**Response to the comment of anonymous referee #1**

**Peer review of "Vertical distribution of sources and sinks of VOCs within a boreal forest canopy"**

A two-year data set, gathered from 2014 and 2016, composed of micrometeorology and chemically reactive species measured in the Norunda station (ecosystem Boreal forest) is analyzed. The investigation focused to identify and attribute the emissions sources of Biogenic Volatile Organic Compounds (BVOC) during different seasons and their diurnal variability. The analysis and the data set are worth to be published since the study helps to advance our understanding on the characterization of these actively species in the boreal forest and for future modelling studies of the canopy-atmosphere interaction. The research presents very interesting results, but I found it a bit unbalanced towards the description and discussion of the vertical and evolution of the emission of the BVOC without connecting to the turbulent transport at the canopy-atmosphere interface.

Below my major and minor comments:

**Major comments**

1.- In my opinion, a fundamental part of the investigation is on how the canopy is interacting with the atmosphere (Figure 2) as well as the potential influence of non-local phenomena on the estimation of these emissions. Here my questions are:

The reviewer's comments are quite well-considered. Frankly, the canopy-troposphere dynamics from the collection of 11 sonic-anemometers active on the tower for the three years of the field-campaign could be prepared for an entire separate paper on the topic.

a) What is the level of coupling between the canopy and the atmosphere? Does it depend on the diurnal boundary-layer dynamics? Is it seasonal dependent? Perhaps here they could give a characterization of the degree of coupling by including a quantification of this coupling using an index such as the decoupling factor (Goldberg et al., Annales Geophiscae 2001, 19 581-587)

1.a:

We investigated the level of coupling between the forest canopy and atmosphere using the decoupling factor as suggested by referee #1 and described in Goldberg and Bernhofer (2001). For the campaign, the decoupling factor typically varied between 0.1 and 0.6. The nighttime-daytime transition did appear to have an influence on the coupling, with peaks in the decoupling factor occurring at sunrise and sunset times. Regarding seasonal dependence, there tended to be more decoupling in spring and fall seasons than in the summer. An example of the decoupling factor (for the spring 2016 period presented in Figure 6) is shown below. Average time of sunrise (solid vertical line) and sunset (dotted vertical line) are indicated. Shaded region indicates the range of sunrise and sunset times during the May 1st to June 30th period.
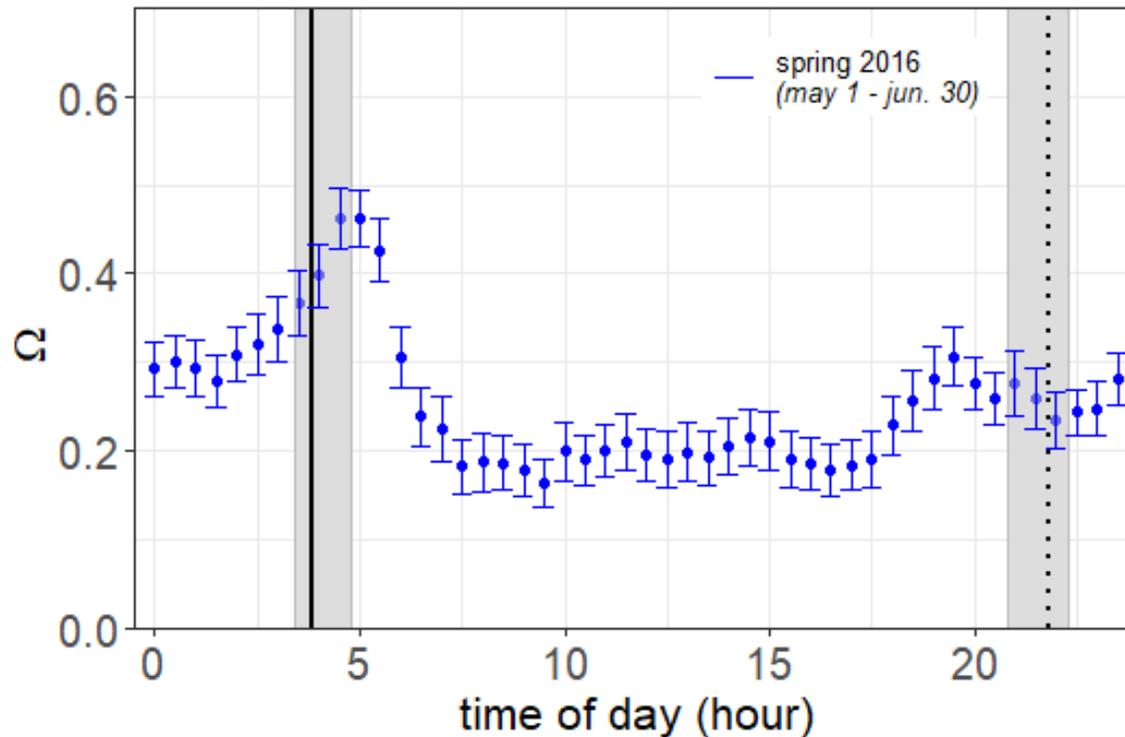
*The authors will add a discussion of the decoupling coefficient to the paper. Worked to produce the equivalent of Fig. 2 from Goldberg and Bernhofer (2001), showing the values of the decoupling coefficient during a 24h-period, that indicate the decoupling behavior seasonally (spring, summer, fall). Points in the figure above show the mean and error bars indicate standard error. We plan to add a final version of such a decoupling figure to either Figure 6 as an additional subpanel or as a separate figure.*

