Response to Anonymous Referee #1

We would like to thank the reviewer for the thoughtful and constructive examination of our paper. Please find below our responses to each comment individually. Please consider that:

A) Green bold: Comments of the Referee
B) Black bold: The response to each referee's comment
C) Red bold: Added text in the manuscript according to referee's comments.

1. Comment: Could you add MetOp-C as well? (Page 2, Line 14)

Response: The MetOp-C satellite is now added to the text (Page 2, Line 14) as follows

"Those observations started in 1995 with the ERS-2 GOME (Global Ozone Monitoring Experiment) instrument (Burrows et al., 1999), followed in chronological order by ENVISAT-SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) in 2002 (Bovensmann et al., 1999), AURA OMI (Ozone Monitoring Experiment) in 2004 (Levelt et al., 2006), MetOp-A/ GOME-2A in 2006, MetOp-B/ GOME-2B in 2012 **and MetOp-C in 2018** (Munro et al., 2016)."

2. Comment: Could you give some numbers? What are the European standards? On how many days the standards are exceeded at this station? (Page 3, Line 20)

Response: The following paragraph is added (Page 3, Line 21), which quotes the European standards for the NO2 values and what is happening in Brussels according to annual reports from the Bruxelles Environment/ Leefmilieu Brussel agency:

"Frequently, the NO_2 concentration monitored by the network of telemetric air quality stations from Bruxelles Environment/Leefmilieu Brussel (https://environnement.brussels/) often exceeds the European standards **upper limits fixed to 40 and 200 µg/m^3 for the NO_2 annual and hourly mean concentrations, respectively. For instance in 2015, the NO_2 annual mean concentrations have been found to exceed those European standards in narrow busy streets in Brussels with an annual mean concentration between 42.5 to 52.5 µg/m^3 (see https://environnement.brussels/). Additionally, as detected by satellite sensors like OMI (Huijnen et al., 2010), NO_2 columns over Brussels are among the highest in Europe."**

3. Comment: Is this number correct? In Kreher et al. (2019) you mention -50°C. Please check again. (Page 4, Line 5)

Response: The remark is correct and the temperature has been corrected to 223 K (Page 4, Line 9).

4. Comment: Could you give some details on MMF performance? What are its strengths/weakness compared to other algorithms? (Page 5, Line 13)

Response: The following paragraph, which describes the strengths/weakness of MMF inversion algorithm, has been added to the manuscript, (Page 5, Line 15:20):

"The main advantages of MMF are (1) the on-line calculation of Jacobians with operation in a logarithmic state vector space, which prevents unphysical negative partial columns to be retrieved, (2) the use of a stable Levenberg-Marquardt non-linear iteration scheme (in replacement to a Gauss Newton scheme) and (3) the fast computing time (5 s per scan for both aerosols and trace gases). A drawback of OEM-based profiling algorithms such as MMF, is that a priori profiles should be carefully chosen in order to avoid biases in the retrieved profiles and columns at altitudes characterized by a low information content."

5. Comment: Is there any reasons why you don't use data from radiosondes instead of standard profiles? Is there any nearby station? (Page 5, Line 18)

Response: When the study started, the pressure and temperature profiles from the US Standard Atmosphere were used because they are easily accessible. However it is true that the Royal Meteorological Institute of Belgium is performing radiosondes and the use of such T/p profiles would be more appropriate as we investigate the seasonality of the MAX-DOAS retrievals. Given the fact that this point has been raised by both reviewers, we decided to redo the complete data analysis using 20-year-based monthly averages of T/p profiles from the ECMWF ERA Interim reanalysis output extracted for Brussels, Uccle. The following sentences have been modified in the manuscript:

"The pressure and temperature profiles are taken from the Air Force Geophysics Lab (AFGL) 1976 Standard Atmosphere (Anderson et al., 1986). The variation of the temperature profiles during one year of measurements is taken into account as an additional error on the profile retrieval."

now becomes:

