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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A modified band approach for the accurate calculation of on-line photolysis rates in stratospheric-tropospheric Chemical Transport Models" *by* J. E. Williams et al.

J. E. Williams et al.

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We would like to thank referee #2 for their extensive review of our paper. We have made substantial changes to the manuscript as a result of this review and feel that we have improved the readability as a result. We answer the suggestions and questions raised by the referee below:

General Remarks:

Many of the referee's comments are related to the style and order in which the results are presented. For instance, the referee objects to the use of appendices in the paper and feels that the details should be moved into the main text. The authors have considered this suggestion and have subsequently moved the information contained in both appendices to Section 3 in line with the referees wishes. Moreover, in a number of cases the referee suggests that we expand the number of Figures in the paper. Although we have added three new figures at the expense of two of those originally included (see below), and considering that there are currently 17 figures in the ACPD version of the manuscript, we feel the inclusion of more figures depicting additional stages of the analysis and additional comparisons (such as the percentage differences which occur between the original and modified band results) is not warranted. The paper has been re-drafted to improve readability, remove forward referencing and to provide more clarity with regard to the discussion of the results.

Specific Comments:

1. Please see the general comment above which outline the modifications related to the appendices.

2. Due to the existing number of plots we feel that the inclusion of additional figures should only be considered if they provide additional information which cannot be elucidated from the figures provided in the original version of the manuscript. To enhance the comparisons we have re-formulated the figures such that the contour plots for both the original and modified band approaches now appear side-by-side. To replace these plots with contour plots of percentage differences between the original and modified methods would mask the magnitude of the associated errors in J values.

3. The term ‘pseudo-absorption’ is used to describe the way in which Rayleigh scattering is included in the photolysis scheme of Bian and Prather (2002). In section 3.1 of their paper, Bain and Prather (2002) state: “An alternative to the full multiple-scattering calculation of the standard model would be to include Rayleigh-scattering effects somehow in a simple exponential attenuation model that depends only on the solar path [Ě] . Thus, we define a pseudo-rayleigh absorption cross section (% of the scattering cross section) that accounts for the loss of photolysis radiation in the lower stratosphere from Rayleigh scattering”.

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4. In order to provide more details as to the motivation behind this assumption we have added the following text to provide more clarity: “A further modification to the band approach is the introduction of a scaling ratio for the first band. An assumption is made in the original band approach that absorption dominates for wavelengths less than 202 nm. This assumption only holds when the single scattering contribution to Fact. can be neglected compared to Fabs. Here the single scattering contribution from a certain model layer scales with the transmission of the atmosphere located above that layer and the amount of radiation deposed in the model layer. Furthermore, the single scattering contribution is proportional to the single scattering albedo, which is the probability that an extinction process is caused by the scattering of radiation. However, this term does not depend on solar geometry. For sza less than 70 deg the single scattering contribution is insignificant because the fraction of radiation deposed in the specific model layer is small and, in combination with the low single scattering albedo higher up in the atmosphere, results in the scattering of light being relatively unimportant. For lower sun, the path length of the solar beam increases markedly which enhances the fraction of radiation deposed in each atmospheric layerĚ.. Therefore, for large slant paths, the scattering contribution becomes a significant part of the total flux in the upper part of the atmosphere. For this reason, it is necessary to use a scaling ratio for sza greater than 72 deg. the first spectral band which accounts for this behaviour. ”

5. The text has been changed accordingly in line with the referee’s wishes.

6. Additional text has been added to the legend of Fig. 1 and text concerning the height of the layer for which the results pertain.

7. We agree with the point made by the referee and provide a more rigorous explanation in the text for the motivation which guided the choice of the new band limits for grids A and B. Therefore we have added the following text: “Due to the shift of the amount of radiation towards longer wavelengths in instances of low sun, the band settings of Landgraf and Crutzen (1998) have to be modified for the band approach to maintain optimal performance at high incident zenith angles.”

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8. This sentence has now been removed from the text as a consequence of the inclusion of a more robust explanation for the motivation for the new band settings.

9. The text has been changed accordingly to: "Figure 1 shows the relative actinic flux t(lamda) = Fact.(lamda)/Fo.(lamda) at 10 km altitude between 300-320 nm normalized to the corresponding value at 310nm across a range of solar zenith angles."

