Dear editors and reviewers,

Thank you very much for your constructive comments and advices on our manuscript. Your positive evaluation and comments encourage us and are great help for us. We have carefully considered every comment, and made the corresponding revisions in the revised manuscript (indicated by the 'tracked changes').

Point to point response is following:

General Comments:

1. The choice and quality of the figures can be significantly improved:

1) For many comparisons, the authors created three figures with actual profiles, absolute deviations, and relative deviations, respectively. They decided to only show the relative deviations in the main text and moved the other figures to the supplementary material. I strongly recommend showing the actually retrieved profiles in the main text and (to keep the manuscript concise) move the relative deviation to the supplementary material (e.g. swap Figure 2 and Figure S8, Figure 4 and Figure S12, and so on…). Furthermore, where possible, I recommend to also indicate the PriAM a priori profile in each subplot. Plots of this kind are easiest to read, provide very complete information (relative and absolute deviations can be readily estimated by eye), and allow to directly perceive potential impacts of a priori biases.

Response: Thank you very much for your suggestions. We have considered your advice to change the figures in the main text. The actually retrieved profiles were moved to the main text, and the relative deviations were moved to the supplementary material. And we also changed the corresponding content in the article.

Concerning your suggestion about including the PriAM a priori profile in each subplot, we did not follow this suggestion.

The main reason is that when we include the *a priori* profile in each subplot, the value of the a priori profile is smaller than the retrieved and true profiles. Thus we finally choose not to add the *a priori* profile in each subplot.

2) Furthermore, the vertical extent of some figures might be enhanced to improve the visibility of profile fine structures particularly close to the surface.

Response: Thank you very much for your suggestion. We have checked every figure and enhanced the vertical extent to make them more clear.

3) Reduce line thickness of the profiles for better visibility where necessary.

Response: Thank you very much for your suggestion. We have checked every figure and reduced the line thickness of the profiles to make it better visibility.

4) Regarding labels and legends:

- the axes tick labels are sometimes wrong (see e.g. horizontal axes in Figure S19)
- please double check units (see e.g. Fig. 6, where O4 dSCDs are given in molec cm-2)
- Assure readability of legends
- Avoid long legend labels (an extreme case is Figure 16, as discussed in the specific comments)

Response: Thank you very much for your suggestions. We have checked every figure and made the corrections.

5) Where possible, consider applying the same horizontal axes limits in different (sub-)plots.
Response: Thank you very much for your suggestion. Please note that if the same horizontal axes limits in different subplots were applied, then the results for the low values cannot clearly be recognized by the reader. Thus we did not use the same limits.

2. Some of the wordings in the discussions are not clear to me or at least hard to follow and particularly the final conclusions should be more quantitative. The corresponding paragraphs are listed in the specific comments. A general issue is that authors seem to use the term "systematic deviations" sometimes for the relative differences (hence, considering the sign of the deviations) and sometimes for the general magnitude (independent of the sign) of the systematic deviations. This should somehow be clarified, probably by consistently using the expressions "differences" and "deviation magnitudes" for the first and the second case, respectively.
Response: Thank you very much for your suggestion. We used the term "systematic deviations" to describe the general magnitude (independent of the sign) of the deviations, including the relative and absolute differences. In order to make it more clear, we have revised the manuscript according to your suggestions. We now use "differences" and "deviation magnitudes" for the first and the second case, respectively, and revised the manuscript accordingly.
We also followed the suggestion that the final conclusions should be more quantitative, see our reply to the specific comments.

3. The authors state, that their findings "explain part of the deviations between the AOD retrieved from MAX-DOAS and sun photometers in previous studies" (P2L5 but also P24L12). First, I do not agree with the word "explain" here, since the presented results simply show the same behavior as observed in former publications. In fact, an actual "explanation" for these systematics has already been proposed by Irie (2008), Frieß (2016), and Bösch (2018): they proposed that biases introduced by the a priori assumptions are responsible for the deviations. Second, Tirpitz et al. (2021) have shown, that, in the case of OEM algorithms, these biases can be accounted for by applying a "partial AOT correction": by taking AVK smoothing effects into account, they estimate the fraction of the AOD that MAX-DOAS inversions are actually able to perceive, and by applying corresponding correction factors, the AOD underestimation observed for MAX-DOAS inversions can largely be removed. Ideally, a "partial AOT correction" should also be performed for the AODs in the presented manuscript (as it is expected to remove large parts of the discrepancies). If the authors think this is out of the scope of their study, they should at least mention the above publications and corresponding explanations/correction approaches.
Response: Many thanks for these suggestions! We now state that our results confirm the results of previous studies (e.g. from Irie et al, 2006, Frieß et al., 2016, Bösch et al., 2018) that part of the deviations between the AOD retrieved from MAX-DOAS and sun photometers can be explained by the biases introduced by the a priori assumptions. We introduced a new sub-section (3.3.) which describes the main findings of our study and relates them to previous studies.
Changes in manuscript:
3.3 Comparison with the earlier studies
In this section we discuss the most important findings of our investigations and compare them to the

results from earlier studies. Especially Bösch et al. (2018) and Frieß et al. (2019) investigated the sensitivity of the MAX-DOAS inversion results using synthetic data. But compared to this study, they used less profile shapes (Bösch et al. 2018) or they restricted their investigations to a set of profiles with fixed combinations of shapes and vertically integrated quantities (VCDs and AOD). Most importantly, in this study, we cover a larger range of VCDs and AODs, including especially high values (AODs up to 5, and NO_2 VCDs up to 10^{16} molecules cm^{-2}), while previous studies used maximum NO_2 VCDs of 2×10^{16} molecules cm^{-2} and 3.5×10^{16} molecules cm^{-2}, respectively and maximum AODs of 1. Also our study investigates the trace gas retrievals for a minimum NO_2 VCD of 0.1×10^{16} molecules cm^{-2}. Using these wide ranges of VCDs and AODs revealed new effects and/or confirmed earlier findings in more detail. The most important findings are:

With increasing AOD the retrieved AODs systematically underestimate the true AODs. The underestimation reaches values of >40% and >50% for AODs of 3 and 5, respectively. The largest underestimation is found for Gaussian profiles, while for exponential profiles with scale height of 0.5 km the smallest underestimation is found. These results confirm results from previous studies with similar findings (e.g. Irie et al., 2008; Bösch et al., 2018; Frieß et al., 2019; Tirpitz et al., 2021). However, in this study, the range of AODs and the variety of profile shapes is much larger, which allows a more detailed interpretation of the results. Interestingly, the underestimation is systematically smaller for MAPA compared to PriAM, which indicates that only a part of the underestimation can be attributed to the missing sensitivity of MAX-DOAS measurements towards higher altitudes. In most cases, the larger effect for OE algorithms is probably due to the smoothing effect.