"The pressure and temperature profiles are prescribed using 20-year-based monthly averaged data extracted from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA Interim reanalysis (see Beirle et al., 2019) for the location of Uccle." (see page 5, Line 26)

and

"The near-surface VMR is then obtained by dividing the concentration of the trace gas (Eq. 4) by the air number density (n_{air}). For the calculation of n_{air}, the pressure and temperature profiles were taken from the AFGL 1976 Standard Atmosphere (Anderson et al., 1986) and are the same as used in Section 2.3.1."

is now:

"The near-surface VMR is then obtained by dividing the concentration of the trace gas (Eq. 4) by the air number density (n_{air}) **derived from monthly averaged temperature and pressure profiles extracted from the ERA Interim reanalysis (see Section 2.3.1)**." (see page 9, Line 24)

6. Comment: How far is the AERONET station away from the MAX-DOAS station? (Page 5, Line 22)

Response: The AERONET station is located on the rooftop of the BIRA-IASB building, at a distance of 180 m from the MAX-DOAS station. The location of the AERONET station as well as the ceilometer are now added to Figure 1.

7. Comment: Is the temperature dependence on NO2 cross section relevant for UV and Vis, or only for Vis? (Page 6, Line 23)

Response: The temperature dependence as adopted by Takashima et al. (2012) is relevant for the Vis fitting window. Although it is not strictly applicable to the UV range, the temperature dependence of the NO_2 cross section is similar in both spectral regions. Therefore, we used the same uncertainty estimate for both fitting windows.

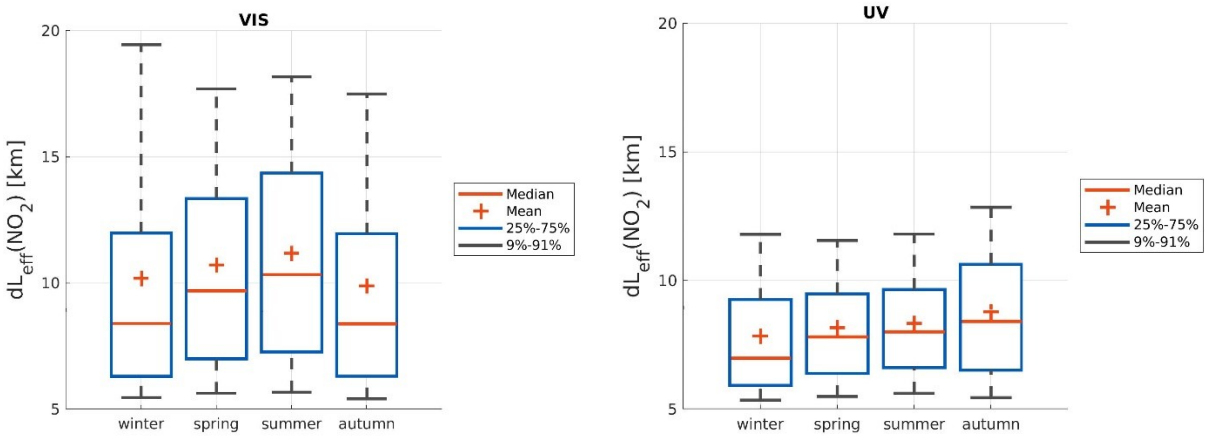
8. Comment: Does the direct use of dLeff only lead to underestimation of NO2 VMR? A factor of three appears quite large. Is a factor of three appropriate for such low elevation angle as used in your study? (Page 7, Line 16)

