Response to Referee #1

We sincerely appreciate Referee #1's effort in reviewing our manuscript, and thank them for constructive comments which will certainly help increase the scientific quality of the manuscript. Below are our answers to the specific comments.

In the following, the original comments are in ***italic bold font*** and our responses are in regular font.

General Comments:

***(1) The authors state that, "Disadvantage of this approach (the relative flux gradient method) is that the vertical source and sink profiles of H2O, CO2, or sensible heat may differ significantly from those of BVOCs resulting in errors in the similarity assumption for K among these different tracers." However, this is exactly the assumption that they are making when they use specific BVOCs as the reference compound. The vertical source/sink profiles for different BVOCs also vary. According to their own data, methanol has a significantly large soil source whereas MBO is primarily emitted from the canopy – so these compounds have significantly different source/sink profiles. In this way, I do not see any difference between using a specific BVOC as your reference compound as opposed to some other scalar such as CO2 or H2O. It is only an advantage here because it can all be done with one instrument*.**

>> We thank Referee #1 for pointing this out and we agree that the advantage of our approach is to use one instrument to estimate the ecosystem scale fluxes for wider range of BVOCs than previously studied. So, we replace those parts by mentioning that the past approach for BVOC fluxes using flux-gradient method requires an additional sensor.

(2) Along these same lines – BVOCs that are reactive can be considered to have very different source/sink vertical profiles (i.e. there is a constant reactive source/sink with height). Patton et al. (Bound-Lay. Met, 2001, 100, 91-129) have clearly shown that the eddy diffusivity of a compound depends directly on its reactivity (or atmospheric lifetime). So you cannot apply an eddy diffusivity derived from a conserved scalar (for example, methanol) to a reactive scalar one that has a lifetime similar to the turbulent transport (example – a reactive sesquiterpene) without incorporating substantial errors. This will apply both to reactive compounds emitted that then decay with vertical transport (e.g., sesquiterpenes) as well as those produced by photochemistry (e.g., oxidation products). The fluxes that are derived via the gradients for these compounds may reflect some relative trends (such as diurnal patterns), but will likely be incorrect as a quantitative measure of how much mass is moving between the ecosystem and atmosphere. This is an especially difficult problem for SQTs since the PTR_MS measures a mixture of compounds that could range over several orders of magnitude in their atmospheric chemical reactivity. In this case, it is nearly impossible to determine a proper eddy diffusivity.

>> Most of the compounds observed in this study have longer chemical lifetimes than the vertical turbulence transport time between two measurement heights (~ a few 10ths of a second). While methanol, isoprene, and monoterpenes have substantially different chemical lifetimes in the atmosphere, our observations showed that eddy diffusivities (K) did not substantially differ from each other, at least for these compounds. As reviewer #1 pointed out, it might be challenging to properly determine K for some very reactive compounds such

as sesquiterpenes which may have shorter lifetimes than the turbulent transport times between measurement heights. When using our universal K, those compounds with very short lifetimes could have overestimated fluxes due to an enhanced gradient driven by photochemical losses between the gradient measurement heights. This is acknowledged now in the text.

(3) I am also concerned about the sampling scheme employed for the flux-gradient method. From Fig. 1 it appears that measurements are made on the lower (9.5 m) gradient level for only 6 min. out of the entire hour. The basic concept of the flux-gradient method is to measure an average concentration over the entire flux period, or, at least, a close representation of that average. I have my doubts that measurements from 12% of the flux averaging period can adequately represent the average concentration over the entire hour due to changing airflows and environmental conditions. Although concentrations from levels within the canopy can be informative, if the major purpose of the study was to report above-canopy fluxes, I would have recommended cycling only between the above-canopy inlets to obtain a better representative average concentration. This is especially true for this particular experimental setup, since the lowest inlet level is well within the roughness sublayer where it is well known that sweep-ejection flows within the canopy can create counter-gradient fluxes which invalidate K-theory (see Cellier and Brunet, Ag. For. Met., 1992, 58, 93-117 for one example – there are many others).

>> This study was focused not only on estimating above-canopy fluxes but also investigating within-canopy chemistry and characteristics of chemical evolution. So, we measured BVOCs from 5 heights (2 inlets below canopy and 3 inlets above canopy).

As the reviewer states, 6-min averaged data cannot be perfectly representative for fluxes averaged over 30 minutes or an hour. To assess the quality of the 6-min. sample's representation of VOC concentration within the one-hour period, we correlated the 6-min averaged concentrations taken from the gradient inlets with the average concentrations from the 30-min sample taken from the EC inlets within the same hour period for m/z 33, 69, and 81. We found the measurements agreed within 10% (slope of the regression line being between 0.9 and 1.1), indicating that the 6-min measurement represents the average concentration within the hour period as reasonably as does the 30-minute sample.

