Author reply to Referee #2

Lisa K. Behrens et al.

March 6, 2018

We thank Referee #2 for carefully reading our manuscript and for the helpful comments which will improve the quality of our manuscript. We will reply to the comments point by point.

Legend:
- referee comments
- authors comments
- **changed text in the manuscript**

Behrens et al. present a study showing the relationship between UV and VIS tropospheric NO2 VCDs and how it can relate to information on vertical distribution of NO2 in the troposphere. Although the paper is well written, it appears to be rather deceiving. I find the paper too qualitative and the reader could expect an attempt to effectively derive some information on the vertical distribution of NO2 from the combination of UV and vis NO2 measurements. The authors provide a number of possible reasons for the differences between UV and vis NO2 VCDs (a-priori profiles, effect of clouds/aerosols, etc) that are all plausible (and speculative) but there is no clear way forward. They almost conclude on the current impossibility to derive profile information. A weak point is that it is difficult to separate possible errors in the retrievals (in the UV spectral fits) from real effects.

We agree with the referee that the title may suggest that combined UV and vis NO_2 measurements would be provided information about the NO_2 profiles in this manuscript which may lead to confusion. Therefore, we will change the title to: "GOME-2A retrievals of tropospheric NO_2 in different spectral ranges - influence of penetration depth". Furthermore, we will add maps with retrieved top-altitudes of NO_2 layer height (Fig. 1) in the revised manuscript in Sect. 3.1. For the altitude retrieval simple box profiles are assumed for tropospheric NO_2. A seasonal dependency of the retrieved al-

titude can be clearly observed in the global maps.

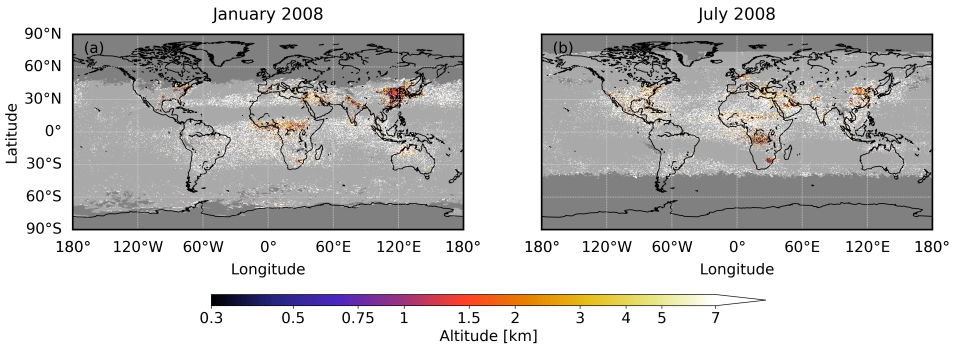

Figure 1: Monthly mean top-altitudes retrieved from the ratio between the UV and blue spectral range. To retrieve the altitude, box profiles are assumed for the tropospheric NO_2. The light grey coloured values indicate values which are below the threshold defined for the ratio (see Fig. 8 manuscript; will be added in a revised version)

Furthermore, we agree that it is difficult to separate possible errors in the retrieval from real effects. With the help of the referee's comments we will make the manuscript more quantitative, by including an error discussion and the discussion of the temperature dependency of SCDs.

To be published in AMT, the following points need to be addressed:

-a comprehensive error analysis on the UV retrievals needs to be undertaken.

Done — we will include the following table in Sect. 2.7 (2.8 in the revised manuscript):

Table 1: Errors for the UV and vis spectral range.

