Atmos. Meas. Tech. Discuss., 2, C1363–C1375, 2010
www.atmos-meas-tech-discuss.net/2/C1363/2010/

Atmospheric Measurement Techniques Discussions

Interactive comment on "Extending differential optical absorption spectroscopy for limb measurements in the UV" *by* J. Puķīte et al.

J. Puķīte et al.

janis.pukite@mpic.de

Received and published: 19 March 2010

Response to Referee #1

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 determined 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.

We thank Referee #1 for the positive feedback on the description of the new approach and the very detailed and constructive comments that helped us much to improve the article.

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.

We agree with the Referee in this point and based on the suggestions we reduced the article length. However we also think that the detailed studies are relevant to fully describe the approach (e.g. we should first demonstrate the improvement in the fit of ozone SCDs and then show the related improvement for the profile retrieval of the minor trace gas BrO). We minimised redundancies in the text (especially in the introduction and by rewriting Sect. 2.2 and Sect. 3). We also moved the description of the Taylor series approach for the AMFs (previously Sect. 2.2.2 and 2.3) to the Appendix. We also restructured Sect. 5 and excluded the side studies w.r.t. the different cross-sections from Sect. 5.3.

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].

We agree with the Referee that the full retrieval approach by Rozanov et al. 2005 needs to be mentioned and discussed. We now describe this method that is not affected by the wavelength dependence of the SCDs in the article accordingly.

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.

We revised the manuscript as suggested by the Reviewer i.e. with respect to the length (by reducing its redundancy and also eliminating or moving some studies to the Appendix) and by a proper credit to the full retrieval approach as well as considering the specific comments by the Referee.

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 intermediate 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.

The Referee is right. Although we say that the problem can be accounted for by different retrievals in the later text of the abstract, this statement is misleading. We reformulate it as follows: 'The Beer-Lambert law is valid only for weak absorptions and narrow wavelength intervals (strictly only for monochromatic radiation)..'

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

We correct as suggested.

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 ..."

We correct: ...absorbers (e.g. ozone), as well as clouds, aerosols...

Page 2923: You discuss different techniques that in some way take into account the wavelength depen-

dence 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.

In general the Taylor series approach is not limited to the applications for limb geometry only. Therefore we discuss it in the context of other approaches that deal with this issue, which were mostly applied for nadir and ground based geometries. We add the reference to Rozanov et al. 2005 and discuss the full retrieval as an approach exclusively for the limb measurements: "For limb observations the so called full retrieval approach was developed (Rozanov et al., 2005), modelling the intensities for all altitudes of one particular limb scanning sequence simultaneously."

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.

Sect 2.1.2 on page 2926 continues the discussion in Sect. 2.1.1. where the standard DOAS was introduced. Therefore the statements here are also meant for the standard DOAS approach. We specify this by adding 'standard' before DOAS : "For standard DOAS applications on satellite nadir geometry"

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.

In our opinion both argumentations are correct and valid and give the same conclusions (as pointed out by the Referee). The question is how one starts the considerations: with absorption strength for certain path possibilities or with the path possibilities for a certain absorption. If we first look on possible paths without absorption (i.e. as determined by scattering only), we see that, after the absorption is included, longer paths will have lower intensity and shorter paths larger intensity. These arguments (preferred in Platt, 1997; Marquard, 2000) are not in opposition to those of the Referee (i.e. considering absorption first and concluding that there are less paths for cases when larger optical depths should be crossed). Both of them in our opinion are generally applicable also for limb geometry. Also, due to the argumentation in the article, the average scattering point (when weighted with intensity) will be shifted towards the observer for stronger absorption thus leading to shorter average light path and lower SCDs, in agreement with the comment of the Referee. We specify the consequence of the shorter average path for stronger absorptions accordingly in the paper by adding: "Therefore the average path will be shorter and SCD lower for stronger absorption."

Page 2926, line 20: I suggest replacing 'In opposite to ...' by 'in contrast to ...'

Corrected as suggested

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 $(\lambda\upsilon)$ be S_i (λ, υ) and A_i $(\lambda\upsilon)$ be A_i (λ, υ) ?

The Referee is right. We corrected this typo.

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

τ is used already for slant optical depth (e.g. in Eq. (11)), therefore we use υ for vertical optical depth.

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.

Yes, it is. We add "vertical" in the respective place.

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

Corrected as suggested.

Page 2929, line 8: '... or AMF at wavelength w and vertical optical depth d.' I think this should read '... or AMF at wavelength λ and vertical optical depth υ (or better τ).' F(λ, υ) is linearized about w and d and approximated for wavelength λ and vertical optical depth υ.

We rephrase (considering that approximation is a generalization for linearization): '...or AMF at wavelength λ and vertical optical depth υ being approximated around wavelength w and vertical optical depth d.'.

Page 2929, equation (6): I suggest replacing '=' by '$\approx$', because the latter is also used in equation (7)

We rewrote the equation as suggested.

Page 2930, equation (8): The right side of the equation should read '$S_0 - S_\lambda w - S_\upsilon d$' and not '$S_0 - S_\lambda d - S_\upsilon w$'

Corrected as suggested.

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

In order to make the description more straightforward we now write this equation directly for one strong absorber s. Now S_s. indicates that S is related to one absorber in contrast to S_υ which is related to the total vertical optical depth.

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_v 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.

