RESPONSES TO REVIEWERS

<u>Ms. Ref. No.</u>: Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-252. <u>Title</u>: Discrete-wavelength DOAS NO₂ slant column retrievals from OMI and TROPOMI

Response to Anonymous Referee #2

This manuscript presents a simplified NO₂ slant column retrieval approach, which makes use of a limited number of wavelengths in an otherwise classical DOAS retrieval framework. The approach is tested on sample data from the OMI and TROPOMI sensors, and results are discussed in terms of their consistency with standard retrievals. It is concluded that retrievals based on strongly sub-sampled spectra (only 10 wavelengths are used) still provide good NO₂ slant columns. Although this result is not surprising as such (given the high quality of the original measurements), the small reduction of the noise on the retrieved slant columns is in my view a bit unexpected and worth pointing out (and possibly explain). So we come with the conclusion that retrieving NO₂ columns from 10 spectral points is feasible.

There are however a number of drawbacks and limitations in doing so, and one may wonder whether the potential advantages of reducing the spectral information would actually compensate these drawbacks. The fundamental motivation behind the study relies on the postulate that reducing the spectral information would allow to simplify instrumental design (of future satellite missions) leading to potentially improved spatial resolution at low cost. Statements along these lines are given at several places in the manuscript, but without any further elaboration, e.g. what kind of instrumental solution could be adopted? More importantly, key requirements on spectral accuracy and stability that would need to be considered for such a design are not mentioned at all. It is basically assumed that spectral performances equivalent to those of OMI and TROPOMI can "easily" be obtained with low-cost imaging systems suitable for integration on small satellites. To my opinion, the lack of such a discussion significantly limits the relevance and impact of the study. I therefore recommend publication only if these questions are better addressed in a major revision of the manuscript.

Thank you for your comment. Our intention was to demonstrate that the concept of retrieving NO₂ using only 10 discrete wavelengths is feasible and that accuracy comparable to existing level 2 NO₂ products can theoretically be achieved. We absolutely agree that in practice the concept is more complex and there are implementation challenges that must be overcome, e.g. co-registration of the spectral bands. While the study was deliberately discussed in generic terms, i.e. independent of any specific instrument solution, we acknowledge that the manuscript would benefit from more discussion about implementation challenges and potential instrument solutions. We have now added such discussion in paragraph 3 of the conclusions. Regarding the key requirements on spectral stability and accuracy, those are the focus of the next study, which we are currently working on to derive such requirements from sensitivity analyses.

Specific comments

Pg. 2, line 25: not all sources of tropospheric NO₂ are anthropogenic in nature. Please complete.

We did not intend to suggest that all sources of tropospheric NO₂ are anthropogenic, but rather that anthropogenic emissions are the main contributor. We have clarified this in the text and completed the statement with examples of natural sources of tropospheric NO₂.

Pg. 2, line 43: in addition to in-situ and satellite techniques, also ground-based remote sensing constitute a key component of the atmospheric composition monitoring system. This includes e.g. the Network for the Detection of Atmospheric Composition Change (NDACC) or the emerging Pandonia/PGN network.

Thanks for pointing this out, also highlighted by Anonymous Referee #1. We have now mentioned the ground-based remote sensing technique and given the suggested examples.

Pg. 2, line 48: current satellite instruments are limited in resolution, but TROPOMI is already doing much better than OMI. This should already be mentioned here, with a mention that ultimate resolutions in the range of 1x1 km² are needed to allow for individual source identification.

Thanks for your comment. We now give the example of TROPOMI instead of OMI, and mention the spatial resolution requirement for point source identification.

Pg. 2, line 56: The current resolution of TROPOMI at true nadir is 3.5 x 5.5 km².

We have now clarified that the stated resolution is at true nadir.

Pg. 3, line 71: the Brewer instrument is cited here as an example for a NO_2 measuring system based on a few wavelengths; however it is well-known that Brewer NO_2 measurements are dramatically lacking sensitivity. This was actually at the origin of the development of the Pandora instrument, which uses simple (low-cost) grating spectrometers to (strongly) improve the quality of NO_2 column measurements.

Thanks for pointing this out. We have added the lack of sensitivity as another drawback of the Brewer spectrometer.

Pg. 3, line 75: what are the "specific viewing geometries" that prevent usage of the NO_2 camera for space applications? Please clarify.

The algorithm used in the AOTF-based NO₂ camera as described in Dekemper et al. (2016) relies on clear-sky pixels being present in the scene for background subtraction. In addition, the sequential sampling of wavelengths poses a limitation to the speed at which they can be registered, making the

retrieval challenging for non-static scenes. These drawbacks make the NO₂ camera unsuitable for nadir-viewing space applications. We have clarified this in the text.