	errors UV	errors vis
total SCDs	1.8×10^{15} molec cm^{-2}	0.6×10^{15} molec cm^{-2}
	(calculated above the Pacific Ocean, see Fig. 4 (manuscript))	
strato. VCDs	7.4×10^{14} molec cm^{-2}	2.1×10^{14} molec cm^{-2}
	(see Fig. 4 (manuscript))	
SSR	40% at 320 nm	5% at 500 nm
	(Kleipool et al., 2008)	(Kleipool et al., 2008)
0.05, increase 0.01	BAMF increases 9%	BAMF increases 11%
	(338 nm, Lorente et al., 2017)	(440 nm, Lorente et al., 2017)
AMF		
cloud fraction	0 – 30%, Boersma et al., 2004	
cloud height	< 10%, Boersma et al., 2004	
aerosols	not included in our calculations → 15%, Boersma et al., 2004	
profile shape	< 15% (regions with little NO_2: > 50%), Boersma et al., 2004	

-section 2.2: the effect of T on Uv+vis NO2 retrievals is not well discussed. NO2 cross sections are varying with T but only one T cross-section is included in the fits (both for UV and VIS DOAS fits). What is the impact on the results and conclusions of this study?

The NO_2 cross sections have a temperature dependency, which differs for the UV and vis spectral range as shown in Fig. 2 and Fig. 3 exemplarily for the two spectral ranges. This temperature dependency can further increase the differences between the two spectral ranges. Figure 4 and Fig. 5 show the altitude dependent sensitivity of the scaling coefficients for the UV and vis spectral ranges. The temperature dependency influences the tropospheric as well as the stratospheric NO_2 measurements. For the stratospheric NO_2 measurements, the sensitivity of the UV NO_2 spectral range is approximately 10% higher than for the vis spectral range. Close to the surface, the difference in temperature sensitivity is up to 10% stronger in the vis spectral

range. For VCDs, the temperature dependency of the NO_2 cross section is scaled by a linear correction factor for both spectral ranges, as suggested in Boersma et al., 2004. In the revised manuscript, we will include this issue in our discussion, but we will move this point to Sect. 2.4 (p. 5, l. 27, manuscript):

[...] **Furthermore, the measurement sensitivity for NO_2 decreases towards the surface. This can be clearly observed in the BAMF (Fig. 2, manuscript). This effect is enhanced by the temperature dependency of the NO_2 cross section. The temperature dependency influences the tropospheric as well as the stratospheric NO_2 measurements (see Fig. 4 and Fig. 5). For the stratospheric NO_2 retrieval, the sensitivity in the UV spectral range is up to 10% higher than for the vis spectral range. Close to the surface, the temperature sensitivity is up to 10% stronger in the vis spectral range compared to the UV spectral range. The temperature vertical sensitivity introduces a seasonal and a latitudinal dependency. This effect is stronger in the tropics than for higher / lower latitudes and in the mid-latitudes it is more pronounced in summer and less in winter.**

[**In combination with ...**]

