

We thank you the reviewers for the careful reading of the manuscript and the positive comments and suggestions.

Response to reviewers comments are in italics.

Reviewer 2.

This manuscript provides a nice analysis of the seasonal variability of NO₂ in the free troposphere at Izana Observatory. There are limited long term free tropospheric NO₂ measurements, due to the challenges in operating instruments in remote mountain top sites and the impact from upslope effects. To overcome this, the authors have applied a recently developed method to determine vmr of NO₂ in the free-troposphere from horizontal MAX-DOAS measurements, using O₄ to infer horizontal path lengths. The measurements benefit from a reduced impact from upslope, which is often observed in the in situ measurements, at the same site. Additionally this method allows for measurements of NO₂ vmr, down to a few pptv. A comparison study with in situ measurements was performed and the data were analyzed under different meteorological conditions along with back trajectories and model results. An Optimal Estimation Method was applied to data, during a period when an air mass with high NO₂ arrived at the station, as a case study to determine the profile of NO₂ in the FT. General comments: Overall this manuscript includes some very interesting results and should be published after revision.

I agree with the comments from reviewer1 regarding the OEM technique. If sensitivity tests on the impact of aerosols on the OEM have not been performed, then the authors may want to consider doing this for the revised manuscript.

We have used the OEM technique to estimate the vertical distribution of the transported NO₂ cloud for a single case. In this particular day (2013, 128 day number), the mean Aerosol Optical Depth above the station at 500 nm was lower than 0.02 which in practice implies a pure Rayleigh atmosphere. We have noticed, however, that there is no comment in the text to this very low AOD conditions, therefore we have added it in OEM section.

Figure 4a shows individual NO₂ measurements from MAX-DOAS for 2011, 2012 and 2013 as a seasonal time series. This figure demonstrates the seasonal variability of the NO₂ in the FT, however, it may also be interesting to see how the NO₂ differs between years, in particular over the spring/summer period when biomass burning may impact the site. Do you observe higher NO₂ levels in the FT during a high fire year?

Summer is the season when large and deep Saharan outbreaks takes place. Since we remove the dusty data there are large gaps on these months which makes difficult a year to year analysis of monthly variabilities. That is why we accumulated the 3-years of data in a single analysis. However, we do observe larger year to year variability in the winter months than in the summer ones. I.e. July means are 41.0, 38.8 and 37.8 pptv for 2011, 2012 and 2013, respectively whereas the August ones are 46.7, 49.5 and 40.4 ppt with a similar sd of about 15. Since we believe that the dataset is still short for this analysis, in this paper we will restrict to the general view.

Specific Comments:

Pg 14476, I5-6: Kleissel et al. (2007) also discusses the impact of upslope on trace gases at Pico.

Kleissl reference has been added.

Pg 14477, l19-20: What is meant here when discussing the slant paths of 0 and 90 degree views cancelling out. Are you referring to the absorption of gas species (except NO₂) and scattering from aerosols between the different views cancelling out? This should be clarified.

We thank the reviewer for this comment. The text was unclear and confusing. We have reformulated the sentence:

“The DSCD is obtained by subtracting the measurement obtained at the zenith (SZA=90°) from the measurement in the horizontal path. In a first approximation, the slant paths contributions of 0 and 90° geometries cancel out and only the signal of the tracers present in the horizontal path remains (See Gomez et al., 2014 for details).”

Pg 14778, l12: How are the AMFs at different SZA calculated here, to determine g and g'?

Again, the text was confusing. We have also modified the text to clarify:

“h is the effective scattering height of the vertical ray. R and R' are the ratio of the mean concentration of the layer divided by the concentration at the level of the station of tracer X and O₄, respectively, and g and g' accounts for their AMF in the zenith geometry (g=AMF(SZA)-1), where SZA stands for Solar Zenith Angle”.

The AMF in the zenith mode for each gas (NO₂ and O₄ in this case) is obtained by using a RTM. We think is not necessary further details since it is a common procedure in MAXDOAS technique.

Pg 14481, l10: Please expand on the negative values and why these were observed. What percentage of the final dataset were negative?

Since the MGA technique is based on the difference in concentration between the zenith and the horizontal spectrum, negative cases might appear occasionally whenever the horizontal path is shorter than the vertical one. Most cases occur are under strong Saharan dust events but those cases are already removed by the previous condition of low AOD. Other cases can be a narrow layer of smoke at the level of the station due to a local wildfire, or a broken cloud in front of the instrument telescope in the horizontal configuration.

The number of negative values remaining after the removal of measurements with AOD>0.1 was of 0.88 % (153 cases out of 17231) of total data. We believe that the contribution to this effect is too low to further extend the explanation in the text.

Pg 14481, l14: Here, do you mean that the OEM was applied to the final year of data (i.e. 2013)? Please clarify.

First sentence is a general one referring to the usefulness of the technique to retrieve estimates of the NO₂ vertical distribution in polluted locations, which is not the case in Izaña. We outline that the OEM technique cannot be used in free troposphere background conditions since the concentration is too low. We have used only to characterize a particular case study in which the NO₂ concentration was very high, but the aerosols remained below the level of the method validity. We believe that the first paragraph of the section 4 is clear enough.

Pg 14486, l22: Can you link the sporadic peaks over 100 pptv to the transport of polluted plumes from North America, either from biomass burning or anthropogenic emissions?

As previously mentioned, a more detailed analysis focus on monthly year-to-year variability and short term cases will be performed in the next future with a larger database.

Technical corrections:

Pg 14475, l28: clouds→cloud. *Corrected.*

Pg 14477, l1: On year 2010→In year 2010. *Corrected.*

Pg 14477, l22: de→the. *Corrected.*

Pg 14478, l7: The later→The latter the. *Corrected.*

Pg 14484, l24: not negligible→non-negligible the. *Corrected.*

References: Kleissl, J.K., R. E. Honrath, M. P. Dziobak, D. Tanner, M. Val Martín, R. C. Owen, and D. Helmig (2007), Occurrence of upslope flows at the Pico mountaintop observatory: A case study of orographic flows on a small, volcanic island, J. Geophys. Res., 112, D10S35, doi:10.1029/2006JD007565. *Reference has been included.*