Interactive comment on “Extending differential optical absorption spectroscopy for limb measurements in the UV” by J. Puķite et al.

Anonymous Referee #1

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Review of

Extending differential optical absorption spectroscopy for limb measurements in the UV

General comments:

This paper describes an extension of an implementation of DOAS to situations, where the atmosphere cannot be considered optically thin any more, and that allows for a more accurate treatment of the wavelength dependence of slant column densities de-
termined as part of the DOAS retrieval. This is achieved by a Taylor-series expansion of the slant column density with respect to wavelength as well as the vertical optical depth of the atmosphere. The approach is convincingly demonstrated to improve minor constituent profile retrievals from (a) synthetic observations and (b) from satellite limb-scatter measurements.

Overall the paper is interesting and well written. Apart from the specific comments below I have 2 more general comments. First, the paper is relatively long, and includes several different sub-studies. The length somewhat distracts from the main approach of the paper, and I ask the authors to consider reducing the size of the paper. Second, a whole category of retrieval approaches (usually called ‘global fit’ or ‘full retrieval’ approaches) are not mentioned at all, except at the very end of the paper. The global fit approach is not affected at all by the retrieval issues dealt with in this study (wavelength dependence of the SCD, problems for non-optically-thin conditions), and is therefore an alternative – existing and demonstrated – to the Taylor-series approach described in the current study. I ask the authors to include appropriate references to the global fit approach, e.g. to the retrieval of minor constituent profiles from limb-scatter observations [e.g., Rozanov et al., 2005].

Furthermore, the manuscript contains many little inconsistencies and issues – discussed in detail in the list of specific comments below – that need to be addressed. Major revisions are required before the manuscript is acceptable for publication. However, that the revisions only require adjustments and improvements of the text, but no additions or corrections of the simulations and retrievals.

Specific comments:

Page 2920, line 4: ‘Therefore, they are strictly valid for weak absorptions and narrow wavelength intervals (strictly only for monochromatic radiation)’

I find this statement problematic, because I don’t think it is true when posed in such a general way. A global fitting approach – where the SCD is not determined as an inter-
mediate product – is not affected by the problems you indicate, but it can also be based on DOAS (it doesn’t have to), and can be applied not only to weak absorptions and also for quite large spectral windows. The problems only appear, when slant columns are determined first, followed, e.g. by the inversion to vertical profiles.

Page 2921, line 18: References are not listed in chronological order

Page 2922, lines 8 – 11: “This AMF depends ... absorbers like e.g. ozone, clouds, aerosols ...”

The sentence may be misunderstood as suggesting that clouds and aerosols are also absorbers. I suggest to change it to: “... of absorbers like, e.g. ozone, as well as clouds, aerosols, albedo ...”

Page 2923: You discuss different techniques that in some way take into account the wavelength dependence of the SCD, but the method most relevant to your application (limb measurements) is not mentioned at all: The full retrieval (or global fit) method applied e.g., by Rozanov et al. [2005] to retrieve NO2, BrO and OClO profiles from SCIAMACHY limb measurements. This method is not affected at all by the issues you’re addressing with you Taylor-series approach and must be mentioned and discussed in your paper.

Page 2926: ‘Limitations for measurements of scattered light’

Here again you suggest, that DOAS applications in general are affected by the problem of the SCD being wavelength dependent, and I believe this is not true. The 2-step approaches, where the SCDs are determined first as an intermediate data product, followed by the inversion of the SCD profiles to vertical absorber concentration profiles, are indeed affected. However, the full retrieval technique – also being a DOAS method – mentioned above is not.

Page 2926, lines 16 – 18: ‘ Longer paths with stronger absorption have a smaller intensity and thus contribute less to the measurement than shorter paths with weaker
absorption’

This is a hypothetical consideration, and does not describe the situation relevant for limb measurements. Isn’t it the case, that the light path for wavelengths with stronger absorptions is actually shorter? At least for limb observations this should be true. For more strongly absorbing wavelengths the LOS optical depth is larger, and therefore, the “average” scattering point along the LOS will be shifted towards the observer – compared to a less strongly absorbing wavelength? I’d expect that the average light path is therefore shorter.