b) Connected to the last remark: What is the relevance of processes of non-local phenomena like entrainment or advection on their analysis? For example, at lines 339-345 they described the diurnal variability of monoterpene concentration: Is this variability entirely determined by biophysical canopy processes or by other processes driven by the atmospheric-boundary layer dyrnamics. As described by Patton et al. (2016, Journal of the Atmospheric Sciences 73, 1621-1624) there might exert non-local processes that can an influence the mean and fluxes of atmospheric composition in and above the canopy.

1.b:

Non-local phenomena to the tower were quite interesting to consider, as it had a clear impact on the concentration measurements. For example, occasional morning spikes in MT concentration at the highest inlet levels following sunrise (coinciding with spikes in $CO_2$ concentration at the same levels) were clearly connected with the mixing out of the nighttime surface layer. Figure 8 from Ouwersloot et al. is an excellent example, as more of these morning transitions were observed to occur between around 8 and 10am. A sharp delineation in the concentration profiles for ozone and monoterpenes can be seen occurring around 8am in Figure 7 as well, coinciding

with the breakdown of the nighttime stable layer above the canopy and development of the convective boundary layer.

The authors believe that the discussion of the Henry's law constant (lines 451 - 456) of monoterpenes also goes quite some way towards addressing the influence of some biophysical processes (i.e., stomatal control of emissions) versus the influence of atmospheric-boundary layer dynamics. In particular, we believe that the large Henry's law constant values for monoterpenes in general rules out the morning behavior in monoterpene concentrations at the highest flux tower inlet levels as being a stomatally-controlled burst phenomena, and rather it being controlled by boundary-layer dynamics as the lowest levels of the forest canopy mix out.

As touched upon above and later in 1.d, the diurnal interplay of biophysical processes and boundary layer dynamics in the observed mean concentrations of BVOCs (particularly for MT as discussed in lines 339-345) is quite clear in Figure 7. While the emission rate of MT is primarily controlled by temperature (Niinemets and Reichstein, 2003), with the MT emission rate actually lower at night due to lower temperatures, the nighttime influence of the collapse of the convective boundary layer and subsequent development of the stable layer above the forest canopy lead to the nighttime being the period of typically highest MT concentrations, with the inability of MT to mix-out of the forest canopy leading to high ambient concentrations.