Another important finding of this study is that the NO_2 profiles are not very sensitive to the aerosol profiles confirming similar findings by Frieß et al. (2019).

Further, it was found that the influence of the assumed asymmetry parameter and single scattering albedo have typically a minor effect on the retrieval results. This is an important result, because usually the optical properties of aerosols are not well known. However, for aerosol inversions, the errors can still be up to 25%. Thus it is still important to use reasonable values for both parameters to minimize the remaining uncertainties. For the NO_2 inversion the influence of the asymmetry parameter and single scattering albedo is smaller, similar as found by Hong et al. (2017).

Another important finding of this study is that the NO_2 VCDs either systematically overestimate (for low NO_2 VCDs) or underestimate (for high NO_2 VCDs) the true NO_2 VCDs. Interestingly, these results are rather insensitive to the shape or the AOD of the respective aerosol profiles. The underestimation for high NO_2 VCDs is a new finding which was not reported so far. It is probably caused by non-linearities in the radiative transport for strong NO_2 absorptions. It can reach deviations of more than –30% for a NO_2 VCD of 10^{16} molecules cm^{-2}. A tendency of an overestimation for small NO_2 VCDs was already observed (for OE algorithms) by Frieß et al. (2019), but not discussed in detail. Our results clearly indicate that the overestimation systematically increases towards small NO_2 VCDs (with deviations >50% for an NO_2 VCD of 0.1×10^{16} molecules cm^{-2}). Here it is interesting to note that similar results are found for different profile shapes. This finding is probably caused by the fact that the trace gas VCD is mostly constrained by measurements at high elevation angles and the fact that the trace gas SCDs for these elevation angles only weakly depend on the profile shape.

Overall, the reason for the underestimation of the retrieved NO_2 VCD for low NO_2 VCDs is not yet fully understood. However, for the OE algorithm it might be caused by the influence of the a priori profile on the retrieval result. Interestingly, in this study a similar underestimation was also found for the parameterised algorithm (which was not observed by Frieß et al., 2019). This finding is currently

unexplained, but might be caused by the different radiative transfer models used for the generation of the synthetic data (SCIATRAN) and in the MAPA inversion algorithm (MCARTIM). This aspect should be further investigated in future studies.
Interestingly, an overestimation of the true NO_2 VCDs (derived from direct sun observations) by the retrieved NO_2 VCDs from MAX-DOAS observations was also reported by Tirpitz et al. (2021) for low NO_2 VCDs (but not for HCHO VCDs).
Another important finding of our investigations confirms the results from earlier studies (e.g. Wang et al., 2017; Bösch et al., 2018). Changing the covariance matrix changes also the retrieval results from OE retrieval as it results in different weighting of a priori and measurements in the inversion.

Specific Comments:

1. P3L12: "scattered sunlight"

Response: Thank you for your suggestion.

Changes in manuscript: Spectra of scattered sunlight are measured at different elevation angles (EAs) by the MAX-DOAS instrument.

2. P3L22: remove "aerosol optical depth (AOD) and" here, since the aerosol profiles are the only primary results of the typical inversion.

Response: Thank you for your advice. It is removed.

Changes in manuscript: P4**.** The MAX-DOAS technique basically utilizes the EA dependence of differential absorption structures of O_4 to derive the vertical distribution of the aerosol extinction (Wagner et al., 2004; Frieß et al., 2006).

3. P4L1: cite Wagner 2004 and Frieß 2006 after "… aerosol extinction."

Response: Thank you for your suggestion. The reference was added.

Changes in manuscript: P4. The MAX-DOAS technique basically utilizes the EA dependence of differential absorption structures of O_4 to derive the vertical distribution of the aerosol extinction (Wagner et al., 2004; Frieß et al., 2006).

4. P5L8-11: Hard to understand. Maybe simplify the sentence at this point: "We compare the aerosol and trace gas profile retrieval results from two MAX-DOAS inversion algorithms (PriAM and MAPA, for details, see below) for different aerosol and trace gas scenarios."

Response: Thank you for your advice. The text was changed to make it more clear.

Changes in manuscript: Here, we compare the aerosol and trace gas profiles retrieved from MAX-DOAS by two inversion algorithms (PriAM and MAPA, for details see below) with the input values (used as input for the DSCD simulations) for different aerosol scenarios. We also investigate the effects of the aerosol extinction and optical properties, including single-scattering albedo (SSA) and the asymmetry parameter (AP), on the aerosols profiles retrieved by PriAM in the UV and Vis.

5. P5L11-12: remove the sentence "For trace gas retrievals…" and improve the explanation in Section 2.1. instead.

Response: Thank you for your advice. It was removed in the P5L11-12 and moved to the section 2.1 P6L12-15.

6. Section 2.1: refer to Figure 1 more often in the text to help the reader understand the strategy, e.g.: P6L1: "A set of atmospheric scenarios (orange box on the very left), …"
Explain the two strategies here already (please do this carefully also by referring to the Boxes S1 and S2 in Fig.1), such that the reader understands the entire figure before moving on.

Response: Thank you for your suggestions.

Changes in manuscript: P6: A set of atmospheric scenarios (orange box on the left side), including variations of the viewing geometries, single-scattering albedos, and asymmetry parameters, was used to simulate the SCDs of traces gases and O_4, which will be described in detail in Section 2.2. The first step was to quantitatively evaluate the effect of different aerosol loads on the aerosol inversion (The upper part of the Fig.1). For that purpose the simulated O_4 DSCDs were used as input for the aerosol profile retrievals. The retrieved and input aerosol profiles were then compared in order to characterize the effect of the aerosol properties (in particular the AODs) on the retrieved aerosol profiles. The second step was to quantitatively evaluate the effect of different aerosol loads on the trace gas inversion (the bottom half of the Fig.1). For the trace gas retrievals, we apply 2 retrieval strategies where either the retrieved (S1, red box in the lower half of Fig.1) or the input (S2, red box in the lower half of Fig.1) aerosol profile is used.

