Response to Anonymous Referee #2

We would like to thank the Anonymous Referee #2 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: Major drawback: Standard atmosphere

The authors use a standard atmosphere for their evaluation. This is quite odd, as the MAXDOAS measurements are taken right at the meteorological institute of Belgium, where I would expect that atmospheric profiles of temperature and pressure would be available on daily basis. The authors discuss the seasonality of their results. This discussion is hard to assess as the seasonality of T and p, which directly affects the O4 concentration (and thus aerosol inversion and thus NO2), is just ignored. Thus the authors at least have to study the impact of the used standard atmosphere on the seasonality of results. But I would highly recommend that the authors redo the complete data analysis for real atmospheric profiles.

Response: We agree with the reviewer. The standard atmosphere has been used because it was easily accessible for starting our study. Taking into account your comment, we decided to redo the complete data analysis using 20-year monthly averages of atmospheric profiles from the ECMWF Interim reanalysis extracted for Uccle, Brussels. The text in the manuscript has been adapted as follows:

"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."

is now:

"**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)

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.

2. Comment: 5/25: For a DoF of 1, there is only 1 piece of information available (i.e. the total column). So for real "profile information" I would expect a threshold of DoF>2 rather than 1.

Response: We agree with the reviewer. The main goal of this manuscript was the use of the total columns. For this reason, a DoF equal to or larger than one was used. As the complete data analysis was redone according to your first comment, a DoF equal to 2 or larger is now used in order to ensure the validity of the MAX-DOAS a-priori profiles (see page 6, Line 2).

The use of a DoF equal or larger than two leads to the reduction of the number of valid MAX-DOAS scans by 4 % for the total time period examined in our study.

3. Comment: 6/3: Is the pyrometer also pointing in the same direction as the MAX-DOAS instruments, or does it have a fixed viewing direction? If the latter, how could it provide information on cloudiness for low elevation angles?

Response: The pyrometer is located on the BIRA-IASB rooftop and it is pointing at the zenith direction. The BIRA-IASB building is located 180 m away from the MAX-DOAS instrument. The cloud filtering applied in this study (only scans for a cloud percentage lower than 80 % are selected) is a first cloud filtering approach to remove scans under high cloud coverage. In the future, the approach developed in Gielen et al. (2014) will be applied, which can be used as a cloud filtering in different elevation angles (in our case, it will be applied for the elevation angle of 2°). In this approach, the color index, which is the ratio between two wavelength radiances, is used to define the sky condition (clear sky, thin clouds and high cloud coverage). First tests have already been done by using the color index in 412 nm and 500 nm as measured in the elevation angle of 2°. For high cloud coverage, 93 % of the total cases, the color index method and the pyrometer give similar result. This indicates that for the present study, pyrometer measurements can be used in first approximation for the cloud flagging of the MAX-DOAS scans

4. Comment: 6/27: Please add a direct comparison of the NO2 results for UV and Vis and compare with the listed uncertainties.

Response: A direct comparison between the NO2 near-surface VMR and NO2 VCD for UV and Vis shows that differences can reach up to 15 %. This source of uncertainty has been added to Table 4, where the total uncertainty in the MMF retrieved NO2 VMR and VCD in the Vis and UV spectral ranges is presented. As discussed into details in the manuscript (Section 4.1), the horizontal sensitivity in the Vis and UV wavelength ranges is different (larger for Vis than for UV), which results to a different horizontal air sampling.

In order to address this comment, the following sentence is added on page 6, line 32:

"Combining all the above-mentioned sources of error, the following uncertainties for NO_2 retrievals are estimated: 11 % and 13 % on NO_2 VMR and VCD in the Vis range, respectively, and 10 % and 14 % on NO_2 VMR and VCD in the UV, respectively. Another source of uncertainty is estimated by comparing the NO_2 near-surface VMR and VCD in the UV and Vis wavelength ranges. The uncertainty is up to 15.5 % of the near-surface VMR and 15.4 % of the VCD. This percentage difference is slightly larger than the above-mentioned retrieval uncertainties. The main origin of this uncertainty is the different horizontal sensitivity in the UV and Vis wavelength ranges and therefore, a different air mass sampling."

5. Comment: 6/29: The subsection heading announces details on the dual scan retrieval. However, large parts of the following text just explain the par method. I propose to add a subsection dedicated to the par method for better clarity.

