

Interactive comment on “First direct measurements of formaldehyde flux via eddy covariance: implications for missing in-canopy formaldehyde sources” by J. P. DiGangi et al.

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The paper presents a study on the exchange of HCHO inside and above a Ponderosa forest focusing on the role of in-canopy sources/sinks. Overall the paper is well written and presents a number of interesting features on atmosphere-biosphere exchanges of VOCs, oxidation products and the role of in-canopy interactions between biogenic emissions, dry deposition, chemistry and turbulence. It indicates through the combined use of gradient, eddy correlation measurements and chamber measurements of HCHO exchange complemented with a box model analysis on the potentially important role of further unidentified sources of reactive compounds but also stresses a further need

C7246

for a revised insight in the removal of species involved. I recommend this article for publication in ACP after consideration of a number of issues I raise.

My main comment (also found below) concerns the representation of the dry deposition and biogenic emission process in the box model that has been applied to support analysis of the observations. This concern might be mainly due to the way these representations have been described and which would profit from a clarified explanation but, when properly interpreted, it comes down to the fact that biogenic emissions and dry deposition are treated here in a separate form. It appears obvious from the results that observed HCHO fluxes (and concentrations) are the result of a subtle interplay between in sources and sinks operating at the leaf/needle up to the canopy scale and that rather than treating leaf/needle scale emissions/dry deposition separately one should preferentially apply exchange approaches, e.g., the compensation point approach in a multi-layer modeling approach.

I cannot provide specific comments on the experimental sections (2.2, 2.3, 2.4) since much of the presented information on the measurements is beyond my expertise so hope that one of the other reviewers can have a critical look at that part of the ms.

Major/Minor comments;

Pp 18732; line13; “but much is not yet understood”, what is much here?? On absolute concentrations, on the diurnal cycle in concentrations, on fluxes, on in-canopy sources/sinks?? You give an example in the next sentence but when you use the term much there should be a number of issues that are worthwhile to shortly discuss.

Pp 18733, line 2, What is meant with “high duty cycle”?

Pp 18739, line 26; “The HCHO ogive...” ??? I am not familiar with the term ogive, could you explain?

Pp 18740, line 13; “mention here (again) that “(note that positive values reflect and upward flux)” to avoid confusion.

C7247

Pp 18741; lines 7-11; You are hypothesizing about what explains the observations of enhanced HCHO concentrations at the lowest measuring height (1.6) in terms of a litter layer source of HCHO. This is followed by a statement about the enhancement of HCHO in the crown layer associated with the oxidation of the emitted VOCs. I could envision that a part of the chemically produced HCHO is also mixed downward and that because of a missing sink there/reduced mixing in that part of the canopy, you could explain these observed enhanced mixing ratios compared to the levels above or?

Pp 18741; I had to read through the discussion on the link between the vertical profiles in the gradient and fluxes a couple of times to really get the point about the role of advection in the exchange regime. I am wondering to what extent an observed nighttime deposition gradient can be reconciled with the measurement of a \sim zero flux. I recall from the measurement sections (and in general) that the EC technology provides direct measurement of the flux down to a friction velocity of \sim 0.2 m s⁻¹. Was u^* at night indeed typically $<$ 0.2 m s⁻¹ implying that you cannot draw any conclusions from the EC measurements and that you need to rely on the measured gradients? And if the nocturnal gradients are significant, pointing at the role of sinks/sources, including the potential role of advection, how can you then conclude that all expected drivers of HCHO exchange (sources/sinks) are linked to the solar cycle? Yes, for the measured HCHO flux you can since you assume that the fluxes are \sim zero but what for the HCHO canopy budget in general. I am also wondering about for example the importance of non-stomatal deposition of HCHO.

Pp 18742; regarding the soil/litter layer HCHO sources/sink measurements; how many samples did you take and did you try to get an indication about the heterogeneity in the soil/litter fluxes?

Pp 18744, line 6; Could you comment a little more on what kind of oxidative chemistry you are referring to and would it be possible to provide some estimate of the order of magnitude of how much this term could potentially contribute to the overall budget?

C7248

Pp 18747; Interesting discussion on the nighttime (non-stomatal) deposition rate. The effort to infer this non-stomatal uptake resistance raises a number of questions/comments. First of all; At line 8 you are referring to $R_{x,night}$ but this term is not included in equation 8. Does it actually refer to the inferred R_c (=RNS) or does it refer to the sum of $R_{a,night} + R_{b,night}$? And how did you calculate these last two resistances? It would require information about turbulence inside the canopy (u^* , wind speed, stability). There is also the issue of the large differences between the nighttime V_dHCHO based on the in-canopy measurements and those based on the boundary layer budget. You simply mention this without discussing the reasons for this substantially higher estimate of V_d based on the BL budget method. Is there any indication about the reasons for this discrepancy? One point that could partly explain this difference is the fact that you inferred V_d is representing an overall smaller effective surface area although with an LAI of 1.9 this would not make a big difference. This discussion on nighttime exchanges/deposition also triggers another thought about the role of boundary layer mixing in the HCHO budget through the influence of nocturnally produced HCHO in the residual layer being entrained in the early morning. We addressed this issue in an analysis of the atmosphere-biosphere and boundary layer exchange of the tropical forests of Guyana (Ganzeveld et al., ACP, 2008) indicating that such a mechanism of entrainment of HCHO being chemically produced overnight could have an impact on the early morning fluxes/concentrations of HCHO. That was for a regime where this is apparently lost of isoprene which is not the case for your site but can imagine that there are other chemical sources of HCHO in the inversion/residual layer that impact the observed exchange, e.g. relevant to this discussion on differences in inferred nighttime removal rates. I actually don't expect an important role of this early morning entrainment since it is apparently not seen in the flux observations.

Pp 18747; The discussion on the inferred daytime dry deposition velocity triggers a critical observation. The fact that the inferred daytime V_d of 0.39 cm s⁻¹ is much smaller than reported in literature is explained in terms of a lower LAI, smaller contribution by the underbrush compared to other sites. Then it is stated that the "deposition term is

C7249

highly dependent on litter emission” but don’t get this since from the explanation on the how the deposition velocity is calculated, this term doesn’t include any emissions. It is assumed that the mesophyll resistance is set a zero and there is no reference at all to the possible role of an HCHO compensation point (now partly considered in the study through the representation of biogenic emissions), that could result in a reduced daytime leaf-scale V_d , for example due to the internal production of HCHO as a function of radiation (?) or.. Is this value of (the canopy-scale) 0.39 cm s^{-1} the effective removal rate based on the summed emission fluxes (soil/litter and canopy emission fluxes) and the estimated V_d based on equation 8 (so without considering the compensation point approach)?? This would make much more sense in explaining an inferred V_d which is substantially smaller compared to other sites where potentially biogenic sources might have been smaller.

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C7250