Reply to referee J. Joiner

Dear Referee,

Below you will find detailed responses to your review of manuscript amt-2013-38, 'Retrieval of aerosol parameters from the oxygen A band in the presence of chlorophyll fluorescence'. The review helped to improve the manuscript and we would like to thank the referee for her effort and time.

Sincerely,

Bram Sanders (on behalf of the authors)

Note: page numbers in reviewer's comments refer to amtd manuscript, page numbers in responses refer to revised manuscript

--- --- ---

This manuscript describes linear retrieval error estimation of aerosol parameters and chlorophyll fluorescence within the oxygen A-band using simulated data. The paper is clearly written, contains original material, and the topic is timely and appropriate for AMTD. The paper could be accepted for publication after revision, if it is properly placed within the context and scope of previous and current works, with all caveats and simplifications fully and carefully explained including implications for a full retrieval algorithm with real satellite data. This simulation study would be more realistic and meaningful if other parameters are included in the state vector that affect absorption in the oxygen A-band as described below.

Major points:

Dr. C. Frankenberg has already written a thorough review. Since he has made many of the same comments that I would have made, I will expand on a few points. Our group has also conducted a full retrieval exercise using simulated data in and around the O2 A-band, and we have also retrieved fluorescence with the GOME-2 instrument using this spectral region. Please see the following reference that went on line shortly after this paper:

Joiner, J., Guanter, L., Lindstrot, R., Voigt, M., Vasilkov, A. P., Middleton, E. M., Huemmrich, K. F., Yoshida, Y., and Frankenberg, C.: Global monitoring of terrestrial chlorophyll fluorescence from moderate spectral resolution near-infrared satellite measurements: methodology, simulations, and application to GOME-2, Atmos. Meas. Tech. Discuss., 6, 3883-3930, doi:10.5194/amtd-6-3883-2013, 2013.

Reply: Reference has been added (p.2,l.9).

In our work, we used a simulated data set consisting of 230,400 different scenarios. We did not assume that the surface pressure was known, nor did we assume that we had a single, perfectly known atmospheric temperature profile. As Dr.

Frankenberg states, these variables need to be included in a retrieval that uses the O2 A-band. The statement on L 20 of p. 3194 would apply only to a minimum set of fit parameters within a simplified simulation environment. In a realistic scenario, these other variables need also to be considered (in addition to other aerosol parameters mentioned in Dr. Frankenberg's review). In our simulations, the fluorescence and surface reflectance also had realistic spectral variations. When all of these parameters and conditions are included, a physically-based non-linear retrieval algorithm becomes quite difficult and in practice extremely problematic to implement.

Reply: We have now included in our retrieval simulations model errors in the single scattering albedo, surface pressure and temperature profile (see p.12-13). Furthermore, we have added calibration errors to the measurement error covariance matrix (next to the noise error). Finally, surface albedo and fluorescence emission in retrieval are described by a second-order polynomial (although we expect based on the literature that a linear wavelength dependence captures most of the spectral variation across the relatively small fit window). Effects of model errors on retrieval are investigated by including model parameters in the state vector with appropriate a priori errors. Thus, reported precision levels are estimates of the sum of retrieval errors due to measurement errors and smoothing errors but also model parameter errors. This approach takes into account that errors in retrieval parameters and forward model parameters may be correlated. We then take these precision values to represent realistic precision levels. Model errors in the aerosol phase function and the presence of more than one scattering layer are also discussed (p.16,l.17-28).

The simulations that the reviewer is referring to explicitly concern fluorescence retrieval. The intended focus of the present manuscript however is the effect of fluorescence on aerosol retrieval (hence the title). We have not made this clear enough, as we apparently have given the impression that the manuscript was more directed towards fluorescence retrieval itself. We have modified the manuscript throughout and now focus more explicitly on aerosol retrieval.

Concerning the reviewer's remark on the retrieval algorithm being non-linear: We have stated more explicitly that we are performing a linear error analysis (p.13,l.8-12). We also mention that if the model is non-linear, convergence may be problematic or multiple minima in the cost function may exist. Investigating the effect of a non-linear forward model is beyond the scope of this paper (see also p.20,l.5-10).

The statement on L.20, p.3194 (numbering amtd manuscript) has been removed. We now simply say 'The main fit parameters are mid pressure of the aerosol layer (P_{mid}), aerosol optical thickness (τ) and surface albedo (A_s).' (p.12,l.18-19) and we then proceed discussing other (model) parameters, such as fluorescence. We also explain that the state vector contains retrieval quantities of interest (aerosol pressure, aerosol optical thickness), other retrieval quantities for which the measurement may contain information (e.g. fluorescence emission), and true model parameters for which the measurement probably does not contain any information (e.g. aerosol single scattering albedo) (p.12,l.9-13). We believe these

formulations are more in line with the purpose of this paper to investigate the effect of fluorescence on O2 A aerosol retrieval.