7. P6L5: "SCDs" are not introduced and this is should also not be necessary. Change "SCDs" to "DSCDs" here and for all following occurrences of "SCDs". Further, pay attention to consistency: either "dSCD" or "DSCD" (e.g. P6L19

Response: Thank you for your reminding. We checked the full text and changed SCDs to DSCD.

Changes in manuscript:

P6: For that purpose the simulated O_4 DSCDs were used as input for the aerosol profile retrievals.

P7. The differences of the simulated O_4 DSCDs by both models are discussed in section 3.1.2.

P11: The final profiles are weighted averages of the best matching profiles for the given trace gas DSCDs.

P15: Because the aerosol properties used in the MAPA LUT (SSA = 0.95 and AP = 0.68) are different from those used for the simulations of the O_4 DSCDs by SCIATRAN (SSA = 0.90 and AP = 0.72), two sets of O_4 DSCDs for SSA and AP (SSA = 0.90 or 0.95 and AP = 0.72 or 0.68) were simulated by MCARTIM.

P15: Using McArtim for the calculation of synthetic DSCDs, i.e. consistent RTM in forward model and inversion, results in much better agreement, in particular for low AOD.

P19: Part of the systematic underestimation of the MAPA AODs for exponential profiles is probably caused by the differences of the RTM (SCIATRAN v2.2) and settings (SSA=0.9, AP=0.72) used for the simulation of the input O_4 DSCDs and for the MAPA algorithm (MCARTIM, SSA=0.95, AP=0.68), see **Fig. S11**.

8. P6L14: "assumed input profiles" for consistency.

Response: Thank you for this suggestion.

Changes in manuscript: P6. Before the effects of different aerosol loads on the retrieval of aerosol and trace gas profiles were analyzed, some basic parameters were prescribed for simulating the O_4 and trace gas SCDs for the 'assumed input profiles' in the RTM.

9. P7L11: “Section 3.1.2”

Response: Thank you for your suggestion. It was changed.

Changes in manuscript: P7. The differences between O_4 DSCDs simulated by SCIATRAN and MCARTIM are further investigated in Section 3.1.2.

10. P7L16: “wavelengths of 360 nm”

Response: Thank you for your suggestion. It was changed.

Changes in manuscript: As standard settings we chose wavelengths of 360 nm and 477 nm, elevation angles of 1°, 2°, 3°, 4°, 5°, 6°, 8°, 15°, 30°, and 90° (the same as the settings in the CINDI 2 campaign, Kreher et al., 2020).

11. P7L18: The described profile shapes were generally used for different investigations, not only as a priori, right? Maybe keep this more general and remove “as a-priori”.

Response: Thank you for your suggestion. You are right. The described profile shapes were generally used for different investigations. We changed the text as suggested.

Changes in manuscript: P8

In the real atmosphere, a large variability of aerosol and trace gas profiles exists. However, we had to limit our profile shapes to typical profile shapes, which occur in the atmosphere. In this study, three different profile shapes were used, which are Exponential, Boltzmann, and Gaussian profile shapes:

a) Exponential profiles: such profiles are typical if the emissions mainly occur at the surface. During transport to higher layers the concentration systematically decreases with altitude. The scale height depends on the atmospheric lifetime and the vertical transport time. The description for Exponential functions of altitude z as follows:

Exponential: $f_E(z) = A_E(h_E) \times \exp(\frac{-z}{h_E})$ with scale height h_E,

b) Boltzmann profiles: Such profiles represent situations, for which a layer is quickly mixed (compared to the lifetime of the species), and there is a barrier for further upwards transport above that layer. Such situations typically occur for well mixed boundary layers. The description for Boltzmann functions of altitude z as follows:

Boltzmann: $f_B(z) = \frac{A_B(h_B)}{1+\exp(\frac{-(z-h_B)}{0.3})}$ with effective profile height h_B.

c) Gaussian profiles: in our study these profiles describe elevated layers. Such profiles represent situations with long range transport of pollutants, which typically occurs above the boundary layer. Elevated profiles might also occur for aerosols and trace gases which are secondary formed, while air is transported upwards. The description for Gaussian functions of altitude z as follows:

Gaussian: $f_G(z) = A_G(h_G, \sigma) \times \exp(\frac{-(z-h_G)^2}{2\sigma^2})$ with peak height h_G, and the full width at half maximum (FWHM) σ.

12. P8L11: “a priori” instead of “a-priori”. Check further occurrences throughout the manuscript.

Response: Thank you for your suggestion. We checked the full text and changed "a-priori" to "a priori".

Changes in manuscript:

P9: For RTM calculations, vertical profiles of the aerosol extinction ε and NO_2 concentration c are generated by multiplying f with the respective *a priori* column:

Table 1 lists the parameters used for RTM, including solar/viewing geometry, a priori AOD/VCD, and parameters for the different profile shapes.

13. P8L16: "were PriAM and MAPA, as listed …"

Response: Thank you for your suggestion. It was changed.

Changes in manuscript: P9. The retrieval algorithms used in the comparison were PriAM and MAPA, as listed in **Table 2**.

14. P10L17: AODs = 3 are included or? So it should be AODs less-equal 3 instead of less than 3.

Response: Thank you for your suggestion. AODs =3 are included. We changed the text as suggested.

Changes in manuscript: P11. It is worth noting that the maximum AOD in MAPA is 3, since higher AODs were not included in the RTM look-up table; therefore, only aerosol scenarios with AOD $\leqslant$ 3 were included in this study for MAPA.

15. P11L1: Sentence seems messed up: multiple references to Table 1 and a bracket out of place. Please correct/rephrase.

Response: Thank you for your suggestion. We corrected the sentence.

Changes in manuscript: P12. In order to limit the number of investigated profiles, first a sensitivity study with PriAM was carried for the selected profile shapes in Table 1 (these best represent the variety of realistic profile shapes).

16. P11L3-8: I do not understand the criteria on which the authors took the decisions here. This needs to be explained in more detail. Currently, I do not see much value in the presented side investigation. Therefore, alternatively, the corresponding investigation might be removed completely, and instead only the four profiles of relevance for the rest of the study might be introduced in the text and in Table 1. This might also avoid some confusion.

Response: Thank you for your suggestions. We think this investigation should be retained, although similar conclusions for the same profile shape were obtained. But the results were slightly different for the same profile in different heights. For example, when the scale heights of the exponential profile are low, the retrieved profiles are close to the input profiles. But for high scale heights the retrieved scale heights underestimated the true high scale heights. So we chose two exponential profiles. This information was added to the text.