*Revisions will be made to address in greater detail in the paper, and in particular to highlight the importance of boundary-layer dynamics.*

c) Also connected with this, much more simple inverse models as presented here show that not only the local turbulent dispersion processes can be important in determining the inferred inversion, but entrainment or chemistry (see Vila-Guerau de Arellano et al., Atmospheric Chemistry and Physics9, 3629-3640). Do they need to take into account these processes in inferring the sources and sinks of BVOC?

1.c

Regarding chemical processes (particularly breakdown and formation in the canopy atmosphere), the comment 1.c is partially addressed by the reply for 2.b. For the compounds investigated by inverse Lagrangian dispersion in this paper, chemical processes such as the breakdown or creation of compounds directly in the atmosphere typically occur on timescales much longer than the canopy mixing timescales used for the Lagrangian dispersion analysis, or friction velocity based mixing timescales, leading to low Damköhler number (Rinne et al., 2012)

For the inferred BVOC sources and sinks, entrainment is not expected to play a large role for BVOC (if not by advection) as the local production in the forest ecosystem is by the largest source locally.

The breakdown of particularly reactive VOCs (i.e., sesquiterpenes) was considered for the inversion model. During PTR-quad-MS signal analysis, however, it was found that the signal-to-noise ratio for the instrument for sesquiterpenes was too low to be useful for the analysis presented in the paper. Likely these processes (entrainment, as well as breakdown and creation of compounds directly in the troposphere) are necessary for a more holistic, generalized approach.

*Will be addressed in the manuscript, including citation to Vila-Guerau de Arellano in revised manuscript.*

d) Are the source layers (lines 205-210 or lines 357-359) influenced by the dynamics of the morning transition (see Figure 8 in Ouwersloot et al. Atmospheric Chemistry and Physics 12, 9335-9353). How much of the diurnal variability described in section 3 depends on biophysical processes and how much on the contribution of the boundary-layer dynamics?

1.d

Yes, it is quite clear during the analysis of the Lagrangian inversion source profiles that the inferred source layers are strongly influenced by the dynamics of the morning transition. This is partly why daytime averages of the model results are filtered to 1.5 hrs after sunrise and 1.5 hours before sunset, as it avoids the night/day transition of the boundary layer. The inversion model typically behaves quite poorly during the relatively rapid morning transition (i.e., numerical instability), as it is using half-hourly averaged data. As mentioned above in 1.b., Fig 8 from Ouwersloot et al. is an excellent example of the impact of the morning transition.

As mentioned in 1.b, Figure 7 for ozone and monoterpenes is an excellent example of the different interplays between VOC biophysical emission processes and boundary-layer dynamics. For instance, whereas isoprene emission is typically modeled as a function of PAR and temperature as fully de novo emission, monoterpene emission frequently has a significant component that comes from plant storage structures, with emission strongly dependent on temperature (monoterpene emission is frequently modeled just as a function of temperature)(Niinemets and Monson, 2013). This is visible in the diurnal behavior presented in Figure 7 for the profile concentrations of ozone and monoterpenes (panels a & b).

Following sunset, ozone concentrations drop as there is no more ozone mixing down from the above canopy atmosphere, yet ambient monoterpene concentrations typically peak at night, as MT emission is not solely dependent on PAR, but typically for the most part on temperature. The role of boundary layer dynamics is also clear, however, since despite MT emission being typically lower at night due to lower temperatures, the fall of the boundary layer height and stable atmospheric conditions leads to a build-up of the ambient monoterpene concentrations, as the MT at night is often unable to mix-out of the canopy.

The authors believe that the discussion of Henry's law constant (lines 451 - 456) of monoterpenes goes some way towards addressing the influence of some biophysical processes (i.e., stomatal control of emissions) versus the influence of atmospheric-boundary layer dynamics. *The authors will extend the discussion of stomatal control as a potential mechanism of biophysical processes to the other BVOCs studied, such as methanol.*

*Will address in revised manuscript by discussing influence on diurnal variability from bio-physical processes vs boundary-layer dynamics.*

e) In several section of the paper (see for instance Figure 11 and the discussion at line 490-500) dew is described as a main process that can influence the chemical transformation of water-soluble BVOCs in forest canopy. This is a relevant processes, but in my opinion, the characterization can be more complete. For instance, the relative humidity is measured a 36

meters which 8 meters above the canopy: Is this a dew situation or a fog situation? Are they measuring negative values of evaporation? Could the authors complete and better quantitation these relation between micrometeorology and BVOC?