Response: Two separate subsections were created:

'2.3 Aerosol and OEM-based profile retrievals' and '2.4 Dual-scan MAX-DOAS retrieval strategy'. In the subsection 2.4, two new subsections were created for the sake of clarity: '2.4.1 The parameterization method' and '2.4.2 Dual-scan MAX-DOAS retrieval in Uccle'.

Also small adjustments have been made to the manuscript, which can be found in Page 7, Line 6 and Page 8, Line 24.

6. Comment: 16/4: Please put this finding in relation to other comparisons (which have also reported a low bias of TROPOMI columns) and add respective references.
Response: In the present manuscript, TROPOMI tropospheric NO2 columns are found to be systematically lower than co-located MAX-DOAS measurements. This finding is in agreement with recent studies such as:

1. The study of Ialongo et al. (2020) which evaluates the TROPOMI NO2 columns in polluted conditions in Helsinki, Finland.
2. The study of Griffin et al (2019) for the Canadian Oil Sands
3. The study of Zhao et al. (2019) for urban conditions in Toronto, Canada.

A sentence is now added in Page 16, Line25 to make the link between our findings and the above references:

"**This finding is in agreement with the recent studies of** Griffin **et al. (2019), Zhao et al. (2019), and Ialongo et al. (2020).**"

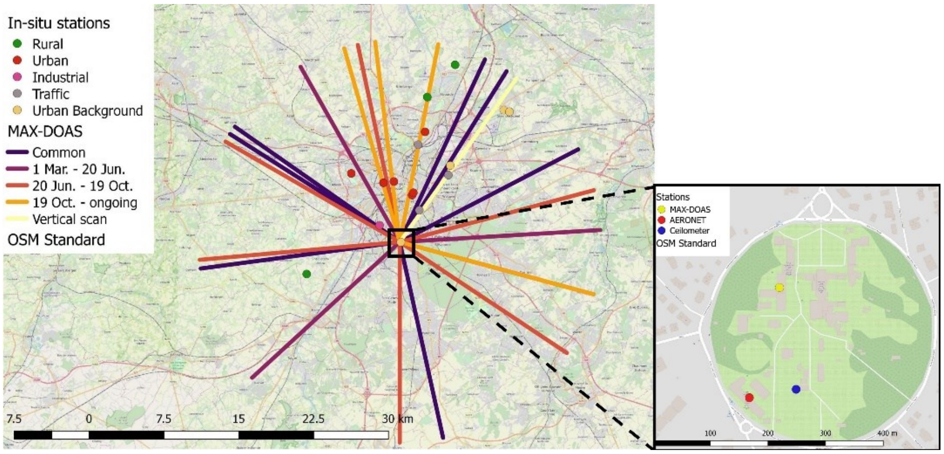
7. Comment: 17/26: I don't agree with the general statement that cloud effects are quasi random and do not cause systematic biases, and I don't see these bold statements supported by the presented measurements. Cloud impact is definitely "complex" and may indeed "lead to positive or negative biases". So they definitely introduce considerable scatter to the retrieved columns. But underneeth this scatter, which might look "quasi random", there are very likely systematic effects as well, which probably could only be quantified with large data samples.
Response: In the present manuscript, we focus on the impact of the a-priori profile in the TROPOMI retrieval, and not on the influence of clouds. For the latter, we refer only to the existing literature. We agree with the reviewer that the cloud effects are more complex than presented here.

In TROPOMI retrieval, the FRESCO-S cloud fraction and height is used. By applying a filter in the qa_value, we exclude pixels with a cloud fraction larger than 0.5. However, pixels with small cloud fractions, which are part of the data analysis, can cause bias on the TROPOMI tropospheric NO2 columns. More precisely, over bright cities, if the TROPOMI albedo value (spatial resolution of 0.5^ox0.5^o) is lower than the real albedo value of the surface, the TROPOMI algorithm tends to give a non-zero cloud fraction to account for the brightness of the pixel. This can lead to an underestimation of the air mass factor (AMF) and consequently, an overestimation of the TROPOMI columns.

