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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.

Anonymous Referee #2

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General comments:

The paper presents an interesting scientific material which is of a great importance for developers of chemical transport models. Although there is nothing really new in the described method, authors do a great job extending an existing model to a wider range of atmospheric conditions. However a very important part of this extention concerning the investigation of the validity of the improved model is done with substantial deficiencies. The contribution of errors arising using a band approach instead of spectrally resolved radiative transfer calculations is extensively discussed and illustrated, although the presentation form is suboptimal, whereas the errors due to usage of a

pseudo-spherical model seem to be treated by author as insignificant. As a results this part of investigation was moved to an appendix accompanied by a quite confusing description which does not allow the reader to draw any conclusion about the validity and quality of this investigation. Moreover, discussion of results and justification of conclusions are not always clear and concise. In general, the paper is suitable to be published in ACP after a moderate revision including a reconsideration of the presentation form and clarifying important issues listed in the detailed comments.

Specific comments:

1. Appendixes should be moved to the main text beacause the material contained here is essential for the subject of the paper. Only technical details which could be skipped by an unexperienced reader are appropriate for a appendix.

2. One of the main subjects of the paper is to demonstrate a reduction of an error due to an improved band structure whereas the error distribution for different species is of minor importance. Thus, it would be much more convenient for a reader if errors of the old and new model were compared in one plot, i.e., each plot should show errors for both old and new model for the same species and the plots for different species can be split.

3. Page 3516: "with Rayleigh scattering being described as a pseudo-absorption" is a very curious formulation. Does it mean that only extinction due to Rayleigh scattering is taken into account?

4.Page 3518: "Due to the strong absorption by O_2 in the spectral range λ = 178.6 - 202.0 nm, the contribution to $F_{act.}$ by scattering in this spectral range is assumed to be negligible for $\theta < 75°$". This statement is not obvious because of very strong Rayleigh scattering in this spectral region. It is not clear why the argument of a strong absorption is valid in this spectral range and not valid at longer wavelength, for example, for strong ozone absorption. Furthermore it is unclear why the scattering is negligible for $\theta < 75°$

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and non-negligible for larger θ (at even stronger absorption!) and why this "negligibility boundary" can be so sharp determined.

5. Page 3518: "purely absorbing atmosphere" - I think it is better to say "non-scattering atmosphere"

6. Fig. 1: It is unclear for which alitude the actinic flux is shown. The explanations to the figure are only valid for a height below the main absorbing layer. For an altitude level within the main absorbing layer the path through this layer always increases with an increasing solar zenith angle.

7. Page 3520: "In other words, the relative amount of radiation is shifted towards longer wavelengths for larger solar zenith angles due to the longer path length of the direct beam through the atmosphere. This effect holds up until a $\theta = 80°$, after which the shift in radiation towards longer wavelengths is weaker. This can be explained by considering the spherical shape of the atmosphere. Until $\theta \approx 80°$ the path of the solar beam through the ozone layer increases with respect to solar zenith angle until the angle becomes so large that the path length actually begins to decrease due to the spherical shape of the model atmosphere." - This is supposed to be a motivation to adapt the band limits for $\theta = 75°$ - $85°$. Following this logic one should keep the same band limits for $\theta > 85°$ as for $\theta < 75°$. But according to Table 1 authors do not go this way. The motivation to do this remains hidden from the reader at least at this point.

8. Page 3520: "In turn, the band limits and δ_i values of the band model become non-optimal, which subsequently results in errors of between 10 - 30% which generally occur in the lowest 10 km of the atmosphere for important tropospheric species (e.g. JH2O2) when the $\theta > 80°$. Moreover, the J values calculated for important stratospheric species such as CFC11 and CH3Br also exhibit relatively large errors for high solar zenith angles, especially around 30 - 40 km, where their photolysis is important." - These sentences can be understood as "This results in an error of 10 - 30% below 10 km and a large error (unspecified!) between 30 - 40 km" which is quite a strange

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formulation.

9. Page 3521: "Figure 1 shows this effect for band 4 between 300 - 320 nm, where the scaling ratios become very large due to dramatic decreases of $F_{abs.}$ for high θ." - Figure 1 does not show any F_{act}/F_{abs} ratios

10. Page 3521: "As a consequence the J values in the middle atmosphere (around 30 - 50 km) may become larger than those calculated higher up the atmosphere for species which exhibit absorbance features below λ = 320 nm, especially for $\theta > 85°$...." - I think, the problem is that J values are wrong, does it really matter whether they are larger than those calculated higher up the atmosphere or not? Why did you split your explanation between 30 - 50 km and lower 20 km? Is the effect in these altitude regions different? What about 20 -30 km?