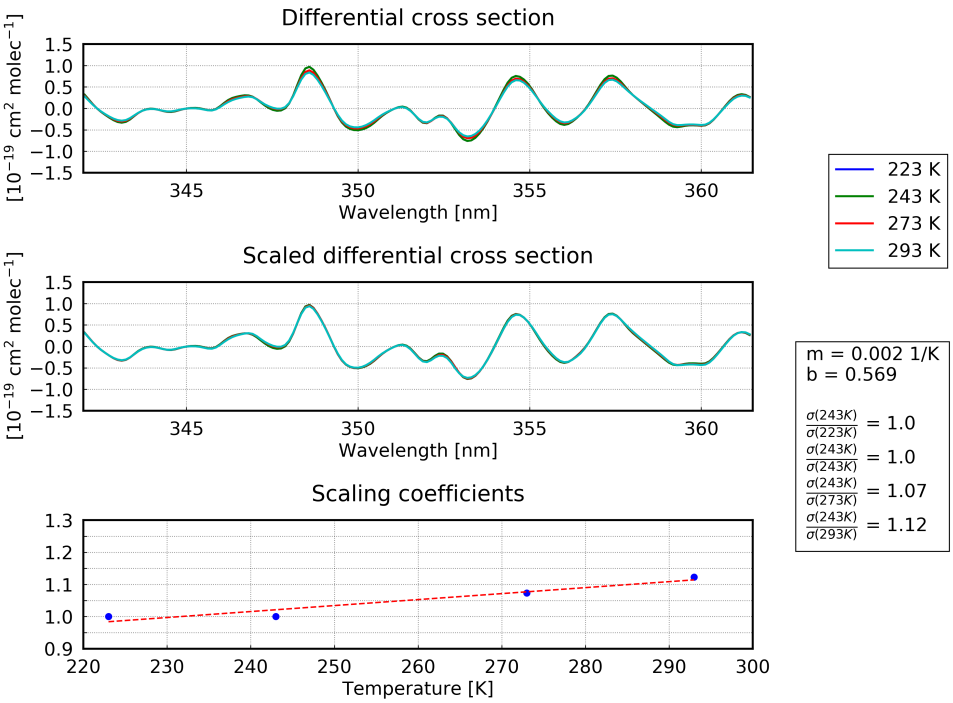

Figure 2: NO_2 cross section and their temperature dependency for the UV spectral range. The scaling coefficient of the temperature dependency is calculated. (will be added in a revised manuscript, supplement)

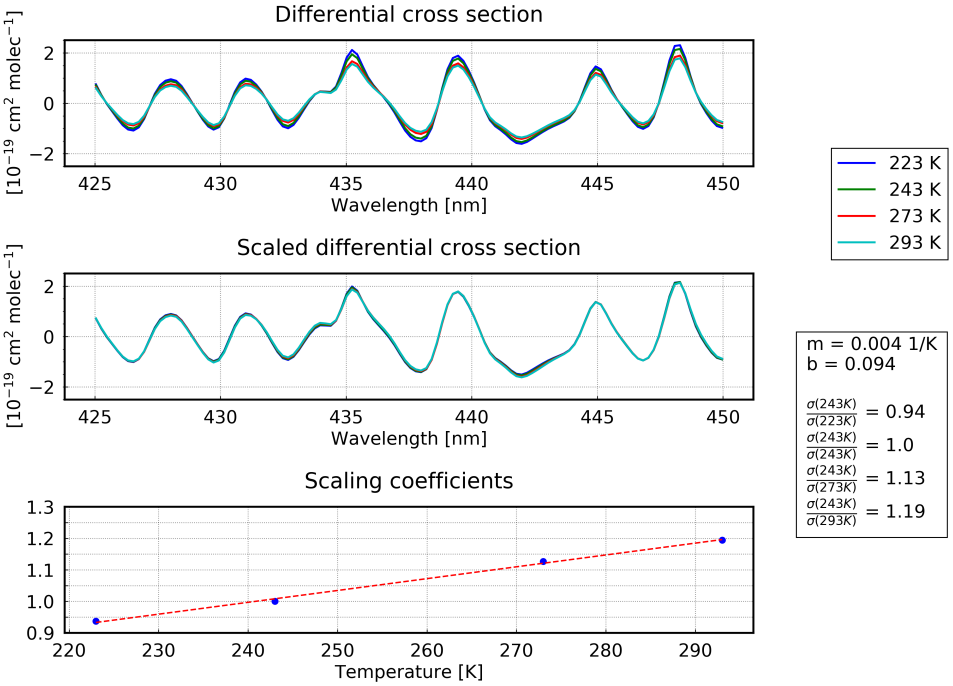

Figure 3: NO_2 cross section and their temperature dependency for the vis spectral range. The scaling coefficient of the temperature dependency is calculated. (will be added in a revised manuscript, supplement)