Referee #1 brings up a valid point, that a more complete quantification of this relation between micrometeorology and BVOC can be performed.

The authors are confident that for Figure 11 this is a dew situation. The conclusion is based on the analysis of temperature and water vapor profiles from forest surface, through the canopy, and to the top of the station flux tower (102 m).

*As there is a substantial collection of sonic anemometers and other equipment on the Norunda flux tower, a profile of the water vapor content in the Norunda canopy as a function of height is also available (0.8 m to top-of-tower). The authors will address referee #1's request in the next revision of the manuscript.*

2.- I found very interesting and complete the application of the Lagrangian dispersion matrix to estimate the distribution of sources and sinks. Here, I have the following questions:

a) Would it be possible to include in an appendix an evaluation of the main variables used in this Lagrangian dispersion matrix? For instance the $s_w/u_*$ (line 211)?

2.a:

Yes.

*Will be included in revised manuscript (Table A1)*

b) I realize that this comment is beyond the scope of the paper, but perhaps it should be mentioned in the discussion. Equation (3) is valid for atmospheric tracers that have a time scale larger than the turbulent transport. As explained by Lamb (1973, Atmospheric Environment 7, 257-263) for chemical compounds with similar or shorter time scales, these theories might be violoted. Do Lagrangian dispersion matriz need to be adapted for compounds with similar Eulerian time scales (see Figure 6c).

2.b

As referee #1 points out for Equation (3), our Lagrangian inversion analysis does not explicitly account for photochemical loss or production mechanisms. As a result, in the analysis they are mixed-in with the inferred direct sources and sinks.

While attempting to adapt the Lagrangian dispersion matrix for highly reactive compounds like sesquiterpenes (typical life times on the order of seconds to minutes) may present an interesting study for future work, it was decided that such an adaptation was not necessary for the compounds presented in this manuscript. Doing so may also introduce substantially more sources of uncertainty for the inversion analysis.

As briefly touched upon in the manuscript (lines 373 – 374), based on the far-field limit of Lagrangian dispersion ($\sigma_z = \sqrt{2\sigma_w T_L(t - T_L)}$ ) described in Raupach (1989), we estimated a turbulent exchange time scale within the canopy of about 50 – 110 s during the daytime. For these turbulent exchange times, most compounds will have lifetimes significantly longer than this (for some examples of typical lifetimes, see Table 11.1 in Niinemets and Monson (2013)).

For sesquiterpenes, this adaptation was not possible for this field campaign. The PTR-quad-MS, following sample transport through the 45m of tower tubing lines (and undergoing oxidation), was not able to resolve the sesquiterpene signal with sufficient signal-to-noise magnitude to include in this paper (it would be interesting for a future experiment to repeat this field campaign design using a more sensitive PTR-ToF-MS instrument model).

*The relevance of tracer time scale for turbulent transport in the context of the Lagrangian dispersion application will be further mentioned in the discussion.*

**Minor comments**

Line 226: What sort of instability?

When the number of measured scalar input layers is equal to the number of source layers being computed, small errors in the input variables (such as the concentration, $\sigma_w$, or $T_L$), may be amplified to extremely large errors in the inferred $S_j$ source profile. This kind of inversion instability is described in Raupach 1989. Such instability issues can be overcome by including redundant concentration information when performing the inversion approach, such that $n > m$. The potential for inversion instability was also another reason that a damped least-squares approach, weighted by solution smoothness (Siqueira et al., 2000), was applied to the Lagrangian inversion analysis (line 241).

*Address by detailing inversion method instability, cite references containing examples.*

Line 231: What is the tension parameter?

