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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.

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Atmospheric boundary layer (ABL) dynamics exert a large influence on atmospheric chemistry, but it is sometimes relegated to a secondary role in the analysis of the behaviour of reactive species within the ABL. Since Butler et al (2008) used the word lessons in their article's title; I would like to add some extra ones motivated after reading their paper. The first one is related to the vertical distribution of isoprene in the ABL. As shown in Figure 3, model results and observations (difficult to see them in the figure) show a large isoprene mixing ratio gradient probably caused first by the fast





isoprene reactivity and second by the entrainment of clean isoprene air masses originated in the free troposphere. Nothing is mentioned about the latter process, which in addition to the isoprene dilution due to the boundary layer growth, it can lead to a decrease of isoprene during the day. Furthermore, they quoted: "It seems from the vertical extent of isoprene mixing that our model does well in simulating the height of the mixed layer..." In relation to this statement, it will be convenient to point out that only conservative variables like potential temperature or an inert compound have to be used to corroborate the estimation of the boundary layer depth, but not reactive species like isoprene.

My second comment is related to the ad hoc used of the intensity of segregation. This variable depends on the co-variance of the turbulent fluctuations between reactive species. Similar to observing turbulent fluxes, in order to measure turbulence variables it is required to use fast response instruments (frequencies higher than 1 Hz) and an adequate data detrending and time averaging. Therefore, their estimation of Is=-0.1 (at which height in the ABL? at what time during the day? under which stability conditions?) is highly debatable. Their additional statement over that the large spatial scales are represented in this estimated value is also not very well supported. Supposing they have assumed the typical turbulent averaging time of 30-minute, there are still missing large spatial scales associated to the low frequencies of mesoscale motions (Sakai et al. 2001).

Moreover, the value used in their model calculations (Is=-0.5) is only based on idealized flows simulated by LES. Although LES is a very good tool to study the segregation process, it does not provide a parameterization of the intensity of segregation to be implemented in one-dimensional models. Similar to the calculation of turbulent fluxes using K-theory, The parameterization of Is should be based on the representation of the co-variances by variables explicitly calculated or prescribed in the one-dimensional model (see possible parameterizations of the variances and co-variances in the atmospheric convective boundary layer proposed by Moeng and Wyngaard 1984, Patton et

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al. 2003 and Vinuesa and Vilà-Guerau de Arellano 2005).

Closely connected with my previous remark, I agree with the comment of Thomas Karl et al. (see interactive comment) that -0.5 is probably an extreme value. As far as I know, only in conditions of high reactant segregation like a chemically reactive plume, this value has been measured (Builtjes and Talmon, 1987 and Vilà-Guerau de Arellano et al., 1990). I will however be very cautious in using confusing terminology like "artifical"; segregation. In my opinion, it is important to clearly distinguish between the control of atmospheric turbulence in mixing reactive species quantified by Is and other boundary layer processes (I guess also important in the work of Butler at al.) like the interaction turbulence-canopy-emission (Patton et al., 2001) or the role of shallow cumulus clouds in enhancing vertical transport of reactants (Vilà-Guerau de Arellano et al, 2005).

As mentioned by Butler at al. (2008) at the end of section 3, the emission heterogeneity can indeed play a role in the segregation of species. However, this discussion should be once again placed in the right context. Krol et al. (2000) did idealized flows with absence of geostrophic wind. LES studies (Avissar and Schmidt, 1998) have found out that the influence of surface heterogeneity on boundary layer dynamics largely decreases if the winds are stronger than approximately 2.5 m/s. Although this wind information is missing in the paper, I can imagine that wind speeds higher than this value were measured during the GABRIEL campaign. Moreover, the potential influenced of the induced mesoscale motions in the boundary layer dynamics, and therefore in atmospheric chemistry, largely depends on the thermal amplitude of the heterogeneity and the characteristic length scale of the non-uniformity (Patton et al, 2005). I am aware of the difficulties in estimating this effect using field measurements, but in my opinion the discussion in the article should be based on more justified arguments.

In concluding, my last lesson learned after reading the paper is that in addition of detailed LES analysis (see comment by Thomas Karl), field campaigns must have a good balance of measurements related to the physics of the atmospheric boundary layer, in

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addition to the important and relevant measurements of atmospheric chemistry. This combination will largely improve our understanding of atmospheric chemistry in the ABL.

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