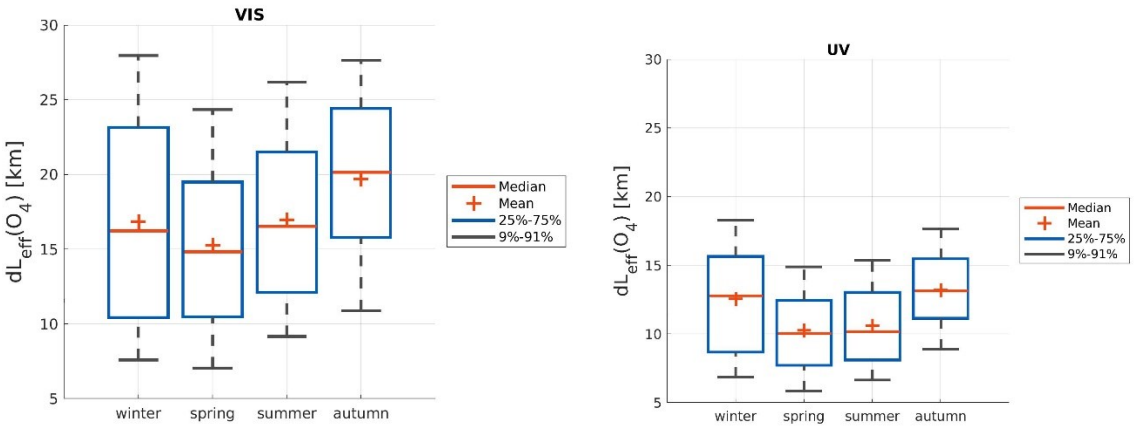
Response: According to Sinreich et al. (2013), Ortega et al. (2015) and Figure 2 in our study, when the correction factors are larger than one, the method becomes highly dependent on the aerosol load. So, these measurements are excluded from the data analysis. Based on that, when applying the appropriate correction factors, the dLeff_NO2 can only be underestimated and consequently, the NO_2 VMR can only be overestimated. When comparing NO_2 VMRs before and after the use of correction factors, they differ by a mean factor of 2 and 1.6 for Vis and UV, respectively. The following sentence has been added on Page 8, Line 1-2:

"**In the present study, where off-axis measurements were performed at 2° elevation in an urban polluted environment, the dL_{eff} of NO_2 was found to be smaller than the corresponding dL_{eff}_O_4 by an average factor of 2 and 1.6, in the Vis and UV wavelength ranges, respectively (Fig. 8).**"

9. Comment: Previous studies have highlighted the importance of properly estimating correction factors, but did any of these studies compare direct use of dLeff (dLeffO4, without fc) with corrected dLeff (dLeffNO2? I would suggest to also compute dLeffO4 (e.g. Seyler et al. 2018) and compare with dLeffNO2. I would be surprised to see a factor of three difference. I suggest to add one plot (dLeffO4 for UV and Vis) to Fig. 8 (b). This would really help to know how essential are such correction factors for urban settings and low elevation angles. (Page 7, Line 26)

Response: For the first part of your comment, please see our response to your previous comment. The figure, which shows the dLeffO4 for Vis and UV, has been added to Figure 8:





As we can see, the difference between the two dLeff (before and after the use of appropriate correction factors) can become quite large and is approximately about a mean factor of 2 and 1.6 for Vis and UV, respectively (Page 13, Line 14). For extreme cases, the difference between the two dLeff can reach a factor of 5 and 4 for Vis and UV, respectively. Consequently, the use of appropriate correction factors under urban conditions and measurements at low elevation angles is critical. The following paragraph has now been added in the text:

"**Similarly, dLeff(O_4), which represents the horizontal sensitivity before applying the appropriate correction factors, is larger in the Vis than in the UV range. The dLeff(O_4) Vis can reach values of up to 28 km, while the maximum value for UV is around 18 km. The difference between dL_{eff} (NO_2) and dLeff(O_4) can become quite large reaching approximately a factor of 2 and 1.6 for Vis and UV, respectively. For extreme cases, this difference factor reach up to 5 and 4 for Vis and UV, respectively.**" (see Page 13, Line 16).

10. Comment: Did you take AOD, asymmetry parameter, and SSA values from the AERONET station? AOD at which wavelength?

Response: The page and line of your comment is not indicated but we assume that it is for the correction factors estimation. As indicated in Sinreich et al. (2013), appropriate correction factors (that are not dependent on AOD) can be calculated when a certain aerosol load is reached in the atmosphere. This aerosol load is reached for AOD in the range of 0.3 - 0.6. In the present study, as indicated in the manuscript, the dAMFs for AOD equal to 0.3 are used for the analysis.

