



Improving satellite retrievals of large tropospheric NO₂ columns

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Introduction

- NO₂ is one of the most important satellite data products
- analysis is performed using the Differential Optical Absorption Spectroscopy method (DOAS)
- in most situations, NO₂ is a weak absorber and the approximations made in DOAS are valid
- under very polluted conditions, NO₂ absorption can become significant (> 5%)
- here, we evaluate some effects present at very large NO₂ columns with respect to their impact on NO₂ fitting quality and the possible use as information source

Column dependent air mass factor

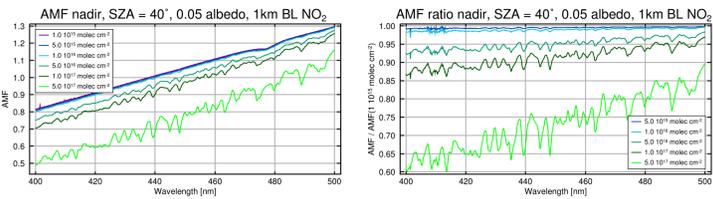


Figure 1: Wavelength dependent NO₂ air mass factors computed for different NO₂ vertical columns in a 1 km thick layer at the surface. Left: absolute values, Right: Change relative to the AMF having the smallest NO₂ a priori

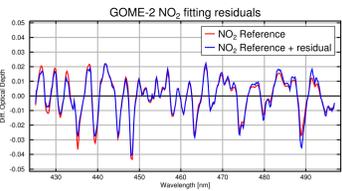


Figure 2: Example of a GOME-2 NO₂ fit result over China for a very large NO₂ slant column. The mismatch in the wavelength dependence of the NO₂ differential absorption amplitude resulting from the change in AMF over the fitting window is evident

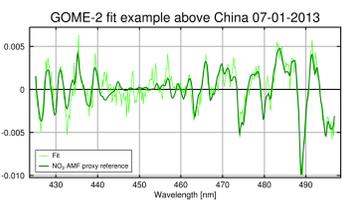


Figure 3: Example of a GOME-2 fit result for the AMF proxy created by linearly scaling the NO₂ cross-section and then orthogonalising it to the original cross-section. The same ground pixel as shown in Fig. 2 is used.

- for an optically thin atmosphere, the air mass factor does not depend on the column amount of the absorber of interest
- in addition, the air mass factor does only smoothly vary with wavelength (wavelength dependence of scattering and surface reflectance)
- in the presence of large amounts of NO₂, the AMF decreases with increasing NO₂ column which will result in wrong vertical columns if not accounted for
- in addition, spectral structures of NO₂ appear in the AMF, further interfering with the NO₂ retrieval
- this needs to be corrected in the AMF application (not further discussed here)
- at strong NO₂ absorption, the wavelength dependence of the AMF (which is always present) will lead to poor fits (see Fig. 2)
- the effect can be compensated by using an additional NO₂ cross-section, scaled linearly in wavelength and orthogonalised to the original NO₂ cross-section
- this AMF proxy can be retrieved from GOME-2 data over polluted regions (see Fig. 3)

Acknowledgements

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- GOME-2 lv1 data were provided by EUMETSAT

Temperature dependent cross-section

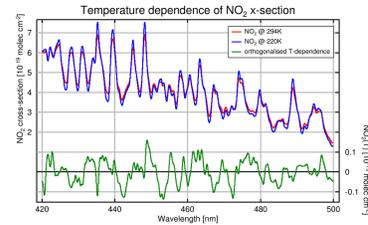


Figure 4: Temperature dependence of the NO₂ cross-section at spectral resolution of GOME-2. The green curve is the orthogonalised differential temperature dependence which can be included in the fit as additional absorber.

- NO₂ absorption cross-section depends on temperature
- effect is relatively large (up to 30%) but mainly a linear scaling
- current retrievals use a low temperature cross-section and correct for the temperature effect a posteriori in the AMF application
- at very large NO₂ columns, the difference in cross-sections can lead to poorer fits
- at least in principle, it can provide information for the separation of BL and stratospheric NO₂

I₀ correction

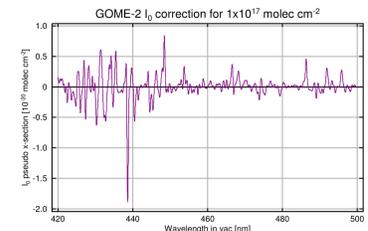


Figure 5: I₀ correction for NO₂ at a slant column of 1x10¹⁷ molec cm⁻² and for GOME-2 spectral resolution. The correction has been calculated by simulating the effect of exchanging convolution and absorption