11. Page 3522: "Although this modification still introduces an error it is much smaller than that calculated without applying such limits" - What about the error introduced if no correction at all is applied for these conditions? How large is it in comparison with the error introduced by the suggested modification?

12. Page 3522: "These limits were applied for all chemical species for instances where $\theta > 85°$" - According to Eq. 2 the scaling corrections are applied to fluxes which are related to species through the spectral bands. Thus this sentence should mean that "These limits were applied to all bands", shouldn't it? If yes, this sentence is contradictory to "No limits were applied to bands 1 or 5 through to 8 under any circumstances." Or did you apply different corrections for the same flux then calculating J values for different species? Or does it mean that there is no species which J values are dominated by the spectral information form bands 5 and 6? Please take into account that your argument "For the latter bands sufficient light penetrates through to the lower layers for $\lambda >$ 320 nm such that the limits are never reached, even at high zenith angles." can not be applied to bands 5 and 6 containing wavelengths shorter than 320 nm.

13. Section 3 should contain the details about model comparisons which are misplaced

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in Appendixes.

14. Page 3524: Please explain what "a standard 1-D column model" is.

15. Page 3524: TM5 is referenced without any explanation. The reference "manuscript in preparation" is absolutely useless for the reader. Please either remove the reference to TM5 or explain in a few sentences the basics of this model.

16. Page 3524: "The final working version was the result of several upgrades to the original code driven by the need to remove the most computationally expensive steps." - This sentence supplies the reader with an ambiguous information. Does it mean that the final version differs from the description in section 2 and 3? Do you want to point out any special differences? Was the original algorithm already described or compared elsewhere and you what to point out that this version is different? Please explain what you really want to say with this sentence.

17. Page 3524: "In Appendix A we have shown that, when applying a modification for the effects of spherical geometry, PIFM-PS is capable of calculating values of $F_{act.}$ within an acceptable error limit across a range of incident θ values up to 90°." - Does it mean that for $\theta > 90°$ the errors are unacceptable?

18. Figure 2: Why do not you show the contributions for $\theta > 90°$

19. Page 3526: "In contrast, due to the overhead O_3 column being relatively low at this altitude and the broad absorption characteristics of $BrNO_3$, contributions are made to J_{BrNO3} across the entire spectral range i.e. bands 1 to 8." - Which altitude do you mean here?

20. Page 3526: "This removes all contributions made by bands 1 to 3 in the lower layers, resulting in a decrease in the J values with respect to height (e.g. Figs. 3a/c)." - I do not understand the logic of this sentence. According to Eq. 1 J is a weighted sum of actinic fluxes in all spectral bands. According to the first part of the sentence the contributions of some bands are removed in lower layers which has to result in

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decreasing J values in this (lower!) layers. How can it result in a decrease in the J values with respect to height?

21. Page 3526: "For $\theta > 80^{\circ}$ the contribution by band 4 increases again due to the sphericity of the Earth8217;s atmosphere. At these geometries the path length of the direct beam through the ozone layer decreases with an increasing solar zenith angle. In turn, the relative amount of radiation is shifted towards shorter wavelengths, which increases the contribution from band 4. The features shown for bands 5 and 6 can also be explained by this effect." - In principle most everything in paper can be explained by the sphericity of the atmosphere. However, I think, the main point here is that the main absorber layer is located in the upper atmosphere. You would not see this effect if the bulk of ozone was situated in the lower layers. Furthermore, an interesting point here why the band 5 and 6 are anticorrelated for $\theta > 75^{\circ}$. You are right, this behavior can also be explained by the sphericity of the Earth8217;s atmosphere but, i think, you have to supply some explanations how this quite different behavior can be caused by the same effect.

21. Page 3526 - 3527: "For JBrNO3 a similar effect is observed near ground level, where the contributions from bands 1 to 3 are screened out meaning that JBrNO3 is principally determined by the contributions from bands 7 and 8." - What about bands 4-6? May be it would be usefull to plot $\sigma(\lambda)$ and $\phi(\lambda)$ at least for extensively discussed species to make the distribution of band contributions better understandable for a reader.

22.Page 3527: "Therefore, truncating the spectral grid at either end of the spectral range for high zenith angles is not possible without introducing large associated errors in either the stratosphere or troposphere." - The discussion in subsection 4.2 do not show any substantial influence of band 1. May be a plot for O_2 should be added to Fig. 2?

23. Page 3528: The errors in J using the standard and modified grids should be

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presented in the same figure, i.e., Figures 3 and 4 should be combined in one plot containing 6 panels.

24. The labels in contour plots in Fig. 5-13 are difficult to see.

25. Page 3528: As pointed out in the item 2 of my comments it would be more convenient to present pairs 5a $\leftrightarrow$ 6a, 5b $\leftrightarrow$ 6b, etc. in the same Figure.

