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Interactive Comment

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 #3 for their comprehensive and thorough review of our paper. We have made substantial changes to the manuscript and incorporated many of the suggestions made by the referee. We address the specific questions raised below:

General Remarks:

The referee highlights the fact that, for overhead sun, the penetration of photons down into the lower stratosphere can be significant for the far UV between 190-202nm as a result of the transparency of the atmosphere in this spectral range. This means that the assumption of a non-scattering atmosphere may introduce an error for the photolysis of certain species. We acknowledge this point but feel that the errors introduced on the

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resulting J values are acceptable considering the magnitude of the J values in the lower stratosphere. For details of these errors the referee is referred to the results shown in original paper by Landgraf and Crutzen (1998) for sza=0 deg. Moreover, applying a scaling ratio would necessitate the calculation of a relevant threshold for band 1 which would require extra processing time when calculated online in a CTM. As a result of this remark we have provided more text related to the reasoning behind the use of a scaling ratio for band 1: " 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 $<$ 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 layer.[Ě] 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 $>$ 72 deg for the first spectral band which accounts for this behaviour."

The wavelength grid used in reference B is that of Brühl and Crutzen (1988) and identical to that used by the band method. The calculations made with reference B utilize identical RT solvers (i.e. PIFM-KY and PIFM-PS) for the ranges in solar zenith angle (SZA) from 72-85 deg and 85-92.5 deg, respectively. These choices allow one to differentiate the error introduced by the modified band method from that introduced by the

2-stream approximation. We have added the following text: “Comparisons are made versus a version of the model which calculates Fact. explicitly for each wavelength bin of the working spectral grid of Brühl and Crutzen (1988), without the use of a look-up table for the temperature dependent absorption parameters (hereafter referred to as reference B). It should be noted that both the “final working version” and reference B both use either PIFM-KY or PIFM-PS to calculate Fact. values for ranges of sza above and below sza = 85 deg, respectively.”

The referee states that it would be interesting to compare reference A against reference B. Unfortunately reference A is so computationally expensive that performing calculations for each wavelength bin on the working spectral grid over the entire model atmosphere is not currently feasible. This is why the Fact values calculated by reference A were interpolated in Appendix B in order to obtain the errors introduced into the J values by PIFM-PS.

Specific Comments:

Page 3517: One aim of the original approach of Landgraf and Crutzen (1998) was to parameterise the J values obtained using a non-scattering atmosphere with respect to the slant optical depth (tau slant). However, this parameterisation introduces some errors which are now removed in the modified band approach due to the calculation of Jabs online using a spectrally fine grid. This enhances the flexibility of the resulting code significantly.

Page 3518: The errors introduced into the resulting J values as a result of this assumption are given in the original paper of Landgraf and Crutzen (1998) and are considered to be satisfactory.

Page 3519: The final error for each respective band is the result of the integral of the errors across the entire range of wavelength bins included in each respective band. By the term ‘canceling out’ we refer the referee to Figure 1. of Landgraf and Crutzen (1998) and the associated text which is provided therein.

ACPD

6, S1752–S1757, 2006

Page 3520: Although the calculation of a scaling ratio across the entire range in sza could be performed online it would also require that a limit is applied for lower levels of the atmosphere to avoid any possible erroneous contribution as a result of exaggerated scaling ratios. This requires additional processor power as currently such limits are only calculated once the sza $>$ 81 deg, which limits the number of columns on a global grid upon which it needs to be applied.

Page 3521: We agree with the referee on this point and have subsequently made changes to the text: “Figure 1 shows the relative actinic flux t(lambda) = Fact.(lambda)/Fo.(lambda) at 10 km altitude between 300-320 nm normalized to the corresponding value at 310nm across a range of solar zenith angles.”

Page 3522: The band method relies on the assumption that the Fact. value scales proportionately with Fabs. to produce a valid scaling ratio within each band interval. However, whenever large associated errors occurred when using the original band method there were correspondingly large scaling ratio present for certain bands. By examining these scaling ratios it was found that ratio’s larger than ˜10 produced substantial errors of between 20-150% in the resulting J values. Such ratios were therefore used as a diagnostic for the appropriate limits for Fabs.. We have added the following text to address this: ”The diagnostic used for the derivation of these Fabs thresholds was the occurrence of large associated errors on the most sensitive J values, which were accompanied by correspondingly high scaling values.”

Page 3524: We agree with the comment made by the referee and have inserted the following text: “As a result, the following comparison pertains to the cumulative error introduced by both the modified band method and by the use of offline look-up tables for the absorption parameters.”

Page 3526: Additional text has been added in line with referee’s comment.

Page 3527: A typo has occurred throughout the manuscript. The range of SZA for which grid A is used should be from 72-85 deg. All the errors shown in Figs. 3 through

ACPD
6, S1752–S1757, 2006
Interactive Comment

to 11 are for the band method used in the operational version of the code using either the original or modified band settings versus calculations performed using reference B. The legend for Fig 3. has been changed to clarify this.

Page 3528: See explanation for Page 3527 given above.

Page 3529: The scaling ratios are not used to scale Fabs during the calculation of Fact but rather to scale the individual band contributions to the final J value. Therefore, even though an exaggerated scaling ratio results in an over-estimate of the contribution from bands 2 and 4 to certain J values, it does not influence the magnitude of Fact which is calculated using tau slant (c.f. Eqn. (4)). For the original band approach no limits were applied during the calculation of the scaling ratios meaning that unrealistically high values were subsequently used in Eqn.(5). This results in photolysis rates which are larger than those calculated by reference B in lower layers. The wavelength chosen for calculating the scaling ratio had minimal effect on reducing such errors.

The accuracy of the modified band approach for new chemical species maybe tested by making comparisons with resulting profiles calculated using reference model B. Such a reference model maybe easily constructed by forcing the RT solver to calculate an explicit Fact. value for each particular wavelength bin of the working spectral grid. No new reference data is needed to perform this step. It should be noted that the results presented here only pertain to the use of the spectral grid of Bruhl and Crutzen (1988).

Page 3531: The inconsistency between the contours shown for sza = 90 deg in Figs. 10 and 11 is due to an error which was made during the comparison of the J value profiles between sza = 85- 90 deg. This has subsequently been corrected. The error budgets for this range of incident angles is now significantly reduced compared to those shown in Fig 10. Moreover we have combined Figs. 10 and 11 so that a continuous contour plot is nowincluded for the range sza = 85-93 deg.

Technical Corrections:

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

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

ACPD

6, S1752–S1757, 2006

Interactive Comment