We thank the reviewer for their helpful questions and comments. The original reviewer questions and comments are shown in italics, while our responses are shown in bold.

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 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; line 13; "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 discus.

We thank the reviewer for pointing out this oversight and have clarified in the manuscript that it is the qualitative and quantitative understanding of in-canopy HCHO production that is currently lacking.

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

We thank the reviewer for pointing out this error and have corrected this to read "high time resolution".

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

We thank the reviewer for pointing out this misunderstanding. The ogive is the cumulative distribution function of the cospectrum, and is defined in Sect. 2.4.2 of the manuscript.

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

We thank the reviewer for this comment and have added this to the manuscript.

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?

We believe that two points make it unlikely that downward mixing explains the observations. The first is that the chamber experiments show evidence of a significant amount of direct soil/litter emission, which agrees with our hypothesis. The second is the lack of a loss process that is lower below the canopy than inside the canopy that changes temporally. There was no observable OH gradient, while the ground solar radiation was often potentially greater than inside the canopy due to frequent gaps in the canopy. However, a difference in deposition may have lead to this enhanced concentration, and we have added this possibility to the manuscript.

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.

We thank the reviewer for pointing out this confusion and have reworded this point in the manuscript to clarify. In brief, the diurnal profiles of the concentration measurements show sharp changes during the two times of day at which sharp changes in wind direction were observed. As the flux data does not show evidence of this dependence, the flux does not appear to be sensitive to changes in the advective airmass.

I am wondering to what extent an observed nighttime deposition gradient can be reconciled with the measurement of a ~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 ~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?*

The flux data presented has all intervals with u_* < 0.2 m/s removed, including the nighttime data. Each nighttime (<6:00 or >18:00) 1 hour average bin shown in Fig. 4a, with the exception of the single point midnight bin, has at least 4 data points, and many have more than 6 points. This demonstrates that the nighttime flux was actually quite close to zero, thus it was reasonable to consider it negligible toward the nighttime canopy deposition. It was then necessary to extract the nighttime deposition velocities/rates from the gradient data. We have further clarified this point in the manuscript.

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 ~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.

By HCHO exchange, we mean vertical exchange at the flux measurement height, above the canopy. We did not mean to imply this held for the overall HCHO canopy budget, and have adjusted the manuscript text to clarify. We have also changed the qualifier from "all" to "most", as non-stomatal deposition is indeed significant and is not treated as tied to the solar cycle.

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?

We only performed a measurement at a single, seemingly representative ground site, but the reviewer is quite correct in pointing out the likelihood of heterogeneity in these sites. We have added language to the manuscript that expresses the limitations of our single measurement.

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?

The higher-order oxidative chemistry to which we refer is HCHO production from the further oxidation of oxidized VOCs (OVOCs). For example, glycolaldehyde and 2-Hydroxy-2-methylpropionaldehyde, two major MBO oxidation products, can further oxidize, resulting in more HCHO production. While the reactivities of any one of these higher order OVOCs are usually small compared to oxidation of the parent BVOC, their

total reactivity may become significant. The situation is further complicated by mixing in and out of the canopy. While mixing may remove OVOCs from the canopy before they can further oxidize, it can also reintroduce air containing OVOCs produced above the canopy. These OVOCs can then oxidize inside the canopy to create additional HCHO. Due to the complexity of this chemistry and transport, it is difficult to estimate the extent to which this may affect the model results, as it would require a model containing the full explicit chemical mechanisms for the BVOCs in question. The sentence in the manuscript was revised to clarify the lack of OVOC oxidation in the model.

Pp18747; Interesting discussion on the nighttime (non-stomtal) 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 Rx,night but this term is not included in equation 8. Does it actually refer to the inferred Rc (=RNS) or does it refer to the sum of Ra,night+Rb,night?