- in the DOAS method, measurements are usually performed at intermediate spectral resolution (0.1 - 1 nm) which do not fully resolve the structures of the solar spectrum
- absorption in the atmosphere is approximated by using convolved cross-sections and the measured (convolved) intensities instead of treating absorption at high spectral resolution and convolving the results
- for strong and structured absorptions, this can lead to systematic residuals (I₀) effect

Application to GOME-2 data

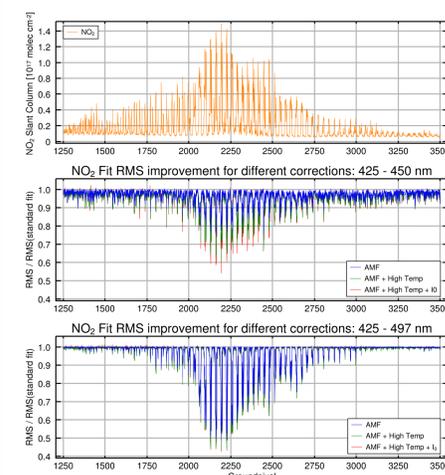


Figure 6: Small section of one GOME-2A orbit over China on January 7, 2013. Top: NO₂ slant columns, Middle: relative change in fit residual when successively applying correction for wavelength dependence of AMF, temperature and I₀ effect in the 425 - 450 nm window. Bottom: As middle but for 425 - 497 nm

- NO₂ slant columns over China can become as large as 1x10¹⁷ molec cm⁻²
- three corrections have been tested on GOME-2 data
 - an NO₂ AMF proxy
 - an NO₂ temperature correction
 - an I₀ correction assuming an SC of 1x10¹⁷ molec cm⁻²
- 425 - 450 nm window:
 - clear improvement of fitting RMS when including corrections
 - AMF and T-effects of similar size, I₀ less important
- 425 - 497 nm window:
 - even larger improvement
 - AMF effect dominates
 - I₀ effect not important (less NO₂ structures and smaller absorption)

NO₂ layer height and AMF proxy

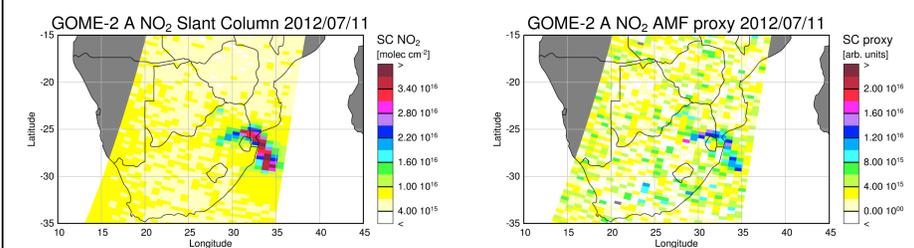


Fig. 8: NO₂ slant columns (left) and AMF proxy fitting coefficient (right) for a large NO₂ plume

Application of the AMF proxy to GOME-2 data shows

- clear signatures of the AMF proxy over all major pollution hot-spots (China, Europe, US, large cities) in monthly averages
- in daily values, the scatter is large outside of very polluted scenes
- for cloudy scenes, the AMF proxy is not found even if large NO₂ columns are present as expected from radiative transfer considerations
- there appears to be an interference over clear water bodies

A case study over South Africa (Fig. 8) shows that

- the AMF proxy tracks the NO₂ plume
- highest NO₂ SCs are found at the end of the plume (elevated NO₂)
- highest AMF proxy values are found close to emission point (NO₂ close to surface)

Conclusions

- in very polluted situations in China, satellite slant columns of NO₂ can get as large as 1x10¹⁷ molec cm⁻²
- under these circumstances, the AMF is no longer independent of NO₂ column which has to be taken into account for the calculation of vertical columns
- because of the large (> 5%) NO₂ absorption, several effects can deteriorate the spectral fit:
 - the wavelength dependence of the AMF
 - the temperature dependence of the NO₂ cross-section
 - the I₀ effect
- All effects can be corrected, reducing NO₂ fitting RMS by up to a factor of 2
- the NO₂ AMF proxy has the potential to provide information on the vertical position of an NO₂ layer

Selected references

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