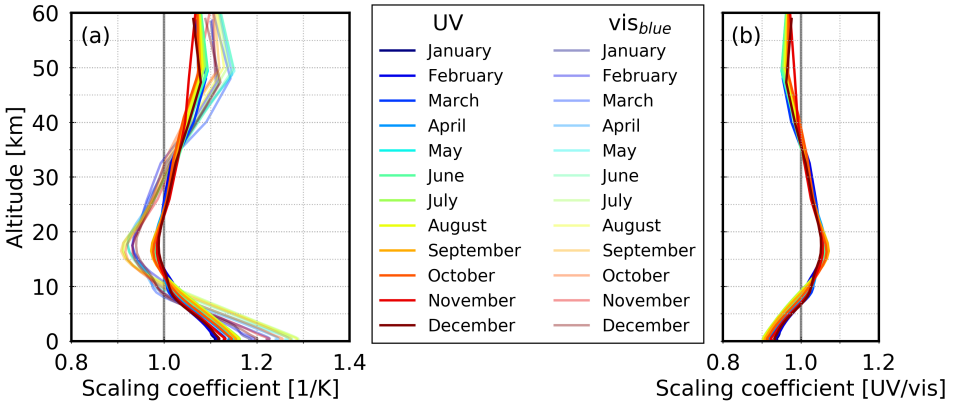

Figure 4: Altitude dependency of the NO_2 scaling coefficient for China. The profiles are calculated for model data simulated with the TM5 model for 2008. (will be added in a revised manuscript, supplement)

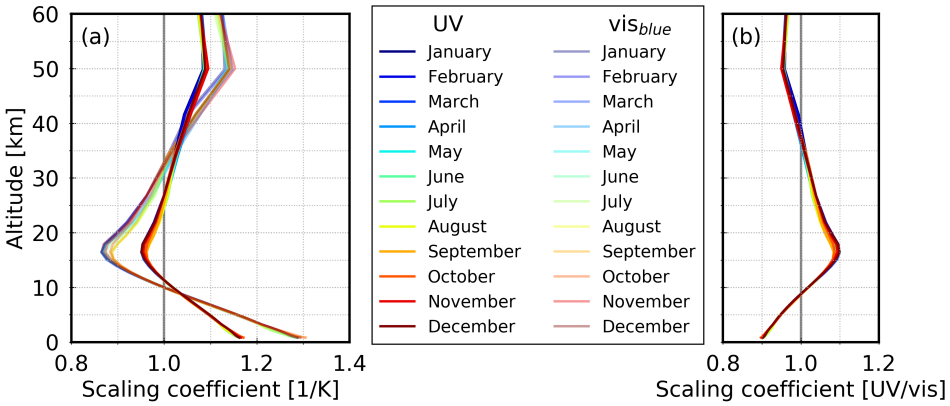

Figure 5: Altitude dependency of the NO_2 scaling coefficient for ASE. The profiles are calculated for model data simulated with the TM5 model for 2008. (will be added in a revised manuscript, supplement)

-section 2.6: what is the uncertainty due to errors in SSR?

This issue has been addressed in the new Tab. 1.

-section 3.3, p14, l20: it is written that VCD differences are small but it is hard to judge as only absolute values for the differences are shown. It would be better to incorporate relative differences as well as proper error calculation (see comment above).

In response to the suggestion of the reviewer, we will show the relative differences in the revised manuscript (see Fig. 6 and Fig. 7). The figures with the absolute differences will be moved to the supplement.

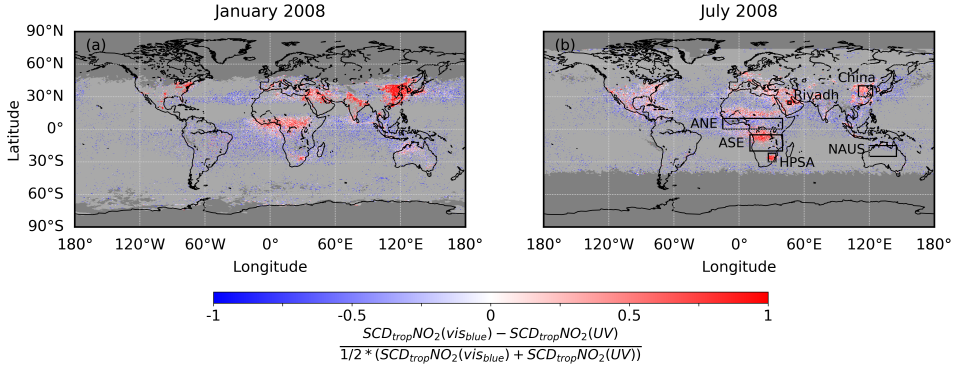

Figure 6: Relative difference between monthly mean tropospheric NO_2 SCDs in the vis and UV spectral range. Differences for (a) January and (b) July 2008. Dark grey shaded area: no NO_2 values available. Light grey coloured values indicate values where the vis NO_2 is close to zero, which have been filtered out.