Page 2926, line 20: I suggest replacing ‘In opposite to …’ by ‘in contrast to …’

Page 2927, equation (3): Perhaps I’m missing a point, but it’s unclear to me, why the $S_i$ and $A_i$ are functions of the product of wavelength and optical depth. Shouldn’t $S_i(\lambda \nu)$ be $S_i(\lambda, \nu)$ and $A_i(\lambda \nu)$ be $A_i(\lambda, \nu)$?

Page 2827, equation (3): Perhaps the equations and the more theoretical parts of the paper are easier to follow if $\tau$ is used for the optical depth rather than $\nu$?

Page 2929, line 3: ‘and/or the optical depth ‘ Is this the total and vertical optical depth? I think it is, and I think it should be explicitly mentioned here, because the reasoning motivates the following linearization about wavelength and optical depth.

Page 2929, line 5: ‘and optical depth’ Also here it should explicitly be mentioned that the total vertical optical depth is meant

Page 2929, line 8: ‘… or AMF at wavelength $w$ and vertical optical depth $d.$’ I think this should read ‘… or AMF at wavelength $\lambda$ and vertical optical depth $\nu$ (or better $\tau$).’ $F(\lambda, \nu)$ is linearized about $w$ and $d$ and approximated for wavelength $\lambda$ and vertical optical depth $\nu$.

Page 2929, equation (6): I suggest replacing ‘=’ by ‘≈’, because the latter is also used in equation (7)
Page 2930, equation (8): The right side of the equation should read ‘$S_0 - S_{\lambda} w - S_{\nu} d$’ and not ‘$S_0 - S_{\lambda} d - S_{\nu} w$’

Page 2930, equation (9): Please use another name than ‘$S_i$’ for the product of ‘$S_{\nu} V_i$’, because ‘$S_i$’ was already used for the slant column density of absorber I in equations (1) and (2).

Page 2932, equation (13): I’m confused by the second factor of the last term on the right hand side. In equation (10) this factor was the optical depth, and in equation (13) it is the absorption cross section. This can only be true if $A_{\nu}$ and $A_{O_3}$ are different quantities. However, this is not explained. Perhaps the index $O_3$ suggests that, but it may also just indicate that the derivative is taken w.r.t. the optical depth of ozone only. Please clarify.

Page 2933, line 4: ‘(in the extent of practical applicability)’. This sounds a little odd. I suggest: ‘at the level relevant for practical applicability’

Page 2934, line 1: ‘UV/VIS’ should just be ‘UV’ as the fit window is clearly outside the visible range.

Page 2934, equation (15): again (as in the case of equation (13)) I’m confused by the appearance of the absorption cross section on the right. Perhaps I’m missing an obvious point. Please clarify.

Page 2934, equation (16): I’m sorry, but I fail to understand this equation, and I’m unable to derive this from equation (9). I guess this is related to the point above, and the fact that it’s not fully clear what ‘$S_{O_3}$’ stands for.

Page 2935, line 21: I suggest adding ‘at some wavelengths’ after ‘gives more that 10% underestimation’

Page 2936, line 10: Add ‘wavelength’ to read ‘for the wavelength variation of SCD’. Otherwise it’s not clear what variation it is.
Page 2937, line 11: Why is it a ‘true simulated’ profile? You’re talking about vertical BrO profiles, not SCD profiles (as a function of TH). The latter have to be simulated, but vertical BrO concentration profiles don’t.

Page 2942, line 17: The text says ‘black’ line, but it’s actually green. I suggest adding ‘dashed’ to read ‘green and brown dashed lines’.

Page 2946, line 22: ‘colder temperatures’ -> ‘lower temperatures’ (Can temperatures be colder?)

Page 2947, line 5: ‘a priori variance of 100%’. I guess you mean the square root of the variance corresponding to 100%?

Page 2947, lines 12/13: Perhaps you can mention briefly what ‘match criterion’ was used here.

Page 2947, lines 14/15: ‘except for measurement at Teresina, where only air mass trajectory modelling was performed’. Why’s that? Was the photochemical correction not necessary there?

Page 2948, lines 15 and following: ‘we found that the latter is by 10% larger in the fit window . . .’. Then you write that using the Fleischmann X-sections about 10% larger absorber columns would be expected. Is it trivial, that a 10% increase of the entire cross-section leads to a 10% increase of the differential structure relevant for the DOAS fit?

