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Atmospheric Chemistry and Physics Discussions

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.

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Krol (2008) has made a detailed and critical Short Comment on our manuscript, Butler et al (2008). Above all we are very pleased to note that he accepts our main point, that our work "may lead to a better understanding of atmospheric chemistry". His major point of contention seems to be our presentation style, which echoes throughout his comment. For instance, we "show too many results" and should "present our findings in a clearer way". Our "9-panel figures" must be enlarged by "600%", and "grossly overestimate the eyesight of many potentially interested readers" while losing the "fundamental questions" in a "mist of plots". Instead we should "focus on the point we wish

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to make".

We essentially agree with most of this assessment. We propose to reduce the current sensory overload by creating an electronic supplement for our manuscript, in which we will place Figures 4–10 from Butler et al (2008), which Krol (2008) has singled out for criticism. In their place in the manuscript we propose to retain single, or at most double panel figures with the existing panels of the model-measurement comparison for OH, and where relevant, for isoprene. These two species best embody the theme we wish to explore with our manuscript: the placing of limits on the extent of our ignorance about the OH-isoprene system in the boundary layer over the Guyanas during October 2005, and by extension, OH-isoprene chemistry in similar regions at other times.

Our proposed solution differs somewhat from that suggested by Krol (2008), that we only focus on 0–2000m. We believe that there is value in presenting a complete comparison of all GABRIEL measurements with the corresponding points from the model. As our hypotheses revolve around OH and isoprene, we will present these comparisons in the manuscript. When we use correlations of other species as support, we will refer to the supplement. We believe it is still very important that the potentially interested reader has access to all of the relevant information. The point is well taken that the boundary layer is the most interesting region for our work, and we believe that this point in fact comes out more clearly when the reader can see for themselves in our figures that there is a clear difference between the blue dots and the other coloured dots. Some readers may also wish to know whether the changes we make to our chemistry scheme have any effects at higher altitudes. We must strike a balance between focusing on the point we wish to make, and allowing the reader to see things for themselves. We must also allow for the possibility that a new pair of eyes may actually notice patterns in our own figures which we ourselves have missed.

Additionally, a restriction of the model-measurement comparison to some arbitrary altitude threshold will also do nothing to improve the readability situation for the isoprene panels. Just about all of the isoprene above the detection limit of the instrument was

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measured at low altitudes during the campaign. This is also the case for a number of other species. Additionally, we will explore further ways in which these plots could be made more readable (eg. a different colour scheme, larger or variable dot sizes, etc. . .).

In addition to this major readability issue, Krol (2008) also raises a number of other points.

In his second paragraph, he uses the existence of other papers on the GABRIEL campaign to cast doubt on whether Butler et al (2008) should be accepted for publication in ACP. In particular, it seems problematic to him to publish "another paper which specifically deals with the global implications". Leaving aside, for the moment, the fact that he has criticised us in his last paragraph for not dealing with the global issues enough, we completely disagree that our paper "specifically deals with the global implications". "Global implications" is in fact the title of Section 4 of our manuscript. There are three preceding sections which do not deal with global implications. We believe we have struck a reasonable balance in our manuscript between presenting work at the scale of the GABRIEL campaign, and work at the global scale. Krol (2008) cites Lelieveld et al (2008) as evidence that our main findings are "already published". This is simply not correct for the OH-isoprene segregation hypothesis, and is only partially true for the OH recycling hypothesis. We present a great deal more scientifically interesting detail in our work, which is necessary given the summary nature of Lelieveld et al (2008). Krol (2008) also refers to other papers based on the GABRIEL campaign which are in preparation and "appear to deal already with the same problem on the smaller scale". This speculation is simply incorrect.

In the paragraph beginning "Figures 4–10 present. . . ", Krol (2008) asks whether OH is always underestimated when isoprene is overestimated. This is a good question, and the answer is yes. He rightly points out that there are a small number of blue dots indicating points where the model-measurement disagreement is small. These points correspond to measurements made at low altitude over the ocean, where there is no

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isoprene. We should add some text to our manuscript to explain this. In this same paragraph, Krol (2008) also makes a number of interpretation errors. We hope that our proposed changes (mentioned above), designed to improve the readability and focus more on the point we wish to make will reduce such errors among future readers. For now we respond to each individual point. Krol (2008) quotes our "dimensionless covariance of -0.1" in order to indicate that the OH-isoprene linkage in the model and its comparison with the measurements is not a predominant issue. In fact the whole basis of our work is the fundamental disagreement between OH in the model and the measurements when isoprene is present. Furthermore, the "dimensionless covariance of -0.1" is in fact our attempt to calculate the intensity of segregation between OH and isoprene from the GABRIEL measurements. This calculation has absolutely nothing to do with our model at all. We reiterate, this number is based purely on the GABRIEL measurements. On line 6 of page 6289 of our manuscript, we do in fact say that this number is "based on the GABRIEL measurements". The discussion surrounding this calculation also refers to segregation between components of the actual atmosphere. Krol (2008) also asks "what then brings the observed OH down compared to the modeled value?". We do not understand this question. The observed OH is actually higher than the modelled value. Again we hope that our proposed changes will reduce the probability that future readers will make such simple interpretation errors, and avoid potential misunderstandings and the resulting tangential discussions.