Explained in more detail in the manuscript's reference *Cleveland et al. 1992* (LOESS function, and it's span setting). The tension parameter (typically referred to as the "span setting" in most reference material) is a setting for the LOESS (standing for "Locally Weighted Least Squares Regression") function in the R programming language. For the LOESS fitting, the fitting at say some point X is weighted toward the data nearest to X. The distance from X that is considered "near" is controlled by the span setting, S. When S is less than 1 (as is the case for this manuscript, which uses 0.75), <u>it represents the proportion of data that is considered to be neighbouring X</u>. The weighting LOESS then uses is proportional to 1-(distance/maximum distance)^3)^3. The default span setting in R, for comparison, is 0.7.

*Described in more detail in revised manuscript, changed description in manuscript from "tension parameter" to "span setting" (line 231).*

Figure 4: Maybe it is conveninet to include zero-line to facilitate the visualization of the figure

*Included a zero-line for Figure 4 (dashed grey). Added mention of zero-line to Figure 4 caption: "Horizontal dashed grey line indicates zero net flux level."*

Line 361: Acetic -> acetic

*Spelling corrected.*

## Response to the comment of anonymous referee #2

This is a very well written paper with a logical, quantitative, and consistent analytical approach to interpret the observations. The observations themselves are impressive largely because of the latest generation PTR instrument that the investigators have deployed. With the very low detection limits for select VOCs, the authors are able to quantify ppt level changes in concentration for many species. There is alot of data here and the authors do a pretty good job in highlighting some aspects of the data set, but there doesn't seem to be a unified scientific impact or conclusion that can be captured by the myriad presented analyses. This reviewer is led with the thought of, And so...what does it all mean?

We thank the referee for the positive comments. The significance of the results arises from detailed understanding of the within-canopy distribution of VOC sources and sinks, of which there are relatively few empirical studies. As mentioned in Karl et al. (2004), predicting the role of the biosphere in the Earth system, and understanding ecosystem-atmosphere interactions, is becoming an increasingly important part of atmospheric science. This manuscript in particular focuses on the ecosystem-atmosphere interactions for the boreal forest system in terms of the emission and deposition of biogenic reactive hydrocarbons.

As referee #2 points out, the use of a PTR instrument allowed us to investigate a range of BVOC compounds, and that are compounds significant for ecosystem-atmosphere interactions and chemistry.

Many BVOCs, such as monoterpenes, act as precursors for new particle formation and growth, influencing air quality and climate (e.g., Seinfeld and Pandis, 2016). Isoprene and other BVOCs play a role in the production and lifetime of tropospheric ozone (Chameides et al., 1992). These BVOC measurements aid our understanding of biological VOC emission processes in the boreal forest ecosystems, such as constitutive and induced emissions (including plant stress detection (e.g., Niinemets and Monson, 2013)). Moreover, these measurements (collected here as a vertical profile of the canopy) also act as tracers in atmospheric transport for studying turbulent canopy exchange (and with the amount of information collected during the 2014-to-2016 campaign, allow us to study it on a seasonal timescale).

These analyses perform the useful role of filling the existing knowledge gap in scales between bottom-up (e.g., chamber measurements of emissions from an individual leaf or branch, which can vary widely between individual plants and populations (e.g., Persson et al., 2016)) and top-down (e.g., total ecosystem-scale flux measurements made from a flux tower above the forest canopy) approaches for studying and understanding BVOC emission and deposition in boreal forest ecosystems.

*The authors will address this concern by improving the conclusion section of the manuscript to detail more clearly the impact of canopy scale processes for ecosystem-atmosphere interactions, and how the findings of the paper address these impacts.*

Application of the met data (w) from the 6 sonic anemometers at heights 4-34m to the observed concentration profiles is an interesting way to get at emissions of the individual VOCs. I very much appreciate the correlation coefficients between CO2 EC flux measurement calculations compared to the model inversion results in Figure 3. The authors rightfully show the limitations of the inverse modeling approach. It does however also lend itself to the robustness of the vertical distribution of the emissions for various species, as shown in Figure 9 and 10. Prior to this generation PTRMS, those lines would pretty much be straight up and down. This leads to the question; what is the significance of the magnitude of the difference in flux throughout the canopy? With the exception of methanol, the other species are not so exciting, and seem to fall within the range of 0. The standard error in shaded purple in figs 9 and 10 does not take into account the uncertainty associated with the inversion model, just the error associated with observations, or cps/amu. Again, the measurements are pretty impressive. How does our understanding benefit from these very precise measurements?

