Atmos. Chem. Phys. Discuss., 8, S1551–S1555, 2008
www.atmos-chem-phys-discuss.net/8/S1551/2008/

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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Received and published: 11 April 2008

In the opening remark to his interactive comment on our manuscript, Vila (2008) notes that "Atmospheric Boundary Layer (ABL) dynamics ... is sometimes relegated to a secondary role in the analysis of the behaviour of reactive species within the ABL". We certainly agree. We are currently not aware of a single global three dimensional chemical transport model which includes a parameterisation of the intensity of segregation of reactants. Vila (2008) mentions three studies (Moeng and Wyngaard 1984, Patton 2003, and Vilà-Guerau de Arrelano 2005) which propose such parameterisations. Although this work has been part of the established literature for over two decades, the

current state of the art in global three dimensional modelling of reactive trace species in the ABL is to ignore any segregation effects. With Butler et al. (2008), we hope to play our part in provoking a rethink of this situation.

Vila (2008) refers to our use of the concept of segregation as “ad hoc”. We agree. This term indeed describes our hypothesis that there was approximately 50% segregation of isoprene and OH in the ABL over the Guyanas in October 2005. This degree of segregation is required for the purpose of reconciling our model with the bulk observations of isoprene and OH made during the GABRIEL airborne field campaign, given that our model appears to be reproducing the isoprene flux and the vertical extent of isoprene mixing, and that the rate constant for the reaction of isoprene with OH is well known.

This point deserves to be emphasised. Our use of a 50% intensity of segregation is based soundly and solely on the unique set of observations made during the GABRIEL airborne field campaign. It is not based, as mentioned by Vila (2008) “on idealised flows simulated by LES”, nor, as indicated by Karl et al (2008) in their interactive comment on our manuscript “on work by Krol et al (2000) and Verver et al (2000)”. Vila (2008) would like to see “discussion . . . based on more justified arguments”, by which he seems to mean “a good balance of measurements related to the physics of the atmospheric boundary layer”. We agree completely. Unfortunately the required measurements made with “fast response instruments” simply do not exist for the GABRIEL period and in general over this or similar regions. We hope that the discussion based on the information available to us will play its part in prompting future work, in both the measurement and the modelling domains, aimed at improving our understanding of the role of ABL dynamics in atmospheric chemistry.

We agree with both Vila (2008) and Karl et al. (2008) that our estimate of 50% segregation seems high in comparison to the established literature. Since there were no direct measurements of this segregation over the Guyanas in October 2005, we will never know how strong it actually was. In Butler et al. (2008) and in our response to Karl et al. (2008), we have suggested three possible mechanisms in which isoprene

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and OH may have been segregated. As well as the previously recognised mechanisms of dry convection and heterogeneity of surface fluxes, we have also suggested that co-location with isoprene of other highly reactive trace compounds may lead to depletion of OH, thus increasing the segregation between isoprene and OH. We believe that future work on this issue should examine all possible mechanisms by which isoprene and OH may be segregated.

In addition to our general response to Vila (2008), we wish to respond in detail to a number of points raised.

In his first paragraph, Vila (2008) makes the very good point that the good agreement between measurements and model which we observe in the vertical extent of isoprene mixing should not be used to conclude that "our model does well in simulating the height of the mixed layer". Should our manuscript be accepted for publication in ACP, we would propose to change the text "height of the mixed layer" to "vertical extent of isoprene mixing", as this is more accurately describes the point we wish to make in that section of our manuscript. Regarding entrainment of free tropospheric air into the ABL, we also propose to extend the description of the ECHAM5/MESSy model to include the treatment of vertical diffusion and the ABL scheme.

In his second paragraph, Vila (2008) questions our calculation of the intensity of segregation between isoprene and OH based on the GABRIEL measurements. We thought we had made it clear in our manuscript that this calculation can not be expected to yield a true value of this intensity of segregation at any particular point in space and time due to the implicit averaging involved in taking measurements from a platform as fast-moving as the learjet used in the GABRIEL campaign. As far as we are aware, this is the first published attempt to calculate an intensity of segregation between isoprene and OH using in-situ co- located measurements of OH and isoprene. Should our manuscript be accepted for publication in ACP, we would certainly be prepared to alter our discussion of what we admit are shortcomings of this calculation. In the same paragraph, Vila (2008) refers to our "additional statement over that the large spatial scales

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are represented in this estimated value is also not very well supported". Regrettably, we do not understand what is meant by this comment, so we are unable to respond.

In his fifth paragraph, Vila (2008) questions the degree to which surface heterogeneity can affect segregation in the ABL in the presence of high background wind speeds. They cite Avissar and Schmidt (1998), who examined the effect of heterogeneities in the surface heat flux. It is not immediately clear to us if these results are transferable to heterogeneities in surface fluxes of reactive trace gasses. The effect of background wind on segregation of reactive species in the ABL seems like an excellent topic for future work involving LES models. Vila (2008) makes a good point that presentation of wind observations from the GABRIEL period may help the portion of the readership with expertise in LES modelling to interpret our results. To that end, 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. Should our manuscript be accepted for publication in ACP, we are certainly prepared to include this information, or possibly even a more detailed analysis of the winds or other available meteorological measurements over the Guyanas during the GABRIEL campaign, at the guidance of the editor.

In summary, we thank Vila (2008) for the detailed and thought-provoking comment on our manuscript. He has made a number of very good points which we are prepared to accept and could lead to changes in our manuscript in the event of its final publication in ACP. It does seem to us, however, that Vila (2008) has made a similar oversight to Karl et al. (2008) in failing to appreciate what might be called the "top- down" approach we have used in identifying the intensity of segregation between isoprene and OH based on their measured bulk mixing ratios and knowledge of their reaction rate constant.

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We would contrast this with a “bottom-up” approach to calculating this term based on fast, co- located measurements of OH and isoprene, supported by a good balance of measurements related to the physics of the ABL. Such measurements are yet to be made, and we agree completely with Vila (2008) and Karl et al. (2008) that such observations are vital in improving our understanding. We hope that our analysis of the unique set of observations taken during the GABRIEL airborne field campaign will play its part in prompting future work, which may one day enable direct measurement of the segregation of chemical reactants in the ABL, as well as LES modelling to better understand the processes involved, and the eventual inclusion of parameterisations of segregation effects in large-scale models.

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