

Interactive comment on “Differential optical absorption spectroscopy (DOAS) and air mass factor concept for a multiply scattering vertically inhomogeneous medium: theoretical consideration” by V. V. Rozanov and A. V. Rozanov

V. V. Rozanov and A. V. Rozanov

alex@IUP.PHYSIK.UNI-BREMEN.DE

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Dear Reviewer, we would like to thank you for spending time to review our paper as well as for helpful comments. Please find below our answers to your comments.

1 General Comments

The paper is generally well written and addresses an important discussion with respect to the applicability of different DOAS variants in weak and strong absorption strength regimes. It introduces DOAS and intercompares four commonly known variants and

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the associated air mass factor concepts within a single mathematical framework. This consistent approach allows for a sensitive judgement of the different assumptions and simplifications made.

There are however the following points of criticism, which are specified in more detail in the specific comments that follow.

a) The paper focusses especially on the DOAS analysis of spectra of multiply scattered (MS) Sun light. Contrastingly, the authors relate the DOAS variants applied to these spectra to the direct light (DL) experiment. Whereas for the DL experiment the Beer-Lambert law can be exploited to linearly relate the trace gas number densities to the logarithmic Sun normalised radiance even for the case of strong absorption, this approach is not valid for multiply scattered light in the case of strong absorption. The functional dependence between the radiance logarithm and the number density is therefore not equation (49) but the solution of the RTE in terms of the radiance as a function of the trace gas number density profile. A suitable representation can be obtained e.g. from the Neumann series Marchuk et al. (1976); Marshak and Davis (2005) or employing the equivalence theorem van de Hulst (1980).

Answer: *We completely agree that equation (49) is not suitable to relate the trace gas number densities to the logarithmic Sun normalised radiance for multiply scattered light, especially in the case of strong absorption. The required relation is provided in our study by equation (50) which represents a functional Taylor series for the intensity logarithm. Here, the intensities $I_{\lambda}(\bar{k})$ and $I_{\lambda}(k)$ are indeed solutions of RTE.*

b) Another striking difference between DL and MSL measurements is the wavelength independence of the slant column density. The reason is, that the light path is the same for all wavelengths in DL measurements, whereas it is different for different wavelengths in MSL spectra. The authors try to relate the MSL DOAS SCD to DL DOAS SCD by compelling the wavelength independence. The suggested SCD resp. AMF definition is unprecise and related to a certain setup of DOAS (especially a certain number of fit coefficients) in a certain wavelength window. It may be different for a slightly different

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fit window.

Answer: *The relation of MSL DOAS SCD to DL DOAS SCD by compelling the wavelength independence is used by several DOAS groups since early eighties and remains up to now the standard approximation to employ the standard DOAS technique when retrieving amounts of atmospheric species from multispectral measurements of the scattered solar light (as discussed in Sec.3 of the manuscript). That is why this approximation needs to be discussed in any case. On the contrary, our general definition of AMF given by equation (70) as derivative of the light path with respect to the geometrical path does not contain any relation to DL DOAS. According to this definition AMF depends on the wavelength and is not related neither to any DOAS setup nor to any fit coefficients. Other expressions for AMF given in the manuscript pertain to different forms of the DOAS equation and might be more approximative as the general one.*

c) The paper focusses on satellite DOAS, but this is not properly reflected by the title. The difference becomes evident when analysing MDOAS UV box air mass factors for the retrieval of tropospheric ozone using DSCDs obtained from ground based measurements.

Answer: *The manuscript focuses on the theoretical investigation and generalization of the DOAS equation and AMF concept. The obtained results and relationships between different DOAS equations are independent of the observation geometry. The satellite geometry, specific wavelength region, and absorbing species are only selected to perform numerical experiments when illustrating obtained theoretical results.*

Furthermore there is a lack of description of other features of the DOAS method, potentially interfering with the SCD retrieval as these are for instance described in Wenig et al. (2005). The paper can therefore not be termed a review.

Answer: *As mentioned above this manuscript is intended to describe the theoretical basis of the DOAS technique, analyze the applicability of different DOAS equations, and derive the corresponding air mass factors. With no doubt it can not be treated as a review of all existing DOAS applications and special features.*

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I encourage the authors to explicitly write more about the separability of DOAS and RTM, since it is a key issue in your paper.

Answer: *Although, the separability of DOAS and RTM is of a great importance in practical applications it does not play any role in our theoretical considerations and, thus, is not a key issue of the paper.*

d) The paper is too long and has too many formulas. It is suggested to merge parts of the text as for example equations (9) and (10) in order to increase the readability.

Answer: *We think that a substantial shortening of the paper will cause difficulties in the understanding rather than increase the readability.*

2 Specific Comments

page 703

Equation (2): you should define l_1 and l_2 although it might be clear.

Answer: *Definition of l_1 and l_2 is added.*

line 21: Why does the atmosphere need to be cloud free? I guess due to an increased scattered light contribution.

