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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Turbulent exchange and segregation of HO_x radicals and volatile organic compounds above a deciduous forest" *by* R. Dlugi et al.

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Introduction

First at all, we would like to express our thanks to Jordi Vila for his thoroughly review and his constructive criticism. To understand our replay to his review it is indispensable to further explain the scope of our paper. The experiment during ECHO 2003 as described in our paper was designed to analyze the capability of an instrumental set-up (LIF, PTR-MS, Sonic Anemometer) for determining turbulent quantities like covariances (flux densities) of OH, HO_2, isoprene etc. within the framework of a sophisticated field campaign. This study was the first one performed to find out if such quantities can be

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calculated from measured data with sufficient accuracy. In doing so, the covariances of chemical compounds were computed and also the magnitude of the segregation term as expressed in our eq. 6 were calculated. However, the latter was not the primary goal of this study because its interpretation needs a more advanced analysis of the complete 2-D (3-D) data set for these specific situations during ECHO. For preparing this study under field conditions, Michael Möllmann-Coers, Michael Zelger, Axel Knaps and Martina Berger analyzed different aspects of the meteorological and turbulent flow characteristics at this site. In addition, this feasibility study for this instrumental set-up was prepared by the detailed calibration of sonic anemometers in the Hamburg wind tunnel as cited in the paper. Theoretical estimations based on field data from ECHO 2002 and laboratory tests whether such an experiment would give results with sufficient accuracy were done by some of the authors. In conclusion, this field study and the paper shows that the applied techniques are suitable to determine turbulent characteristics even for very low concentrations of OH and HO_2 together with BVOCs. Within this context also terms like the segregation term were computed to further illustrate the performance of the equipment and to underline that the accuracy of measurements is high enough for determining such quantities. The results can be compared to data from literature, but actually we have not the full required information to decide on the contribution of different processes especially to the segregation term. Therefore, we find we should be cautious in interpreting our results in front of a detailed analysis of the governing physical a chemical processes.

Reply

In the following we refer to points made by the Referee:

1. The fluxes are determined for the reference layer $7m$ above the canopy. The discussion of the meaning of a flux density (simply called a flux) in relation to the terms of the balance equation clarifies that a point measurement can never be applied to identify gradients of field quantities or divergences of fluxes and second-rank tensors. Eq. 5 shows that we have the possibility to estimate the influences of chemical sources and

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sinks on this flux. A divergence cannot be quantified without spatial resolved measurements. The estimation for the influence of some sinks on fluxes of OH and HO_2 are given in chapter 5.2. The layer thickness of 1m is chosen because it relates to a live time of about $1s$ if the magnitude of w in Fig. 6a is considered. Local OH appears to be locally produced and destroyed and the flux is in the order of chemical sinks. But there are chemical sources and sinks for OH which work on larger scales, and, therefore, advection may influence this local flux. Unfortunately, such effects cannot be computed from a single point measurement. For HO_2 the high NO mixing ratio causes a significant chemical sink. We will point out this aspect more distinct in a revised version.

2. "Well mixed" will be canceled in the revised version.

3. As discussed in the introduction, we did not perform this special analysis focused on special requirements for quantifing processes affecting segregation. But we know from site specific studies by Schaub (2007) and the non-cited paper by Aubrun et al. 2004 (Physical modelling of an inhomogeneous finite forest area in a wind tunnel – comparison with field data and Lagrangian dispersion calculations. Agric. Forest Meteorology, 129, pp 121 – 135) that the inhomogeneous distribution of sources of BVOCs cause smaller isoprene fluxes at the main tower than nearer to source area. This agrees with findings from the same field site by Spirig et al. (2005) and by us. Therefore, we only cited the work of Krol et al. (2000) because they considered similar situations. We agree, that Patton et al. (2001) found comparable values for the segregation intensity for the reaction OH + isoprene above the canopy. However, in contrary to our situation, he modeled the exchange with a homogeneous source distribution and found that intermittent turbulence caused inhomogeneous mixing and a nonzero segregation intensity. Although it was not in the scope of the paper, we will add these remarks to our revised version.

4. We cited Butler et al. (2008), because they described how they performed measurements on OH and isoprene. They described that they used the data to estimate the

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intensity of segregation. They assumed, their presented values to underestimate the real intensity. We cannot decide which processes were dominant during their study. In addition, the analysis of aircraft measurements differs from that of tower measurements.

5. In the introduction we mentioned some gaps of knowledge and the sentence "In order to close some gaps ..." is a connection to the general questions posed during ECHO.