Page 2949, line 20: ‘The comparison at Aire sur l’Adour shows systematically lower values for SCIAMACHY at lower altitudes than for the ballon observations.’ This sentence doesn’t seem to describe the plot well. Overall the agreement is quite good, it’s the best of the 4 comparisons shown, and the Taylor-series approaches show agreement to within 10%. Therefore I wouldn’t emphasize the systematically lower values for SCIAMACHY too much. If you were talking about the lowest panels, OK.

Page 2950, line 9: ‘. . . is planned to be presented in a publication by Rozanov et al.'
Future plans for a publication that is already 5 years old?

Page 2950, lines 11 – 14: ‘Note that for all compared BrO profile retrievals the agreement with the profiles obtained with the IUP Bremen global fit approach is improved when applying the Taylor series approach compared to standard DOAS’

Here, the global fit approach is finally mentioned. It should be explained that the improved agreement is expected, because the global fit approach intrinsically correctly considers the wavelength dependence of the SCDs.

Page 2950, line 19: ‘Therefore the Lambert-Beer law is not applicable…’ That’s a very strong statement and I believe it’s untenable in this general form. Of course the Lambert-Beer law still holds, and it’s the basis of your analysis. However, some applications based on wrong assumptions will lead to erroneous results.

Page 2951, lines 18/19: ‘…that the agreement … is very similar …’ An agreement can be good or bad, but not similar.

Page 2953, equation (A1): The first factor of the first term on the right hand side should be $S_{0,O_3}^\star$.

Page 2953, line 9: I suggest adding ‘broadband’ to read ‘neglecting the term describing the broadband dependency on wavelength which is skipped in comparison to Eq. (18).’ Otherwise the sentence is difficult to understand, because the retained term also represents a wavelength dependence of the SCD.

Typos etc.:

Page 2923, line 14: ‘of the SCDs FROM optical depth and also FROM wavelength’ -> ‘of the SCDs on optical depth and also on wavelength’

Page 2926, line 3: ‘For DOAS applications on’ -> ‘For DOAS applications in’

Page 2926, line 13: ‘functions from wavelength’ -> ‘functions of wavelength’
Page 2935, line 16: ‘near to the peak’ -> ‘near the peak’
Page 2937, line 2: ‘when appropriate’ -> ‘when an appropriate’
Page 2938, line 1: ‘Fig. 4)’ -> ‘Fig. 4’
Page 2939, line 2: “atmospheric condition’ -> ‘atmospheric conditions’
Page 2939, line 24: I suggest deleting ‘the’ in ‘of the BrO is’
Page 2940, line 5: ‘schema’ -> ‘scheme’
Page 2941, line 18: ‘As comparison’ -> ‘For comparison’
Page 2943, line 16: ‘have very small’ -> ‘have a very small’
Page 2944, lines 10/11: ‘even larger relative discrepancy’ -> ‘an even larger relative discrepancy’ or ‘even larger relative discrepancies’
Page 2944, line 22: ‘near to peak’ -> ‘near the peak’
Page 2944, line 26: ‘the later’ -> ‘the latter’
Page 2946, lines 16/17: ‘... spectral features ... is well accounted ...’ -> ‘... spectral features ... are well accounted ...’
Page 2946, line 20: ‘studied possibility’ -> ‘studied the possibility’
Page 2949, line 22: ‘was modelled’ -> ‘were modelled’
Page 2951, line 4: ‘Applying the Taylor series approach on’ -> ‘Applying the Taylor series approach to’
Page 2951, line 11: ‘prevail’ -> ‘prevails’
Page 2951, line 11: ‘and light path’ -> ‘and the light path’
Page 2951, line 23: ‘with standard DOAS’ -> ‘with the standard DOAS’
Page 2952, line 1: Add ‘the’ to read ‘... AMF modified DOAS the Taylor ...’

Page 2955, line 1: ‘as function of’ -> ‘as a function of’

Page 2956, line 4: ‘Besides of ‘ -> ‘Beside’


Page 2958, line 29: ‘Schemltekopf’ -> ‘Schmeltekopf’

Page 2959, line 31: ‘55 N’ -> ‘53 N’

Page 2961, First line of Table 1: ‘Settings for AMFs comparison’ -> ‘Settings for AMF comparison’

Page 2965: ‘method’ -> ‘Method’

Page 2978, caption Fig. 9, line 4: ‘using ozone profile’ -> ‘using the ozone profile’

Page 2978, same line: ‘also simulation for’ -> ‘also a simulation for’