

We thank the referees for their comments and suggestions and the careful reviewing of our manuscript. They have helped us to improve greatly our draft. We have followed the most of their suggestions and we have changed the text of the manuscript accordingly for a better understanding of our work.

Our reply to your questions/comments is in blue.

Referee #1,

General comments:

This paper describes a method to derive average free tropospheric concentrations of NO₂ and Ozone from a passive DOAS instrument and validates the results with independent measurements. This technique can also be applied for other species measureable by DOAS. I suggest publication only after major revisions to the introduction/method section since there are significant shortfalls in the author's rationale. I do not doubt the applicability of the described method. However, I don't think that a single scattering radiative transfer code (p. 8242, ll. 4-20) can be used to confirm that a single scattering geometric approximation is valid. I'm certain that the single scattering assumption is a valid one here because of the fact that those measurements are comparable to airborne MAX-DOAS measurements and studies have been published comparing multiple-scattering radiative transfer calculations with geometric approximations (e.g. Baidar et al. (2013) though for nadir column measurements only).

In the text it is stated that the method is valid on stations where the aerosols concentration is low (page 8250, line 9). Free troposphere stations are the case. In particular, Izana AOD, out of Saharan intrusion days, is below 0.05 and typically 0.02. Under these conditions we believe that the single scattering approximation is appropriate. The decision is supported by the similar results obtained when using the O4-MGA method, in which optical paths are directly measured without any scattering assumptions,

The method is based on the simple assumption that when evaluating a spectrum in the horizontal direction (elevation=0°) using the zenith spectrum as reference, the vertical components cancelled out and only the horizontal contribution remains. We have tried to clarify this point in the corrected manuscript.

In view of this, the lengthy discussion about ground-based geometrical approaches (p. 8237, l. 27- p. 8238, l.18) seems to be out of place since these measurements are always much more affected by a multiple scattering regime than measurements at a higher altitude in the atmosphere and hence a geometric approach in these cases yields poor quality results.

Our method is an adaptation of something that has previously been published and we believe that credit and description to previous works must be provided. In addition, we find this background useful for readers who are not familiar with these techniques.

I would suggest to discussing your method in comparison to airborne MAX-DOAS methods.

We understand the reviewer interest on comparison with airborne MAXDOAS since the geometry can be much the same, but we prefer in this first publication to limit the comparison to the “in situ” instrumentation. The observed differential absorption is not large and major effort has been carried out to ensure a great thermal stability (spectral drift <0.02 pixels) and minimize the detector noise. However, we will take into account the reviewer suggestion for a next future comparison.

My second major criticism is that this paper does not include any error estimates.

The following paragraph has been included in the text:

Even though the differential absorption signal between the zenith and the horizon is small in the unpolluted free troposphere, measurements are of good quality. Instrumental detection limit as estimated by Stutz & Platt (2008) is of 4×10^{13} and 8×10^{16} molecules.cm⁻² for NO₂ and O₃, respectively, for the typical root mean square error of the residuals of 2.5×10^{-4} DOD. The contribution of the fitting error in O₄ is negligible (0.1%). Major uncertainties come from calculation of the optical path since the absolute accuracy of the O₄ cross-sections is uncertain. (Wagner et al., 2002, 2009, Clemer et al 2010).

More specific comments/suggestions:

* The pronounced diurnal cycle in the DOAS measurements for NO₂ as well as for ozone (Fig. 7,9) could probably be explained if you reproduce Fig. 5 for a SZA of 85deg. Another guess would be that the single scattering approximation is not valid at such large SZA anymore and I would recommend using stricter selection criteria for the SZA.

The fact that the method is not valid near twilights has now been made clear in the text. Sza 85° is probably too large for the approach used. We agree with the reviewer that the limit should be lower, between 75° and 80°. We have now limited the plotted data to 80°. However the observed diurnal cycle cannot be due to errors in path calculations due to single scattering approach since the diurnal shape in O₃ and NO₂ is not the same, even though they are analyzed in the same spectral range. Further tests are on-going to figure out how much of the observed diurnal cycles is real.

* O4-MGA: O4 has been used in several studies to estimate the optical path for MAX-DOAS measurements before. Please cite appropriate literature. Sinreich et al. (2012) were not the first and only ones.

Wagner et al., 2004, Sinreich et al., 2005, Frieß et al., 2006 and Clemer et al., 2010 have been included in the text

* P. 8249, l. 4-6: What are the detection limits of the NO₂ monitor and for the DOAS instrument?

TECO manufacturer provides a detection limit of 50 ppt in 300s average for NO₂. Following Platt and Stutz (2008) formulae, DOAS instrument has a typical detection limit below 4×10^{13} molec.cm⁻² for NO₂ along the path, which is, assuming no error in optical paths estimation, of less than 1ppt. This information has been included in the error paragraph.

* P. 8240, ll. 18-19: It would make more sense to call it a path average than the 'concentration at the level of the station' since the aim is to obtain FT concentrations.

Following the reviewer suggestion 'concentration at the level of the station' has been changed to 'average path concentration'.

* P. 8239, l. 15: Is it clear-sky above the sea of clouds or are the clouds only towards the north, but not the rest of the island? This is confusing.

Trade winds induce a layer of clouds in the North face of the island often known as "sea of clouds". It is located below the inversion layer, typically at 800 to 1500 masl depending on the season. Above that height, clear skies prevail. We have tried to clarify in the manuscript by reformulating the section 1.1 on the Izana station atmospheric conditions. MAX-DOAS instrument is pointing towards North. This has been clarified in the text.

* Fig. 2 is misleading in helping to understand the calculations in section 1.2 since SZA1 is not the real SZA in this off-axis geometry. The real SZA for the sketched geometry would be $SZA1 + \gamma$. However, I assume your method only works if γ is small and $h \ll h_s$. If the sketched geometry would represent the true geometry, then the true SZA of the off-axis direction would depend on the distance d and hence f would be a function of d and with that your whole argument would fall. I suggest redrawing the sketch with a flat Earth's surface.

To avoid misunderstanding, figure 2 has been redrawn using a flat Earth's surface, as both reviewers suggest.

* If you use f' and d' for the AMF-MGA calculations, then you should also use c_{st}' .

The notation for c_{st}' has been changed for coherence with the used notation for the other parameters.

* P. 8241, l. 5-6 and p. 8247, ll. 15-20: what temperatures do you use for the calculation of the O4 concentration?

It is not clear for us if referee refers to the temperature of the used cross-sections or to the air temperature at the station altitude. In the first case, we use cross-sections of Hermans at 298K (see Table 1). In the second case, the temperature of the station (286° K) has been taken from the standard atmosphere for tropical latitudes.

Both diurnal and seasonal temperature variability in the tropics is very small compared to higher latitudes and thus, the expected impact on optical paths estimation is negligible.

* Fixing the rel. azimuth in the simulations to 0 (p. 8246, l.24) while the telescope points to the North, seems to be a poor choice.

RT calculations have been performed under actual azimuth conditions for O3 and NO2 when standard profile is considered. When considering photochemical variation of NO2, a fix relative azimuth of 180° has been used instead. Azimuth dependence would complicate these calculations and however no appreciable difference is observed in our results when azimuth dependence or fix relative azimuth of 180° is considered.

* Last, but not least: English syntax problems have to be addressed throughout the manuscript, especially the use of articles. Here is an example, the sentence (p.8237, ll.16-17): On one hand, unlike the in situ measurements, MAX-DOAS integrate optical paths over

few tens of km. should read: On the one hand, unlike in situ measurements, MAX-DOAS integrates optical paths over a few tens of km.

English has been revised throughout the text by native speaker.