Author reply to Referee #3

Tim Bösch et al.

November 5, 2018

We thank Referee #3 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
- author comments
- old text from the manuscript
- **changed text in the manuscript**

In this paper, Tim Bösch et al. presented a new MAX-DOAS profile inversion algorithm, named as BOREAS. As the author noted, the algorithm could be the first one which retrieve profiles based on optical depths of absorbers. The algorithm is well verified through sensitivity tests with synthetic data and comparisons with various collocated independent data during the CINDI-2 campaign. In general the scientific topic is meaningful.

Thank you very much for the positive comment.

However I think the authors should clarify two major points before publication:

1) The unique feature of the BOREAS algorithm is doing inversion based on optical depths of absorbers. The approach allows including DSOT at different wavelengths in a profile inversion. However the authors do not discuss the improvement of doing inversion with optical depths compared to with slant column densities. If there is no a considerable improvement, innovativeness of the algorithm is doubtable.

The authors would like to clarify that inversion with optical depths is only applied to the aerosol retrieval. In order to make this clear, the following lines will be added to the abstract:
The aerosol profile retrieval is based on a novel approach in which the absorption depth of O_4 is directly used in order to retrieve extinction coefficient profiles instead of the commonly used perturbation theory method. The retrieval of trace gases is done with the frequently used optimal estimation method but significant improvements on how to deal with wrongly weighted a priori constraints and for scenarios in which the a priori profile is inaccurate are presented.
The aerosol approach was chosen in order to avoid the standard perturbation theory method. We believe that the perturbation of an a priori profile, the calculation of the weighting function matrix, the comparison of model and measured dSCD as well as the subsequent perturbation of the next layer, are time consuming and maybe complex steps which can be avoided when utilizing the optical depths. However, it should be noted that for simple atmospheric conditions, larger differences of the profiling results to other community algorithms are not expected. The generally good agreement of algorithms will be presented in the upcoming papers of Frieß et al. (2018) and Tirpitz et al. (2018).

2) In section 4, the author demonstrates that profile inversion can be improved if a priori shape is used to scale a pre-calculated AOT and VCD. However the improvement only works when a priori profile is similar with the true atmospheric profile. How do we know whether or not the a-priori shape is close to the real atmospheric profile for real measurements? If a wrong priori shape is assumed, does the method

even cause larger deviations of retrieved profiles and AOT from the truth?

The knowledge of the true atmospheric profiles is not necessary in order to apply a priori pre-scaling. If the true profile is similarly shaped as the a priori profile, a clear improvement is achieved. When the true profile has a different shape, the pre-scaling does not deteriorate the results. This can be seen in Figure A2 on the example of box profiles for aerosols and in Figure A9 for NO_2. Especially the B3 scenario shape is not even close to an exponential profile. Even though no clear improvement can be observed, no deterioration can be seen either. Additionally, the number of iterations for the aerosol retrieval is reduced when using an a priori profile closer to the true atmospheric conditions.
In order to clarify this, the following lines will be adapted in Section 4.1.1 in the manuscript:

However, no improvement is found when the shape of the a priori profile deviates strongly from the true atmospheric condition (see box profiles in appendix A).
However, no improvement is found when the shape of the a priori profile deviates strongly from the true atmospheric condition but no deterioration either (see box profiles in appendix A).

Specific Comments:

1) In section 3, the authors do not clarify that how the algorithm deals with negative values which could be retrieved.

The aerosol profile retrieval works in the logarithmic space and thus no negative values can be retrieved. The optimal estimation based trace gas retrieval works in the linear space. Here, negative values are possible, especially when the regularization ratio is wrongly weighted. However, a priori pre scaling leads to a better weighting of a priori constrains in addition to the usage of a regularization parameter $g \neq 1$. Negative values are still possible but rare. While negative values are meaningless from a chemical/physical perspective they can be understood as a representation of uncertainties in the respective altitude region from a mathematical point of view.
The following line will be added to Section 3.1 in the manuscript:

Equation 15 is solved in the logarithmic space in order to avoid negative values.

Additionally, Section 3.2 will be adapted:

Equation 18 is solved in the linear space. Negative values are possible but can be avoided by applying appropriate a priori constraints.