Changes in manuscript: P12: For the exponential profiles, two height parameters were chosen, because for both height parameters systematically different results were obtained: when the scale heights of the exponential profiles are low, the retrieved profiles are close to the input profiles. But for high scale height, the retrieval underestimates the scale height of the exponential profiles.

17. P11L19: "In this Section the effect…"

Response: Thank you for your suggestion. We made the correction.

Changes in manuscript: P12. In this Section the effect of different AOD on the retrieval of aerosol profiles are presented for a scenario with SZA = 60°, RAA = 120°, SSA = 0.9, and AP = 0.72. Note that similar results were found for different scenarios for both PriAM and MAPA.

18. P13L8-11: I cannot follow. On the one hand, the relative deviations (I guess it should be the "relative deviation magnitude") increases with AOD but then it does not? Please clarify.

Response: Thank you for your remark. The sentence "But the relative deviation magnitude does not increase with the increase in AOD." was removed.

19. P14L17: Also add the scale height. It might be useful to add a "default a priori profile"-row to Table 1 with the relevant properties.

Response: Thank you for your suggestion. We added the new Table 4, with information about the *a priori* profile and the *a priori* covariance.

Changes in manuscript: P16. Here it should be noted that an exponential shape with an AOD of 0.2 and a scale height of 1.0km was used as universal *a priori* profile in this study.

Table 4. Parameter settings used in the general PriAM retrieval for aerosol and NO_2 profiles.

Parameters		
a priori profile	Aerosol :	exponential shape with an AOD of 0.2 and the scale height of 1.0km
	NO_2:	exponential shape with the VCD of 1.0×10^{15} molecules cm^{-2} and the scale height of 1.0km
Sa_ratio	Aerosol :	0.1
	NO_2:	0.5

20. P15L1-3: It might be interesting to briefly discuss the motivation for this approach and particularly its relevance for real measurements, where there is basically no information on the vertical distribution or the AOD prior to the inversion. Do have any strategy in mind on how real retrievals might be improved e.g. by iteratively adapting the a priori profile (which is btw. quiet arguable as it violates the OEM principle)? It might be worth discussing this at least at some point (Conclusions?)

Response: Thank you for your suggestion. A discussion of the implications of this study for real observations is added at the end of the paragraph.

Changes in manuscript: P16. In order to investigate the importance of the *a priori* profile for the aerosol profile retrieval, the influence of the *a priori* profile was analyzed by changing the *a priori* profile to different aerosol profile shapes.

P17. This provides a possibility for real measurements to obtain more accurate aerosol profiles if independent information on the *a priori* profiles is available, e.g. from Lidar observations and sun photometers.

21. P15L8-9: "no effect" doesn't seem right here. According to Figure 4, for the Boltzmann and the Gaussian input profiles it has "little effect", while for the exponential input profiles there are significant differences right? Please clarify.

Response: Thank you for your suggestion. The text was corrected.

Changes in manuscript: P16. However, increasing the AOD of the universal (exponential) *a priori*

profile exhibited only little effect on the inversion results of the Boltzmann and Gaussian shapes.

22. Section 3.1.3: What are the values of the off-diagonal elements of Sa? Btw. the default a priori covariance information also be included in Table 1.

Response: Thank you for your suggestion. The values of the off-diagonal elements of Sa are the square of the a priori profile. In order to better explain Sa, a new symbol (Sa_ratio) is introduced. We added the new Table 4 (see above), which contains the description of the *a priori* profile and the *a priori* covariance.

Changes in manuscript: P17

The Sa is the covariance matrix of the *a priori* profile (N×N), and its diagonal elements are the square of the *a priori* state uncertainties with the off-diagonal elements calculated from the Gaussian function with the correlation length of 0.5 km (Frieß et al., 2006).

The diagonal elements of Sa for the aerosol profile were set as the square of the *a priori* profile uncertainty. The standard settings for the *a priori* profile uncertainty were 10% of the *a priori* profile. To describe this ratio, a new symbol (Sa_ratio) is introduced (see Table 4).

Table 4. Parameter settings used in general PriAM retrieval for Aerosol and NO_2 profiles.

Parameters		
a priori profile	Aerosol :	exponential shape with an AOD of 0.2 and the scale height of 1.0km
	NO_2:	exponential shape with the VCD of 1.0×10^{15} molecules cm^{-2} and the scale height of 1.0km
Sa_ratio	Aerosol :	0.1
	NO_2:	0.5

23. P16L1: what does "correlation" mean here? Correlation coefficient?

Response: Thank you for your remark. It was the correlation coefficient.

Changes in manuscript: P17. For the exponential profiles with a scale height of 0.5 km, the correlation coefficient between the retrieved and input aerosol profiles decreased with increasing Sa.

24. P16L5-6: Shouldn't this be the other way round? Also, it is not ideal to talk of "limits" in the context of OEM approaches. I propose: "This is due to the fact that biases towards the a priori profiles are reduced with increasing Sa values."

Response: Thank you for your suggestion. We changed the text accordingly.

Changes in manuscript: P18. In particular, the retrieved surface extinctions and scale heights could be improved by increasing the Sa. This is due to the fact that biases towards the *a priori* profiles are reduced with increasing Sa values.

25. P16L7: Give an approximate altitude for "upper layers".

Response: Thank you for your suggestion. The approximate altitude for "upper layers" was above 2.0 km.

Changes in manuscript: P17. When the Sa values were too large, however, the retrieved aerosol profiles in the upper layer (approximately above 2.0 km) were more unstable.

26. P16L8-11: In my opinion, this is the major finding of the section. However, it only considers the correlation coefficient. What about the actual agreement (e.g. in terms of RMSD). This is probably

the most important quantity to minimise.

Response: Thank you for your advices. We calculated the RMSD according to your suggestions. We found that the RMSD was the smallest when the correlation coefficient was the highest. And we also quantified the RMSD in the manuscript.

Changes in manuscript: P18. The highest correlation coefficient was found when the diagonal elements of Sa were set to the square of 20% of the *a priori* profile for the Boltzmann profiles and exponential profiles with a scale height of 1.0 km at AOD of 5.0, with the smallest root-mean-square deviation (RMSD) of 0.54 and 0.50 (averaged of 360nm and 477nm for each shape), respectively. For the Gaussian profile, the correlation coefficient was highest with the diagonal elements of Sa in 50% of the *a priori* profile. The smallest averaged RMSD of 0.55 was also found for this scenario with values of 0.58 at 360nm and 0.52 at 477nm, respectively.

