Response to referee #3
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We would like to thank reviewer #3 for the constructive comments that aided us to improve our manuscript. In this document we provide our replies to the reviewer's comments. The original comments made by the reviewer are numbered and typeset in italic font. Line, page and figure numbers in the reviewer's comments refer to the original manuscript. Following every comment we give our reply.

We provided a revised version of the manuscript in which all changes are indicated: Newly added sections are typeset in red. In our reply we give page and line numbers that refer to the revised manuscript, unless otherwise stated.

Major comments:
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1) In the beginning of Section 2.2 the authors state on p. 4924 that the state vector of the retrieval comprises a total of six components (total ozone column c, the surface albedo A s , spectrally linear dependence δA s of the surface albedo, the amplitude a for the linear scaling of the ratio of Raman-scattering reflectance to the Rayleigh-scattering reflectance (using a pre-computed look-up table), a spectral shift Δλ s of the solar spectrum, and a spectral shift Δλ ISRF of the instrument spectral response function. It is not clear to the reader how LINTRAN is used to compute the associated partial derivatives of the radiance (forward model) function with respect to the latter spectral shift parameters. Also, for the determination of the parameter a of the state vector, a look-up table is employed. How does this fit in the LINTRAN forward model? The authors are asked to provide more detail about how these partial derivatives are evaluated with LINTRAN.

Changed: One page 6, lines 176 – 185, the revised manuscript provides more details about the derivatives of our forward model with respect to the parameters to be retrieved. Here, we start the discussion with the fact that the radiative transfer solver LINTRAN provides derivatives with respect to optical properties of the model atmosphere, like absorption optical depth and Lambertain ground albedo. For this, the calculus relies on the forward-adjoint perturbation theory of radiative transfer. Details on this subject go beyond the scope of this manuscript and the interested reader is referred to corresponding literature (e.g. Landgraf et al., 2001; Hasekamp and Landgraf, 2001; Landgraf et al., 2004).

2) p. 4925, equation (9): The meaning of the bold-face quantity K i col is not clear. Is it a matrix or a vector? Or is it the i-th component of a vector? From the middle part of equation (9) it seems that ∂F i /∂c is a scalar quantity. Please clarify in the manuscript.

Changed: Eq. (10): Reviewer is correct, K_i^{col} has to be normal font, because it is the derivative with respect to the total column at the i-th wavelength, hence the i-th component of a vector.

3.1) p. 4926, lines 9-11: "Consequently, when the correct relative profile is used for the scaling approach, the retrieved column can be interpreted as an estimate of the true column." The reviewer has two questions to this sentence: How should one know about the correct relative ozone profile? Such a case only occurs in a "validation situation" for which measured ozonesonde or ozone lidar profiles are available for the satellite retrieval case (i.e. for both the actual time and geolocation for which satellite measurements and ground-based validation data are available). And even for such measurements there will be some observational error so that the knowledge of the "correct

relative
profile" refers to a hypothetical situation.

Adjusted, page 7, line 237 – page 8, line 241: The sentence was not well formulated and changed in the revised version to “Borsdorff et al. (2014) discussed the meaning of e_n in terms of the profile scaling approach. Interpreting the effective column c_{eff} as an estimate of the true column, e_n represents the error made by the choice of the reference profile ρ_{ref} to be scaled in the inversion. Obviously, when the reference profile represents the correct relative vertical trace gas distribution, e_n vanishes.”

In this part of the paper, we summarize the interpretation of the null space error of the profile scaling approach from a theoretical point of view. Borsdorff et al. 2014 showed that the null space error describes the error of the inversion approach due to the particular choice of the reference profile. Only in case the reference profile represents the correct relative vertical distribution of ozone, a scaling of this profile estimates correctly the total amount of ozone. This is an important conclusion because it motivates one of the science questions of our manuscript. We also agree that this situation may only happen in the most optimistic case for particular validation sites but cannot be considered to be true in general. This is the starting point for our discussion in Section 5.

3.2) In the retrieval of the total ozone column for a particular geophysical location, one does not know in advance the vertical profile of the ozone concentration; also its column (resulting from vertical integration over the profile) is not known. Therefore, even if we take the relative profile shape as a priori, the outcome of the retrieval turns out to be an estimate for the total ozone column. The authors are asked to clarify the above sentence on p. 4926 so that it reflects the real retrieval situation in which the correct relative profile shape is not known.