Commenting on our simulation in which isoprene emissions have been replaced by CO, Krol (2008) states that "isoprene or other intermediates in the isoprene-CO oxidation chain are taken away from the system, most probably by transport". Larger CO mixing ratios in the run with isoprene replaced by CO are also due to the fact that carbon emissions are concentrated into a single species, rather than being partitioned over a much larger number of species. The end point of this discussion, however, is a reasonable one: that vertical transport plays an important role in this system. We have already discussed this issue at some length in our response to Vila (2008). It is clear that future work in this area will involve detailed simulations with Large Eddy Simulation

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(LES) models and ideally also the appropriate micrometeorological measurements.

Krol (2008) states that he misses “sensitivity calculations that deal with (vertical) transport”. In response we note that we have used a global three dimensional model of atmospheric chemistry and climate. Using such models it is not possible to explore the sensitivity of the system to every possible process. The computational expense is simply too large. This is a very common limitation of all such studies. The standard approach is to pick a reasonable subset of the parameter space to which the sensitivity is explored, while ensuring that other important parameters at least appear reasonable. In our case we have chosen to examine the sensitivity of our modelled system to the chemistry of our model. This makes sense for the GABRIEL campaign, due to the detailed measurements of chemical composition taken during the campaign. Detailed meteorological measurements, unfortunately, are almost completely lacking. In our manuscript we already state the importance of vertical mixing of isoprene, and conclude that the vertical mixing extent in our model is well represented based on its reproduction of the vertical chemical gradients.

The suggestion by Krol (2008) that we compare OH primary production based on the measurements with primary production from our model is a good one. We would propose, however, to retain the comparison with the original terms in the electronic supplement. Regarding clouds, October is the dry season in the Guyanas, and although there were occasional showers and storms present, skies were generally quite clear.

In his paragraph dealing with our treatment of the intensity of segregation between isoprene and OH, Krol (2008) notes that the mechanisms which lead to segregation “are not well separated in the analysis”. This is of course completely true. Unfortunately the measurements necessary to separate these mechanisms are simply not available. We have also dealt extensively with this issue in our response to Vila (2008).

In the same paragraph, Krol (2008) develops the interesting idea that variations in the isoprene mixing ratio alone could be used to determine the intensity of segregation be-

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tween OH and isoprene, due to the dominance of the OH + isoprene reaction in the OH loss. Unfortunately, as we increase the complexity of the chemical scheme and include more explicit and realistic representations of intermediate isoprene oxidation products, this dominance of the loss term by isoprene diminishes. His point that "measurements of the entire chemical composition of an air mass" would be useful in determining OH recycling terms is of course correct, but such comprehensive measurements are not yet available. A steady state analysis using the available observations from GABRIEL will form a part of Kubistin et al (2008, manuscript in preparation).

In his final paragraph, Krol (2008) requests numbers on the effect of our findings on the methane lifetime. We would be happy to make this calculation and provide these numbers in future versions of our manuscript.

Krol (2008) has made an error in fact in his final paragraph by stating that we performed a four year integration. We have not. As mentioned in our methodology section, the longest integrations we have performed are one year (plus spinup). Perhaps this is why he strangely regards our work as "impressive in the amount of computational effort spent".

This last paragraph of Krol (2008) also includes the interesting statement that "many questions arise again that are interesting but deserve a more thorough discussion than can be given in this paper". We make two fundamental points in our manuscript: that we require OH recycling in our model during the isoprene oxidation chain in order to match the GABRIEL OH measurements; and that given a good representation of the OH sink, we require a reduction in the OH + isoprene rate constant, for which we believe segregation of reactants to be the most likely cause. These two basic points by themselves are not "too many subjects". We believe that they belong together in the same paper due to the very close linkage between the chemistry of isoprene and the chemistry of OH in the remote tropical terrestrial boundary layer, as we have demonstrated in our manuscript. In the process of developing these two hypotheses, we touch on a number of other issues which are not fully explored. This is a perfectly normal part of the

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process of the development of a field of scientific inquiry. It is rare that a single study encompasses the entirety of a whole field. Rather, knowledge is built incrementally. Studies build on previous work, and suggest avenues for future work. Some studies open up new areas of investigation, some fill in the gaps. Our work has used the unique set of measurements obtained during the GABRIEL airborne field campaign to develop hypotheses about isoprene oxidation in the remote tropical terrestrial boundary layer. Future work will examine in more detail the mechanisms by which isoprene oxidation may recycle OH, and the mechanisms by which OH and isoprene may be segregated. Based on future work, these mechanisms may be discovered, or we may be found to have been wrong. This is the way in which our work will play its part in contributing to a better understanding of atmospheric chemistry.

In summary, we thank Krol (2008) for pointing out to us several shortcomings in the presentation style of our manuscript. Should our manuscript be accepted for publication in ACP, we shall endevour to present our material in a clearer, more concise way.

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