Atmos. Chem. Phys. Discuss., 9, C7530–C7535, 2009
www.atmos-chem-phys-discuss.net/9/C7530/2009/

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.

J. Vila (Referee)

jordi.vila@wur.nl

Received and published: 27 November 2009

Detailed observations of micrometeorology and atmospheric compounds taken above a deciduous forest are presented and discussed. The novelty of the research lies in the calculation of second-order moments (fluxes and co-variances) of the atmospheric compounds. These observations can shed light on the role of turbulence above the canopy in transporting and mixing reactants. I found particular interesting the calculation of the intensity of segregation, a variable which quantifies the turbulent mixing of reactants, and consequently their reactivity rate. Recently, different values of the intensity of segregation have been used, in my view, out of the context and without very

much justification to explain the disagreement between OH observations and modeling studies. Therefore, the observational evidence presented here is at the right time and relevant. The measurements and further data treatment are explained and discussed with rigor. However, I found the physical and chemical discussion of the reactant flux and co-variance a bit sketchy and sometimes inexact. I am confident that the authors can improve this imbalance by elaborating more on these aspects. The reader will indeed appreciate it. Below, I include my comments.

Abstract

1.- As they carefully explained at page 24445 and it is mentioned in the conclusions, the radical OH is a short lived species compared with the turbulent transport. As such, one should expect that there is hardly any contribution of the OH flux divergence to the OH budget. I understand that their measurements indicate that the co-variance between the fluctuations of the vertical velocity (w) and OH is not zero. How important is this flux? Can they establish if OH is transported by turbulence or it is locally produced and destroyed? And more precisely, is the flux divergence of OH relevant for the budget of OH? I will therefore recommend a clarification in the abstract, page 24445 and in the conclusions of the meaning of the observed flux for a radical and fast chemical species.

Introduction

2.- p. 24426. Why do they mean by well mixed troposphere? Above the canopy, the atmosphere can also be stably stratified.

3.- p. 24447. The explanation of the segregation of species is confusing. The segregation of species is driven by different processes and phenomena. In short, the chemically reactive species are segregated due to: (a) turbulent coherent structures in the convective boundary layer (Schumann U., Atmospheric Environment 23, 1713, 1989), (b) vertical thermal stratification (Galmarini S., Journal of Applied Meteorology 36, 944, 1997), (c) intermittent turbulence influenced by canopy (Patton E.G., Boundary-layer meteorology 100, 91, 2001) and (d) spatial horizontal heterogeneity of the BVOC emis-

sions (Krol M.C. et al, 2000) and point source emissions (Builtjes P.J.H., Boundary layer meteorology 41, 417, 1987). The segregation coefficient therefore depends on entirely different factors and I think it is convenient to clarify it to the reader and place their observations in the right context.

4.- p. 24428. Is Butler et al (2008) measuring co-variances? Can these observations quantify the segregation intensity? Are their results comparable to the observations presented in the current study?

5.- p. 24428. Could they be more specific about the gaps of knowledge?

Flux measurements

6. p. 24431 Do they assume horizontally homogeneity on their measurement site? How uniform is the forest site, both from surface conditions (sensible heat and latent heat flux) and BVOC emissions?

7.- p. 24432. In addition to the Damkholer number, the influence of turbulent transport on the reactivity depends on the ratio of the reactant concentration (see Appendix at Fitzjarrald D. et al, Atmospheric Environment 17, 2505, 1983 or Vilà-Guerau de Arellano et al., Journal of Geophysical Research 100, 1397, 1995). This ratio can be very relevant for the reaction between the abundant isoprene and the scarce radical hydroxyl. Could they elaborate more on this issue at section 2.2?

8. p.24432. The intensity of segregation can be also positive if the species are premixed, see Toor (I. Eng. Chem. Fundamentals 8, 655, 1969) and if the reactants are transported together.

9.- p. 24434. Closely connected with my comment 6, how important is the spatial variability of the BVOC emissions for their interpretation of the measurements?