Other specific comments:

(1) page 25086 – description of the profiling system. What is the residence time in each of the inlets and were any inlet tests conducted to see if losses of the heavier or oxygenated compounds were occurring. Bouvier-Brown et al., (2009a) reported losses of ~ 40% for linalool from an 18-m long inlet line as well as some losses for several other compounds. Here, you have 30 m lines – although the residence time may be less.

>> Tube wall loss is expected, particularly for some sticky compounds. Under the measurement conditions in this study, the residence time of the sample air in the tube was ~2.3 s as written in Section 3.1. The tubes used in this study were Teflon and Bouvier-Brown et al. (2009a) used Silcosteel with 50^{o}C heating. So it is hard to argue how the recovery changes due to the effects of residence time, material, or temperature. However, based on lab experiments, we found that continuous high purging flow (~10 L/min) minimizes tubing wall loss of compounds presumably because the wall becomes saturated by the compounds. In addition, all tubes were the same length in this study, thus the loss rate should be very similar for each inlet.

Therefore, we expect tubing loss is not a substantial error for estimating the EC flux and vertical gradient for most compounds reported here, except possibly the sesquiterpenes, and we mentioned this information in the text.

(2) page 25088, lines 13-15. Were the mixing ratios of the 5 monoterpenes used in the calibration tank reflective of the typical ambient concentrations? This is probably not too critical as the ion-molecule rate coefficients are all typically within 10-20% of each other.

>> We used 3 monoterpenes (a-pinene, 3-carene, d-limonene) in a calibration tank which were mixed with similar ambient fractions as the 3 dominant monoterpenes at the site. We agree that the concentration calculation should be within 20-30% of each other as we indicated Section 2.2.2, and therefore do not believe this to be a critical issue.

(3) Page 25093, lines 15-19. The authors report higher SQT mixing ratios than the previous study of ambient concentrations (Bouvier-Brown, 2009a) and suggest some were missed in that study. Yet, Fig. 11 shows that the canopy-scale fluxes presented here underestimate the flux compared to scaling up of branch-level data (Bouvier-Brown, 2009c). This seems contradictory since the 2009a study measured the same SQTs as the leaf level study and quantified losses in their sampling procedure for those specific SQTs. The scaled-up emission calculated from the branch level should be for the same set of SQTs that were measured in the 2009a study.

>> Flux and concentration are different terms. The flux estimated by K-theory is not dependent on the absolute concentration in the ambient air, but rather is dependent on the vertical concentration gradient (dC). So, the flux will not increase by elevating background concentration in the ambient air. In addition, the Bouvier-Brown et al. study (2009a) was focused on the ambient SQT measurement, and they also pointed out that ambient SQT mass contribution to total terpene + methyl chavicol mixing ratio was smaller than that they studied with branch enclosure (2009c). They discussed two main loss mechansims: 1) rapid loss via oxidation in the ambient air; 2) sampling loss for a few highly reactive species with O_3. For example, in figure 4 methanol concentration is higher than isoprene+MBO, but isoprene+MBO flux is larger.

In the comparison between branch level emission and canopy scale emission for SQTs as shown in figure 11, we understand the reviewer's point that the same set of SQTs should be used. However, it is not possible to separate specific SQTs in PTR-MS measurement since the instrument can only detect the total sesquiterpenes at m/z 205, while the set of SQTs reported in Bouvier-Brown et al (2009a) was measured by GCMS allowing individual species (but not the total) to be observed. In addition, the scaled-up emission from the branch level observation (shaded area in figure 11) was calculated using Eq. (6), and was based on basal emission rates reported by Bouvier-Brown et al and temperature data from this study. We do not expect basal emission rate changes significantly even if considering missing SQTs by GC measurement in the scaled-up emission estimation.

(4) page 25094, line 10. You are limited by how many ions you can measure fast, but also by sensitivity. It would be quite difficult to measure SQTs (204) by EC even if you only did this ion! The main reason to do gradients is that it gives you the ability to signal average and increase your detection ability.

>> Thank you for this comment. Yes, we agree that SQTs are hard to measure by PTR-MS due to low transmission efficiency in the quadrupole mass filter. To overcome this, we assigned a longer dwell time for SQTs (m/z 205) for vertical gradient measurement to achieve a lower detection limit as mentioned in Section 3.1. Also, in Table 1 we added detection limits of all species measured.

(5) page 25094, eqn.4. Eddy diffusivity is typically parameterized as a function of height. Please state the height at which you are calculating the eddy diffusivity.

>> We used two heights (17.8m and 9.5m) for the term 'dz', and it has been mentioned for Eq. 5 since we used it to calculate K_{univ}.

(6) page 25095, lines 19-22. You do not show the comparisons between the K's determined by the 3 different species (only the diurnal averages). But this is an important part to the story – this shows that, regardless of individual source/sink profiles, you can use a single Kuniv to describe the eddy diffusivity – i.e., that your similarity assumption works in this situation! If you use EC flux and gradients to compute Kuniv over a given hour – it seems a bit circular to then go back and say that the gradient fluxes agree with the EC fluxes. Then the fluxes should only show variability only due to the timing mismatch (i.e, the hourly gradient flux does not completely overlap the 30 min . EC flux). One further question here – did the authors compute eddy diffusivity and/or gradient fluxes between the two upper gradient levels (13.6 and 17.8 m). This would be a good internal check.