We thank the reviewer for pointing out this confusion. In this case, we intended x to be a generic variable, simply denoting that it can be either a or b (i.e. $R_{a,night}$ is R_a averaged over the nighttime hours and $R_{b,night}$ is R_b averaged over the nighttime hours). We have changed the manuscript to say this explicitly. This also points out an error in Eqn. 8: R_c should actually read R_{NS}, as this is only meant to denote the non-stomatal component of R_c. This has also been corrected in the manuscript.

And how did you calculate these last two resistances? It would require information about turbulence inside the canopy (u, wind speed, stability).*

The calculation of R_a and R_b were calculated by standard literature methods, using u_*, wind speed, and stability corrections as described in the supplement (Sect. S4).

There is also the issue of the large differences between the nighttime VdHCHO 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 Vd 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 Vd is representing an overall smaller effective surface area although with an LAI of 1.9 this would not make a big difference.

In response to this comment, we have added the following text to the manuscript: "The discrepancy between this work and the literature likely lies in the different assumptions on which either models is based. The boundary layer method assumes similarity between HCHO and ozone deposition and usually depends on literature estimates of ozone deposition. This method also assumes that deposition is the only nighttime loss process and there are no production processes. Finally, the boundary layer method is based on a single measurement and assumes a continuous concentration throughout the boundary layer. The gradient method used in this work makes no assumptions on the HCHO profile, as it is measured directly, and does not depend on literature ozone deposition. The gradient

method also estimates nighttime production and loss via the model terms. For this, we chose to neglect NO_3 chemistry due to the low NO_x concentrations at this site. However, the gradient method has limitations in that it is much more dependent on direct emission measurements/estimates and assumes the canopy gradient is well represented by the available measurements (in this case, 4 heights)."

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 mechanisms 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.

We thank the reviewer for pointing out the effect of morning entrainment, or lack thereof, in our data. We also find the lack of an entrainment effect at this site curious, but we believe it is supported by our concentration and gradient measurements. The temporal width of the morning peak is typically ~1 hr, which means it is probably too wide to be due to the entrainment of overnight produced HCHO above the canopy. Additionally, evidence in entrainment would show in the gradient data, specifically at the beginning of the rise as the HCHO starts mixing down. Concentrations for all levels at the beginning of this rise seem to increase at the same rate, implying a source mostly consistent over all levels, as we would expect from the oxidation of fresh emissions. Finally, as the reviewer points out, we did not observe a sharp downward HCHO flux as convection increased in the morning as we would expect from HCHO entrainment. We have added text to the manuscript pointing out the lack of this effect.

Pp 18747; The discussion on the inferred daytime dry deposition velocity triggers a critical observation. The fact that the inferred daytime Vd of 0.39 cm s-1 is much smaller then 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 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 Vd, 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 Vd based on equation 8 (so without considering the compensation point

approach)?? This would make much more sense in explaining an inferred Vd which is substantially smaller compared to other sites where potentially biogenic sources might have been smaller.

The HCHO compensation point varies considerably by tree species and the environment of the tree. While this is a more accurate representation of the tree stomatal emission/deposition, we did not feel we had enough information to treat this explicitly, thus we treated the stomatal emission and deposition separately.

However, we have performed sensitivity analyses where we neglect stomatal deposition (assuming that our branch emission measurements are the net effect of stomatal emission and both stomatal and cuticular uptake) and/or non-stomatal deposition (assuming the litter emission measurements are the net effect of litter/soil emission and ground deposition). This also carries some additional assumptions, such as neglecting tree trunk cuticular deposition. However, even with all "deposition" terms turned off, the model could only account for ~1/4 of the noontime HCHO flux. This indicates that this effect, while likely important, is still insufficient to explain the missing HCHO flux.

We have added a brief discussion of compensation point and this analysis to the manuscript. Additionally, we have added to our conclusions the need for experiments to achieve a parameterization of compensation points, emission and deposition rates for the leaves and soil as functions of temperature, radiation, and humidity.