We value referee #2's appreciation of the utility of the CO2 EC flux and inversion model result comparison (displayed in Figure 3), as it was useful to apply the Lagrangian to other tracers in the forest canopy and to test the inversion model procedure for BVOCs, for which we unfortunately did not have EC flux data available for direct comparison purposes.

The significance of the flux differences throughout the canopy is that it reflects the differences in source and/or sink strengths as a function of height in the forest (from near surface to canopy top). This reflects key differences in the emission and deposition processes for the studied BVOCs throughout the boreal forest canopy. It enables us to investigate emission and deposition processes from top-of-canopy down to the surface vegetation, litter and even soil processes, collectively. While the dynamics of methanol sources and sinks (as shown in Figure 11) are quite engaging, the other species are also exciting in their own fashion. For example, from Figure 9, while monoterpene sources are predominantly inferred from the inversion analysis to be in the canopy during spring and summer, another strong source of emissions is identified near-surface during the fall, attributed to needle litter (line 465). This is a relevant finding, as it <u>quantifies the differences in MT emissions between different positions in the forest ecosystem by season (e.g., Hayward et al., 2001)</u>. Depending on management practices, this implies that considerable influence on ecosystem emissions may arise.

The use of shaded purple in Figures 9 and 10 was to highlight the range of mean seasonal output from the inversion model for estimating the mean source/sink strength per layer for each of the prescribed seasonal periods. The systematic and unknown uncertainties of the inversion model can be difficult to quantify without making assumptions. This is one of the reasons the CO2 EC flux and inversion model comparison was performed.

Our understanding of ecosystem-atmosphere exchange of BVOCs benefits from these measurements by differentiating between different sources in the forest as a function of height.

This addresses the gaps in our collective knowledge between leaf-scale chamber measurements of BVOC emission and landscape-scale eddy-covariance BVOC flux measurements performed above ecosystem canopies.

One technical question; with a 3/8" OD teflon tube, that means the ID is at most 1/4", and possibly less, like 3/16"; very thin wall PFA or PTFE is fragile and easily kinked, so I am not sure what wall thickness the sample tubing is; but with a pumping speed of only 20 SLPM per line, that's an approximate residence time of 45 seconds per line. This is, as the authors note, long enough for chemical loss. Did the authors try to shorten the lines at the lower elevations to see if the responses changed? I note all sample lines were consistent in length at 45 m; so I assume also that the sample lines at the lower elevations were coiled up within the trailer/shed? Why was this presumed to be a better choice than minimizing sample line length, residence time, and pressure drop by shortening the lengths that could be shortened?

The PTR instrument was housed in a permanent shelter next to the flux tower, approximately 11m away from the tower's base. As a result, since the tallest inlet height was 33.5m, the longest PFTE tube length was 45m. A total length of 45 m was therefore the minimum tubing distance that could be used for the highest BVOC inlet. The reviewer is correct to assume that all four PTFE tubing lines were 45m in length, and that excess PFTE tubing from sampling lines for inlets at lower elevations was coiled up within the instrument shelter. The four PTFE tubing lines were made the same length so that there would not be a systematic bias in the sampling from each height due to unequal chemical loss during transport through the tubing. The tubing losses will vary not only with tube length, but for each different length of tubing, will also vary over a range of temperatures, compounds, humidity levels, ect., during the measurement campaign, and so to reduce systematic bias it is typically safer to keep all Teflon tubes the same length if possible. The sampling tubing dimensions and sampling flowrate are consistent with other BVOC investigations (e.g., Holst et al., 2010), including those involving large tubing lengths and/or BVOC canopy profile measurements (e.g., Karl et al., 2004; Rantala et al., 2014). The PTFE-Teflon tubing ID is, as Referee #2 supposes, 1/4". No kinking or damage was observed during installation nor at the completion of the 2014-2016 field campaign.