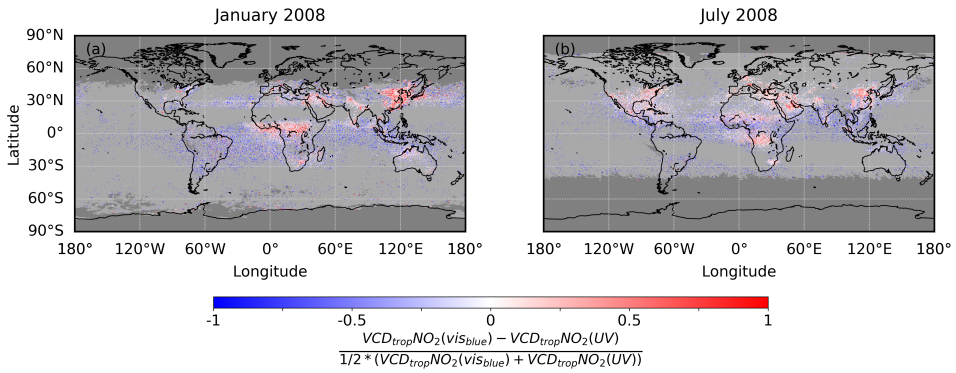

Figure 7: Relative difference between monthly mean tropospheric NO_2 VCDs in the UV and vis spectral range. Differences for (a) January and (b) July 2008. Dark grey shaded area: no NO_2 values available. Light grey coloured values indicate values which are filter out. The same filter as for Fig. 6 is used.

-p15: it is not clear how the CTM profiles should be changed to reconcile the vis, uv and modeled VCDs.

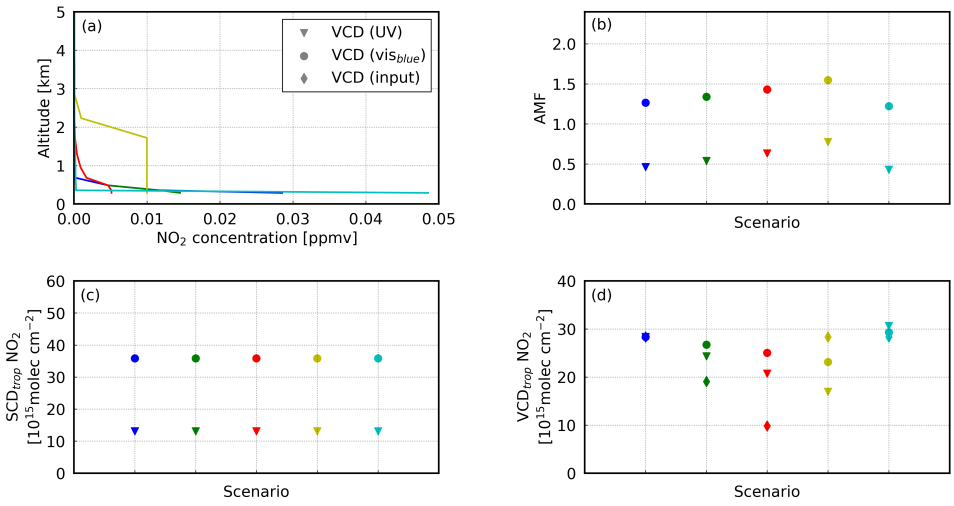

Figure 8: SCD, VCD, and AMF for different NO_2 profiles, calculated for one scenario in January over China with a SZA of 66°. Blue: true profile, the SCDs are calculated for this profile. Other color: changed input profiles for AMF calculation leading to changes in the retrieved VCDs.