11. Comment: Why did you not use data from weather station for the calculation of nair? (Page 9, Line 7)

Response: As mentioned above, the data analysis was re-done replacing the AFGL 1976 Standard Atmosphere by more realistic profiles obtained using 20-year-based monthly averages of temperature/pressure profiles from the ECMWF ERA Interim reanalysis extracted for Uccle, Brussels. The text in the manuscript has been adapted accordingly (see our reply to your comment n°5 above).

All the figures and discussion results have been adjusted to the new data analysis results. When comparing the NO_2 VCDs, NO_2 near-surface VMRs and MLH(NO_2) derived using the revised atmospheric profiles with those obtained with the initial climatology, we report the following changes:

Value	Percentage difference
NO2 VCD Vis	Winter: -7.1 % Spring : -7.7 % Summer: -8.5 % Autumn: - 6.5 %
NO2 VCD UV	Winter: -7.2 % Spring : -9.2 % Summer: -10.2 % Autumn: -8.6 %
NO2 VMR Vis	Winter: -1.8 % Spring : -5.4 % Summer: -9.6 % Autumn: -5.4 %
NO2 VMR UV	Winter: -0.9 % Spring : -5.3 % Summer: -10.1% Autumn: -6.1 %
AOD Vis	Winter: 0.6 % Spring : -2.5 % Summer: -6.3 % Autumn: -5.8 %
AOD UV	Winter: 2. 9 % Spring : -2.5 % Summer: - 8.0 % Autumn: -6.0 %
MLH (NO2)	Winter: -5.9 % Spring : -2.7 % Summer: 0.5 % Autumn: -1.7 %

The percent differences are calculated against the initial data (AFGL 1976 Standard Atmosphere).

12. Comment: Did you include uncertainties arising from the use of AFGL profiles instead of data from weather stations? (Page 9, Line 25).

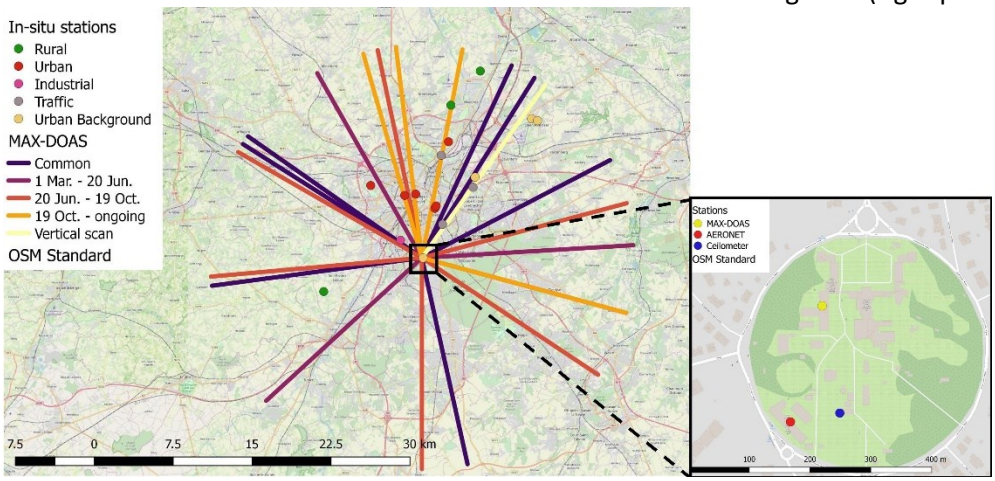
Response: As mentioned above, the whole analysis was redone using climatological temperature/pressure profiles based on ECMWF reanalysis data. An estimation of the impact on NO2 near-surface VMR and VCD when using the ECMWF climatology instead of US standard atmosphere is presented in the previous comment n° 11. Unfortunately, temperature/pressure profiles from weather stations were not easily accessible so, these uncertainties could not be estimated.

13. Comment: Throughout the whole manuscript you are using the terms MLHNO2 and MLHMAXDOAS but actually, as I understand, the two terms refer to the same parameter? I suggest to use only MLHMAXDOAS?