27. P16L17: AOD less-equal than 3 (?)

Response: Thank you for your question. The AOD is less-equal than 3. We changed the text accordingly.

Changes in manuscript: P18. Note that only the results for AOD $\leqslant$ 3.0 were derived from MAPA. Also the slopes, intercepts, and correlation coefficients are shown in **Fig. 7**.

28. P17L8: Remove double full stop.

Response: Thank you for this hint. It was removed.

29. P17L13: comma after "AOD"

Response: Thank you for this hint. The comma was added after AOD.

Changes in manuscript: P19. Especially for low AOD, the AODs retrieved by PriAM are closer to the input AODs than those retrieved by MAPA.

30. P17L14-19: I would expect the different ways of how a priori information is incorporated in the two retrievals as a major reason: for PriAM this is of course the a priori profile and the a priori covariance. For MAPA a priori assumptions are incorporated in the form of prescribed profiles described few parameters. This might be added as another potential reason.

Response: Thank you for your advice. The information was added.

Changes in manuscript: P19. The different incorporated methods for providing the a priori information is also a potential reason for the differences between the two retrieval algorithms. Prescribed a priori profile and the *a priori* covariances are used in PriAM, while a priori assumptions are incorporated in MAPA in the form of prescribed profile shapes by the chosen parameterization.

31. P18L5: Would be helpful to give the applied values for SAA and AP in brackets here again.

Response: Thank you for your suggestion. It was added.

Changes in manuscript: P20. First, a single aerosol profile was used to simulate the O_4 DSCDs for different SSA (0.8, 0.9, 1.0) and AP (0.68, 0.72) values (See **Table 1**).

32. P18L7: "…using the "correct" SSA and AP values (hence, the same values as they were applied in the corresponding O4 DSCD simulations)"

Response: Thank you for your suggestion. We changed the text accordingly.

Changes in manuscript: P20. Next, the simulated O_4 DSCDs were used to retrieve the aerosol extinction profiles by PriAM using the "correct" SSA and AP values (hence, the same values as they were applied in the corresponding O_4 DSCD simulations).

33. P19L1: What do the numbers "0.01 to 1.5" represent? I guess these are absolute deviation magnitudes in the extinction coefficient? Please clarify and add units if necessary.

Response: Thank you for your remark. The umbers "0.01 to 1.5" represent the absolute deviations of the extinction coefficient. This information was added to the text.

Changes in manuscript: P21. The effect of incorrect SSA and AP values on the aerosol profiles retrieved by PriAM increased with increasing AOD with the absolute deviations of the extinction coefficient increasing from 0.01 to 1.5 km^{-1} as the AOD increased from 0.1 to 5.0.

34. P19L5: Add the 5 VCD values in brackets here again.

Response: Thank you for your suggestion. It was added.

Changes in manuscript: P21. First, the effects of different aerosol extinction profiles on the trace gas profile inversion for 5 NO_2 VCDs (0.1×10^{16}, 0.3×10^{16}, 1.0×10^{16}, 3.0×10^{16}, and 10.0×10^{16} molecules cm^{-2}) were examined using aerosol profiles with 4 AODs (0.3, 1.0, 3.0, and 5.0) (AOD = 5.0 was not included for MAPA).

35. P19L6: Add one sentence here again on "S1" and "S2" to remind the reader of approximate meaning.

Response: Thank you for your suggestion. The information was added.

Changes in manuscript: P21. Two strategies (either the retrieved (S1) or the input (S2) aerosol profiles served as input for the retrievals of the NO_2 profiles) were employed to retrieve the NO_2 profiles (see **Section 2.1**).

36. P19L21: Shouldn't this be "Fig 10" instead of "Fig. S10"?

Response: Thank you for this hint. All the figures were changed due to the suggested changes (the retrieved profiles are now shown in the main text and the relative deviations in the supplementary material). Thus also all figure numbers in the manuscript are changed.

37. P19L19: "…, the magnitude of absolute deviations between the retrieved …"

Response: Thank you for your suggestion. The text was changed accordingly.

Changes in manuscript: P22. For the same aerosol conditions, the magnitude of the absolute deviations between the retrieved NO_2 profiles and the input values increase with increasing NO_2 VCDs.

38. P19L20: "… with increasing NO2 VCDs. However, the relative deviations…"

Response: Thank you for your suggestion. The text was changed accordingly.

Changes in manuscript: P22. However, the magnitude of the relative deviations stays constant (**Fig. S20**).

39. P19L19 – P20L1: I would say, the relative deviations generally increase with AOD, don't they?

Response: Thank you for this hint. The content of P19L19-P20L1 is to introduce the effect of NO_2

VCD on inversion. But the effect of AOD is exactly what you said. The relative deviations magnitude generally increase with AOD for the same NO_2 VCD. It was added at the end of P20L1.

Changes in manuscript: P22. For the same aerosol conditions, the systematic deviations between the retrieved NO_2 profiles and the input values increase with increasing NO_2 VCDs, while the magnitude of the relative deviations increases slightly with the increase of AOD for the same NO_2 VCD.

40. P20L1: What is meant by "The systematic deviations here"? The largest deviation magnitudes? Absolute or relative? Please clarify.

Response: Thank you for your remark. It means the largest deviation magnitudes. We changed the text accordingly.

Changes in manuscript: P22. The largest deviation magnitudes between the retrieved NO_2 profiles and the input NO_2 profile for the exponential NO_2 profiles with scale height of 0.5 km were mainly found below 1.0 km. The largest deviation magnitudes between the retrieved NO_2 profile and the input NO_2 profile appeared below 2.0 km for the other three profile shapes, with the maximum deviation magnitude occurring at 1.0 km and 0.2 km.

41. P20L8-13: Do these findings apply for both algorithms or only for PriAM? Please clarify.

Response: Thank you for your suggestion. These findings apply only for PriAM. This was made clear in the text.

Changes in manuscript: P22. The smoothing effect of PriAM overestimates the NO_2 concentrations around 500 m to compensate for the underestimation of the NO_2 concentrations above 1.0 km. In other words, PriAM yields another solution for the ill-conditioned problem in order to achieve convergence between the retrieved and measured SCDs under the control of the *a priori* profile and its covariance.

42. P20L13 Remove either "uncertainty" or "covariance"

Response: Thank you for your suggestion. We changed the text accordingly.

Changes in manuscript: P22. In other words, the PriAM yields another solution for the ill-conditioned problem in order to achieve convergence between the retrieved and measured SCDs under the control of the *a priori* profile and its covariance.

