

**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 manuscript by DiGangi describes formaldehyde, HCHO, flux measurements with a new LIF instrument over a pine forest. A well thought through introduction and description of the experiment are presented. The data is analyzed to extract vertical fluxes above the canopy throughout the day. Enclosure experiments of leafs and soil are also presented that supplement the flux data. The authors use a chemical box model to further analyze the data and to test our understanding of HCHO chemistry in a forest environment.*

*In general this is a very interesting manuscript. The presented data is unique and should add to our knowledge on HCHO chemistry. The experimental description, as well as the determination of the fluxes is clear and well written. The box model part of the manuscript, on the other hand needs some revisions as it is often difficult to understand how the model calculations were performed. Consequently, I recommend this manuscript for publication in ACP after some revisions. Below are my detailed comments:*

*Page 18734, lines 0 - 10: Please add the cell length and the air volume of the cell.*

**We thank the reviewer for this suggestion and have added this information to the manuscript: “The separation between the White cell mirrors was ~25 cm, but only ~6 cm of that is in the ~6 cm x ~5 cm x ~6 cm central region through which ambient air was flowed and the fluorescence was excited. The remaining cell volumes were purged with air from a zero air generator.”**

*Page 18734, line 8, . . . : I am not sure if SLM and SCCM are units understood by all readers. I suggest not using abbreviations, but instead writing them in full word. Alternatively, define the units the first time they are used.*

**We thank the reviewer for this suggestion and have added definitions the first time that these abbreviations appear.**

*Page 18734, line 15: Is a weekly calibration of the instrument sufficient? Most other instruments calibrate more frequently. Also it appears that with a 4 week long measurement period only four calibrations were performed? How statistically significant is the statement that the calibration only varied by 2.5%?*

**Weekly calibrations were chosen for a variety of reasons. Many instruments require frequent calibrations due to fast changes in sensitivity, such as the presence of interferences, changes in laser parameters, or changes in reagent concentrations. Using our online/offline sampling method (see below), we have never found any evidence of interference. Additionally, our signal is continuously corrected for both changes in laser power and wavelength.**

**The primary contributions to changes in our instrument sensitivity are changes in alignment and the degradation of optics. Alignment changes affect the background, and therefore limit of detection, but have little effect on the laser power-normalized sensitivity of the instrument. These events may happen over short timescales, but are easily detected and corrected/eliminated if necessary by monitoring the input and output laser power from our detection axis. Optics degradation increases the background and decreases the sensitivity of the instrument, but happens slowly (weeks-months) and is easily characterized by weekly calibrations.**

**Laboratory testing using this instrument has shown that its sensitivity is typically highly reproducible. The 2.5% variability between calibrations is likely dominated by variability in our permeation source, rather than any instrumental variation.**

*Page 18735, line 7: Please clarify what you mean by “offline and online sampling”.*

**We thank the reviewer for pointing out this lack of clarity. In the manuscript, we have defined online sampling as measuring the fluorescence signal while tuned to a wavelength with a high HCHO absorption cross-section, while offline sampling is defined as measuring the fluorescence signal while tuned to a nearby wavelength with a relatively low HCHO absorption cross-section. However, this definition is unnecessary for the paper and now simply refer to them as the laser positions on and off of the absorption feature.**

*Page 18736, lines 5-6: I do not agree with the approach that “OH concentration was assumed to be equal to half the detection limit ( $2.5 \times 10^5 \text{ molec cm}^{-3}$ ).” In a statistical sense values below the detection limit are undistinguishable from 0. The only information one can extract from values below the detection are upper (detection limit) and lower (zero) limits for OH concentrations. These limits should then be propagated in any calculations using the OH data.*

**The reviewer presents a valid point. We performed sensitivity analyses with our model where we varied the nighttime OH concentration from the OH CIMS detection limit ( $5 \times 10^5 \text{ molec/cm}^3$ ) to 0. However, this effect was negligible ( $< 5\%$ ) compared to the measured noontime HCHO flux and does not significantly impact the manuscript. We have added a discussion of this to the manuscript.**

*Page 18741, line 3: Please explain this more clearly. The decrease in HCHO concentration after 8 am is caused by the rise of the boundary layer. Together with the increasing surface flux, this leads to a peak at 8:00.*

**We thank the reviewer for this point and have clarified it in the manuscript. While the rising edge of this peak is likely due to increasing BVOC emissions in the canopy, the falling edge seems to correlate more closely to changing wind direction than an increase in convection.**

*Page 18741, line5: This could also be explained by a smaller HCHO loss, for example due to weaker photolysis below the canopy. Please comment on this possibility.*