2) Line 10 on page 11: aerosols can impact the sensitivities of MAX-DOAS measurements to trace gas profiles, especially at high altitudes. The optimal settings of inversion parameters could depend on aerosols. Therefore sensitivity tests should also be done under typical aerosol conditions, especially a heavy aerosol load.

We fully agree with the reviewers suggestion. However, the BOREAS algorithm participated in a comparison study for synthetic data and real data which will pe published by Frieß et al. (2018) and Tirpitz et al. (2018), respectively. Especially in the first study, various combinations of aerosol and trace gas profiles (also with a heavy aerosol load) are investigated in order to answer this question. Therefore, we would like to avoid repeating similar investigations on this matter in this manuscript.

3) Figure 4 (left): It is hard to identify which color curves are corresponding to individual true profiles. Please try to mark them.

As suggested, the linestyles were changed and the pre-scaled a priori profiles were included (as asked by reviewer #2). The following four figures were adapted for this purpose and will be added to the revised manuscript.

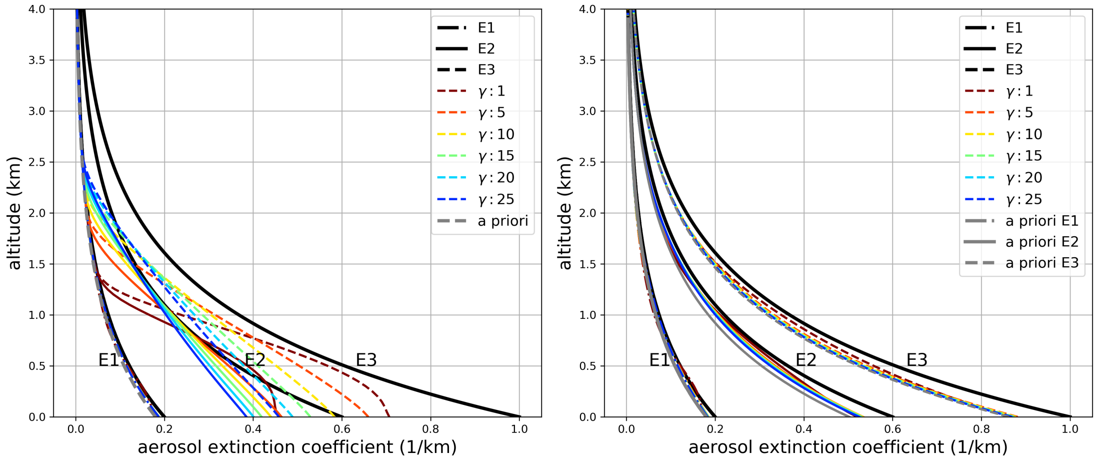

Figure 1: **(Changed version of manuscript Figure 4)** Retrieval results with a fixed (left) and a pre-scaled a priori profile (right) with varying Tikhonov parameters γ for SNR = 3000. Small γ values mean less smoothing of the resulting profiles.