Changed: On page 8, lines 244 – 268, we have added the text “Consequently, two different conclusions with respect to the interpretation of the profile scaling approach can be drawn: (1) aiming for an estimate of the true column, it has to be stressed that accurate a priori knowledge on the relative vertical distribution of ρ_{ref} has to be provided. In that case, the column averaging kernel is not needed for a proper data interpretation. This interpretation is adapted by Lerot et al. (2010) and further elaborated by Lerot et al. (2014), where the reference profile is updated during the iteration using the empirical correlation between the total amount of ozone and its vertical distribution (Ziemke et al., 2011). (2) Alternatively, one can focus on the information provided by the measurement and consider Eq. (14) as the definition of the retrieval product, where the total column averaging kernel describes a weighted altitude integration of the vertical ozone profile. Here, the effective null space error is not part of the error budget of the product and the retrieval depends much less on a priori profile information. However, the proper data use requires detailed knowledge and application of the column averaging kernel.

The comparison of both views on the data product in the context of the product validation is one aspect of this study. For validation purposes, following the first interpretation the retrieved column can be directly compared to total ozone columns inferred from ground-based spectrometer measurements, which are recorded routinely as part of a global measurement network, while for the second interpretation the vertical distribution of ozone needs to be known. For the latter, ozonesonde measurements can be used. However, due to fewer observation sites and less frequent measurements, a corresponding validation is limited in its temporospatial coverage. On the other hand, the advantage of this approach is the minor dependence of the data product on the a priori knowledge of the vertical ozone distribution. Important applications, like the assimilation of the total ozone column in global and regional models, preferably deal with information purely coming from the measurements and thus try to minimize the effect of ozone knowledge originating from a priori data. For such applications, the effective column together with its total column averaging kernel forms a well suited data product.”

4) p. 4926, lines 14-16: Based on Borsdorff et al. (2014) it is stated that the regularization associated with profile scaling is identical with a Tikhonov regularization procedure of the first order employing an infinitely strong regularization strength. What is the meaning of "first order" Tikhonov regularization? Is this associated with employing a first-order approximation for the first derivative in the smoothing operator $L\ n-p$ *?*
The reviewer has some difficulties with the concept of the "infinite regularization strength". What does this really mean? Generally speaking, with the λ parameter in the cost function one may enforce weak or strong regularization. But what happens for infinitely strong regularization in the expression for the cost function (i. e. the norm which needs to be minimized)? How can it be minimized if λ goes to infinity? Isn't there the risk of over-smoothing the problem at hand? - Please discuss in more detail in the revised manuscript.

Changed: On page 8, lines 271 – 272, we have added the remark "Interpreting the profile scaling approach as a particular case of a regularized profile retrieval using Tikhonov regularization of the first order (i.e. using the first derivative in the regularization matrix) with an "infinitely strong" regularization, Borsdorff et al. (2014) showed that the gain matrix reduces to a gain vector g^{col} representing the fitted ozone column,….". Here Borsdorff showed explicitly that for the limit (γ → ∞), γ is the regularization parameter, only the first singular vector and so the scaling of the reference profile is adjusted by the inversion. Certainly, the reviewer is right that generally over-smoothing, i.e. a too strong regularization, leads to the fact that not all information available in the measurement is used. For ozone retrieval in the UV, this would be the case if the spectral range is extended to shorter wavelengths and more profile information becomes available. However in our study, we restrict ourselves to the spectral range of 325nm – 335nm, which is known to be sensitive only to the total ozone column. Thus, we do not see the risk of over-smoothing in this case.

5) Regarding the profile scaling approach to finally retrieve total ozone columns, the reviewer has the following questions:
Generally speaking, a difficulty exists to find the derivative of the simulated radiance with respect to the total columnar ozone, since the radiative transfer model (it is presumed that this is also the case for LINTRAN) requires partial ozone columns for each of its model layers. Therefore, in order to find the Jacobian matrix of the radiance at the TOA (top of the atmosphere) for total column retrieval, one has to define a suitable map between the partial columns in each model layer and the total ozone column. With the forward model, one can always compute the full Jacobian matrix of the TOA radiance with respect to all possible changes of the partial ozone columns (i. e. in all model layers). What is not known is, in which manner each of these partial derivatives will contribute to the Jacobian of the radiance for the total column. Consequently, there are infinitely many ways to force a change, say, by 1 DU ozone, of the total ozone column. The simplest way to treat this situation is to "force" a scaling of the initial profile. The authors call this initial profile there the "reference ozone profile".
Questions: How does LINTRAN calculate the full Jacobian matrix of the TOA radiance with respect to the total ozone column? Does LINTRAN compute new full Jacobian matrices for each iteration step (Gauss-Newton iteration for solving the minimization problem)?