43. P20L21: Start a new paragraph before "The NO2 profiles…"

Response: Thank you for your suggestion. We added a new paragraph.

44. P21L4-6: What is meant by "singular values". Outliers in single layers? I do not see such things in the figures. Please clarify.

Response: Thank you for your remark. The "singular values" mean the outliers in some layers, which obviously deviated from the true values. This information was added to the text.

Changes in manuscript: P23. An interesting phenomenon was the occurrence of some singular values (outliers which deviate from the true values in some layers) in the upper layers of the retrieved profiles for low NO_2 VCDs (mainly for NO_2 VCD $< 1 \times 10^{16}$ molecules cm^{-2}).

45. P21L17: "…value of the Sa diagonal…"

Response: Thank you for this hint. Sa was added.

Changes in manuscript: P24. As standard value of the Sa diagonal elements for retrieval of NO_2

profiles, we used the square of 50% of the *a priori* profile.

46. P23L5-8: Similarly as for the AOD (see general comments) I would suspect a priori biases as the reason for the systematic deviations. It might be out of the scope of the study, but I encourage the authors to try a corresponding correction: Convolute the input profiles with the retrieval AVKs, recalculate the VCD from the smoothed profile, and compare this "a priori bias-corrected" true VCD to the retrieved VCDs. Do the systematic deviations disappear?

Response: Thank you for your suggestion. Concerning your suggestion about convoluting the input profiles with the retrieval AVKs, we did not follow this suggestion.
The main reason is that the spatial resolution of the input profile (<200m) and retrieved AVKs (200m) is different. So it can't convolve directly.

47. P23L20: add VCD range here

Response: Thank you for your suggestion. The information was added.
Changes in manuscript: P30. In addition, a series of NO_2 scenarios was assumed with the same profile shapes and various VCD values (from 0.1×10^{16} to 10.0×10^{16} molecules cm^{-2}).

48. P24L5: Please provide at least some order of magnitude or a range of deviations.

Response: Thank you for your suggestion. The information was added.
Changes in manuscript: P30. However, for most cases the deviations caused by wrongly assumed AP and SSA were found to be rather small compared to other uncertainties. The maximum relative deviation was generally found around 1.0km with the values of about 25%.

49. P24L12-14: See general comment regarding systematic deviations between MAX-DOAS and sunphotometer observations. Please discuss the actual reasons here and give credits to the corresponding former publications.

Response: Thank you for your suggestion. The information was added.
Changes in manuscript: P31. Such a systematic underestimation has also been found in several previous studies (eg. Irie et al., 2008, Frieß et al., 2016, Bösch et al. 2018, and Tirpitz et al., 2021). The systematic deviation between MAX-DOAS and sun photometers is partly caused by the missing sensitivity of MAX-DOAS observations for higher altitudes, especially for optimal estimation algorithms.
We also added a new sub-section (3.3), see above.

50. P24L17: "…in the RTM model. It…"

Response: Thank you for your hint. We changed the text accordingly.
Changes in manuscript: P31. For MAPA, part of the differences between input and retrieved AODs can be explained by the differences in the RTM model.

51. P25L4-6: This is very likely due to the corresponding reduction of a priori biases. Might be added as a potential explanation.

Response: Thank you for your suggestion. We changed the text accordingly.
Changes in manuscript: P31. The main reason is probably that the corresponding *a priori* bias was reduced.

52. P26L7: What does "single outliers" mean. Single profiles? Single layers? Please explain further.

Response: Thank you for your remark. It means single outliers in same layers. We changed the text

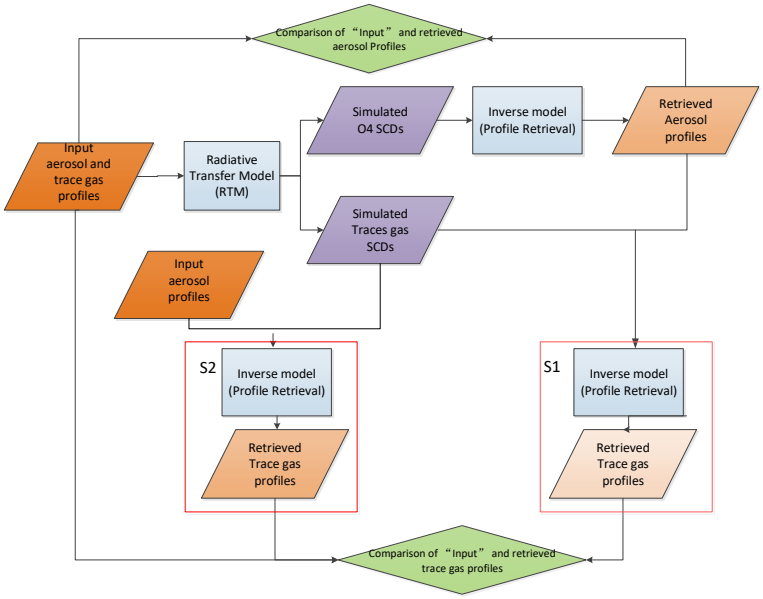
accordingly.

Changes in manuscript: P33. The increase of the Sa values did not improve the inversion results for high AODs, but instead lead to the occurrence of single outliers in some layers.

53. Figure1: Change upper green diamond to "Comparison of "input" and retrieved aerosol profiles". Change lower green diamond to "Comparison of "input" and retrieved trace gas profiles"

Response: Thank you for your suggestion. We changed the figure accordingly.

Changes in manuscript:



54. Figure 2 caption: "where the retrieved AOD exceeds 2". Shouldn't this be "3"?

Response: Thank you for this hint. We changed the text accordingly

Changes in manuscript:

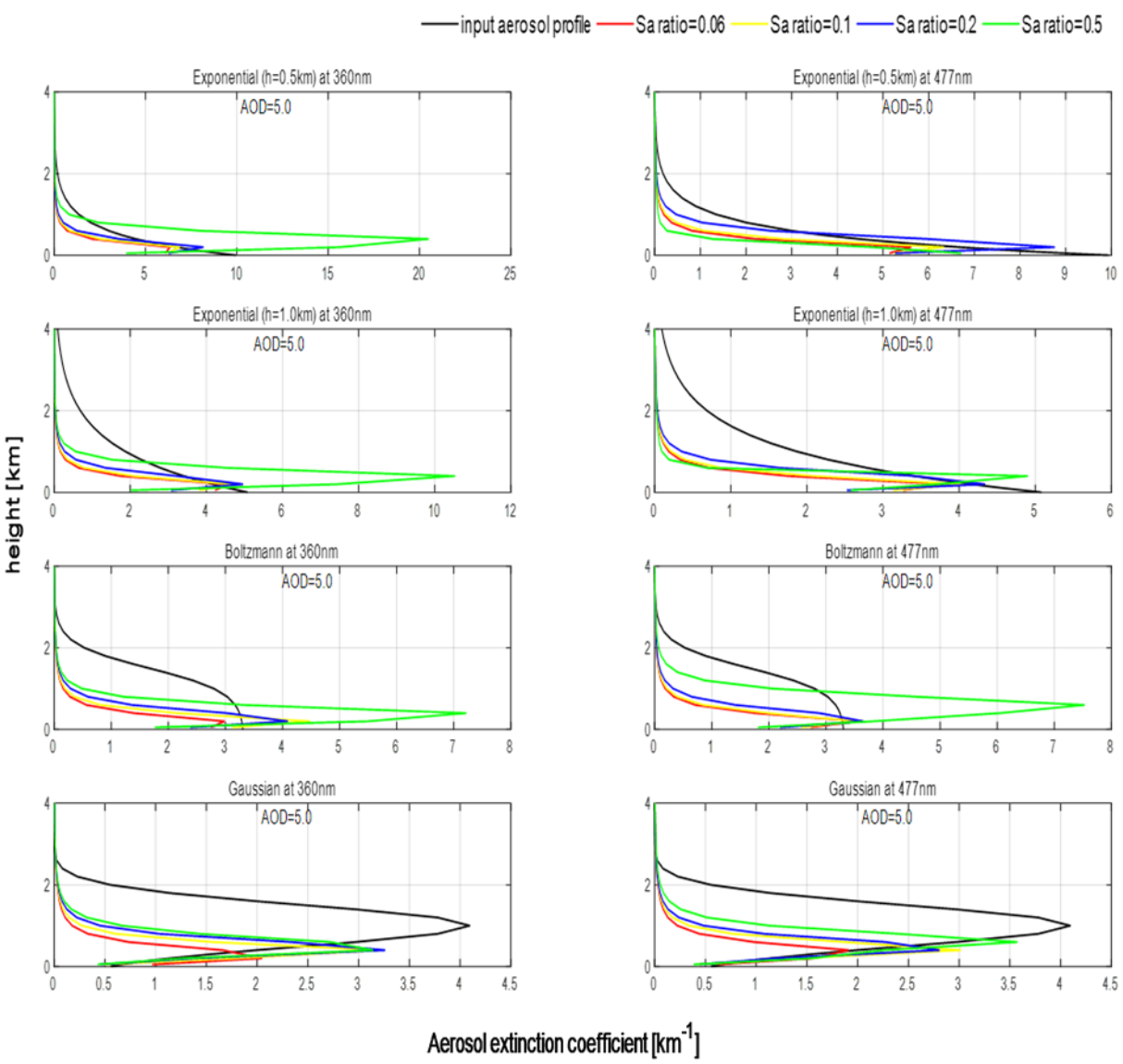
Figure 2. Comparison of the aerosol profiles retrieved by PriAM and MAPA for 360 nm (first line) and 477 nm (second line) and the corresponding input aerosol profiles for (a) exponential shape with h = 0.5 km, (b) exponential shape with h= 1.0 km, (c) Boltzmann shape, and (d) Gaussian shape.

The red and blue curves indicate the results from PriAM and MAPA, respectively. The corresponding relative deviations and absolute deviations are shown in Fig. S8 and Fig. S9, respectively. Note that MAPA by default flags cases where the retrieved AOD exceeds 3, thus the high aerosol scenarios are missing for MAPA.

55. Figure 5: Legend: add a square root over "Sa" or a square to the numbers (0.06^2, 0.1^2, …).

Response: Thank you for your suggestion. The Sa_ratio is introduced in the question 22. Here we used the Sa_Ratio.

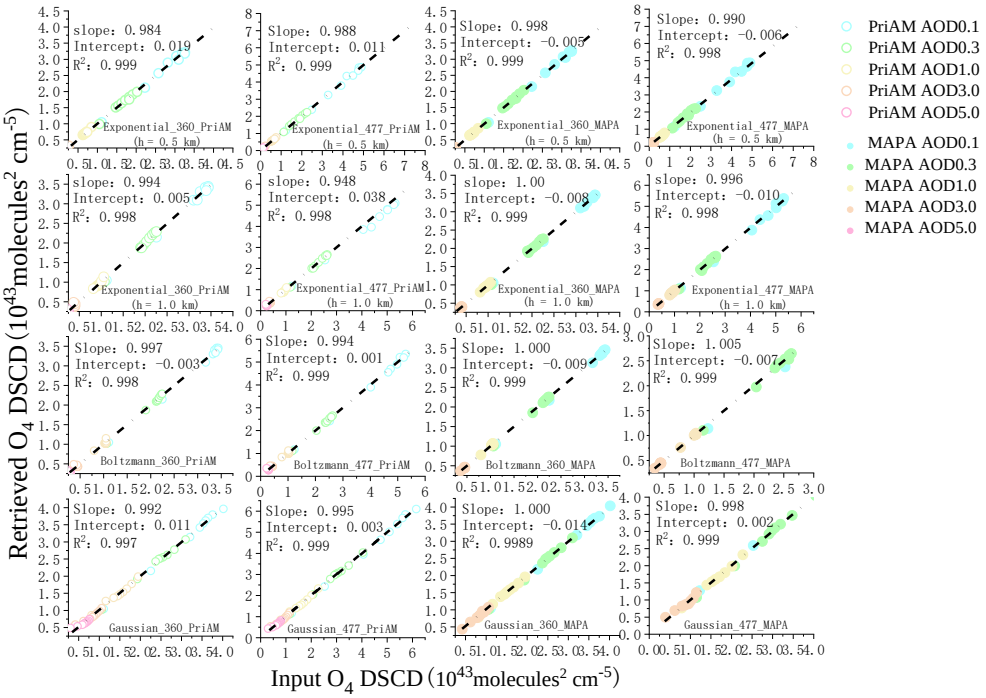
Changes in manuscript:



