

We would like to thank the reviewer for their constructive comments. We have amended the manuscript to account for the technical suggestions, the most serious of which are discussed herein. We have added a section to the paper addressing the limitations to operationalization for this technique which have been identified by both the reviewers.

The major drawback of the presented method comes from the difficulty in the selection of an appropriate reference sector: can this selection be done in an automated way based on pre-defined criteria for the location of the reference sector(s) and the level of NO₂ the latter can contain (e.g. how to automatically reject reference sectors contaminated by pollution transport events ?) ?

We agree that the reference sector selection can be a significant source of uncertainty in this retrieval, particularly due to potential tropospheric contamination. The reference sector selection could potentially be improved through a priori information provided by a chemical transport model (CTM), which would indicate regions where high pollution would be expected. A similar approach was applied in the stratosphere-troposphere separation performed by previous iterations of the OMNO₂ algorithm (Bucsela et al, 2006), in which the stratospheric VCD was inferred from regions where the GEOS-CHEM predicted low annual tropospheric NO₂ columns.

Identifying tropospheric contamination from pollution transport events would admittedly be more difficult. One possible solution would be to preliminarily select a small longitude range in the Pacific as the reference sector and then use the reference spectra to fit SCDs over the rest of the Pacific. While decreased in magnitude, contamination from transport events should still be observable. Through iterating the longitudinal range and masking problematic regions, it should be possible to identify the optimum reference sector for a global retrieval.

A discussion of this has been included in the new section on operationalization.

Another point is the time difference between measurements in the reference sector and in other regions: should a reference sector be defined on daily, weekly, monthly or yearly basis? In the case of monthly or yearly selection, what is the impacts of the stratospheric NO₂ seasonality and possible instrumental degradation on the retrieved tropospheric NO₂ columns?

Figure 13 shows that the SCD retrieval uncertainty has a seasonal variation of $\sim 1 \times 10^{14}$ molec cm⁻², which provides an estimate of the potential uncertainty resulting from using an annual reference. This uncertainty will be exacerbated by the seasonal variation in the stratospheric NO₂ column observed over the Pacific, which makes using an annual reference unlikely to offer any significant advantage.

For reference spectra derived over shorter timescales the number of cloud-free scenes available in the reference sector is a contributing factor, which may only be partially mitigated by improving the instrument spatial resolution. For regions such as the Intertropical Convergence Zone (ITCZ) and high latitudes, excessive cloud cover means that a daily reference cannot always be taken. For such regions it may be prudent to use a several-day average spectrum to compensate for this effect.

Because of these factors it is likely that using daily-to-weekly reference spectra would minimise retrieval uncertainty caused by the temporal frequency of the reference measurements. For a single instrument, a daily reference would be the best possible choice, while using a constellation of

instruments would also help to account for the diurnal cycle as well. These points have been added to the limitations section in the paper.

How to deal with cloudy scenes?

Spectra from ground pixels exceeding a certain cloud fraction threshold cannot be used to form the reference spectra, as the cloud top height introduces a potential bias in the assumed stratospheric column. Spectra measured over cloudy areas would also need to be robustly calibrated in order to avoid spectral shifts arising from inhomogeneous entrance slit illumination (Voors et al, 2006).

For cloudy ground pixels observed elsewhere the SCDs would need to be corrected through computation of an appropriate air mass factor (AMF), similar to those computed for existing retrievals of NO₂ (e.g. Boersma et al, 2004). The AMF would incorporate forward model parameters such as cloud fraction, pressure, and albedo; many of these can be derived using the O₂-O₂ column fitted in the DOAS fit (e.g. Accareta et al, 2004) through analysis of the 477 nm absorption peak. These points have been added to the limitations section in the paper.

References:

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