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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 #3

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This paper is an important followup on the paper of Landgraf and Crutzen (1998) that extends the algorithm for the online calculation of photolysis rates in stratospheric-tropospheric chemical transport models to twilight environments. The modified algorithm enables more accurate global chemistry modeling by improving the accuracy of the 2-stream approximation with a modification for pseudo-sphericity and by introducing new band parameters and scaling ratios in the original approach.

This paper presents a useful contribution to the field of global photochemistry simulation and can be accepted for publication in ACP after the corrections as recommended follows.

General Remarks:

The algorithm treats tropospheric and stratospheric atmosphere as a purely absorbing atmosphere when wavelength (λ) is less than 202.0 nm and solar zenith angle (θ) is less than 75°. This may induce a large error in photochemistry for the species whose cross sections are large around 190-202 nm such as N2O, CF2Cl2. This is because this wavelength range is between O2 S-R bands and O3 Hartley bands and the sun light can penetrate the atmosphere to the low stratosphere where scattering flux is no longer negligible. For example, total scattered flux could occupies up to 70% of total solar flux during this wavelength range at 16 km and overhead sun in the clear sky atmosphere and 0.3 surface albedo (Figure 3 in Bian and Prather 2002). I would suggest that the authors introduce a scaling ratio for band 1 even when θ is less than 75°.

Please clarify reference B. How many bins have been used in reference B? Does reference B account for spherical geometry of the atmosphere using PIFM_KY at 80°$<$ θ $<$ 85° and PIFM_PS at θ $>$ 85°? In addition, since reference B is used to be a standard to evaluate modified band approach, it is important to examine reference B first against an idea case of reference A.

Specific Comments:

1. page 3517 line 11-14: Why is the parameterization of Jabs as a function of the slant path of the total overhead O3 and O2 no longer needed since the modified band approach does not provide the new way to calculate Jabs? 2. page 3518 line 9-11: The contribution to Fact by scattering could not be negligible for θ $<$ 75° at certain atmospheric conditions. See general remarks. 3. page 3519 last 2 lines: Why are biases in the individual band contribution cancelled out, not summed up? 4. page 3520 line 4-7: The conclusion that the maximum amount of radiation is shifted away towards λ of weaker absorption i.e. away from the O3 absorption at the low sun may not held for band 1 and 2 due to O2 absorption (see your table 1). 5. page 3521

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line 26-27: Figure 1 shows Fact(λ)/F(λ0) for band 4 and 5. I can not tell from this figure that the scaling ratios become very large due to dramatic decreases of Fabs for high θ. 6. page 3522 line 8: What is the criteria in your choice of a minimum value for Fabs for band 2 to 4? 7. page 3524 line 5-13: Reference B does not include the use of a lookup table for temperature dependent σ and φ, while the stratospheric-tropospheric version of TM5 calculates photolysis rates (J) with temperature dependent σ and φ at a resolution of 5°C. Therefore, the J value discrepancies between reference B and TM5 comprise the errors introduced not only by the band approach, but also by temperature perturbed σ and φ. 8. page 3526 line 10-11: Here it can be seen that JO3 is principally determined by contributions originating from band 3. You may miss to indicate the layer of 65 km for this sentence. 9. page 3527 line 14 and line 18: Why are two sets of wavelength range used for grid A (i.e. 75-85° in line 14 and 72-85° in line 18) ? 10. page 3527 line 22-25: Are the J values in Figure 3a and 3c calculated with reference B or with the operational version of the photolysis scheme in conjunction with the original band settings? As I pointed out in general remarks, the large errors for J(cfc12) and J(n2o) at the atmosphere 30-40 km using original band approach are most likely induced by neglecting scattering flux in band 1. 11. page 3528 line 8-12: Why is the error of J(cfc12) at 72°$<$ θ $<$ 75° improved so much in stratosphere by using the modified band approach (Figure 7d) than by the old band approach (Figure 5d)? In my understanding, the modified band approach does not do anything when θ is at this range. 12. page 3528 last line and page 3529 line 1-2: I do not quite understand this explanation. Since band 4 has a strong absorbing ability, if the contribution of band 4 is overestimated, the sunlight will be underestimated at low atmosphere. This results in a decrease in J values of tropospheric species. Why does the original method, which overestimates the band 4 contribution, give higher J-values than reference B? Also, if an overestimation of the band contribution from band 4 using the original method is the major reason for a significant error when θ =82°, why is scaling ratio of band 4 unchanged from the old band approach to the new band approach (see Table 1) ? 13.

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page 3529 line 8-13: This may pose a limitation for application. It is not easy for a user to test the accuracy of a new species since the evaluation needs a reference data to compare. 14. page 3522 line 18-19: Why does it have two uncertainty ranges? 15. page 3531, Figure 10 and Figure 11: Why does Jclno3 improve so much when θ $>$ 90°? Why are the errors not consistent around θ = 90° between Figure 10 and Figure 11?

Technical Corrections:

1. page 3514 line 12: Please give full name for PIFM when you mention it at first time, so as to other abbreviations. 2. page 3530 line 22: Change the phrase "As with zenith angles $<$ 85" to "As with zenith angles $>$ 85". 3. page 3533 line 19: Change "blue" to "purple" for Jno2 in Figure 12. 4. page 3534 last line2: Change "compared to the clear-sky scenario" to "compared to full cloud scenario". 5. page 3535 line 4: Table 2 should be Table 3. 6. page 3537 line 7 and line 11: θ=72-85° should be 75-85° (see Table 1). 7. page 3549 line 14: Miss wavelength range for Koppers and Murtagh (1996). 8. page 3552 figure 1: change y-label t(λ)/t(λ0) to Fact(λ)/F(λ0). 9. page 3554 figure 3: Please enlarge the legend. 10. page 3556 figure 5: In the caption "The variation in the errors associated with the J values calculated using the band approach", please add "original" before the band approach.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3513, 2006.