I am concerned about the statement on p. 3199, "We have noticed in our work on the O2 A band that retrieval precision significantly deteriorates for very specific combinations of ..." parameters. This warrants further investigation. The spectral shape of the aerosol Jacobian varies quite a bit with all parameters, but particularly with surface albedo, aerosol optical thickness (AOT), and aerosol height. The shape of the AOT Jacobian shown in Fig. 2 should be explained. More insight can be obtained by examining the high spectral resolution Jacobian shown in Frankenberg et al. (2011a). There, one can see different behavior for saturated and unsaturated O2 lines. I believe this effect is coming from enhanced absorption due to scattering between ground and aerosol layer. This will increase absorption in line wings or in unsaturated lines, but has no effect on saturated lines where the aerosol brightening effect will cause the Jacobian to have the opposite behavior. Our own simulations (not published) show that this enhanced absorption effect is not significant under all conditions. It decreases with AOT, aerosol layer height, and surface albedo. For example, In cases of low AOT, height, and/or surface albedo, the AOT Jacobian spectral shape will be more similar to those of surface albedo, surface pressure (not included as a state variable in the simulations here), and aerosol height. This may explain in part the mentioned instability under certain conditions and necessitates more analysis. Is there any evidence of the so-called critical surface albedo effect or is this speculation?

Reply: We have observed in our O2 A band simulation studies that for specific scenarios, derivatives can suddenly become strongly linearly dependent and precision deteriorates. However, it is important to note that this is characteristic of O2 A band aerosol retrieval in itself and not so much the result of a simultaneous retrieval of aerosol and fluorescence parameters. If fluorescence is not fitted and is absent in the simulation, we observe the same thing. A small clarification has been added on p.18,l.4-6. We think we should mention this observation, since the paper discusses O2 A aerosol retrieval, but we also think that it is not needed to discuss it any further, as we focus on the effect of fluorescence on O2 A aerosol retrieval.

The suggestions on the difference between saturated and unsaturated lines made by the reviewer are interesting and we will take note of it in our continuing studies of the O2 A band. We agree that the specific shape of the jacobian for the AOT can affect convergence.

Concerning the reviewer's remark on the critical surface albedo: The critical surface albedo is that particular ground albedo for which the continuum reflectance does not depend on AOT. The critical surface albedo depends for example on the aerosol single scattering albedo. For high spectral resolution data, one should in principle be able to distinguish between different AOTs from absorption in the O2 A band. However, one may still expect retrieval from high spectral resolution data to be more difficult for a critical surface albedo case than for a non-critical surface albedo case. From our retrieval simulations, it indeed seems that critical surface albedo cases typically have relatively flat chi-square

functions or suffer from multiple minima. But we must also remark that sometimes conditions that cannot be labeled as critical surface albedo cases show a similar behavior. This is the reason why we wrote: 'These singular cases often occur for optically thin layers over land and may be related (but not limited) to situations of a so-called critical surface albedo (e.g. Seidel and Popp, 2012).' (p.17,l.32-p.18,l.2)

The paper also mentions the neglect of rotational-Raman scattering (RRS). Our calculations of RRS in the presence of aerosol show similar behavior (please note that the title of our RRS paper has changed since the publication of your manuscript); RRS has a complicated dependence on aerosol parameters and surface albedo for the same reasons discussed above regarding absorption. The effects of RRS are more costly to compute than radiances because they involve convolutions with RRS spectra.

Reply: We have done retrieval simulations indicating that the neglect of RRS has a very small effect on retrieved aerosol parameters. The effect of neglecting RRS on retrieved fluorescence parameters has still to be investigated.

The title of the RRS paper has been updated in the reference list.

Note also that the very low radiances within the O2 A-band and large contrast with the continuum make O2 A-band observations susceptible to biases due to instrumental effects such as non-linearity (e.g., seen in SCIAMACHY deep solar lines in the reference listed below and the GOSAT zero-level offset problem also discussed in several papers) as well as stray light contamination.

Reply: A multiplicative offset (calibration error) has now been included in the error analysis. An additive offset may be due to, for example, stray light in case of grating spectrometers. Stray light may be fitted, provided an accurate stray light model is available from calibration. Effects of non-linearity remains to be investigated

This O2 A-band retrieval problem is extremely non-linear and also computationally costly if on-line radiative transfer calculations (including RRS) are required; a table lookup approach appears to be intractable for this problem. Discussion of these implementation details for a realistic physically-based retrieval algorithm would be appropriate in this work.