As shown in Fig. 8, the profile shape has an influence on the retrieved VCDs, if the assumed and true profile shape do not agree. Exemplarily for one scenario over China in January with a SZA of 66°, the VCDs are calculated. To avoid possible measurement errors in the retrieved SCDs, the SCDs are also calculated with the radiative transfer model SCIATRAN. The "true" NO_2 profile is the blue curve in Fig. 8 a. Additionally, the NO_2 profiles were changed (colour coded) and AMF (Fig. 8 b) were calculated for these changed profiles which introduce differences in the retrieved VCDs (Fig. 8 d). The two profiles with lower NO_2 values than the true NO_2 profile close to the surface and with a slightly higher PBL and a smoother decrease of NO_2 values (red and green) for lead to a similar situation as observed in our study. The observed VCDs are higher for both spectral ranges than model VCDs and the VCDs for the UV spectral range are lower than for the vis spectral range. For a scenario with a much higher PBL, a constant mixing in the PBL (yellow), and a sharp decrease above, the model values are higher than the retrieved values which we have not observed in our study. For lower PBL with higher NO_2 values (cyan), the differences between the spectral ranges and the model VCDs are less pronounced. Therefore, our observations suggest that compared to the real NO_2 profiles, in the TM5

model the NO_2 is higher in the atmosphere with lower surface concentration values.
We will include Fig. 8 in the supplement and discuss this issue in more detail in the revised manuscript.

-p16, l25-26: the reading nearly suggests that it could be better to use UV retrievals but it is quite unlikely given the larger uncertainties.

We will change the sentence as follows:
For ASE and ANE during biomass burning season, the UV NO_2 VCDs and the model VCDs agree quite well, while the vis NO_2 values are higher. It should however be kept in mind that in the vis retrieval, both the uncertainties and the contribution of the a priori are lower, making these results more reliable.

-p18, l35: It is stated that 'concurrent measurement of the same air mass from different view geometries could yield insight on aerosols and vertical distribution' but these measurements do not exist.

We gave an example for developments for possible space-borne measurements in the future. Furthermore, there are already concurrent measurements of the same air mass. Therefore, there might be already the possibility to retrieve these kind of information from GOME-2 measurements using the instruments on the MetOp-A and MetOp-B. For example on the 01 January 2013 (before reduction of spatial resolution of GOME-2A) for a pixel of South Korea, the GOME-2A instrument has an overpass time of 2:04 UTC with a SZA of 61.86°, a LOS of -43.26°, and a RAA of -46.66°, whereas GOME-2B has an overpass time of 1:17 with a SZA of 66.84°, a LOS of 18.41°, and a RAA 137.35°.
We will change the sentence as follows:
Future developments in space-based observation of the Earth's atmosphere could however increase the capability to retrieve vertical NO_2 distribution. For example, concurrent measurements of the same air mass from different viewing geometries could yield insight on aerosol types and vertical profiles which might be also possible for GOME-2A and GOME-2B observations.

Minor comments:

-p2, l34-35: I find misleading that the examples on past studies on vertical profiling are mostly unrelated to NO2 retrievals as given in the present paper (which considers optically thin atmosphere as for NO2).

The reviewer is right that we gave examples which are unrelated to NO_2. However, a similar method has so far not been used before for an optically thin gas. Therefore, we used examples for ozone which is an optically thick gas. We will point out this differences more clearly and change the paragraph as follows in the revised manuscript:
[...] **In consideration of this fact, knowledge of the vertical distribution of NO_2 can be gained by combining measurements at different wavelengths. The idea of using the penetration depth in the UV to determine vertical profiles of ozone was first proposed by Singer and Wentworth (1957). The use of the temperature dependence of the Huggins absorption bands coupled with penetration depth was proposed to retrieve information about the vertical profile of ozone in the troposphere (Chance et al., 1997). Here, will use a similar method for the optically thin trace gas NO_2.**

-p3, l13: ..path within the NO2 layer relative to the vertical path.

