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 1.

General comments:

The Gil et al.'s paper presents interesting results and is reasonably well written. The scientificfic quality of the paper has been improved by adding a Section on the Optimal Estimation Method. However, a lot of questions remain unanswered regarding this method: On which criteria did you decide not to include aerosols in the OEM retrieval? Did you perform sensitivity tests on this?

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 was no comment in the text to this very low AOD conditions, therefore we have added it in OEM section.

What is the impact of the a-priori profile on your retrieved profiles, especially on the vertical extension of the NO2 layer?

We noticed a mistake in the text (section 4). The a priori NO_2 profiles used in the paper were not obtained from the standard atmosphere,but from hourly profiles provided by a photochemical box model (Denis et al., 2005) derived from the SLIMCAT 3-D chemical transport model (Chipperfield, 2006) for the location and day. This profile is the best estimation that can be used for his purpose. Changes of plus/minus 20% in the a priori result in mean changes of 15 % in the results.

The section 4 of the manuscript has been rewritten to include this information.

A brief description on the main sources of uncertainty of the OEM is still missing. All these points should be discussed in the revised manuscript.

The section 4 of the manuscript has been revised and extended to include the requested points.

Specific comments:

Page 14479, lines 15-29: The Thalman and Volkamer (2013) O4 cross-sections are used in the test on the impact of the temperature dependence of the O4 cross-sections. Since this cross-sections data set is now considered by the DOAS community as the reference O4 cross-sections, what would be the impact of using them on the MGA approach results instead of Hermans et al. (1999)?

Following the reviewer's comment we have computed the optical paths obtained by using the Thalman&Volkamer and compared them with the Hermans ones. Results show differences of only 0.5 ± 0.6 % (larger in Hermans) which has a negligible influence on the obtained results.

Page 14482, line 9: a references hould be added for the Gaussian correlation functions.

The following references have been included in the text:

Barret, B., De Maziere, M. D., and Demoulin, P.: Retrieval and characterization of ozone profiles from solar infrared spectra at the Jungfraujoch, J. Geophys. Res. 107(D24), 4788, doi:10.1029/2001JD001298, 2002.

Friess, U., Monks, P. S., Remedios, J. J., Rozanov, A., Sinreich, R., Wagner, T., and Platt, U.: MAX-DOAS O4 measurements: A new technique to derive information on atmospheric aerosols: 2. Modeling studies, J. Geophys. Res., 111, D14203, doi:10.1029/2005JD006618, 2006.

Why did you choose a correlation length of 300 m?. What is the impact of this correlation length on the vertical extension of the NO2 profiles presented in Fig. 3 ?

*After several tests on the retrieval, gamma value has been chosen to be 300 m, corresponding to the value (between 0.1 and 1 km) that maximizes the DFS (trace of **A**), for the overall retrieval as well as for the altitudes closer to the station (2.3-2.6 km)."*

Technical corrections:

Page 14478, line 5: 'specie' -> 'species'. *We think the singular is appropriate since the formulae refers to a only one tracer at a time*

Page 14500, Fig. 4: The y-axis legends in the three plots should be identical, e.g. 'NO2 concentration (pptv)'. *Corrected*

Page 14485, line 1: You should refer to Sect. 4 instead of Rodgers et al. (2000). *Rodgers reference has been removed from this place.*

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 afterrevision.

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 ananysis 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, l5-6: 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=90º) from the measurement in the horizontal path. In a first approximation, the slant paths contributions of 0 and 90^{o} 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 O_4, 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 (NO_2 and O_4 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 NO_2 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 NO_2 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.*