Pg. 4, line 116: describe in short the interpolation method used by Bucsela, and its added value for this study

The method used by Bucsela et al. (2006) calculates the interpolated spectrum using the high-resolution solar reference spectrum as follows:

$$F(\lambda + d\lambda) = \frac{F(\lambda)}{[F_0(\lambda + d\lambda)/F_0(\lambda)]}$$

Where *F* is the measured spectrum, F_0 is the solar reference spectrum, λ is the original wavelength grid of *F*, and $\lambda + d\lambda$ is the new wavelength grid. In Bucsela et al. (2006) the irradiance spectrum is interpolated onto the radiance wavelength grid, whereas in our work we interpolate the radiance onto the irradiance wavelength grid to match what is done for the OMI and TROPOMI L2 products.

This method is an improvement over other approaches (e.g. linear or spline) as it reduces interpolation errors related to the sampling rate. However, this improvement is not expected to be significant for instruments like OMI and TROPOMI where undersampling is not a problem. We have updated the text with this clarification.

Pg. 5, line 123: this introductory paragraph is a bit misleading. To my understanding the critical aspect of selecting appropriate spectral channels for NO_2 fitting is not related to the complexity of the radiative transport, but only to the nature of the differential cross-sections and the presence of interfering species.

We agree with your assessment in the case of traditional DOAS NO₂ fits. However, discrete-wavelength DOAS is more sensitive to scattering and albedo effects than traditional DOAS because the polynomial models the broadband component of the reflectance less accurately. This is why the fitting interval must be narrow enough to minimise the effect of the broadband component. The cross sections and the interfering species are still key aspects but in the case of discrete-wavelength DOAS the complexity of the radiative transport also plays an important role. Nonetheless, we acknowledge that the paragraph is misleading and have updated it.

Pg. 5, line 131: replace "mean optical depth" by "differential optical depth" (or difference in optical depth)

Corrected to "differential optical depth".

Pg. 6, Figure 2: how important is it to include liquid water cross-sections in the fitting? In the spectral range of interest, this cross-sections seem to be very unstructured and may correlate strongly with the polynomial function.

We did some tests and concluded that including the liquid water cross section does not make much difference. However, we included it in the fit to match the reference retrieval settings as closely as possible.

Pg. 6, line 144 (very minor comment): the choice of "discrete wavelength DOAS" as a name could in fact be questioned, since fundamentally all DOAS schemes use discrete wavelengths (it is just that in your case, their number is smaller)

We agree, good point. We considered different names and concluded that "discrete-wavelength DOAS" was the one that best described the retrieval approach while still being clear and short. Nonetheless, we welcome suggestions for alternative names that might be more suitable.

Pg. 9, line 199: how can local variations in surface albedo explain differences between retrievals from same satellite pixels? Please clarify the meaning of this statement.

As we discuss in a previous comment, discrete-wavelength DOAS is more sensitive to albedo effects than traditional DOAS. Therefore, we think that one possible explanation for the bigger retrieval differences in the smaller pixels might be related to the different sub-pixel variability of the albedo due to the size of the pixel. In other words, we would generally expect less albedo variability within smaller pixels and this might mean that some stronger spectral features might be present compared to the bigger pixels. These strong spectral features would result in higher retrieval differences between DW-DOAS and the reference level 2 products. We have clarified this in the text.

Pg. 13, Figure 5: why such a discontinuity in the NO₂ map of 30 Oct 2005 (at 20°S)?

This discontinuity is also present in the reference QA4ECV NO₂ Level 2 product (see <u>http://temis.nl/airpollution/no2col/no2regioomi_qa.php?Region=9&Year=2005&Month=10&Day=30</u>). We don't exactly know what causes it but it is not present in the tropospheric NO₂ column map, so it is likely a combination of stratospheric NO₂ and processes/elements involved in the air mass factor (AMF) calculation such as atmospheric scattering.

References:

Bucsela, E., Celarier, E., Wenig, M., Gleason, J., Veefkind, J., Boersma, K., and Brinksma, E.: Algorithm for NO₂ vertical column retrieval from the ozone monitoring instrument, IEEE Transactions on Geoscience and Remote Sensing, 44, 1245, <u>https://doi.org/10.1109/TGRS.2005.863715</u>, 2006.

Dekemper, E., Vanhamel, J., Opstal, B. V., and Fussen, D.: The AOTF-based NO₂ camera, Atmospheric Measurement Techniques, 9, 6025–6034, <u>https://doi.org/10.5194/amt-9-6025-2016</u>, 2016.