Not changed. This information is already given in the manuscript. Please see Eq. (10) for the calculation of the Jacobian for the total column. Here, the Jacobian is indeed calculated at each iteration step.

6) Question related to the use of the ozone climatology to define a reference ozone profile:

On p. 4927, the authors write that, for a particular geo-location, the Fortuin and Kelder (1998) climatology for ozone is used for finding the appropriate reference ozone profile with which the iterative retrieval (Gauss-newton iteration) is initialized. The Fortuin and Kelder (1998) ozone climatology provides on a monthly basis a single ozone profile for each 10 degree latitude band. And further on the same page it is stated that "Retrievals are performed for three different reference ozone profiles, the US standard ozone profile (NOAA, 1976), the corresponding profile extracted from the climatology by Fortuin and Kelder (1998), which provides ..." The reviewer's questions are connected with the wording "corresponding ozone profile" in the above sentence.
- How does the condition number of the Jacobian matrix K change if one takes either
i) the US standard atmosphere ozone profile, or ii) the corresponding Fortuin Kelder climatological ozone profile, or iii) the measured ozonesonde profile as the respective reference profile?
- How does the dimension of the null-space of K change in either of these situations i), ii) and iii)?
The reason for the above questions is that, with more realistic initial ozone profiles, both the condition number of K as well as the null-space of the associated linear operator can be modified in favor of a more reliable retrieval result for the total ozone column. The authors are asked to discuss this point in the revised version of the manuscript.

Not changed: We assume that the reviewer mean the condition number of K_{prof} as defined in Eq. (10). The condition number of K_{prof} does not change significantly due to the choice of the reference profile and also not the dimension of the null space. The reason why the null space error changes for different choices of the reference profile is different. Here the different choices of the reference profiles determine the shape of the singular vectors v_i of K_{prof}. For Tikhonov regularization of first order in the limit $\gamma \rightarrow \infty$, the inversion adjusts only the component of the first singular vector, which has the relative shape of the reference profile. Consequently, the better the reference profile approximates the real profile the better the total column can be estimated. This explains why the null space error differs for different choices of ρ_{ref} and why the null space error is smallest in our study for the measured ozonesonde profile. Borsdorff et al., 2014, discuss these theoretical aspects in detail and the appropriate reference is given in the manuscript.

7) A question related to the global mean bias and standard deviation of the suggested total column profile scaling retrieval:
How does the suggested profile scaling approach compare globally with the GOME-2 results reported by Loyola et al. (2011) who refer to a global mean bias and standard deviation of -0.28 +/- 0.7. If one considers the results of dataset 2 in Table 1, the mean bias of the authors' results may turn out to be similar to the Loyola et al. (2011) case. For Brewer spectrometers Loyola et al. (2011) find a larger global bias of -1.22.
i) How is the global performance of the suggested algorithm when considering the Brewer stations?
ii) Is there any solar zenith dependence of the presented results, and is the solar zenith angle dependence similar for all ground-based spectrometers used in the comparison?

7.i) Not changed: We do not distinguish between Dobson and Brewer instruments in the validation and hence we did not adjust the revised manuscript. However, when only using Brewer stations in the validation of the effective column approach, we obtain global bias in the order of about -0.1%.
7.ii) Not changed: Here we refer the reviewer to Table 2 which contains solar zenith angle dependences of a sub set of validation stations. The table shows that the solar zenith angle dependencies vary from instrument to instrument and overall, the largest solar zenith angle dependencies are found for Dobson spectrometers.

8) Total number of figures:
A total number of 19 figures in this manuscript is considered to be too high. The authors are asked

the reduce the total number to, say, 15, either by leaving out certain figures, or by suitably combining the information of some figures into a single one.

Changed: The number of figures has been reduced to 13 in the revised version of the manuscript. Some information has been moved into tables and figures showing redundant information have been removed. The following figures have been removed from the original script: Fig. 3, Fig. 5, Fig. 6, Fig. 8, Fig. 13, Fig. 16. The top panels of Fig. 12 have been removed.

Minor comments:
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p. 4918, line 15: Typo, "Futhermore"
p. 4920, line 2: Typo "measurments"
p. 4966, Figure 14, caption: Typo "(Right panel) Same as right panel ..."
p. 4967, Figure 15, caption: "... and the Lambertian surface albeo 0.1
All typos have been corrected in the revised version of the manuscript.