Response: The two terms (MLH_NO2 and MLH_MAXDOAS) refer to same parameter. The term MLH_MAXDOAS has been used in the manuscript.

14. Comment: How far is the ceilometer away from the MAX-DOAS station? (Page 11, Line 10). I suggest to include the position of AERONET and ceilometer stations in Fig. 1, if the position is other than for the MAX-DOAS instrument.

Response: The AERONET instrument, as mentioned above, is located 180 m away from the MAX-DOAS spectrometer and the ceilometer is located 160 m away from the MAX-DOAS location. The locations of both AERONET and ceilometer instruments are now included in Figure 1 (right panel):



15. Comment: Can you give some details on cloud screening? Did you use cloud-screened pixels only? (Page 12, Line 16)

Response: As clouds in the atmosphere can strongly affect the MAX-DOAS trace gas retrieval, a cloud filtering approach is applied to the MAX-DOAS scans by using a co-located thermal infrared pyrometer. In the present study, only MAX-DOAS scans with a total cloud-cover fraction less than 0.8 (80 %) are selected for further analysis. By applying this upper limit, scans under fully cloudy conditions are rejected (Page 6, Line 9).

Concerning the TROPOMI cloud filtering, in the present study, only pixels with a quality assurance value larger than 0.75 (QA>0.75) are included in the validation. This filter removes pixels with a cloud radiance fraction larger than 0.5. This has been added in the text (Page 12, Line 32).

16. Comment: Please also include a few sentences discussion about dLeffO4 and compare with dLeffNO2. (Sect. 4.1, Fig. 8).

Response: After comparing dLeffO4 with dLeffNO2, the following paragraph has now been added in Section 4.1 (see our reply to comment n°9 above):

"**Similarly, dLeff(O_4), which is the horizontal sensitivity before applying the appropriate correction factors, is larger in the Vis than in the UV range. The dLeff(O_4) Vis can reach values of up to 28 km, while the maximum value for UV is around 18 km. The difference between dL_{eff} (NO_2) and dLeff(O_4) can become quite large by approximately a mean factor of 2 and 1.6 for Vis and UV, respectively. For**

extreme cases, this difference factor can be up to 5 and 4 for Vis and UV, respectively." (see page 13, line16).

17. Comment: Did you use 11:00 UTC because of TROPOMI overpass? If so, please add this information. (Page 13, Line 5)

Response: The TROPOMI overpass over Brussels is between 10:00 and 12:00 UTC. For that reason, we have chosen to present the MAX-DOAS results at 11:00 UTC. The following sentence is now added in Page 13, Line 22:

"**The choice of presenting the MAX-DOAS measurements at 11:00 UTC is based on the TROPOMI overpass time, which varies between 10:00 and 12:00 UTC.**"

18. Comment: Is it really only up to 200 m? I would suppose values up to 350 m for dLeff = 10 km and EA = 2_ and also I expect differences for UV and Vis channels, according to Wang et al. 2014 AMT. (Page 15, Line 6).

Response: The above comment is correct. In the manuscript, we used the 200 m as the lowest altitude of the OEM-based retrieval. In the comparisons with in-situ stations, the parameterized near-surface concentrations were used. The estimation of the parameterized MAX-DOAS near-surface measurements correspond to a layer expanding from the surface to a height equal to h=dLeff(NO2)*sin(elevation angle). For an elevation angle equal to 2^{o} and for two characteristic horizontal sensitivities of 12 km and 8 km (Figure 8) for the Vis and UV, respectively, the altitude corresponds to 420 m and 280 m for the Vis and UV, respectively. Overall, for the Vis, the horizontal sensitivity values vary from 5 to 20 km, which corresponds to a height variation between 175 - 700 m. Similarly, for the UV, the horizontal sensitivity values vary from 5 to 13 km, which corresponds to a height variation between 175 - 450 m. The following sentence on page 15, line 24 has been modified as follows:

"Secondly, the in-situ stations are located typically at 3-10 m altitude, while the MAX-DOAS near-surface measurements correspond to a layer extending from the surface to approximately 420 m altitude for the Vis range considering a horizontal distance of 12 km and from the surface to 280 m altitude for UV based on a horizontal distance of 8 km (see corresponding $dLeff_{NO2}$ in Fig. 8). Hence, the MAX-DOAS near-surface concentration is not fully representative of the in-situ surface concentration, as reported in Kramer et al. (2008)."