Answer: *From the theoretical point of view the approach remains the same also for a cloudy atmosphere. However, due to a strong influence of clouds and usually unknown cloud parameters, in practical applications the air mass can not be calculated in a usual manner any more. To account for unknown cloud coverage various approximation have been developed discussion of which, however, is outside the scope of our paper.*

page 707

Equation (12): Does this definition require a constant absorption cross section?

Answer: *This definition does not require a constant absorption cross section.*

page 713

lines 4 to 7: The wavelength dependence might formally be neglected but it will propagate into the lowermost polynomial coefficients, won't it?

Answer: *Depending on that whether the wavelength dependence of SCD is neglected*

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or not different DOAS equations are obtained. Certainly, the polynomial coefficients in different DOAS equations do not need to be the same. This, however, is inessential as the polynomial coefficients are auxiliary parameters.

Please discuss how “greedy” the polynomial is, and how far a wavelength independent SCD definition will be related to the polynomial coefficients. (as for example stated in line 6, on page 740).

Answer: Clearly, the polynomial accounts for the entire broadband component of the spectral signal. However, even a smooth wavelength dependence of a multiplicative term (a term in a product of two wavelength dependent terms) can not be approximated for by an additive polynomial.

However I can not clearly see a benefit of this SCD definition, because the β_k in equation (103) can only be obtained through computationally expensive calculations.

Answer: Equation (103) provides an approach for accurate calculation of the wavelength independent air mass factor for a particular measurement setup. In practical applications, however, more simple approximations are used, e.g., the air mass factor at the central wavelength of the spectral window. The benefit of Eq. (103) is that it can be used as a reference to validate various approximations.

page 716

lines 1 to 3: Please discuss differences between tropospheric ozone UV box air mass factors calculated according to definitions (32) and (57) in combination with (87). What are the implications for retrievals of profiles of strongly absorbing trace gases especially using DSCDs obtained from ground based measurements?

Answer: We do not see any reasons to discuss difference between air mass factors calculated using different expressions. We would like to remind that as demonstrated in the manuscript each AMF has to be considered along with the corresponding DOAS equation. Therefore, the impact upon the retrieval can only be investigated for different DOAS equations with corresponding air mass factors performing end-to-end numerical experiments whereas pure comparisons of air mass factors are not meaningful. This

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investigation, however, is a subject of a separate publication.

page 719

line 6: After introducing $L_{\lambda,j}$ you use it only on the next three pages.

Answer: This is done to avoid an additional superscript ('c' or 'd') in $\ln I_{\lambda,j}(k)$ i.e., $\ln I_{\lambda,j}^c(k)$ or $\ln I_{\lambda,j}^d(k)$ which we need in Sec. 4 of the manuscript only.

page 720

line 1: What exactly is the slant optical thickness when regarding scattered Sun light?

Answer: The slant optical thickness is minus logarithm of the sun normalized intensity (see e.g., right-hand side of Eq. (28)).

If one uses box air mass factors to calculate it in a case of strong absorption, how does it differ from $-L_{\lambda,j}(k)$?

Answer: Using box air mass factors one can only calculate the slant optical thickness of gaseous absorbers. In the framework of our theoretical consideration $-L_{\lambda,j}(k)$ is the sum of the gaseous absorbers, aerosol and Rayleigh slant optical thicknesses.

Of course it is a problem to use the same terms for direct light and scattered Sun light measurements, or not?

Answer: The problem is just that the same notations are used for different quantities.

page 722

Equation (49): This is not the functional relationship between the number density profile of a gaseous absorber and the logarithmic Sun normalised radiance in a MS atmosphere. The correct relationship can be obtained e.g. through the Neumann series or approximately through the equivalence theorem.

Answer: You are right, Eq. (49) is not the functional relationship between the number density profile of a gaseous absorber and the logarithmic Sun normalized radiance in a MS atmosphere. It is also not used in this sense in the manuscript. The valid relationship is provided by Eq. (50) which is the Taylor series expansion of the intensity logarithm.

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page 725

Equation (59): If think instead of k and \bar{k} you wanted to write p and \bar{p} . The expression is generally interesting for other Jacobians as for example derivatives of the logarithmic radiance w.r. to aerosol properties.

Answer: Yes, you are right. We have corrected Eq. (59) as suggested.

page 732

Equation (75): right side of 3rd equation symbol: I think it has to be $d\ln(I(\lambda))$.

Answer: Unfortunately, the font used by AMTD is suboptimal. The symbol you mean is not the normalized intensity $I(\lambda)$ but the path length $l(\lambda)$.

lines 11 to 13: The sentence is problematic and has to be clarified, since the S_λ can be obtained through DOAS, but when obtaining it by RTM the light path information is contained in the $w_{\bar{k}}(\lambda, z)$.

Answer: Of course information about photon paths is contained in $w_{\bar{k}}(\lambda, z)$. This results however from the solution of the RTE whereas in Eq. (12) photon path distribution must be supplied explicitly.

