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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of small-scale variations in isoprene concentrations on atmospheric chemistry over a tropical rainforest" *by* T. A. M. Pugh et al.

Anonymous Referee #1

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The intensity of segregation is inferred by combining relative fast isoprene concentration measurements with OH-box model calculations. The research presented here is an attempt to provide a method to calculate this variable due to its difficulty in directly measuring or calculating it. However, in my opinion, the method is not suited for this study since it is still misses the intrinsic nature of the intensity of segregation. Therefore, I have serious doubts on the validity of the Is-values presented and the related discussion. I will elaborate further on these points below.

1.- Equations (2) and (6) in the manuscript give the definition of the intensity of segregation (Is). The essential contribution of Is is the co-variance between isoprene and

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OH. This is a turbulent quantity that is calculated as a second-moment of the reactant concentration distribution to quantify concentration fluctuations between both species. As the authors mentioned, currently only fast measurements (Dlugi et al., 2010) or large-eddy simulations (Vinuesa et al, 2003) are able to estimate it directly since they are able (within certain limitations) to capture all the essential spatial and temporal scales.

The analysis of the evolution of the co-variance equation enables me to elaborate further into this point ((Vilà-Guerau de Arellano et al., Journal of Atmospheric Chemistry 16, 145, 1993), Verver et al. Journal of Geophysical Research 105, 3983; 2000). Taking Verver et al., (2000) as a reference and by analyzing the co-variance equation (see equations 3 and 4), one can notice that the co-variance of C5H8 and OH is determined by non-linear contributions of mean and turbulent-chemistry terms. The equation shows also the relevance on the scales interaction in governing the covariance. For instance, on the equation right-hand-side, the first two terms quantifies the role of turbulent flux and mean concentration gradients in determining the C5H8'OH'. Similar non-linear interactions are present in the chemistry term at equation 4.

My main concern with the paper under evaluation is that their methodology misses these important contributions in the determination of the C5'H8'OH'co-variance. By using a box-model in calculating OH two relevant processes are omitted: the influence of turbulence/chemistry (at all scales) and (b) the non-linearity in the interaction turbulence-chemistry. Although the assumption that supported their method (page 18205) are well thought and the use of relative (but discrete) fast isoprene observations captures partially some of this information, in my opinion there are not sufficient to guarantee a proper estimation of Is. As Vinuesa et al. (2003) showed in the budget calculation (Figure 7) all the terms of the co-variance equation contribute to the Is evolution, i.e. either they are measured directly with fast response instruments (> 1 Hz), calculated directly with large-eddy simulation models or represent in form of a parameterization the relevant terms.

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2.- I have also serious concerns on the calculation of the numerator of equation (7). First, and closely related to the above point, the production and loss terms of OH do not take into account all the chemistry and turbulent fluctuations. Second, the calculation of the bulk values depend on surface dynamic forcing and reactant emissions, the boundary layer depth evolution, the exchange between the boundary layer and the free troposphere and advection. I am therefore very surprised that the authors are able to reproduce the intensity of segregation of the German mixed forest (Dlugi et al., 2010) with almost the same numerical set up as the one imposed to reproduce the tropical forest experiments (page 18210). Third, in my opinion, reaction C5H8+OH accounts for approximately 60% of the OH destruction, and therefore it is not sufficient to account for all the the effects since the potential chemistry-turbulence fluctuations by other reactions are neglected.

3.- Another strong drawback in using the box model is their independence of height. As shown by Patton et al. (1997) at figure 4, the co-variance depends strongly with height due to its dependence on fluxes and mean concentrations (co-variance budget equation). I understand that this is partially included in the isoprene concentrations, but still the box model omits all the height dependent fluctuations related to OH (and other related species at equation 7) and the non-linearties between turbulence and chemistry. This dependence of height is more pronounced close to the canopy and therefore needs to be included.

In concluding due to the filtering of all the relevant turbulent and chemical spatial/temporal scales and the absence of height dependence (in particular close to the surface or canopy) I have serious concerns in using (partially) a box-model to estimate intensity of segregation. As a closing example of one of these doubts, at lines 5-10 at page 18214, I think the result of Is=0, in the particular numerical experiment of the OH-recycling, is an artifact of the method used, and in consequence requires further confirmation.

In my opinion, and in absence of fast response simultaneous observations of isoprene

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and hydroxyl radical, the authors should use methods to represent/parameterize the co-variance between isoprene and OH (see Verver et al. (2000) or section 6 in Vinuesa et al. (2003)). Notice that Verver et al. (2000) used a combined approach using a box-model and second-order closure modeling to reproduce the intensity of segregation between isoprene and OH in the Amazonian region.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 18197, 2010.