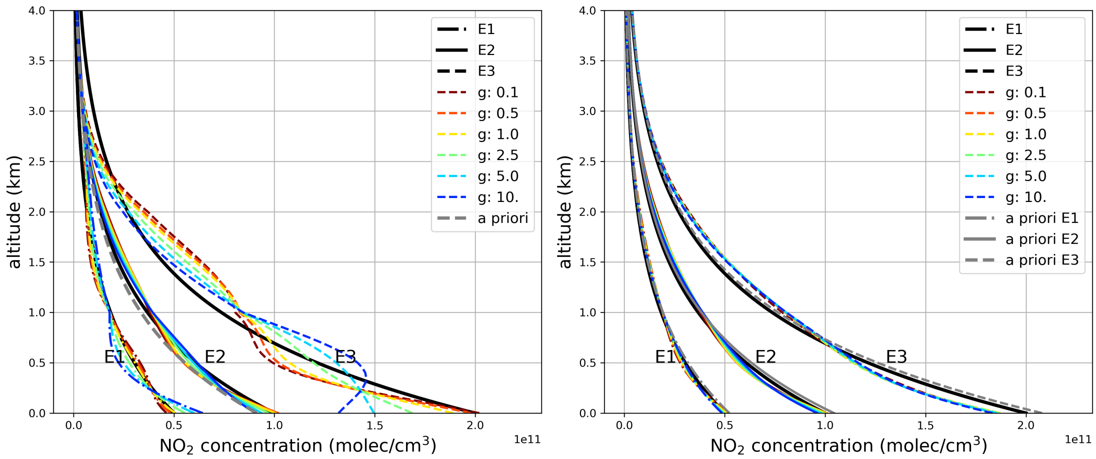

Figure 2: **(Changed version of manuscript Figure 7)** Retrieval results with a fix (left) and a pre-scaled a priori profile (right) with g factors. Small g values mean less measurement weighting for the resulting profiles.

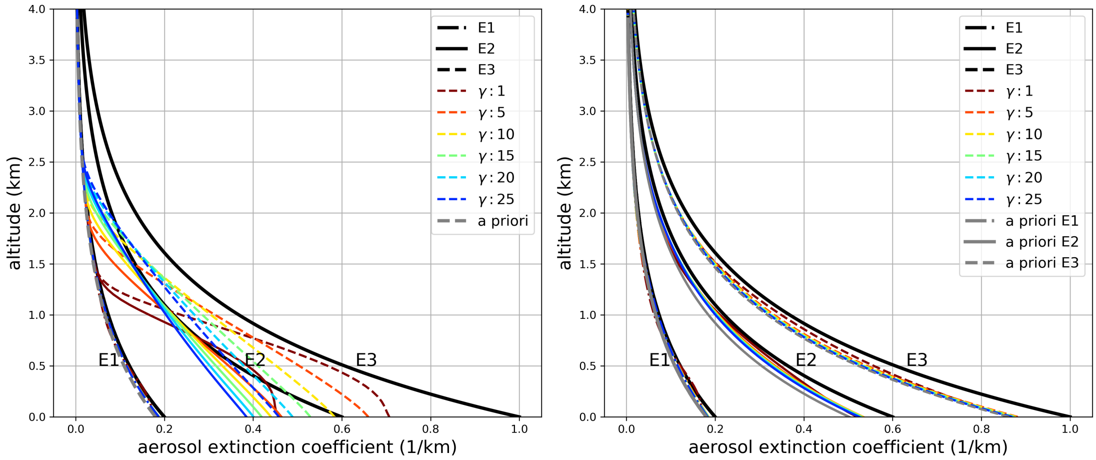

Figure 3: **(Changed version of manuscript Figure A2)** Retrieval results with a fix (left) and a pre-scaled a priori profile (right) with varying Tikhonov parameters γ for SNR = 3000. Small γ values mean less smoothing of the resulting profiles.

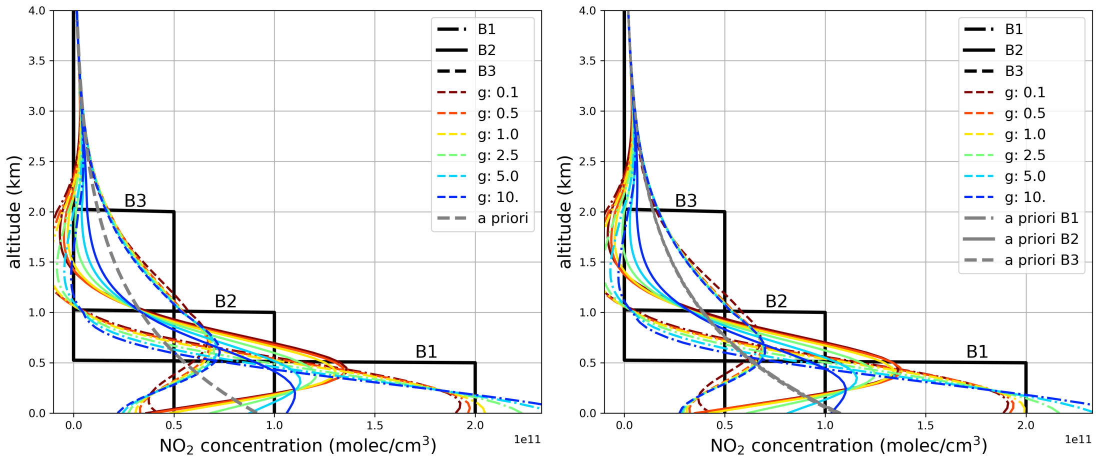

Figure 4: **(Changed version of manuscript Figure A9)** Retrieval results with a fix (left) and a pre-scaled a priori profile (right) with g factors. Small g values mean less measurement weighting for the resulting profiles.