This factor is a product of the derivative of AMF w.r.t. the optical depth of ozone and the VCD of ozone (in analogy to respective term in Eq. (9) for SCD). The cross-section arises when dividing VOD by the VCD (because the last one is used to be multiplied with the derivative of AMF w.r.t. the optical depth of ozone). This reasoning is shown for the last SCD term of Eq. (7) in Eq. (9) and we thought that it would be redundant to write this equation again for the AMF. Equation (13) we get if we consider only the absorption of this one strong absorber (so the summation over other week absorbers is not performed) and write Eq. (10) for the AMF of this strong absorber. In order to make the article more straightforward for application in DOAS, we rewrite the Sect. 2.2. showing the Taylor series approach for SCD and its relation to the [slant] optical depth. We motivate there also the approximations e.g. the neglection of minor absorbers. The approach and applications of the method for the AMF description we move to Appendix. We explain the relation between A_v and A_{O_3} by discussing more clearly the strong absorber assumption before Eq. (A2) (Eq. (13) in the old manuscript) and by adding: 'Note that A_{O_3} is a product of A_v and the ozone VCD.'

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

We rewrote the respective text, so this formulation is not occurring anymore.

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

Corrected as suggested

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.

Please see the response for the next point (and also the answer to the comment on

Eq. (13) , few lines above), which will probably clarify the confusion. Similarly as for the case with AMF: The cross-section arises if the VOD is expressed by the product of VCD and cross-section (because the VCD is used to be multiplied with the derivative of SCD w.r.t. the optical depth of ozone) as shown in Eq. (9).

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.

There is a typo in Eq. (16). Eq. (16) is explaining $S_{O_3,O_3}\sigma_{O_3}$, the last term of Eq. (15). Therefore the cross-section should be added also on the right side of the Eq. (16). Also, the replacement of S_i with S_s in Eq. (9) (Eq. 8 in new manuscript) and additional overbrace for VOD and explanation after the equation: "where S_s is the product of S_v and the VCD of the absorber s, and σ_s its cross-section.", should clarify the interpretation. Writing the expression for the optical depth of absorber a, the second index (i.e. a) appears. If $s = a = O_3$, we get S_{O_3,O_3} (means $S_{s,a}$ compare Eq. (10) and (11) in the new manuscript). With these modifications in the manuscript and the more detailed explanations of the points above, we think that the confusion is now avoided in the manuscript.

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

Corrected as suggested

Page 2936, line 10: Add 'wavelength' to read 'for the wavelength variation of SCD'. Otherwise it's not clear what variation it is.

Added as suggested

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.

We meant 'true profile used for simulation'. We skip 'simulated' as suggested by the Reviewer.

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'.

Changed according to the suggestion of the Reviewer.

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

Corrected as suggested

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

The Reviewer is right. We change this formulation to: 'a-priori uncertainty'.

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

We included the match criterion as specified in Dorf et., 2006 as suggested.

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?

In the meantime the photochemically corrected profile is also available for the Teresina measurements and we included it in the article.

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?

In fact we included a low order polynomial in the fit in order to check the increase of the differential structure. However we admit that the relation between different cross-sections and the profiles is not so trivial because the differential structure itself varies with wavelength and this may explain why the actual differences in the profiles are not 10% as pointed out also by Reviewer #2. Our opinion is that this study and discussion is leading away from the main subject of the article and is not affecting the main

conclusions of our study. Therefore we decided to skip this side study noting in the introduction of Sect. 5: "in comparison to the simulation studies larger errors are expected for real measurements. These include effects of the temperature dependency of cross-sections, the spectral calibration, the Ring effect and instrumental problems. For the comparison of measurements from different instruments, trajectory modelling and photochemical correction for balloon measurements contribute to additional uncertainties. Also note that for the balloon retrieval, different cross-section (Wahner et al., 1988) for the DOAS fit was used and the retrieval was performed in another fit window.".

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.

We agree with the Referee and rewrite the sentence as follows: 'For wide altitude ranges, the discrepancy is within 25%, for certain altitudes also below 10% and always within 40% for both Kiruna cases ... and 10% at Aire sur l'Adour in France ...'

Page 2950, line 9: '... is planned to be presented in a publication by Rozanov et al. (2005)' ? Future plans for a publication that is already 5 years old?

This is a typo due to missing reference in the publication list. There is a manuscript by Rozanov et al. in preparation. We correct this.

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.

We point this out as suggested by the Reviewer adding: The improved agreement is ex-

pected, because the full retrieval approach by itself 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.

We correct for this after "Therefore the Lambert-Beer law is not applicable" adding "without accounting for the changes in the light propagation with wavelength....".

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

We correct this as suggested.

Page 2953, equation (A1): The first factor of the first term on the right hand side should be S^*_{0,O_3}.

Corrected as suggested

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.

We thank for this correction and implement the suggestion.

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'

Corrected as suggested

Page 2937, line 2: 'when appropriate' -> 'when an appropriate'

This expression is skipped in the new manuscript

Page 2938, line 1: 'Fig. 4)' -> 'Fig. 4'

This bracket closes the bracket that is opened at last line in previous page.

Page 2939, line 2: 'atmospheric condition' -> 'atmospheric conditions' ?

We agree (if taking into account that it is done also for other simulated cases) and corrected this point.

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 22: 'pp.,2005' -> 'pp., 2005'

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'

We implemented the technical corrections above as suggested and thank again the Reviewer #1 for the detailed comments in order to improve the manuscript.

Interactive comment on Atmos. Meas. Tech. Discuss., 2, 2919, 2009.