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Interactive Comment

Interactive comment on "Improved simulation of isoprene oxidation chemistry with the ECHAM5/MESSy chemistry-climate model: lessons from the GABRIEL airborne field campaign" by T. M. Butler et al.

T. M. Butler et al.

Received and published: 10 June 2008

General comments

As indicated in our previous comments, we are very appreciative of the comments of Anonymous Referee 1, who in a very short comment, recognises the usefulness of our work, expresses no major problems, and provides some corrections: two minor grammatical corrections; a missing reference; and a query about one of the minor isoprene oxidation products. The comments of Anonymous Referee 2, on the other

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hand, are in stark contrast to this, using more words to say far less, criticising not only our manuscript itself, but also our response to one of the Short Comments. The major criticism of Anonymous Referee 2 of our actual manuscript appears to be our use of a global model, which they believe we fail to justify. The Referee lists three points in their review which they assert would add value to our use of a global model. We have shown in our detailed response to their review that one of their suggested points has in fact already been covered in our manuscript and that another of their suggestions is not relevant for the problem we address. We acknowledge that Anonymous Referee 2 has a good point regarding the effect of our changes to the isoprene oxidation scheme on the global methane lifetime; however, in our opinion this is the only useful modification we could make based on this review. The suggestions by Anonymous Referee 2 for alternative modelling platforms are not helpful because none of them would be able to reproduce the observed variability in the isoprene mixing ratios, which is one of his criticisms of our use of a global model. We note that Anonymous Referee 1 explicitly refers to our use of a global model, and expresses no major problems with our work.

As well as the two Anonymous Reviews, our manuscript also attracted three Short Comments during its open discussion phase. In general, these Short Comments provided good feedback on our manuscript which we believe will lead to an improved revised manuscript, should we be invited to submit one. In particular, the two Short Comments by Karl et al. and Villa have exposed a problem with our communication of our methodology for estimating the intensity of segregation between isoprene and OH over the Guyanas during the GABRIEL period. In both cases, the Short Comment authors have missed the point that our estimation of the intensity of segregation is based on the observed bulk concentrations of both isoprene and OH. The GABRIEL measurements themselves are not suitable for directly determining the intensity of segregation, and our study is not capable of determining which mechanisms may be responsible for this segregation. Both of these determinations are already flagged in our paper as interesting topics for future work. Although neither of the Anonymous Referees, nor Krol in his Short Comment have made the same mistake in interpreting our work, we

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nevertheless think that this point deserves some clarification in our revised manuscript.

The Short Comment by Krol provides useful feedback on our presentation style. We note that none of the other four comments on our manuscript have mentioned our use of hard-to-read multi-panel figures, but we nevertheless agree strongly with Krol that this aspect of our manuscript can be significantly improved. In his Short Comment, Krol also makes a number of detailed points, to which we have responded in our detailed response to his comment. Some of these detailed points are also addressed below in our list of proposed changes to our manuscript.

In summary, we believe that neither of the two Anonymous Reviews or any of the three Short Comments on our manuscript have exposed any flaws in our methodology or in our conclusions; however all five comments have contributed by varying degrees with helpful suggestions for improving the manuscript. These suggestions range from reorganising the figures, to clarifying some points in the text, and providing additional information which may be of interest to various specialist communities. In the rest of this final response, we outline the changes we would propose to make in our final revised manuscript, should we be invited to submit one.

Proposed changes

Following the suggestions of Krol, we propose to move Figure 2 and Figures 4–10 into an electronic supplement. Figure 2 will be replaced in the manuscript by a single panel figure showing the model-measurement comparison for OH primary production. Figures 4–6 and 8–10 will be replaced by two-panel figures showing only the model-measurement comparison for OH and isoprene. Figure 7 will be replaced by two two-panel figures, one showing the model-measurement comparison for OH and isoprene, and the other showing the vertical profile of the OH budgets with production of n=0 and n=2 artificial OH radicals respectively (from Equation 2 in our manuscript).

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In response to Anonymous Referee 1, we propose to add a reference to Thornton (2002) to our introduction (line 28 of page 6275)

Thornton (2002) have also noted inconsistencies in the modelled HO_x (OH + HO_2) budget in NO_x -poor environments in the presence of isoprene.

