Response to referee #1

We thank the referee for the positive review of our manuscript. Below we give detailed responses to the major comments and questions posed in the review.

GENERAL COMMENTS

One weakness of the paper is that the J-values are evaluated indirectly, through their impact on observed ozone distributions, rather than directly, by comparison with observed J-values under particular conditions. The paper would be more valuable if the authors were able to demonstrate clearly that the J-values and their variation were more realistic with the new scheme (which is of immediate concern to readers interested in adopting the scheme) before they start comparison with other observations (which is of greater concern for those running the TM5 model). There is some qualitative comparison with observed J-values in Section 3, but a more quantitative comparison with observations (or even with best-guess J-values calculated by a more complex scheme) is needed here.

For a full analysis of the performance of the MBA against a full solution of the RT equation we refer the referee to the study of Williams et al. (2006), which quantifies the resulting errors for a number of different J-values and shows the improvement of the MBA in a more constrained column model. Comparisons against direct measurements of J-values made on specific days are heavily constrained by the scarcity of suitable data for our chosen evaluation year of 2006. We have chosen this simulation year so that our manuscript can be compared directly to the benchmark paper of TM5 published last year (Huijnen et al (2010)). However, in order to provide some limited validation we have added a multi-year comparison of the resident concentration of OH vs JO1D against measurements made at the Meteorological Observatory Hohenpeissenberg (Rohrer and Berresheim (2006)) as an additional figure and supply associated text. This shows that the MBA performs somewhat better than the BA for this diagnostic.

The paper would be of greater value to readers if the authors could extract more general conclusions about the strengths and weaknesses of the approach. The study demonstrates and quantifies differences in trace gas budgets for one particular atmospheric model versus a simple off-line approach, but it is not clear how widely applicable the results are. What general benefits might other users of the scheme expect to see?

Again we refer the referee to the performance analysis presented in Williams et al. (2006) regarding the strengths and weaknesses of the approach. In summary the calculation of the full RT equation for 7 designated wavelength bins and use of the scaling approximation results in relatively low errors in the troposphere compared to a full RT solution for each respective wavelength bin which cannot currently be employed in large-scale models. There will be differences when applied in other models due to different resolution effects but we cannot quantify these in the present study as this paper acts more of a performance analysis of TM5 as mentioned by the referee. That there are no parameterized absorption co-efficients such as those need in other approaches means that the approach is relatively flexible and easy to change for users who do not specialize in RT. This is already mentioned in the summary.(pg 2302 lns 18-25).

Given the importance and computational expense of photolytic calculations, it would be useful to have some statement of the computational costs or benefits of this approach compared with both simpler and more detailed calculations. Without this it is not possible to judge the computational merits of the approach adopted.

Introducing this more explicit approach does increase the computational cost of a simulation but here we show that this is warranted by the improvements in the trace gas distributions in the troposphere as demonstrated in this manuscript. The actual runtime increases by ~15% when compared to a version of the model using the more simplified parameterized approach (the BA). This increase is highly dependant on the method of parallelization applied in the model, how the approach is implemented and the scalability of any model with respect to allocated processors. We now include a sentence on this in the text. *"In its current form the application of the MBA results in a ~15% increase in run-time, although this is dependent on the method of parallelization used and the scalability of any large scale model."*

SPECIFIC COMMENTS

p.2283, l.24: "truncated (optimized)"; it would be better to choose one word or the other. Is truncation the only optimization applied here?

The word "optimized" is now removed. The removal of the first band in the MBA (Williams et al, 2006) covering the wavelength range 178.6-202.0nm and the removal the effects of refraction at high solar zenith angles are not necessary can be considered to be a further optimization steps. This is explicitly mentioned later on in the same paragraph.

p.2284, l.1-3: The logic of the wavelength bin grouping is not presented here; it would be useful to include a brief statement to explain why these groupings were chosen.

The original band intervals are adopted directly from Landgraf and Crutzen (1998) and were chosen so that errors in calculating J-values are minimal when using the Band Approach. We now include the sentence: "*These band intervals were chosen in order to minimize errors in the final J-values (Landgraf and Crutzen, 1998)*.

p.2284, l.8-10: Similarly, it would be helpful to indicate briefly why the shifted wavelength bins are used. How does higher SZA lead to the need for this (greater attenuation at short wavelength end of bin) and how have the size of the shifts been decided?