10. Allowing J values to be higher in the middle atmosphere than at the top would play havoc with the chemical tracer fields when applying the method in a full chemistry CTM, especially for species such as O3, whose concentration is highest around 15-25km. The reference to the altitudes at which maximal errors occur has been removed. The explanation of why limits need to be applied to the scaling ratios has been re-written thus: "The most important assumption made in the band approach is that the Fact at any altitude within a specific wavelength band scales with its direct component (Fabs), where the scaling ratio is assumed to be wavelength independent within a spectral band. This assumption has enhanced importance when the amount of incident radiation varies strongly within a spectral band (e.g. band 4). For such instances the approach holds for situations where the diffuse radiation is governed by the single scattering contribution of solar light at the same altitude. In instances where the diffuse component is governed by contributions which originate in other parts of the atmosphere this approximation breaks down. This is the case for low sun and at wavelengths of strong to moderate O3 absorption (less than 305nm), where the direct flux in the lower atmosphere is very small. Here the diffuse radiation mainly originates from scattering at higher altitude levels. A consequence of this is the occurrence of J values which can be larger in the either the lower or middle atmosphere compared to the top of the atmosphere for high incident angles. Thus it would be more appropriate to use the scaling ratio calculated at higher altitudes for the lower levels. However, for the numerical implementation we apply a limit on the resulting scaling ratios where Fabs falls below a selected threshold value, which effectively means using a scaling ratio calculated for higher altitudes."

11. In instances where no limits are applied during the calculation of the scaling ratios the associated errors budgets are only slightly better than those shown for the original band settings for the most sensitive species. Therefore, the critical step in achieving a significant reduction in the associated error for such species is the application of a threshold value for Fabs when computing the scaling ratios. We address this point with the following piece of text: “The dramatic reduction in the associated errors is mainly due to the application of limits on the scaling values as can be seen when comparing Figs. 12a and b. (i.e. the reduction in error due to the use of grid A alone is minimal).”

12. This sentence simply means that limits used for calculating the scaling ratios are applied to all chemical species once the incident zenith angle becomes greater than 85 deg. It does not mean that scaling ratios are applied to all bands. The reader can clarify this by referring to the information provided in Table 2 of the manuscript. The offending sentence had been moved so that this section of text now reads: “For species which exhibit strong absorption characteristics for wavelengths less than 320 nm (e.g. O3, HNO3) limits are needed for sza greater than 81 deg. These were applied to bands 2 and 4 for the species highlighted in red in Table 2, with no limits being applied to bands 1 or 5 through to 8 for sza less than 85 deg. For the latter bands sufficient light penetrates through to the lower layers for wavelengths greater than 320nm such that main assumption used in the band model never fails, even at high zenith angles. Once the sza becomes greater than 85 deg, limits were applied for all chemical species for band intervals 1 to 4 using the (Fabs./Fo) thresholds given in Table 4.”

13. Please see the reply to the general remarks given above.

14. A standard 1-D column model is one where a physical quantity, in this case Fact., for any particular layer depends on the spatial distribution of that layer within the atmospheric column. For clarity we replace this description with “The development and testing of the modified band approach was performed using a standard one-dimensional column model atmosphere.”

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15. All references to the 3D global CTM TM5 have been removed from the manuscript in line with the requests of the referee.

16. In order to address this point we have moved the description of the version of the code to which the results pertain to one section and have changed the text thus " Moreover, for further brevity we limit the discussion below to the errors introduced when using a “final working version” of the modified band approach. This “final working version” was the result of several upgrades made to the fully explicit code driven by the need to remove the most computationally expensive interpolation steps. Therefore, a look-up table for the temperature dependent absorption parameters was produced using a resolution of 5 deg C over the temperature range 180-340 deg C and indexed using the temperature of each atmospheric layer.”

17. The information included in Appendix A has been moved to Section 3 therefore the offending sentence has been removed from the text.

18. Careful analysis of the figure shows that band contributions are given for the entire range in SZA discussed in the manuscript.

19. Information related to the altitude of the model layer has been added in the text.

20. The screening of the far UV by molecular O2 and O3 essentially makes the contributions by bands 1 through to 3 very small for BrNO3 in the lower layers (see Figure 2). Therefore, during the summation in Eqn(5) the contributions made to the final J values due to these first three bands is near to zero. For the top of the atmosphere no such screening occurs meaning that the contributions due to bands 1 to 3 to the final J values maybe non-negligible.

21. This section has been changed so as to provide more clarity in response to the query made by the referee:” In turn, the relative amount of radiation is shifted towards shorter wavelengths, which increases the contributions made by bands 4 and 5. As a result, there is a corresponding decrease in the percentage contribution made by band

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6 to JO3 (-> O1D). The actual percentage contributions are weighted by the absorption characteristics of molecular O3. ”

21(again): The text has been changed accordingly to clarify this issue :” For JBrNO3 a similar effect is observed near ground level, where the contributions from bands 1 to 5 are screened out meaning that JBrNO3 is principally determined by the contributions from bands 6 through to 8. ”. Furthermore, we do not feel that the inclusion of the absorption characteristics for BrNO3 is necessary as such data is readily accessible in the reference literature.

22. The J value for O2 relies on absorption co-efficients which are calculated using the parameterization of Koppers and Murtagh (1996) for a non-scattering atmosphere meaning no scaling ratio is applied to band 1 during the calculation of JO2. Therefore, we feel that the inclusion of a plot of JO2 in Figure 2 is not warranted and would be meaningless. The importance of band 1 to species such as N2O and CFC12 can be seen by considering the information provided in Tables 1 and 4, which show the band limits and the spectral range in which the subset species absorb.