In response to the Short Comment from Vila, we propose to alter the text on line 15 of page 6277

Turbulent vertical transport and boundary layer mixing processes are treated using an eddy diffusion method (Roeckner et al., 2003).

In response to the Short Comment from Vila, we propose to alter the text on line 26 of page 6279

...it seems that our model does well in simulating the vertical extent of isoprene mixing over both Suriname and French Guyana.

In response to the Short Comment from Krol, we propose to add a sentence at line 11 of page 6280

The small number of points in Figure 4 for which good model-measurement agreement is observed correspond with measurements taken over the ocean to the East of French Guyana, where no isoprene is present.

In response to Anonymous Referee 1, we propose to clarify that acetaldehyde is indeed a product of isoprene oxidation by adding the following text to the end of the sentence on line 14 of page 6281

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...and the inclusion of previously neglected production pathways of acetaldehyde.

In response to Anonymous Referee 1, we propose to make the following grammatical correction at line 15 of page 6282

Similarly, the OH concentration is increased over previously isopreneemitting regions and decreased elsewhere.

Since the publication of our original manuscript in ACPD, we have slightly refined our "bottom-up" calculation of the intensity of segregation. We propose to alter out text from the sentence beginning on line 6 of page 6289.

Based on 5s average OH measurements, and approximately 2s average isoprene measurements falling inside these 5s OH measurement windows, we calculate an intensity of segregation between OH and isoprene of -0.13, which corresponds to a reduction in the effective rate constant of 13%. Due to this 5s averaging time combined with the high speed of the aircraft...

Should we be invited to submit a revised manuscript, we will also replace all other occurrences of our previous bottom-up estimate of -0.1 with our newer estimate.

In order to clarify our "top-down" approach of calculating the intensity of segregation, we propose to add the following text around line 27 of page 6289

We note that this method of determining $< I_{s,A+B} >$ should be considered a "top-down" approach, in which we effectively treat $< I_{s,A+B} >$ as the single unknown in Equation 5.

In his Short Comment, Vila has noted that the wind speed can determine the influence of surface heterogeneities on the boundary layer. In response to his comment, we propose to add the following text to the end of Section 3.

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LES studies (Avissar and Schmidt, 1998) have found that the influence of heterogeneity in the surface heat flux on boundary layer dynamics largely decreases if the winds are stronger than approximately 2.5 ms⁻¹. We have examined the routine observations taken at the observing station "SJMP", which is located at Zanderij International Airport, the base used by the learjet during the GABRIEL campaign. Filtering these data for observations taken during the campaign between the hours of 12Z and 20Z (the times of day during which GABRIEL flights were made), we find (based on 59 observations) that the average wind speed was 3 ms⁻¹, with a standard deviation of 1.5 ms⁻¹. Calm conditions were reported 8.5% of the time. The influence of wind speed on the effect of heterogeneities in the surface flux of reactive trace gasses remains an interesting topic for future investigation.

In response to the Short Comment by Krol and the Review by Anonymous Referee 2, we propose to add some discussion of the methane lifetime to line 8 of page 6291

For our MIMvK simulation, we calculate a global methane lifetime of 7.3 years. Our calculated methane lifetime is 0.1 years shorter than the lower uncertainty bound of the methane lifetime quoted in the IPCC-AR4 (8.7 ± 1.3 years, Denman et al 2007). Compared with a 26-model intercomparison (Stevenson et al 2006), our methane lifetime is 1.1 standard deviations below the model ensemble mean methane lifetime of 8.7 years. For our MIM2+Slow run, we calculate a methane lifetime of 7.0 years. The effect of all of our changes to the isoprene oxidation mechanism on the global methane lifetime is a reduction of 0.3 years, or 4%. This represents 23% of the standard deviation among the 26 models from the study of Stevenson et al. (2006).

In response to Anonymous Referee 1, we propose to make the following grammatical correction at line 17 of page 6291

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For PROPHET98 and PMTACS-NY, we correctly simulate the existence of low- NO_x environments.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6273, 2008.

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