

Anonymous Referee #2:

We thank the reviewer for his useful and constructive remarks. As described below, we have modified the manuscript as suggested and clarified the text where necessary. We hope that the revised manuscript has improved in respect to the original paper. Please find a rebuttal against each point below.

1)

Review of Tropospheric nitrogen dioxide column retrieval from ground-based zenith-sky DOAS observations Tack et al.

This paper is describing a new algorithm for measuring tropospheric NO₂. The paper contains strong emphasis on the error analysis. This emphasis is negated by the use of a scalar radiative transfer program instead of a vector program for sky radiances. The authors need to demonstrate that the scalar errors are much smaller (doubtful) than the measurement errors described.

Below is a partial review pending a discussion of the use of a scalar radiative transfer calculation. In my opinion, the measurement portion of this paper is good, but the analysis using LIDORT should be justified.

A scalar RTM (LIDORT) has been used here instead of a full vector RTM including polarization such as VLIDORT (Spurr, 2006). The main reason is that past studies have demonstrated that the light path length (and therefore the AMF, since for AMF only light path length matters) in an optically thin atmosphere is weakly affected by polarization. This is in contrast to total intensities which are more affected by these effects. For instance, Clémer et al. (2010) showed that neglecting polarization can give rise to a systematic error of up to 15% for the intensities while the impact on slant columns is significantly less (5%). This was very recently confirmed by the work of Hilboll et al. on *the influence of polarization on box air mass factor for UV/vis nadir satellite observations*, recently presented at the EGU 2015 General Assembly. According to this study, not accounting for polarization effects in the AMF calculation leads to systematic underestimation of NO₂ tropospheric VCDs of about 1-2% for Europe and China. A 1-5% uncertainty due to polarization is therefore small compared to our total error budget which is of about 30% or even more in the case of low tropospheric VCD values. A more practical reason why we used the scalar LIDORT RTM and not its vector version VLIDORT is that our interface for AMF calculation and profile retrieval (bePRO) is currently based on the scalar LIDORT RTM. Coupling bePRO to VLIDORT would require significant additional work which is beyond the scope of the present study. Moreover using VLIDORT would strongly increase the computing time, which is an important issue when time-series of several years have to be processed. Nevertheless the possibility of coupling bePRO to both LIDORT and VLIDORT is something we plan in the future and sensitivity tests on the polarization effects will be then performed.

In the revised manuscript, this point is clarified in section 3.2.2 (P9 L15):

It should be noted that in this study, polarization is not taken into account by means of a full vector RTM, such as e.g. VLIDORT (Spurr, 2006). The main reason is that past studies have demonstrated that the light path length (and therefore the AMF, since for AMF only light path length matters) in an optically thin atmosphere is weakly affected by polarization, in contrast to total intensities which are more affected by these effects. For instance, Clémer et al. (2010) showed that neglecting polarization can give rise to a systematic error of up to 15% for the intensities while the impact on the slant columns is significantly less (5%).

2)

P938 L4 In the case of GB ZS-DOAS <change to> In the case of the GB ZS-DOAS

Corrected.

3)

P938 L14 Since more than three decades -? For more than three decades

Corrected.

4)

P938 L23 increasing therefore the <change to> increasing, therefore, the

Corrected.

5)

P940 L5 10 June to the 21 July 2009 <change to> 10 June to 21 July 2009

Corrected.

6)

P940 L23 Gaussian? Did you measure this or assume the shape?

The spectral characteristics of the instrument have been fully characterized in the lab. In particular the instrument slit function which is close to a Gaussian has been determined using a monochromatic emission light source (HgCd).

Clarified in Section 2 of the revised manuscript (P5 L10):

The Gaussian shaped ***instrument's slit function, which has been determined using a monochromatic emission light source (HgCd), has a spectral resolution of 0.4 nm full width at half maximum (FWHM) and 0.9 nm FWHM for the UV and visible channels, respectively.***

7)

P941 L4 The configuration of the instrument allows to <change to> The configuration of the instrument permits measurement of

Corrected.

8)

P942 L11 is given here while the <change to> is given here, while the

Corrected.

9)

P942 L9 Is the use of QDOAS the source of the Gaussian slit function?

In QDOAS different analytical functions can be used to approximate the instrumental slit function (eg Gaussian, Lorentzian, Voigt, asymmetrical Gaussian). A Gaussian function was used here, as this was found to approximate at best the instrumental slit function measured in the lab (see remark 6).

P942 L16 They account for respectively the <change to> They account for, respectively, The

Corrected.

11)

P942 L25 expressed as SD <change to> expressed as standard deviation SD

Corrected.

12)

P943 L19 What is the effect of using a scalar RT that neglects polarization? The slant optical depth should be in error.

Please see our answer to remark 1.

13)

P946 L5 LIDORT is a scalar radiative transfer program, so it has the same problem

Please see our answer to remark 1.

14)

P948 L7 the opposite of the y-intercept <change to> the negative of the y-intercept

Corrected.

15)

P949 L1 The photochemical model has its own unquantified uncertainties. For polluted areas, the difference between 6.2×10^{15} and the diurnal variation is small compared to the tropospheric values. The value 6.2×10^{15} seem high for extrapolation to zero AMF. The authors need to comment on this.

It is important to note that the value 6.2×10^{15} for the RSCD is a **total slant** column while the values of the diurnal variation in Fig.4 are **stratospheric vertical** columns. Maybe we are wrong as the remark is not completely clear to us, but it seems that Anonymous Referee #2 has mixed up total SCDs with stratospheric VCDs.

The uncertainty on the simulation of the NO₂ diurnal cycle by the photochemical model and the assimilation with the retrieved stratospheric VCDs at sunrise and sunset is estimated at 20% and is discussed at P954 L28.

16)

P952 L22 troposphere is decreasing fast <replace with> troposphere is decreasing Rapidly

Corrected.

References

Clémer, K., Van Roozendaal, M., Fayt, C., Hendrick, F., Hermans, C., Pinardi, G., Spurr, R., Wang, P., and De Mazière, M.: Multiple wavelength retrieval of tropospheric aerosol optical properties from MAXDOAS measurements in Beijing, Atmos. Meas. Tech., 3, 863–878, doi:10.5194/amt-3-863-2010, 2010.

Spurr, R.: VLIDORT: A linearized pseudo-spherical vector discrete ordinate radiative transfer code for forward model and retrieval studies in multilayer multiple scattering media, J. Quant. Spectrosc. Radiat. Transfer, 102, 316–342, 2006.