Done.

-p3, l15: environmental effects is too vague.

Done — we will change the sentence as follows in the revised manuscript:
AMFs are calculated by radiative transfer models, which take into account the viewing geometry and environmental effects, e.g., scattering processes in the Earth's atmosphere, SSR and the vertical distribution of trace gases (Platt and Stutz, 2008).

-throughout the manuscript, the author often use the word 'visibility' to express the idea that NO2 signal is more clear 'visible' in some spectral range. It is ambiguous as it might be interpreted by 'visible wavelength'.

In revised the manuscript, we will replace the word 'visible'. Then, it will be only used for the visible spectral range. However we think, it is not necessary to replace the word 'visibility'.

-p3: the example of bAMFs should be presented here.

We will move Fig. 2 to the introduction as suggested and refer to this figure in the later chapters in the revised manuscript.

-p3: the effect of BRF on NO2 retrievals is not developed enough as possible reason between UV and vis

Done — we will change the paragraph as follows in the revised manuscript: **..... For wavelengths in the UV, the BAMF in layers close to the ground is considerably smaller than for the vis spectral range (Fig. 1, revised manuscript; Fig. 2 manuscript). This effect is even less pronounced for longer wavelengths. In general, BAMFs for longer wavelengths have a smaller dependency on altitude compared with BAMFs for shorter wavelengths (Burrows et al., 2011). The altitude of highest sensitivity further depends on the solar zenith angle (SZA). For increasing SZAs, the altitude of highest sensitivity moves upwards to the stratosphere. Furthermore, the surface spectral reflectance (SSR) depends on the wavelength, and therefore, the SCDs are influenced by the SSR (Burrows et al., 2011). Generally, for the UV and vis spectral range the SSR is quite low between 2 and 30% depending on the surface type except for snow or ice (Burrows et al., 2011). For these kind of surface types, the SSR is lower in the UV than in the vis spectral range. For smaller SSR, the UV shows a stronger decrease and for larger SSR (e.g. snow) the UV shows stronger increase towards the surface thus the UV SCDs decrease or increase compared to the vis SCDs. Therefore, the SSR can strengthen the effect of the Rayleigh scattering which can be further increased by higher SZAs. Additionally, aerosols influence the measurements and also the visibility of NO_2 is influenced by the presence of aerosols (Burrows et al., 2011). Depending on the type and the optical thickness of aerosols the influences on the measurement differs.**

-p3, l34: sofar –> so far.

Done.

-p5, l 15: "strong absorption lines" –> "strong differential absorption lines"

Changed as suggested.

-section 2.5: for the SCDs stratospheric correction, are the averages performed for both UV and VIS data separately?

Yes, they are calculated separately for both spectral ranges — we will change the sentence as follows:
For the SCDs, we use the "reference sector method" (Richter and Burrows, 2002; Martin et al., 2002) for both spectral ranges separately, in which a monthly average of SCDs measured over a presumably clean area above the Pacific (180° E to 210° E) is subtracted from all measurements per latitude band.

-p10, l16-17: this is a bit contradictory. If it is below the detection, then how meaningful is the 0.6 SCD ratio?

Yes, that is true — we will change the sentence as follows:
Finally, Fig. 8 (manuscript) shows SCD ratios over the well known shipping lane leading from South India to the Strait of Malacca.

-p13, l8, first word: India–>China?

No, the paper from Hilboll et al. (2017) is about air pollution in India.

-p13, l18: SZA is lower–> SZA is higher?

Yes — Done.

Additionally as suggested by Referee #1, we will change two main points in the revised manuscript:

1. We will add discussion about stratospheric NO_2 and show that it is so far not possible to improve the stratospheric NO_2 retrieval by using different wavelength ranges.

2. We will discuss the possibility of an additional fitting window in the green spectral range. The fitting window in the green spectral range

has a higher sensitivity to the lower troposphere. However, in the green spectral range interferences with the surface are clearly visible and therefore in the revised manuscript, we will include only a case study for China.

References

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