23. In order to enhance the comparison we have moved the error profiles obtained using the original and modified band settings side-by-side in one figure as requested by the referee.

24. The colouring of the contour diagrams has been redone using more suitable colours which have subsequently made the contour legends more legible.

25. Please see the response made to point 2.

26. The text has been changed accordingly: ”Figures 11a-h shows the resulting contour plots for the variation in the errors due to both the original and modified band approaches. To aid comparison the results obtained using both settings are placed side-by-side. The corresponding contour plots for the tropospheric chemical subsets are shown in Figs. 12a-p, respectively.”

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27. We feel that this is a common way of referring to previous work and do not really see the need to reproduce the figure in our own article. The journal in which the original study was published is readily accessible and is not grey literature.

28. Although the effect of ground albedo on the performance of the band approach was not investigated in the original manuscript of Landgraf and Crutzen (1998) we have now removed the figure in line with the suggestions of the referee and given a brief description of the results of this sensitivity test instead. The new text reads “The effect of ground albedo on the resulting errors was tested for those photolysis rates which are most important near to the ground, as this is where the largest perturbation of the radiative flux occurs due to enhanced reflection. For all instances the surface is assumed to behave as a Lambertian Reflector i.e. a homogeneous surface. It was found that the performance of the modified band approach is fairly robust across the range of ground albedos from 0.01 through to 1.0, and, in general, the errors remain rather constant for chemical species contained in the tropospheric subset. Therefore, it can be concluded that the modified band approach can be used with confidence over a diverse range of reflecting surfaces.”

28.(again) The results for incident zenith angles 85-90 deg and 90-93 deg have now been re-analysed and placed side by side in one continuous plot. The inconsistency which occurred between the original figures was due to an erroneous comparison which was made for the range 85-90 deg. The new figure shows much lower error budgets for the chemical species chosen for the Figure.

29. This section has now been re-written and the figure showing the effect of aerosol on the J value profiles has been replaced with one showing a contour plot of the error budgets associated with a selected number of sensitive chemical species. Therefore, the sentence mentioned by the referee has been replaced.

29.(again) This reference to another paper has been replaced with a brief summary of the original figure : “For the tropospheric species the effect of cloud on the J values is

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similar to that shown in Landgraf and Crutzen (1998), and is briefly summarized here. The attenuation of light directly above the highest cloud layer (at 7-8km) causes a substantial increase in the magnitude of the J values at the cloud top (approx. 50%). This amplification gradually disappears with altitude until the J values are approximately equal to clear sky conditions at approx. 20km altitude. There is a corresponding decrease directly below the cloud layer (approx. 20%), which again gradually disappears at altitudes of 2-3km. The lower cloud layer effectively screens the lowest layers resulting in decreases in the certain J values of upto approx. 80%. Those species which absorb below wavelengths less than 320nm are the most affected due to the importance of enhanced scattering in the UV spectral region. The effects above 40km are minimal due to the cloud being situated much lower down the atmospheric column."

30. Due to the fact that the appendices have been merged into the main text many figures have been renumbered. This typo has subsequently been corrected.

31. This section has been re-drafted and the location of the sentence checked.

32. Please see the reply to point (30) given above.

33. Essentially the model atmosphere does not include cloud layers for sza greater than 85 deg. due to the numerical instability issues. Moreover, due to the incident zenith angle using the cloud information for a particular column will not provide the correct coverage (i.e.) the attenuation of the incident beam will be calculated incorrectly. We feel that the errors introduced as a result of this omission will be minimal when considering the daily integrated effect on the chemical fields. A new figure has been added to show the associated error budgets for the four stratospheric species chosen for Fig.7. in the presence of both cloud and aerosol.

34. The description of the spherical reference model has been significantly expanded to provide more details related to the contents and accuracy of the model.

35. More references have been added in line with the comments of the referee.

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36. We have updated the text to: “In turn the intensity field I can be determined using a Picard iteration scheme. However, for the three-dimensional spatial problem a large number of characteristic lines are needed, which hampers any numerical implementation. Here Rosanov et al. (2001) and Doicu et al. (2005) have shown that the number of characteristic lines can be reduced significantly using the symmetries of the model atmosphere and the solar illumination. In this study we utilize as reference the model of Walter et al. which is based on this solution concept. The model has been verified with comparisons made against Monte Carlo simulations for the reflected intensity field and has shown an agreement of better than 2% employing in total 2.4 x 105 characteristic lines. For the actinic flux within the atmosphere we expect a similar or even higher accuracy.”

37. We have significantly expanded the text for this section and provided more mathematical equations in order to give the reader more information regarding reference model A.

38. There figures will be changed accordingly so that the height of the model atmosphere is equal to that used for the analysis.

39. The typo has been corrected in the text

Technical Corrections:

The suggestions and errors made by the referee have all been incorporated.

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