Reply: Issues related to the forward model being non-linear were already mentioned on p.19,l.13-18. We now also explicitly state that we restrict ourselves to a linear error analysis and that investigating convergence and occurrence of multiple minima is beyond the scope of this paper (p.13,l.10-14). Finally, we have added a paragraph discussing the computational speed (p.19,l.6-12).

In short, due to all of these issues, we decided to implement and test a principal component analysis approach as described in our paper as opposed to a physically-based approach where one attempts to disentangle all of the parameters that have

significantly correlated effects on radiances. Using thousands of observations for "training" with both simulated and actual satellite data (including cloud-contaminated pixels with real GOME-2 data, because cloud-contamination is a serious issue for any of the instruments mentioned in the paper), we are apparently able to model the O2 A-band complexity with a reasonable number of principal components. However, in our simulations, we do not get particularly good fluorescence retrievals when using only the O2 A-band wavelengths with a fitting window similar to the one used here. We get a much better fluorescence retrieval for similar spectral resolution using a fitting window of 715-747nm which is dominated by filling-in of solar Fraunhofer lines, although there is some filling-in of H2O lines and H2O absorption must be accounted for.

In more recent studies since our paper was submitted, we find that removing the Aband wavelengths from the fitting window results in not much if any decrease in retrieval noise with GOME-2 data. We compare better with GOSAT when the oxygen A-band wavelengths are removed from the fit (relatively small biases in the tropics are removed). We also tried to retrieve fluorescence with GOME-2 using just a small fitting window around the A-band similar to the one used in this paper. We obtained poor results (noisy retrievals as in the simulations and biased with respect to GOSAT).

Reply: The intended focus of the manuscript is really the effect of fluorescence on aerosol retrieval. In the revised manuscript, we refrain from any remarks concerning the optimal way to perform fluorescence retrievals (except a remark on computational efficiency on p.5,l.8-11).

The point we want to make in our manuscript is that an error analysis indicates that retrieval precision of aerosol parameters is acceptable even when fluorescence is included in the fit. Retrieving fluorescence parameters together with aerosol parameters will hardly deteriorate retrieval precision of aerosol parameters as suggested by the simulations investigating the dependence of retrieval precision on the a priori error in the fluorescence emission.

I disagree with the statement on L28 of p. 3198 that "there is no reason... to determine fluorescence from spectral regions outside the O2 A band." If given the choice, based on our simulations and experience with GOME-2, we would choose regions outside the O2 A-band over the O2 A-band for fluorescence retrievals. I agree with Dr. Frankenberg that such statements in a published paper can be very dangerous if used to justify a particular choice of expensive space-based hardware. I also agree that it would be nice if we could use only the A-band for fluorescence retrievals since there are several available instruments with this band. That is why we did give it a try. But in the end, it did not work very well, at least with our principle component approach that apparently works well with radiances outside the O2 A-band.

Reply: See previous point. In the revised manuscript, we refrain from any remarks concerning the optimal way to perform fluorescence retrievals.

We also looked at the effects of spectral resolution and found not much improvement using the O2 A-band region for fluorescence retrievals with higher spectral resolution, consistent with your results. However, when using the fitting window outside the O2 Aband, we got significantly better results at 0.3 nm spectral resolution as compared with 0.5 nm. Therefore, the effect of spectral resolution on fluorescence retrievals depends upon the fitting window used.

Reply: The focus of this paper is the effect of fluorescence on aerosol retrieval. Therefore we prefer to restrict ourselves to the O2 A band.

Minor points:

L8, P3196: The use of "fluorescence yield" is confusing. The fluorescence emission should depend on solar zenith angle, but not necessarily the fluorescence yield which is sometimes used synonymously with fluorescence (quantum) efficiency.

Reply: Changed to fluorescence emission (p.14,l.25).

The following reference should also be used for GOSAT fluorescence retrievals:

Joiner, J., Yoshida, Y., Vasilkov, A. P., Middleton, E. M., Campbell, P. K. E., Yoshida, Y., Kuze, A., and Corp, L. A., 2012: Filling-in of near-infrared solar lines by terrestrial fluorescence and other geophysical effects: simulations and space-based observations from SCIAMACHY and GOSAT, Atmos. Meas. Tech., 5, 809-829, doi:10.5194/amt-5- 809-2012.

Reply: Reference has been added (p.2,l.9).