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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Segregation in the Atmospheric Boundary Layer: The Case of OH – Isoprene" *by* Ralph Dlugi et al.

Anonymous Referee #1

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Dlugi et al. present new data on the issue of segregation between isoprene and OH, and put these in context of previously published work. Their data were obtained in the Amazon and largely confirm previous studies on segregation. The paper could provide new insights on the topic of segregation in the Amazon, but there are a couple of issues that should be addressed before any possible publication.

The manuscript is unnecessarily long (60 pages) - as it stands, the manuscript could be significantly shortened and more focused on the important findings. Vast parts read like a review article. Detailed descriptions of previous studies (section 2.1.) could be significantly reduced and rather be included as a citation. The lengthy discussion of previous studies keeps the authors from describing important details about the ATTO site itself, which is essential to the interpretation of the presented analysis. Section 2.2

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Discussion paper

therefore lacks clarity.

What is the immediate footprint of the surroundings? What is the main wind-direction, is there a variation in isoprene emissions surrounding the site? Have the authors looked at sector dependent isoprene emissions? From their assumptions it appears the site is characterized by a rather homogenous isoprene emission source, but it would be good to show this. What QAQC criteria were incorporated for the interpretation of turbulence measurements? I would have also expected to see an overview on latent, sensible and momentum fluxes as well as other important micrometeorological quantities such as Bowen ratio, Obukhov length etc.

The key instrumentation relevant to this article are HOx and isoprene measurements. The frequency of isoprene measurements was 1 Hz, so one would expect a loss in high frequency variability. Further, damping through a 40+ m line has to be expected. The method of inferring a lag-time by comparing water vapor fluctuations through such a long line bears a potential problem, because water vapor retention is expected to be much larger than that of small hydrocarbons. It is also not clear what pressure drop was produced by the 5 um filter. One way to ensure that this analysis is not prone to any substantial bias would be to compare the covariance functions between vertical wind, isoprene and water. I am also missing information on the determined delay/lag time. Overall I am concerned that some (significant?) part of isoprene variability might have been lost due to the experimental setup? Have the authors done any co-spectral analysis?

The authors present the issue of underestimating modelled OH in the tropical atmosphere as a main cause to look into the subject segregation. There are some reports of a possible overestimation of OH inferred from LIF instruments. Several recent studies (e.g. Liu et al., 10.1126/sciadv.aar2547 2018) have concluded that there is no gap between modelled and observed OH in Amazonia within the experimental uncertainty. The cited study by Kaser et al. actually also shows this, as the total impact of different chemical recycling schemes in their study seemed to be quite small. It would

strengthen the manuscript to point out differences in OH measurements during this and previous campaigns, as well as commenting on conclusions of the above papers. In this context it is not clear whether there have been any changes to the presented LIF OH measurements since Lelieveld et al., 2008. At least a reference to a recent validation or intercomparison paper would be warranted. A recent chamber study (Kanaya et al., 10.5194/acp-12-2567-2012, 2012) suggests about a 50% uncertainty (bias) for the measurement of OH in low NOx, isoprene dominated environments. If for example LIF OH measurements were subjected to an offset problem, it would probably not impact the presented analysis of this paper, but if there was a problem associated with a sensitivity bias it certainly would. I am wondering whether this could explain some of the different trends shown in Fig. 4.

The derivation of some of the simplifications is poorly explained – eq. 19: why would only one triple term be important in the analysis here? RES, RE and REis are not well explained – I assume REis refers to term I in eq. 21. In general, I miss a thorough analysis of error propagation in context of the presented equations (e.g. eq 21). Many terms are dropped because they are supposedly small, yet the impact of the experimental limitations is not rationalized well in context of the variance budget of isoprene. I suspect that a significant amount of variance of isoprene might not be accounted for due to spectral attenuation. It also appears that the data availability is rather thin – I only count about 16 individual data points for the analysis presented in Fig3. Within the uncertainty of the analysis, I wonder whether this is enough to draw some of the presented conclusions after considering a thorough analysis of the propagation of errors (ie. systematic and random).

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