4) Section 4.2: The author claim that total error of aerosol and trace gas profile inversion contains the three parts. However the level of converge, namely differences of modelled and measured SOT, could also contribute some errors. The converge level also depends on the maximum number of iterations.

The RMS between measured and simulated dSOT or dSCD differences for the aerosol or trace gas retrieval, respectively, is a complicated quantity as it does not say a lot about the quality of the retrieved profiles. If constraints on the measurement are chosen too weak, very good agreement can be achieved between measured and modelled slant columns but the resulting profiles often show strong oscillation because of the ill-posed problem of profile inversion. Local minima in the respective RMS quantities exist but their analysis is, especially for aerosols, time consuming and the interpretation depends strongly on the actual atmospheric conditions. Due to this issue, we do not suggest the usage of these quantities for an error analysis with respect to the quality of the retrieved profiles.

5) Why is the smoothing error smaller at altitudes $> 2\,\text{km}$ than that near the surface? We can expect higher uncertainties at high altitudes because MAX-DOAS profile inversion is not sensitive to high latitudes well.

For the retrieval of aerosol profiles, higher altitude a priori variances were set to smaller values in order to reduce issues near the grid boundary where the sensitivity is lowest but small instabilities might lead to strong oscillations. The small errors should not be misinterpreted as lower uncertainties in the specific altitudes but are deliberately suppressed.
The following lines will be added to Section 4.2 in the manuscript to explain this issue:
Note that the errors are nearly zero for higher altitudes because of the height depending variances which were chosen in order to reduce possible retrieval instabilities in altitude regimes where the sensitivity is lowest.

6) Line 5 on page 22: The definition of a-priori variance actually increase constraint of a-priori in the inversion at high altitudes. Do you follow the same definition in the synthetic test? The definition could impact the conclusions of discussions on the optimal settings of Tikhonov, a-priori shape scaling.

For the depicted synthetic scenarios a similar definition was chosen. We agree that this is an additional constraint but the optimal Tikhonov parameter is generally not strongly affected by this choice as the variances are only of minor importance when changing the Tikhonov parameter drastically. The a priori pre-scaling is not negatively affected by this choice. Also a constant variance would yield in a clear improvement under the assumptions discussed in the corresponding sections (4.1.1, 4.1.2).

In addition the definition could also cause that the inversion can not retrieve lifted layers of aerosols and trace gases well. Actually the problem can be seen in Fig. 16 and Fig. 19.

We agree partially. The definition reduces the sensitivity in higher altitudes as we force the result stronger into the direction of the a priori profile. With the assumption of an exponentially shaped a priori, we further assumed the main load/concentration to be close to the surface. While the aerosol retrieval is still able to retrieve elevated layers due to the applied iterations, the trace gas retrieval will fail in retrieving the proper maximum value and altitude. However, in theses cases, the profiling results will still show elevated features as can be seen in Figure 16 but not in Figure 19. Unfortunately, both Figures show two difficulties. Figure 16 depicts elevated layers (08:00 and 10:00 UTC) in altitudes where the chance of a successful retrieval is low, which is independent from the a priori variance. Only the change to another a priori profile would result in a clearly better retrieval response. On the other hand, Figure 19 shows elevated layers with such a small vertical extent that the vertical resolution of MAX-DOAS profiling is simply not sufficient.
The following lines will be added to the manuscript (Section 4.3) in order to clarify that the choice of a height depending variance is of minor importance for the results:

The a priori variance was decreasing with altitude from 1.5 at the surface to 0.01 at 4 km.
The a priori variance was decreasing with altitude from 1.5 at the surface to 0.01 at 4 km. This definition was chosen to improve the profiling results for lower altitudes where the main aerosol load/concentration can be expected and in order to suppress instabilities at the upper grid boundary where the sensitivity is lowest.