26. Page 3528: "The corresponding contour plots of the associated error calculated using grid A, as well as a scaling ratio for band 1 and limits for δ_i in the lower layers, are given in Figs. 7a-d and 8a-h, respectively." - I do not see any "scaling ratio for band 1 and limits for δ_i in the lower layers" shown in Figs. 7a-d and 8a-h.

27. Page 3529: "The sensitivity of the band method to the values of σ and ϕ can be elucidated by comparing Fig. 4a with Fig. 5 in Landgraf and Crutzen (1998)." - I do not think it is a good idea to ask the reader to compare plots form different papers. You should either remove this sentence or show the plot from [Landgraf and Crutzen, 1988] in your paper.

28. Figure 9: Why did you put the albedo plots between the plots for different solar zenith angles? It is very well known that the effect of ground albedo on the radiative field decreases with an increasing solar zenith angle. Thus the solar zenith angle of 80° can not be considered as optimal to check the dependence on ground albedo. From the discussion it is also unclear if such an investigation for small solar zenith angles was already done by *Landgraf and Crutzen* and what were the results. If such an investigation was already done and the original model was found insensitive to the ground albedo for small solar zenith angles, it makes no sense to repeat this for larger solar zenith angles because one expects a weaker effect in this case.

28. Figure 10 and 11: Why did not you plot the values for $\theta = 85° - 90°$ and $\theta = 90° - 93°$ on the same plot. There seems to be a discontinuity at 90° please explain the reasons for that.

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29. Page 3533: "These effects cause differences of ≈8722;7% and ≈-2-20%, respectively, with the largest difference being exhibited for JNO2 (blue solid line in Fig. 12)." - Blue solid line in Fig. 12 shows JN2O5 whereas JNO2 is shown by the magenta line.

29. Page 3533: "This behavior is somewhat different to the original results shown in Landgraf and Crutzen (1998), where calculations were performed using the DISORT method for $\theta = 0^{\circ}$ and an albedo =0%." - If you want to make a comparison please provide the old results from *Landgraf and Crutzen* (1998).

30. Page 3533: "Identical calculations for $\theta = 80^{\circ}$ using DISORT revealed that similar reductions as those shown in Fig. 10 occur." - Figure 10 has absolutely nothing to do with the current discussion. May be you mean Fig. 12? Please add some explanations if you really mean Fig. 10.

31. Page 3533: "The associated errors for the tropospheric subset with respect to the incident angle are very similar to those shown in Fig. 8 when compared to reference B, and therefore, are not shown or subsequently discussed." - This sentence seems to have no connection to the discussion above and below. Please check if it is on the right place here.

32. Page 3534: "For the **tropospheric** species the effect on the J values is similar to that shown in Fig. 7 of Landgraf and Crutzen (1998) and therefore is not reproduced here, although the effects are more marked as a consequence of the higher zenith angles. The effects above 40 km are minimal due to the cloud being situated much lower down the atmospheric column. Generally there is an enhancement in J values for the **stratospheric** subset of between 2-12%, with those species exhibiting absorption in the visible region being affected the most." - Are you talking about stratospheric or tropospheric species? The values for stratospheric species have to be shown as well. It is not enough to say "similar" considering a quite different range of solar zenith angles.

33. Figure 13: Why do not you show the entire range of solar zenith angles? As mentioned before, the corresponding values for stratospheric species need to be shown as

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well.

34. Page 3537: "This results in a significant reduction in the errors as compared against a full spherical reference model." - From the description in Appendix A it is unclear is the reference model was really fully spherical.

35. Page 3539: "A more sophisticated method of representing sphericity is the pseudo-spherical approximation (e.g. Walter et al., 2004)." - Citing only the model developed by one of the coauthors although a variety of other similar model exits is inappropriate.

36. Page 3540: "Therein, the singly scattered contribution of the radiation field is calculated analytically, whereas the multiply scattered contribution can be computed via a two-dimensional Picard iteration." - "two-dimensional Picard iteration" does not exist. Please explain that you mean here. How many Picard iterations were performed?

37. The model description is insufficient to understand which kind of model was used. Furthermore it is unclear if the refraction was taken into account.

38. Fig. A4 and A5: The fluxes are plotted up to 100 km although according to description in section 4.1 the model ranges up to 80 km only.

39. Page 3541: "At 610.0 nm the direct beam is much less attenuated, which results in a less marked decrease in the F_{act} with respect to altitude (see Fig. 4)." - Do you mean Fig. A4?

Technical corrections:

1. Page 3516: "geo-metric" should be "geometric"

2. Page 3540: "Heavy-Side" in inappropriate. It is "Heaviside" function, named for Oliver Heaviside

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