The authors did not have the opportunity to test the influence of varying the tubing length, due to time constraints for the field campaign and project, however, multiple other investigations have explored the impact of varied Teflon tubing length on BVOC measurements (e.g., Kim et al., 2009; Kolari et al., 2012).

Referee #2 is right that for highly reactive compounds such as sesquiterpenes, the tubing length can have a significant impact on sampling losses. For example, Figure 6 in Kim et al 2009 (page 107) shows the beta-caryophyllene loss through 40 m of Teflon tubing as a function of temperature. For temperatures below 16 C, practically no SQT can be expected to make it all the way through without experiencing significant losses.

For the monoterpenes, isoprene and those other compounds investigated in this manuscript, which are far less reactive than SQTs loss enhancement due to increased tubing lengths are likely negligible.

For example, according to Kolari et al. (2012), a 50-m-long tubing with a flow of 1 l min$^{-1}$ causes a loss of only a few percent of a signal as compared with a 2-m-long tubing. Table 2 in Kolari et al. (2012), which includes the exact or similar atmospheric compounds investigated in our campaign, shows this quite clearly. Hence, compounded by the fact that our flow rate was 20 times higher than that used in Kolari et al. (2012), one can assume that the losses for the studied compounds in the present system would be negligible. The uncertainty of the systematic bias in the profile measurements between different heights, however, if tubing was made as short as possible but of unequal lengths, would have been more of a concern, as the differences in losses between lines would be much more difficult to address. Particularly for applying a Lagrangian inversion approach to estimate the source layer strengths, where small systematic errors between layers may lead to substantially larger errors in the inferred source profile (e.g., Raupach, 1989; Warland and Thurtell, 2000), such a varying systematic bias between the profile layers is something to be avoided.

I would like to have read more about the repercussions of the methanol observations on canopy (and above) chemistry; the lifetime of methanol is long enough where it could serve as a dominant VOC player in the area.

The authors will expand the discussion of the repercussions of the methanol observations. The role of methanol on local ambient canopy air chemistry is likely negligible relative to far more reactive isoprene, monoterpenes, and sesquiterpenes. Considering the typical lifetimes of methanol (usually on the order of days to weeks) the vast majority will likely mix-out of the canopy before having a chance to chemically react. It is above the forest canopy, in the free and upper troposphere, where methanol is likely to have a greater role in the ambient air chemistry. In particular, through its influence on tropospheric oxidative capacity and air pollution through its participation in the cycling of ozone and the OH radical.

Acetone and acetaldehyde, two other water-soluble VOCs studied in the manuscript, may also have roles in the chemistry of the upper troposphere as well (e.g., Mckeen et al., 1997), in particular the HO$_x$ cycle. The methanol results show the variety of source/sink behavior that can occur in the forest canopy, highlighting this for other water-soluble compounds (acetone, acetaldehyde, ect.) as well.

*Will expand the discussion of the repercussions of the methanol observations on in-canopy and above-canopy chemistry, as well as the discussion of other water-soluble compounds.*

The methodology and data set presented in this paper are very good and very sound. If the sampling site were more active, the results might be more interesting. To me the most interesting emission is methanol, but other than a description, not much thought is put in to the chemical impacts of such a flux. This reviewer is thus led to think that while robust, the paper is not super scientifically intriguing.

The chemical ramifications of isoprene and monoterpenes for tropospheric chemistry (ozone and aerosol formation & growth) are more relevant for boreal forest ecosystems than the immediate impact of methanol fluxes observed (though the methanol emission may have an impact on the

above-canopy atmosphere as well). A concise description of the relevance of the campaign measurements and the core conclusions is key.

*In the manuscript, the introduction and conclusion will revised to further highlight relevant findings, and the discussion of the chemical impacts of methanol and similar fluxes will be expanded.*

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