**We believe that photolysis is an unlikely contributor, as the ground solar radiation was often potentially greater than inside the canopy due to frequent gaps in the canopy. However, we have added text to the manuscript outlining that differences in non-stomatal deposition may have played a role.**

*Page 18741, line 19: SO<sub>2</sub> is not a very good tracer for air mass change due to the fact that its atmospheric levels also strongly depend on the presence of SO<sub>2</sub> sources. H<sub>2</sub>O, CO, or CO<sub>2</sub> would be better suited for making this argument.*

**We thank the reviewer for this comment, and have added CO<sub>2</sub> and H<sub>2</sub>O as species that do not correlate with HCHO flux, further enforcing that advection is not significant for HCHO flux. Unfortunately, CO measurements were limited during the time period that HCHO flux was measured, so this comparison was not possible.**

*Page 18741, line 28: Was water vapor added to the air flow into the chamber? If not, how does this impact the leaf processes?*

**Dry air was added to the chamber, but the tree naturally emits a significant amount of water. The RH in the chamber was typically between 20 and 45% RH, perhaps even a little higher than needles outside the bag were experiencing. We do not expect that this had a significant impact on the results. We have added this information to the manuscript.**

*Page 18743, chapter 4: In principle it would be more appropriate to use a 1D model, but for what the authors like to achieve here the 0D model may be sufficient. However, the explanations in this chapter are somewhat confusing. The authors have several parameters available at different altitudes in the box, while others were only measured at one altitude. It is often difficult to distinguish altitude dependent terms from those that are not. I propose to add (z) to every term that is altitude dependent and explain this more clearly throughout this chapter. Equation 3 implies that all terms were calculated altitude resolved first and then integrated. Is this really the case? This should be explained more clearly. The impact of the vertical integration on the results should also be discussed.*

**We thank the reviewer for pointing out the lack of clarity with respect to the vertical dependence of the model. The idea of the model is to use the mass balance of HCHO inside of a 0D box, neglecting horizontal advection, to determine how much must be vertically leaving the box, which is the HCHO flux. As the goal of the model was to model the vertical flux out of the top of the box, it was unnecessary to have this vertical 1D resolution. While we do use conceptually a 0D box, we have the advantage for some species of vertically resolved data inside of this box. We used this data to more accurately calculate the**

chemical production and loss. As we only care about the total amount of HCHO in the box, we then integrate vertically over this production/loss to determine the HCHO change in the total box, thereby reducing the data from 1D resolution to 0D. We have added a paragraph to the beginning of this section about the model justification to address this point and have added '(z)' to vertically resolved species throughout the manuscript to clarify.

*Page 18743, equation 3: Please change FHCHO in equation 3 to FHCHO(h) to indicate that the flux at the top of the box is meant.*

**We thank the reviewer for pointing out this lack of clarity, and have added this to the manuscript.**

*Page 18744, section 4.1: I am assuming that the [Ox] term was considered to be constant in altitude. This should be mentioned here. How does the unknown OH vertical profile influence the results of the chemical production? One could expect lower OH below the canopy as all photolytic OH sources will be less efficient.*

*Page 18745, section 4.2: How does the assumption of altitude independent OH impact the results?*

**We did assume that OH was constant in altitude, and stated this on page 18745, line 4. Additionally, gradient measurements were performed over multiple days using the OH CIMS instrument, and found no significant gradient throughout the canopy. O<sub>3</sub> concentrations were vertically resolved, and the manuscript was adjusted to clarify this point.**

*Page 18745, section 4.2: How was the integration of photolytic loss over altitude performed? Was the photolysis frequency or the photolytic loss rate ( $J_{\text{HCHO}}[\text{HCHO}]$ ) integrated?*

**The photolysis loss was calculated by integrating the calculated photolysis loss rate ( $J_{\text{HCHO}}*[\text{HCHO}]$ ) from ground to flux measurement height. However, the lifetime estimates in this section were calculated by integrating the calculated photolysis loss rate constant ( $J_{\text{HCHO}}$ ). This has been clarified in the manuscript.**

*Page 18746, section 4.4: The manuscript nicely shows that there is a considerable HCHO gradient between in and below the canopy. Should the resistance model thus not be applied separately for the canopy region (using the canopy HCHO levels) and the soil (using the below canopy HCHO levels)?*

**The calculated nighttime non-stomatal resistance ( $R_{\text{NS}}$ ) includes any present soil deposition. However, we have no way of separating this soil NS resistance from the canopy NS resistance. As a result, there is an implicit assumption that the soil laminar sublayer resistance ( $R_{\text{b,soil}}$ ) is similar to the canopy laminar sublayer resistance ( $R_{\text{b}}$ ). This assumption has been explicitly stated in the manuscript.**