6. We refer to the introduction and our discussion on point 3.

7. The ratios can be estimated e.g. from Fig. 6c and 5a. The source of isoprene is largest some hundred meter away from the tower. Aubrun et al. (2004) and Schaub (2007) found a distance between 200 – 600m for the site. So isoprene reacts with OH during the transport. This changes the concentrations and the ratios and more reaction products are found at the tower than near sources (Schaub 2007). Komori et al. (1991) and others discuss that these ratios and also correlation coefficients depend on the distance from emission sources. Therefore, the relevant ratios may be those at source area and not at the tower.

8. We completely agree, but this requires an even more detailed analysis of the governing processes and initial conditions near the sources and between sources and measuring points. As mentioned above, this must be the part of a still more advanced study.

9. We refer to our comment on 1 and 3.

10. The lower and the highest frequencies are not captured. Measurements are available in the range $0.002 < f < 0.2Hz$. For high frequencies scaling theories for the inertial subrange and for dissipation are available. This allows the application of error estimates. With the lower frequencies we are within the production range for eddies from the mean flow and from interactions of obstacles and the mean flow. It is known,

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that a number of different processes act together in flows near or inside canopies which cause hairpin vortex packets (e.g. Hommema S.E. and R.J.Adrian, 2003, Packet structure of surface eddies in the atmospheric boundary layer, BLM, 106, pp. 147-170; Robinson, S.K., 1991, Coherent motions in the turbulent boundary layer, Ann Rev. Fluid Mech., 23, pp 601- 639). By this action, spectra of u,v,w or T show significant variations from one time interval to the next especially if short averaging intervals have to be chosen. This is intensively discussed for example by Finnigan, J.J. et al., 2003, A re-evaluation of long – term flux measurement techniques. Part 1: Averaging and coordinate rotation. BLM 107, pp. 1-48. They found that spectra for short averaging time periods deviate significantly from those averaged over 1 hour up to 4 hours. They also found the maximum of the frequency multiplied co-spectra near $f = 0.01Hz$ and significant parts of the flux below $0.002Hz$ depending on the quantity being considered. These findings agree with our results and cited results by Beier and Weber (1992), but we only used spectra to estimate $f(max)$ for error analysis. Without any very detailed additional analysis of the interaction between the emitting canopy and the flow and the time dependent behavior of all parameters a rough presentation of spectra and a rather incomplete interpretation with respect to the term Is would have no scientific content. Note, Jonker et. al. did their study for the convective boundary layer and we had – in contrast - slightly unstable conditions.

11. These are formula by Wyngaard (1973) and Mann and Lenschow (1994) as cited by Finkelstein and Sims (2001).

12. If we would have reviewed and discussed the segregation in detail, we would have cited these important publications, but also work done before by John Seinfeld's group. Please note, that influences of inhomogeneous mixing in the atmosphere were analyzed since the 40's by cloud physicists to better describe the broadening of the droplet spectra.

13. See comment on 3.

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14. In the introduction (see above) and in point 1 we discussed the scope of the paper. As mentioned, we had slightly unstable conditions and, therefore, found no significantly skewed distribution for w. This agrees to results of a number of other field studies (e.g. Chu, C. R. et al, 1996, Probability density functions of turbulent velocity and temperature in the atmospheric surface layer. Water Resources Research, Vol 32, No.6, pp. 1681-1688).

15. Water and Carbon Dioxide were measured with LICOR 6262. This is a closed path instrument. In addition, an open path instrument was used. Both were calibrated against standards and in the field against psychrometers. The results from this data analysis have to be prepared for publication in a separate paper with another leading author. Note, that latent heat fluxes from both instruments very often significantly differ from each other. Comparable effects are known also for Carbon Dioxide fluxes as already discussed e.g. by Finnigan, J.J. et al., 2003 (s.o.)

16. See our comments to 10.

17. See our comments to 1.

18. As mentioned above, we had performed a measurement at one point. From these measurements we have no hints for the time interval 6 hours presented, that the influence of entrainment into the ABL could be identified at the site.

19. We follow your argument, that this formulation can be misinterpreted. We shall change the text in a revised version and point out that a) fluxes are influenced by chemistry and b) discuss the importance of that process.

20. In principle we should better call these velocities transfer velocities. They are calculated for a layer and better correspond to the experimental situation above the canopy. Such velocities are often significantly higher than deposition velocities.

21. These requirements need a rather extended additional analysis of all chemical compounds and their distribution and transport in the considered volume. We agree,

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that we add our remarks from point 3 to the paper as mentioned above.

22. The research reported by Butler et al. (2008) was analyzed by them. They reported a value of 0.1 and mentioned, that this value may be a minimum value. Our and their experiment were done separately and we have no access to their data and more experimental details than given in their paper.

23. The statement will be modified. We regret a typing error in line 14 ($< 5\%$ instead of $< 15\%$). We note that this segregation is not insignificant but a small value. We will add the discussion that it may be larger below canopy top if we consider the results of Patton et al. .

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 24423, 2009.