The high angle grid has been defined after performing a comprehensive error analysis against a full solution of the RT equation for each spectral bin of the spectral grid of Brühl and Crutzen (1988). This error analysis has already been presented in Williams et al. (2006) therefore for brevity we reference this article in the text. For large vertical slant columns the small amount of direct incident flux reaching the lower troposphere creates scaling ratios which are unrealistically large when adopting the grid defined in Landgraf and Crutzen (1998). Shifting selected scaling wavelengths towards the red helps reduce the errors in the final J-values for solar zenith angles > 71°. We already provide a reason for using the high-angle grid on l10-12. It is not our intention to reproduce the evidence again in this manuscript.

p.2284, l.15-23: This paragraph needs to be clearer about the methods used. It claims a "full solution of the radiative transfer equation" here, but notes in a later section that it uses a two-stream approach and that "the Mie-scattering component is not included in this study" (p.2286, l.17). If this is a two-stream solution for clear and cloudy conditions that accounts for isotropic scattering (only) from cloud droplets and aerosol, then this should be stated very clearly here.

We now add "*Only the isotropic scattering component introduced by both clouds and aerosols is accounted for using plane-parallel geometry*".

p.2285, l.22: It isn't helpful to show zonal mean r_eff in Fig.S1 averaged over locations where there are no clouds, as the reader can't tell whether higher values indicate larger droplets or just fewer cloud-free locations. Can you plot r_eff averaged over locations with clouds only?

We acknowledge the reviewers point and now present instantaneous horizontal projections at two different pressure levels (945 hPa and 500 hPa) where the averaging does not affect the r_eff values.

p.2286, l.11: Are the aerosol fields described here climatological (i.e., prescribed offline and not explicitly transported in the model)? If so, please state this.

The 'climatology' (i.e. aerosol type) is defined using the land/sea mask, using aerosol properties taken from the parameterization of Shettle and Fenn (1979). A single generic aerosol type is adopted for the Free Troposphere. The aerosols are not included as transported tracers, therefore their atmospheric profiles are fixed. We modify the text thus:
For aerosols in the boundary layer, either a marine or rural aerosol type is prescribed according to the whether the land fraction is > 30% of the total grid cell area, where the optical properties for each type are taken as defined in the parameterization of Shettle and Fenn (1979). Here the influence of relative humidity on aerosol size via deliquescence (and thus isotropic scattering) is also accounted for, although no interaction with cloud liquid droplets (scavenging and washout) is included. The aerosol profile is therefore fixed throughout the simulations (i.e.) the aerosols are not transported. At higher altitudes (above the boundary layer) and at the poles a free tropospheric aerosol type is prescribed throughout the column which exhibits less absorption and higher scattering.

p.2286, l.17: If Mie scattering is neglected, state clearly that scattering from aerosols is assumed to be isotropic (if this is indeed the case). If not integrated, how many orders of scattering are considered?

The additional information given regarding the type of aerosol scattering accounted for in the radiative transfer calculations given on pg2284 addresses this point.

p.2286, l.27: some clarification is needed for "interpolated". What measures are taken to ensure that the total flux is maintained?

We use the interpolation method provided with the Tropospheric Ultra-Violet (TUV) radiation code (http://cprm.acd.ucar.edu/Models/TUV/) which is commonly employed in the atmospheric sciences community for radiative transfer studies.

p.2287, l.12-13: It may be more relevant to plot the fractional change in sec(SZA) in Fig.S2 rather than the absolute change in SZA, given that this is proportional to the difference in slant column.

We present the SZA as absolute changes on request of the Editor.

p.2287, l.17: if scattering is isotropic, insert "isotropic".

Now inserted.

p.2287, Eqn.5: The terms are reversed here: snow should be 0.7, ocean and bare soil low (0.01) and vegetation perhaps 0.05? It is strange to have land surface albedo lower than that for the ocean as suggested by Figure S3, please check these numbers. What wavelength interval were these chosen for?