3 Technical Corrections

page 699

line 7: "applied DOAS" ! applied the DOAS

line 21+22: "extention" ! extension

Answer: Done.

page 701

line 1: "This" ! These

Answer: Done.

page 705

line 3: "are unknown at this point polynomial coefficients" ! are polynomial coefficients, which are unknown at this point

line 4: "Clearly, this" ! This

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lines 10 to 11: "the rapidly [. . .] is usually" ! $\sigma_\lambda^d(l)$ is usually

Answer: Done.

page 706 line 5: "As clearly seen," ! As can be seen on the right side of equation (10)

line 10: "trough" ! through

Answer: Done.

page 707 line 18: "coarse" ! course

Answer: Done.

page 709 line 16: "is so-called" ! is the so-called

Answer: Done.

page 713 line 9: "one have to" ! one has to

line 11: "necessary" ! necessarily, "of the scattered" ! of scattered

Answer: Done.

line 18: "in course" ! in the course

Answer: The sentence is rephrased.

page 714 line 1: "who have introduced" ! who introduced

Answer: Done.

page 716 line 9: "As clearly seen," ! Therewith

Answer: Done.

page 719 lines 18 to 19: "as a sum of slowly and rapidly varying with the wavelength components" ! as a sum of two components, respectively varying slowly and rapidly with the wavelength

Answer: Done.

page 721 line 17: "arbitrary differentiable" ! arbitrary but differentiable

Answer: Done.

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page 722 lines 10 to 11: "As clearly seen, at each wavelength, λ , the intensity logarithm" ! As formulated in (49), the intensity logarithm at each wavelength λ

line 16: "Considering" ! Regarding

lines 16 to 17: "can be also obtained" ! can also be obtained

Answer: *Done.*

page 726 line 18: "previos" ! previous

Answer: *Done.*

page 727 line 18: "of the second" ! of second

line 21: "extention" ! extinction

Answer: *Done.*

page 732

line 11: "As clearly seen, S_λ coincides with" ! This means that S_λ is equivalent to

Answer: *Done.*

lines 11 to 13: A major [...] without a knowledge of photon paths. I believe that this sentence does not make sense, since the knowledge about the photon paths is included in $w_k(\lambda, z)$.

Answer: *Please refer to the answer to your specific comment*

page 732 line 16: "is the Fredholm" ! is a Fredholm

Answer: *Done.*

page 733 lines 1 to 2: "for the i-th layer bordered by altitudes z_{i-1} and z_i " ! associated with the altitude layer $[z_{i-1}, z_i]$

Answer: *Done.*

page 735 line 4: "As clearly seen, " ! As can be seen here,

Answer: *Done.*

page 738 line 14: "rewitten" ! rewritten

Answer: *Done.*

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page 739 line 1: "As clearly seen, Eq." ! Eq. **line 16:** "covert" ! convert

Answer: *Done.*

page 740 lines 3 to 4: "Replacing [...], we have:" ! Replacing in this equation the wavelength dependent air mass factor $A_j(\lambda)$ by an constant value A_j , which is currently unknown, we have:

line 15: "spectral window that is in line" ! spectral window. This is in line

line 17: "A more convenient for a practical use equation" ! A practically more convenient equation

Answer: *Done.*

page 741 lines 10 to 11: "Thus, [. . .] of equations:" ! Thus, the complete DOAS procedure to retrieve the vertical column is represented by the following system of equations:

line 21: "is clearly seen" ! has been revealed

Answer: *Done.*

page 742 line 12: "summarize" ! summarizes

Answer: *Done.*

page 744 line 2: "under assumption of a" ! assuming

Answer: *Done.*

page 745 line 6: "where the [...] given by" ! where the weighting function for the entire atmosphere $W_j(\lambda)$ is given by

line 19: "in 425" ! in the 425

Answer: *Done.*

page 746 line 16: "derivative" ! the derivative

Answer: *Done.*

page 748 lines 20 to 21: "calculated [. . .]" ! calculated assuming the absorption cross section to be σ_λ^c instead of σ_λ .

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lines 25 to 26: "its smoothly [...], σ_{λ}^c " \rightarrow σ_{λ}^c

Answer: *Done.*

page 750 lines 8 to 9: "Here, [. . .] given by" ! Here, $W(\lambda)$ is the variational derivative of the intensity with respect to the gaseous absorber number density integrated over the entire atmosphere and is given by

Answer: *Done.*

page 752 line 12: "for a priori ozone" ! for an a priori ozone

Answer: *Done.*

page 753 line 7: "For a sake of" ! For the sake of

lines 13 to 14: "an error canceling is occurred" ! error canceling occurs

lines 16 to 18: "The similar behavior" ! A similiar behavior

Answer: *Done.*

page 754 line 2: "resulted" ! resulting

line 3: "in retrieved vertical" ! in the retrieved vertical

line 6: "that" ! which

Answer: *Done.*

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