The following sentence has now been added in the manuscript to address the reviewer's comment on the complexity of cloud characterization and filtering in satellite data (see Page 18, Section 4.4.1):

"**This cloud information is used as an input in a cloud-correction scheme applied to NO_2 retrieval (van Geffen et al., 2019). Cloud-induced errors are complex and can lead to positive or negative biases on the tropospheric NO_2 column resulting to considerable scatter to the retrieved columns, especially for small cloud fractions.**"

8. Comment: Table 1: Please add the different sets of azimuth angles to Fig. 1, color coded for the three time periods.
Response: Figure 1 has been modified and different sets of azimuth angles corresponding to the three time periods are now shown with different colors:



9. Comment: Table 4: Please add results from a direct comparison between NO2 UV and vis - is the difference within the listed total uncertainty range?

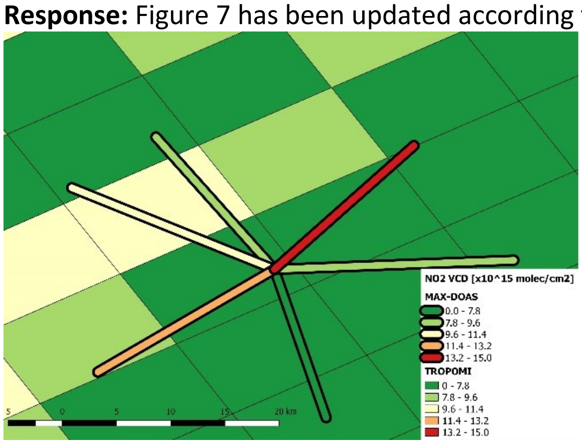
Response: As mentioned in the manuscript (see Page 6, Line 34), the difference between NO2 Vis and UV is about 15.5 % in the NO2 near-surface VMR and 15.4 % in the NO2 tropospheric VCD. This difference is slightly larger than the listed total uncertainty range. These values have been added in Table 4.

10. Comment: Fig. 1: Please add the different sets of azimuth angles, color coded for the three time periods. What is the meaning of the symbols on top of the azimuth direction lines?

Response: Figure 1 has been updated according to your recommendation. The meaning of the symbols on top of the azimuth direction lines showed the direction (from the MAX-DOAS instrument to the maximum horizontal distance). For the simplicity of the figure, the symbols were excluded and simple lines were used.

11. Comment: Fig. 7: The landscape background is not helpful here, but rather disturbing. I propose to show no background, but instead real color coded VCDs for both TROPOMI and MAXDOAS.

Response: Figure 7 has been updated according to your recommendation:



12. Comment: Fig. 12: Several MAX-DOAS VCDs have no errorbar. I assume that for these days no std could be calculated. I propose to also show the mean/typical uncertainty of single VCDs derived from the MAX-DOAS inversion, which would probably be more consistent for the time period. This might be added as second error bar with e.g. light color.

Response: The MAX-DOAS VCDs that have no error bar correspond to days where no standard deviation (std) can be calculated. Following reviewer's comment, we have added the typical MAX-DOAS inversion uncertainty (reported in Table 4) on each data point with grey light color.

13. Comment: Fig. 15/16: Please add the corresponding a-priori profiles used in the TROPOMI retrieval, and add a discussion of this comparison.

Response: The corresponding a-priori profiles used in the TROPOMI retrieval have been added to Figure 15 and 16:

Figure 15

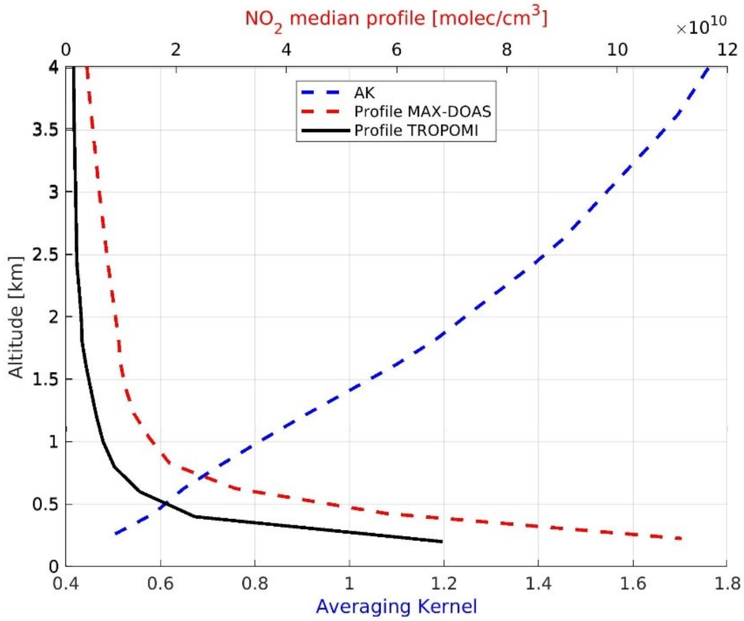
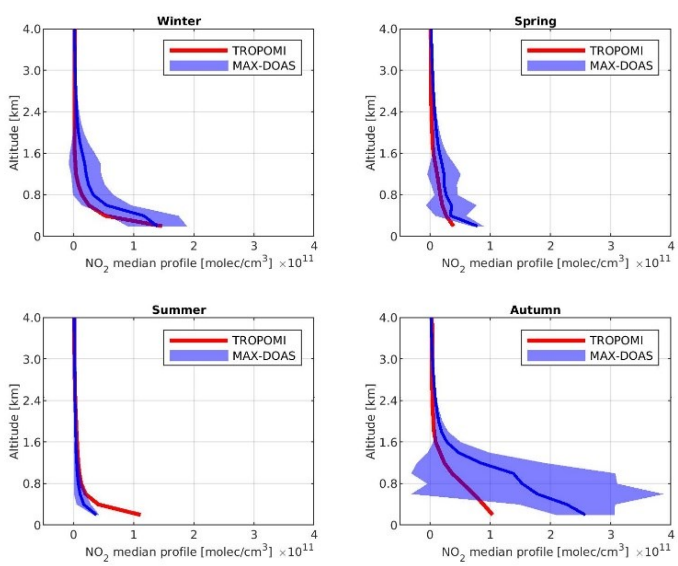


Figure 16



In order to address this comment, the following sentences are added:

“Figures 15 and 16 present a-priori NO_2 profiles used in the TROPOMI retrieval. One can see that the a-priori TROPOMI NO_2 profiles are lower than the median MAX-DOAS NO_2 profile (except for the example summer day in Fig. 16). In the first kilometers above the surface, the difference between both profiles is about 35 % in Fig. 15 and can be up to 80 % for the example autumn day in Fig. 16. ”
(Page 19, Line 24)

14. Comment: Minor comments:
1/18: "concentrations" should be "volume mixing ratios"
3/26: "eventual" should be "potential" or "possible"
6/2: Skip the comma between "measurements" and "strongly"
6/10: "Smoothing error"
6/30: Please define what exactly is meant by "near surface VMR"
8/29: "are equal" should be "are similar"
9/21: "close to zero" is not unphysical

Response: The above technical corrections have been implemented in the revised manuscript.

References:

Gielen, C., Van Roozendael, M., Hendrick, F., Pinardi, G., Vlemmix, T., De Bock, V., De Backer, H., Fayt, C., Hermans, C., Gillotay, D., and Wang, P.: A simple and versatile cloud screening method for MAX-DOAS retrievals, Atmos. Meas. Tech., 7, 3509–3527, doi:10.5194/amt-7-3509-2014, 2014.

Griffin, D., Zhao, X., McLinden, C. A., Boersma, F., Bourassa, A., Dammers, E., Degenstein, D., Eskes, H., Fehr, L., Fioletov, V., Hayden, K., Kharol, S.K., Li, S.-M., Makar, P., Martin, R.V., Mihele, C., Mittermeier, R.L., Krotkov, N., Sneep, M., Lamsal, L.N., terLinden, M., vanGeffen, J., Veefkind,P., and Wolde, M.: High-Resolution Mapping of Nitrogen Dioxide With TROPOMI: First Results and Validation Over the Canadian Oil Sands, Geophysical Research Letters, 46, 1049–1060, https://doi.org/10.1029/2018GL081095, 2019.

Ialongo, I., Virta, H., Eskes, H., Hovila, J., and Douros, J.: Comparison of TROPOMI/Sentinel 5 Precursor NO_2 observations with ground-based measurements in Helsinki, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2019-329, in review, 2019.

Zhao, X., Griffin, D., Fioletov, V., McLinden, C., Cede, A., Tiefengraber, M., Müller, M., Bognar, K., Strong, K., Boersma, F., Eskes, H., Davies, J., Ogyu, A., and Lee, S. C.: Assessment of the quality of TROPOMI high-spatial-resolution NO_2 data products in the Greater Toronto Area, Atmos. Meas. Tech., 13, 2131–2159, https://doi.org/10.5194/amt-13-2131-2020, 2020.