

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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Received and published: 15 March 2010

General Comment

We thank Referee 2 for the very careful reading of our paper and also for the very helpful discussion and comments for the preparation of the revised version. This review – also like the first one - discusses the importance of the estimation of statistical and systematic errors. Both reviewers mention that the averaging interval for flux calculation of 600 s is short compared to the common 1800 s. Initiated by the comments of the referees, we did some additional analysis especially on the low frequency loss caused by a limited length of the time series as discussed below.

In our paper we mainly discussed the error estimation on the high frequency part of the covariances (basis for flux calculation). We estimated the errors for the loss of the contributions above 0.2 Hz by the influences of different effects (see paper: Table 1). Our error estimation for this frequency range is larger by a factor of about 2–3 than the results based on an Ogive analysis by Spirig et al. (2005) from the same experimental period at the same site.

The signals with lower frequencies are sampled with lower probability, and, therefore, have higher statistical uncertainty. In our study we presented data for 600 s averaging intervals for different measuring periods between about 40–60 min. After each of these periods, the calibrations of the LIF and the PTR-MS were checked as discussed in the paper. For these larger periods we additionally calculated covariances which now cover also frequencies f in the range $0.0004 \text{ Hz} < f < 0.2 \text{ Hz}$. The covariances from the 600 s averaging intervals within each of these periods cover a range of $0.0016 \text{ Hz} < f < 0.2 \text{ Hz}$. Therefore, the low frequency loss of the shorter averaging intervals can be estimated relative to each of the longer periods. We find a mean loss of the flux contribution below 0.0016 Hz compared to the larger frequency range of 22 %. But we know from studies of J.J. Finnigan et al. (BLM 107, 1–48, 2003) or the cited work by N. Beier and M. Weber (1992), that the contribution to the flux below $f < 0.0004 \text{ Hz}$ can be between 10–20 %. Therefore, we conclude that this “low frequency loss” of fluxes from the 600 s intervals is at least $22 \% + 10 \% = 32 \%$, and, therefore, in the same order as the loss at high frequencies. We mention again that we chose this 600 s interval to allow for reliable detrending during a period of high variability of net radiation and photolysis frequencies (see Figure 1). We also had to prove under field conditions the negligible change of the base line especially for LIF (see Figure 4). Therefore, we have chosen the given periods and the intervals of 600 s within as a compromise. This will be introduced in the revised version.

Comments to special topics as mentioned by Referee 2

To Abstract: ECHO will be defined in the abstract.

To p.2, l.2-p.4, l.4 from bot.: The comments will be considered in the revised version.

To p.4, l.3 from bot.: The measurements show that the quantity "Photolysis frequency" (dimension: s^{-1}) decreases from canopy top to the ground. Therefore, the main contribution to the OH-production from O_3 photolysis (see cited reference: J.H. Seinfeld, S.N.Pandis, 1998) will be reduced inside the canopy.

To p.5, l.6-p.5, l.8 from bot.: This will be considered in the revised version.

To p.5, l.3 from bot.: Yes, as mentioned on p.7, l.5-6.

To p.5, l.3 from bot.: We will add the definition together with a reference in Appendix A.

To p.6, l.9-p.7,l.3 from bot.: These comments will be considered in the revised version.

To p.8, l.5: This equation will be corrected.

To p.8, l.9: We will use "unmixed" in the revised version.

To p.8, l.11-p.8, l.4 from bot.: Both comments will be considered in the revised version.

To p.8, bot.: In the last 12 years, many studies (especially on the determination of carbon budgets), have chosen a coordinate system with reference to the local mean streamline at measuring height. In this system, the vertical coordinate is not parallel to the vector of local geopotential and gravity. Therefore, we described the choice for the coordinate system which avoids such complications mentioned. In the cited reference, Sun (2007) described the consequences of the choice of streamline oriented coordinate systems for the determination of fluxes and budgets.

To p.9, l.8-p.10 top: These comments will be considered in the revised version.

To p.10, l.7: Spirig et al. (2005) expanded the data from the highest frequency measured of about 0.3 Hz to higher frequencies. They mentioned that they found the largest contribution to the flux from their Ogive analysis in the range $0.067 \text{ Hz} \leq f \leq 0.1 \text{ Hz}$.

To p.10, l.14-p.14, l.8 from bot.: These comments will be considered in the revised

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version.

To p.14, l.7 from bot.: We will summarize and shorten these two sentences.

To p.15: The discussion of the results obtained by Finkelstein and Sims will be shortened.

To p.16, top: Yes. Will be changed.

To p.19, middle: At this site emissions at the tower mix with advected air mass from some sources 50-300 m away from this site. This seems to be the reason for fluctuating mixing ratios of isoprene as shown in Fig. 6c and 6d . It is also important to mention that isoprene emission is influenced by radiation so that the leaves at canopy top emit stronger than leaves deeper in the canopy. Radiation fluctuations are shown in Fig. 1.

To p.20, top: This topic was also mentioned by Referee 1 and will be discussed in more detail in the revised version in the following way: The volume of 1m^3 around the measuring point is chosen by different reasons. According to Eq. 5 the vertically integrated quantity $Q - S$ can be compared to the flux divergence. We integrated over 1 m height. The lifetime of OH is about 0.2s as estimated from measured chemical compounds at the site. If we take into account the fluctuations of the vertical velocity of about ± 1 m/s and the magnitude of the horizontal velocity (see Fig. 6a, 3) OH is produced and destroyed in a volume of about 1m^3 around the measuring point. The lifetime of HO_2 is about 20 s. Therefore, the local HO_2 -flux is not influenced by chemical reactions if the volume of 1m^3 is considered. But on a larger spatial scale (e.g. between the measuring point and canopy top or the maximum of HO_2 mixing ratio in the canopy) this flux is significantly modified by chemical reactions. Due to its lifetime of about 25 min, the isoprene flux is not influenced. We will add this discussion together with tables which show reaction rates for the specific compounds and the estimated lifetimes.

To p.20, bot.: This notation "convective" comes from fluid dynamics and will be clarified.

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To p.20, l.9: The high frequency loss of the sensors above 1 Hz should not be important for the calculation of segregation intensity. But the contribution from the range 0.1 Hz – 1 Hz should contribute to I_s . From the analysis of data for the NO + O₃ reaction as presented in the cited paper of G. Kramm and F.X. Meixner (2000) we know that this contribution is below 14 %. But this is only an empirical and not a general result. We expect that the contribution from the low frequency part of this segregation intensity is larger. The calculation of I_s is comparable to the calculation of fluxes (see above) which shows that the loss at the low frequency end is about 30 %. Therefore, the segregation intensity for 600 s intervals may be underestimated by about 44 %, a value obtained from our calculation of I_s for reaction OH + isoprene for the 40 min period instead of 600 s (see above for fluxes).

To p.21, bot.: The distance between the measuring volume of the sonic anemometer and the inlets of PTR-MS and LIF are mentioned here.

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

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