10.- p. 24440 The authors discussed that the 10-minutes sampling interval is not enough to capture all the contributions to the fluxes and co-variances. Since measurements analysis is very complete, it can be very interesting to show the energy

(co-)spectra of the reactants understudy and if it is similar to the one found for inert species. In that respect, Jonker et al. (Journal of Atmospheric Sciences 61, 41, 2004) analyzed the spectra of reactants calculated by using large-eddy simulation numerical experiments. They found large modifications in particular at the lower frequencies. Have the found similar results with their observations?

11.- p. 24441. Which other formula is compared to Eq. 8 and the one given by Finkelstein and Sims (2001)?

Data analysis

12.- p. 24442. As far as I know there are previous research that have quantified the intensity of segregation in the atmosphere, both experimentally (Builtjes P.J.H., Boundary layer meteorology 41, 417, 1987) and in fine-scale turbulent modeling of the convective boundary layer (Schumann U., Atmospheric Environment 23, 1713, 1989)

13.- p.24442 or in the section Results. I missed in the data analysis or in the results an explanation of the contribution of the footprint to the reactant flux calculation. Could they give an estimation of this contribution to their site in the day understudy?

Results

14.- p. 24442-24443 I think here it is convenient to elaborate more on the role of the interaction between atmospheric turbulence and the canopy presence. For instance, (a) Are the temperature ramp structures entirely driven by the canopy presence? What are they role in the injection and mixing of the species in the surface layer, and in consequence in their measurements site? and (b) Under convective conditions, one should expect a probability distribution function positively skewed for the vertical velocity? Why do they find a normally distributed one?

15.- p. 24444. Could they include a description of the latent heat flux evolution? Their data set is very complete and in my opinion it will be use in future studies, so it is worth to include all the micrometeorological information to become as complete as possible.

16.- P. 24444. As I mentioned in my comment 10, I think it is convenient to support their statements on the underestimation of fluxes for reactive species by calculating and discussing the co-spectra. Here, they claimed that these underestimation is mainly at the higher frequencies. However, Jonker at al. (2004) shows the large variations of the co-spectra of the reactants occur at the low frequencies (normally the most energetic). In my opinion, this discussion requires to be further elaborated and supported.

17.- p. 24445 See my comment on the interpretation of the flux for fast chemically species.

18.- p. 24446. In the analysis of the isoprene budget, it is mentioned that the main sink of isoprene is through reaction with the radical OH. However, there are other processes which play an important role in the isoprene-budget. The dilution and exchange of air masses with the residual layer has a strong effect on the isoprene budget (see for instance Vilà-Guerau de Arellano et al., Atmospheric Chemistry and Physics 9, 3269, 2009). This process can also be relevant in the interpretation of their observations above canopy. Could they comment on this point?

19.- p. 24446. Here it is mentioned that the OH flux divergence do not play a role on the OH budget. So, as commented previously, how important are the OH flux measured?

20.- p. 24447. Do they have an explanation of the larger deposition velocities compared to other studies? Is this because the reactivity of the species? Or is it driven by the canopy flow conditions?

21.- p. 24448. As mentioned in my comment 3, the intensity of segregation depends on several different physical ad chemical processes and surface conditions. In my opinion, here they have to become more concrete and specific and discuss the behavior of the co-variance among reactants in the context of their measurement site and physical-chemical conditions.

Conclusions

22.- p. 24448 How accurate is the value reported by Butler et al. 92008) and compared with other studies? In my view, the values reported in their paper are not based on the measurements of the co-variance as expressed by expression (6).

23.- p. 244449. The authors concluded that the vertical flux and segregation is notable, but they have a relative small effect on the OH-budget. This statement is confusing and misleading. For instance, if the segregation intensity is as they measured between 10-15 %, one should expect a slow down in the reaction rate due to the heterogeneity of the reaction. I realize that this is not straight forward since it depends on other reactions, but I think these sentences need to be rewritten to avoid confusion on the OH- and isoprene budget interpretation.

Figures

24.- Caption Figure 5b. It should be HO2.

25. Caption Figure 6b. It should be m/s.

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