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Geoscientific
Model Development
Discussions

Interactive comment on "The application of the Modified Band Approach for the calculation of on-line photodissociation rate constants in TM5: implications for oxidative capacity" *by* J. E. Williams et al.

Anonymous Referee #2

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GENERAL COMMENTS

This manuscript describes the effect of the implementation of the Modified Band Approach (MBA) on simulated photolysis rates and budgets of chemical species in the TM5 chemical-transport model. It includes a presentation of the basic principles of the MBA, and the way that it is implemented in the model. The discussion of the results demonstrates that there are sizeable changes in the simulated photolysis rates, and that some of the species are simulated better when applying the MBA. The manuscript is well written, certainly suitable for GMD, and the findings are worth documenting for

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the reference of future TM5 and/or MBA users. However, there are some general and specific comments that I would like to make, which I believe would lead to significant improvements of the manuscript.

The main general concern that I have relates to the fact that there is not sufficient discussion on why the photolysis rate simulations are actually different when applying MBA as compared to BA. There is a fairly thorough description of the MBA scheme, but even in this model description section (Section 2), the major differences of the two schemes are not clearly stated. Later, there is a quite thorough analysis of the chemical drivers of differences in the two TM5 model versions, but, before that, there is not much discussion on the physical drivers that may be making photolysis rates drastically different in some cases. There is no need for a very detailed analysis of each individual driver, but some more discussion (and perhaps evidence) could enhance the value of that section.

For example, the representation of clouds is being mentioned a few times as a potential source for differences, but rather speculatively, and without making the reader understand why the cloud treatment may have had such an effect and why the new treatment is better. I believe that a more focused discussion on such aspects could be rather illuminating, especially for the readership of GMD.

SPECIFIC COMMENTS

ABSTRACT

Line 15: "which induced. . ." : Are we sure ozone changes are the main reason? I would expect that the changes in the photolysis of ozone are also important.

Line 15: Does the negative sign refer to the changes in high northern latitudes in Fig. 4a? If yes, that is not so important, and thus the $\pm$ in here is a bit misleading.

SECTION 1

Page 2281, Line 8: Why is longitude not mentioned?

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Page 2281, Line 15: What about surface reflections (albedo)?

Page 2282, Line 5: Sentence starting “J–values. . .” seems a bit out of place here.

Page 2282, Line 21: Liu et al. (2006) is probably the reference for which the authors intended, so it is correct here, but there is a different one listed in the References section (the 2009 paper). See the full 2006 citation at the end of this review.

Also, note that the Voulgarakis et al. (2009) GMD paper that is already in the reference list could also be cited at this point, as it also demonstrated strong improvements due to the use of an online scheme.

Page 2282, Line 25: What about chemistry–climate models?

SECTION 2

Page 2283, Line 20–23: The sentence starting with “One. . .” is not enough for the user to understand all the differences and advantages of MBA compared to BA. Since this is not clearly done in another part of the manuscript either, I would recommend that at this point there should be a small paragraph outlining the main differences/advantages of MBA as compared to BA (not in terms of results, but in terms of structure, principles, user–friendliness etc).

Page 2284, Line 16: If I understand the method it correctly, it would be clearer if you add “single” before “pre–defined”.

Page 2285, Second half of the page: I understand that LWC is the only input to the photolysis calculations, when it comes to clouds. But it is not clear where this LWC comes from. Please specify.

Page 2286, Paragraph starting at Line 11: So, for let’s say, the free troposphere of a fully oceanic region, there will be a single optical depth value used everywhere?

Page 2286, Line 17: So, only isotropic scattering is being considered? Please clarify.

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Page 2286, Lines 20: Look–up tables of what? Please clarify.

Page 2287, Lines 3 and 5: Please add "," after "For this" and "For the BA".

Page 2287, Lines 23–24: The albedo values used in the equation do not seem fully consistent with the surface types.

Page 2289, Lines 1–15: Are these stratospheric ozone values used for the radiative transfer and photolysis calculations as well?

Page 2289, Lines 18–20: What does "either. . .or" mean in this case? When are GEIA and when are ORCHIDEE emissions used? Please clarify.

SECTION 3

Page 2290, Line 5: Please add "(BA and MBA)" after "approaches".

Page 2290, Line 7: Please add "," after "effects".

Page 2290, Line 10: It would be better to rephrase to ". . .namely the photolysis of ozone to produce excited oxygen (O1D) atoms J(O^1D). . ." and change JO3 to J(O^1D) throughout the manuscript, in order to be more specific/accurate.

Page 2290, Lines 10–13: Perhaps rephrase to "Comparisons were also performed for J–values of species that are important hydrogen oxide (HO_x) and nitrogen oxide (NO_x) reservoirs, such as formaldehyde (CH_2O) and peroxy–acetyl nitrate (PAN) (see Supplement...)". Note: remember to change "peroxyl–" to "peroxy–".

Page 2291, Lines 7–9: (Please also see my general comment) How do we know that clouds and aerosols are responsible? This needs some more discussion and convincing evidence. To strengthen this argument (although this may not be enough), the authors could also cite past studies, which have demonstrated that clouds and aerosols can drive large changes in photolysis rates in large spatial scales (e.g. zonal mean). Suggested references (see at the end of this review for full): Liu et al. (2006) JGR, Voulgarakis et al. (2009) ACP, Martin et al. (2003) JGR, Tie et al. (2005) JGR.

