCS-based NO2 camera manuscript by Kuhn et al.

Anonymous Referee #2

Referee comment on "The NO_2 camera based on gas correlation spectroscopy" by Leon Kuhn et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-298-RC2, 2021

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.

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.

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.

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.

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.

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.

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?

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

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.

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.

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.

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.

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.

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).

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.

Spelling, typos:

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

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'

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

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.

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

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

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