19. Comment: Because you mention that one azimuthal MAX-DOAS measurement samples air masses along several kilometers, what about the correlation between MAX-DOAS (geometricapproach, e.g. using 30_ measurements) and TROPOMI? (Page 16, Line 17)

Response: The NO2 VCD derived by MMF in the main azimuthal direction vs TROPOMI (for the whole time period) have a correlation coefficient of 0.47 and slope equal to 0.36, while for the NO2 VCD derived by applying the geometrical approach vs TROPOMI has a correlation coefficient of 0.47 and slope equal to 0.39. We can expect similar correlation coefficient and slope values when using the geometrical approximation since, when comparing the NO2 VCD derived by MMF with the NO2 VCD derived by the geometric approach, we find a good agreement between both datasets (R=0.95 and slope= 0.94).

20. Comment: Actually I do not see improvement for summer. (Page 17, Line 4)

Response: The term improvement is referring to the correlation coefficient values, which increases from 0.57 to 0.77 for summer. On the other hand, the slope value is almost constant for this season (it varies from 0.49 to 0.58). A summary of the regression analysis parameters derived by the three validation exercises can be found in Table 7. The corresponding paragraph has been adjusted (Page 17, line 26) as follows:

"Results displayed in Fig. 14 show that the agreement between TROPOMI and MAX-DOAS datasets is significantly improved, especially in terms of correlation (R in the 0.65-0.82 range). Owing to the improved spatial coincidence associated to the use of dual-scan MAX-DOAS data and the better spatial coincident criterion between TROPOMI and MAX-DOAS data, the scatter in the data points is also substantially reduced during all seasons and especially in winter. Another interesting feature is the improvement of the slope values, observed in all seasons (slopes in the 0.41-0.71 range). During seasons with a more homogeneous NO_2 field, the improvement of the slope values is less pronounced than during seasons, like winter and autumn, where the NO_2 field can be highly inhomogeneous."

is now:

"Results displayed in Fig. 14 show that the agreement between TROPOMI and MAX-DOAS datasets is significantly improved, especially in terms of correlation (R in the **0.60-0.85** range **instead of 0.25-0.72**). Owing to the improved spatial coincidence associated to the use of dual-scan MAX-DOAS data and the better spatial coincident criterion between TROPOMI and MAX-DOAS data, the scatter in the data points is also substantially reduced during all seasons and especially in winter. Another interesting feature is the improvement of the slope values, observed **in winter and autumn (slopes of 0.81 and 0.61, respectively, instead of 0.33)**. During seasons (spring, summer) with a more homogeneous NO_2 field, the improvement of the slope values is less pronounced than during **seasons where the NO_2 field can be highly inhomogeneous (i.e. winter and autumn).** During spring, the slope value is reduced despite the better correlation. Overall, TROPOMI still underestimates MAX-DOAS measurements by about 40-50 %."

21. Comment: Again, not clear which cloud fraction you used for cloud screening. (Page 17, Line 19)

Response: See our reply to comment n°15 above.

22. Comment: Is it mean or median? (Page 18, Line 30) Because in the conclusion you state that you are using median MAX-DOAS profiles as a priori. (Page 20, Line 17)

Response: Indeed, the MAX-DOAS profiles that have been used as a priori profiles in the satellite retrieval are median MAX-DOAS profiles. This has been corrected in Page 18, Line 30. An example of a median MAX-DOAS profile during summer can be found in Figure 15.

23. Comment: Technical corrections:
- associated with (Page 11, Line 29)
- Two modifications are introduced (Page 16, Line 24)
- Some words (e.g. : : : algorithm is based on NO2 : : :) are missing in the first sentence of Sect. 4.4.3 (Page 18, Line 19)

Response: The three technical corrections have been included in the revised manuscript.