We have now corrected the albedo values used for the different types of land surface in the equation. The fact that vegetation has an albedo of 0.01 and ocean has an albedo of 0.05 means that the aggregated albedo over land can be lower than over the ocean depending on the type of land use. We redefine the color bar to provide more detail of the differences. As mentioned in the text the albedo values are spectrally independent therefore used across the entire spectral range, where the value in the UV is adopted (therefore there is no decrease towards the visible). In the future we will consider applying albedo values which vary with wavelength.

p.2292, l.7-16: It would be helpful to provide some indication of the origin of the differences in the TOA spectra used. Does this reflect measurement issues or some natural variability? Is one of them more appropriate than the other for the general user?

Differences between the solar spectra tested here related to the radiometric accuracy and the resolution at which the TOA spectrum is provided, where the spectral bins used in the MBA in the UV spectral region around the absorption maximum for ozone are 0.2nm in width. The Thuiller et al (2003) spectrum is only provided on a 1nm resolution. Therefore, the Dobber et al. (2008) spectrum is more appropriate for this application. We now expand on this in the text.

p.2293: As noted in the general comments above, it would be valuable to have a clearer comparison with J-value measurements illustrated here. The qualitative comparison presented here is weak; a better quantitative comparison is needed, even if it just contributes to an additional figure to show how the J-values are better under particular conditions.

We refer the referee to the answer given to the general comment above.

p.2296, l.14: How well does TM5 capture the diurnal cycle of ozone? This can be important when comparing monthly mean ozone, as nighttime biases may mask the effect of any changes due to photolysis during daytime.

In that we do not apply a diurnal cycle for e.g. road transport NO_x emissions means that TM5 will have some deficiencies with respect to the changes in NO_x that can occur throughout the day. By over-estimating the release of NO during the night there will most likely be enhanced titration. Therefore, any nighttime bias is likely to reduce the monthly mean value. We feel that using the 3hrly output data to assemble the monthly mean values is the best approach we can currently take for performing this comparison without altering the emissions introduced into the model, which out of the scope of this manuscript. We add additional information in the text regarding this point.

Table 4: The budget for CH2O presented here doesn't balance; there's a large sink term missing. Or perhaps deposition is expressed in Tg(C) instead of Tg(CH2O)?

We thank the reviewer for noticing this error which has now been corrected.

Table 6: The caption is not clear. What are the masses shown here? They aren't consistent with the previous tables.

The numbers relate to the total mass of each species oxidized by OH and the percentage change in the mass oxidized between the BA and MBA. We have changed the table legend to provide more clarity.

Figure 1: The information shown here is fine, but the presentation needs to be clearer. Given the difficulty of spanning 3 orders of magnitude on the J-value color scale, I suggest a monochromatic (or perhaps dichromatic, 0-5, 10-750) scale where the intensity is represented by the saturation (larger values shown by stronger colors). On the difference scale, the blue colors should be paler. The figure would also be more legible if the individual panels were larger, as in Figure 2; the axis labels are not needed except on the left hand and bottom plots.

We do not agree with the reviewer on this point. A clear distinction between blue (lower) and red (higher) values is already visible. There is also a clear distinction between the positive and negative differences presented with the current colorbar. The darkest blue colour represents the maximal negative differences and can be used to clearly differentiate between the 0-10% and 10-20% decreases in J-values. Legibility of the colourbar and the titles means we are limited with regard to the size of each panel, especially after the figure is shrunk in the final GMD paper.

Figure 6: The colors of the BA and MBA lines should be swapped so that they are consistent with figs 5, 7 and 8. The captions should state what the error bars represent.

The colors have been changed so they are identical to figures 5, 7 and 8. We now give details regarding what the error bars are representative of.

Fig S2: panels are arranged left/right, not top/bottom. It would be helpful to mark the terminator on the figure (SZA differences at night are of no concern) and to center on the daylight half of the globe (either show 12 UTC or center map at 180E for 24 UTC).

We now centre the figure at 180°E and also show the actual SZA calculated at 24;00 in the MBA so the reader can see the relevant part of the differences.

All technical corrections have been addressed

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