56. Figure 6: Units should be molec^2 cm^{-5}

Response: Thank you for this hint. We changed the text accordingly

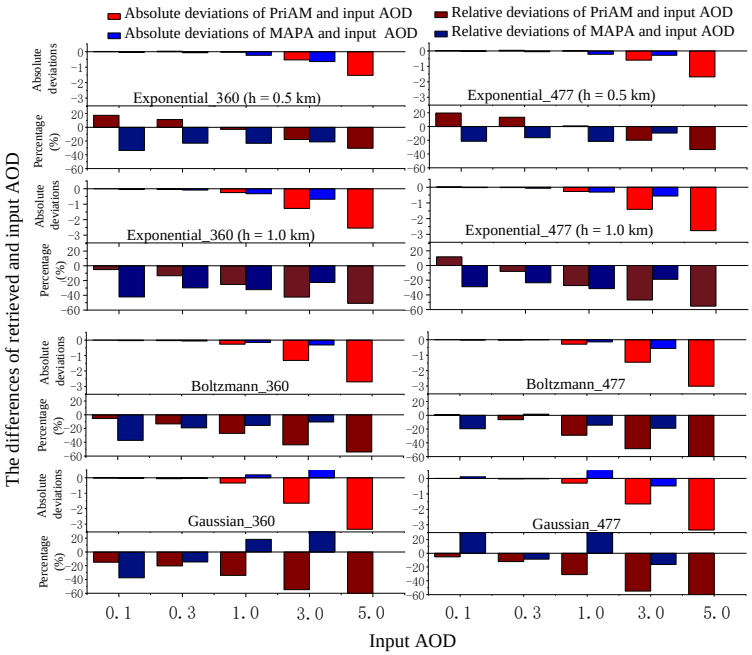
Changes in manuscript:



57. Figure 7, legend: remove typo “deviatiobs”

Response: Thank you for this hint. It was corrected

Changes in manuscript:



58. Figure 10, end of caption: “…shown at the top.”

Response: Thank you for this suggestion. We changed the text accordingly.

Changes in manuscript: **(now Figure 11)**

Figure 11. Retrieved NO_2 profiles by PriAM and MAPA for scenario S1 (see text) for aerosol profiles with 3 selected AODs (0.3, 1.0, and 3.0) and 5 NO_2 VCDs for of (a) exponential shape with

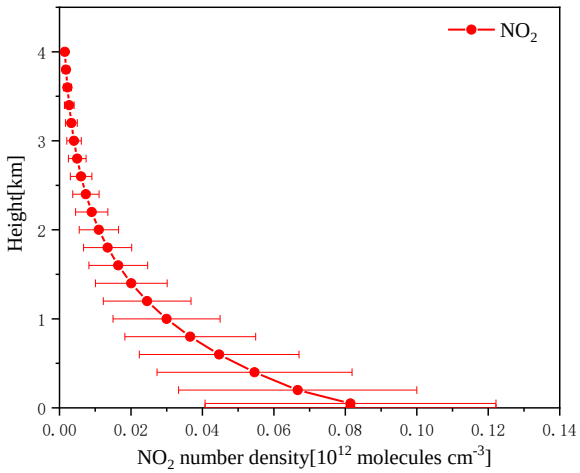
h = 0.5 km, (b) exponential shape with h = 1.0 km, (c) Boltzmann shape, and (d) Gaussian shape.

The solid and dotted colored lines refer to the AODs and algorithms shown at the top

59. Figure 13: Might be useful to add the default a priori uncertainty as error bars or shaded area.

Response: Thank you for your suggestion. The a priori uncertainty was added.

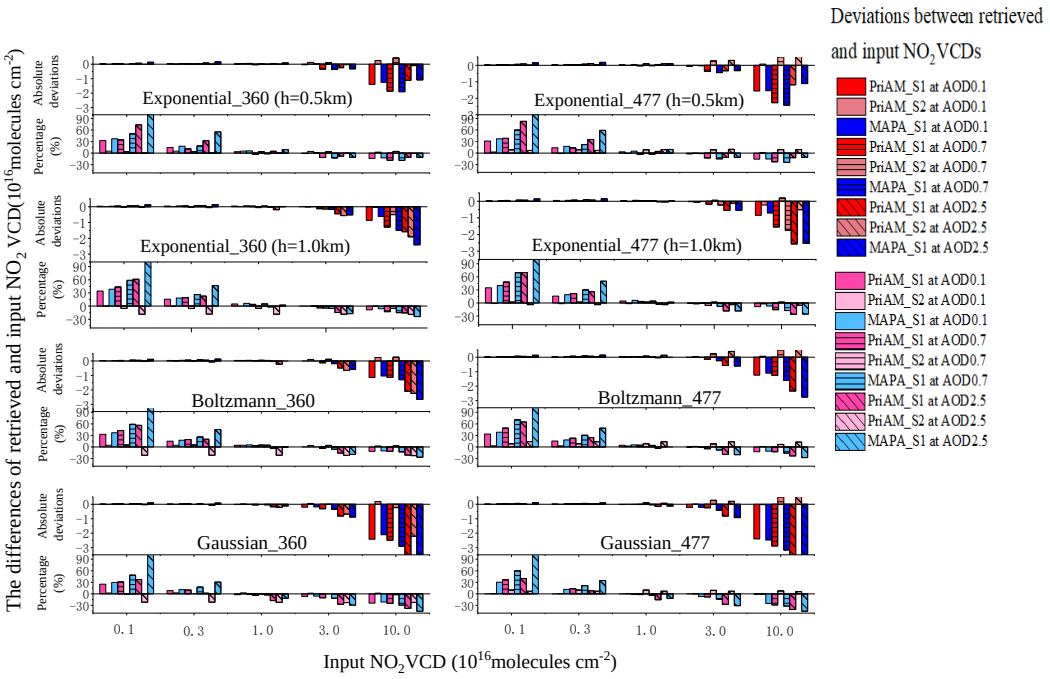
Changes in manuscript:



60. Figure 16: Legend is extremely bulky making it harder for the reader to understand the figure. One column might be enough since absolute and relative deviations are shown in separate subplots. Furthermore, the legend might be equipped with a title like "Deviations between retrieved and input NO2 VCDs". Labels can then be reduced to something like "Priam_S1, AOD 0.3".

Response: Thank you for your suggestion. The figure was revised accordingly.

Changes in manuscript:



Thank you for taking care of our manuscript.

Kind regards,
Xin Tian
E-mail: xtian@ahu.edu.cn

Corresponding author: Yang Wang, Pinhua Xie,
E-mail address: y.wang@mpic.de; phxie@aiofm.ac.cn;