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Page 2291, Lines 25–29: This sentence needs rephrasing.

Page 2292, Line 17: Please add "," after "surface".

Page 2292, Line 23: What is AFGL? Please explain.

Page 2292, Line 25: Please change "For the tropics" to "For the region lying in the tropics".

Page 2293, Line 12: Please change "Finally" to "Also".

Page 2293, Line 13: Please add "," after "OH".

Page 2293, Up to line 22: It would be nice to actually see some of these photolysis rate comparisons with measurements here, and also the improvements (?) seen when using MBA instead of BA . The discussion about these feels a bit too brief, taking into account how crucial the accurate representation of photolysis is in such a study, and also what the readership of this journal would expect.

Page 2293, Lines 20–23: See general comment and also comment on Page 2291, Lines 7–9.

SECTION 4

Page 2294, Line 14: Please change sentence starting with "The corresponding reductions in surface NO_x show that increases in surface O3. . ." to "The corresponding reductions in surface NO_x and the increases in O3 loss via photolysis imply that increases in surface O3. . ."

Page 2295, Lines 7–12: STE is actually too small, compared to past multi–model intercomparisons (e.g. Stevenson et al. (2006)). Please state this clearly. Is this feature of ERA–interim an improvement compared to the operational analyses or not?

Also, net chemistry (Prod.–Loss) is too small. But it is worth commenting on the fact that MBA changes both Prod. and Loss towards the right direction (based on Steven-

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son et al., 2006).

Ozone burden and lifetime look ok.

Page 2295, Line 12: Maybe "recycling of NO_x" is what you meant to write?

Page 2295, Lines 25–28: Less titration or more efficient production, as mentioned earlier?

Page 2296, Line 19: "especially during DJF": I would also say during spring.

Page 2296, Lines 22: Please make "over estimate" one word.

Page 2296, Lines 24: S3 is for the albedos. Please change.

Page 2297, Lines 8: Also, please mention reduced photolysis rates as a potential reason for higher HNO_3.

Page 2297, Lines 7–10: I would say that a possible explanation is that reduced photolysis of NO_2 in the free troposphere in MBA leads to more NO_x, which then translates to more HNO_3. Please comment and change text if needed.

Page 2297, Lines 10–13: Do we understand why PAN is reduced in MBA?

Page 2297, Line 27: Please make the sentence end like "...with a negative bias in most of the cases.", as the bias is not seen for all the months/locations.

Page 2299, Lines 15–17: Yes, but chemical production and loss change by roughly the same amount, in relative terms, while production is larger in absolute terms, so how can it be that the burden is increasing? Please comment.

Page 2300, Line 4: Please change "anthropogenic emission" to "anthropogenic CH2O emission source".

Page 2300, Line 6: Please add "almost" before "inverted".

Page 2300, Line 8: Please add "to" after "difficult".

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SECTION 6

Page 2301, Lines 16–20: Some possible drivers of the changes seen when moving from BA to MBA are mentioned here, but it would be nice to know which of them may have played the most crucial role. This relates to the general comment made at the beginning. Also, this list of differences could be mentioned clearly at the beginning of the model description (see comment on Page 2283, Line 20–23).

Page 2301, Last paragraph: Are there any plans to add more photolysis reactions in the mechanism? If yes, which?

TABLES/FIGURES

Table 2: Not sure what "MBA/BA" ratio mean. If it is a ration how can it often be negative (assuming these quantities do not change sign)?

It would be helpful to have the loss term below the production one, for immediate comparison. Same for Tables 3 and 4.

Table 5: It would be very useful to add a line with mean (mass weighted) OH concentrations and their changes.

Table 6: I cannot see any blue color.

Figure 3: Please add a legend instead of the lines in the parentheses in the caption.

Figures 4a,b: The blue color in the stratosphere for ozone is misleading. Also, the units are not clear. Finally, 0.0 and 0.1 appear twice on the colorbar.

Figures 7 and 8: Please use consistent colors with Figures 5 and 6. Also, please add legend.

I would also say that the fonts used in the x/y titles and panel titles is much better in 7 and 8 than in most of the previous figures. Please increase font size wherever it looks too small.

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SUGGESTIONS FOR ADDITIONAL REFERENCES:

Liu, H., et al. (2006), Radiative effect of clouds on tropospheric chemistry in a global three-dimensional chemical transport model, J. Geophys. Res., 111, D20303, doi:10.1029/2005JD006403.

Voulgarakis, A., O. Wild, N. H. Savage, G. D. Carver, and J. A. Pyle (2009), Clouds, photolysis and regional tropospheric ozone budgets, Atmos. Chem. Phys, 9, 8235-8246, doi:10.5194/acp-9-8235-2009.

Tie, X., S. Madronich, S. Walters, D. P. Edwards, P. Ginoux, N. Mahowald, R. Zhang, C. Lou, and G. Brasseur (2005), Assessment of the global impact of aerosols on tropospheric oxidants, J. Geophys. Res., 110, D03204, doi:10.1029/2004JD005359.

Martin, R. V., D. J. Jacob, R. M. Yantosca, M. Chin, and P. Ginoux (2003), Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, J. Geophys. Res., 108(D3), 4097, doi:10.1029/2002JD002622.

Interactive comment on Geosci. Model Dev. Discuss., 4, 2279, 2011.