>> Although we had already briefly mentioned the comparisons between the K's in the text (page 25095, line 19 – 21), we have added Table 2 to show the slopes and R-squares between each K (i.e. K_{m33} vs K_{m69}, K_{m69} vs K_{m81}, and K_{m81} vs K_{m33}). In addition, we tried to compute eddy diffusivity using the two upper sampling levels, but much less data passed data quality control since concentration differentials were too low (i.e. K-theory requires well resolved gradient between 2 heights), so it was hard to perform a comparison with such a limited data set. Thank you for the suggestion, though, which leads to the conclusion that a minimum distance between two measuring heights above the canopy should be allowed in order to accurately enough measure concentration gradients suitable for k calculation.

(7) page 25096, lines 17-21. The authors report observing both emission and deposition for several ions (masses 71, 113, 151, etc.) that were monitored. The reported fluxes were very small and variable. Not only would this be sensitive to not measuring a representative concentration at the lowest inlet height, but could also be sensitive to wind flow patterns and turbulence changes. Small changes in horizontal and vertical advection could easily be reflected in the observed small gradients and imply a vertical turbulent flux.

>>We used the data only when the differences in concentrations were above the instrumental detection limit. The emission (/deposition) fluxes generally indicate higher concentration at lower (/higher) height since most K_{univ} values were positive. Some bad conditions of advection or turbulence were filtered out in QA/QC procedures (e.g. stationarity, block average, tilt angle of anemometer) before calculating K_{univ}. Thus, we think 'small and variable flux' is real flux and it can be estimated by K-theory.

(8) Page 25100, line 25-30. It is a bit presumptive to assume that the majority of SQTs emitted at this site are highly reactive and would be the cause of the measured lower emission rates. The major identified SQT from prior leaf level and ambient measurements

was a-bergamotene. Its reaction with ozone has not been studied to my knowledge. Only estimates of its rate constant have been reported (Bouvier-Brown et al., 2009a) with a fairly moderate reaction rate (giving similar atmospheric lifetimes for reactions with both ozone and OH). If the SQTs at this site are reactive and you are using a passive-scalar eddy diffusivity (see General Comment #2), then your discrepancy reported here is even larger, since chemistry is causing you to measure too large of a concentration gradient and overestimate the emission flux.

>> As reviewers #1 and #2 pointed out, SQTs flux by K-theory may have high uncertainty due to chemistry that causes bigger concentration gradient between two measurement heights. As we accept two reviewers point we add a sentence below to address possible overestimation of sequiterpenes emission flux.

“Moreover, assuming the majority of sesquiterpenes are very reactive with hydroxyl radical and/or ozone, sesquiterpene flux estimated by K-theory may be considered an upper limit due to chemical loss between two measurement heights”

Response to Referee #2

We would like to thank Referee #2 for the constructive comments and corrections which will certainly help increase the scientific quality of the manuscript. Below are our answers to the specific comments.

In the following, the original comments are in ***italic bold font*** and our responses are in regular font.

Specfic Comments:

1) I don't understand the argument not to use CO2 and H2O or H (sensible heat) fluxes to determine the eddy diffusivity K which then are used to estimate fluxes of other compounds (like BVOCs) from a concentration gradient above the canopy. The authors state that CO2, H2O or H sources and sinks are much different from those of BVOC, and that it would be much better to use BVOCs instead to obtain K. However, this is not completely true, as there are different sources and sinks for the variety of BVOCs as well. For example, the authors themselves discuss that e.g. sesquiterpenes (SQT) are very reactive and interact on short timescales within the canopy, while they did not found this for other compounds (e.g. monoterpenes, MT). Furthermore, they themselves mention different production processes (growth for methanol, temperature controlled emission from pools for MT, temperature and light controlled processes for m/z 69) and speculate about different sources (e.g. emission from soils for methanol). It rather would be interesting to see, if K-values obtained from CO2, H2O or H-fluxes would lead to higher variability compared to Kuniv averaged from Km33, Km69 or/and Km81.

>> As all reviewers pointed this out, we have decided not to mention source profiles of different scalars. This was because: 1) We did not actually compare with other non-reactive compounds commonly used previously such as CO_2 and H_2O; 2) Our measurement demonstrated that K values derived from direct measurement of BVOCs having different sources and chemical lifetimes are in a good agreement. We remarked in the text that our goal was also to demonstrate that a universal k can be calculated using the same instrument, since other scalars are not always available at a field site.

2) I don't know which additional data from that specific site would be available, but instead (or, in addition) using the (disjunct) EC-fluxes to invert their Eq 4 and to resolve for K, they could also use K=0.4 z u* with u* from the sonic (e.g. Foken, Stull). And as I guess that the sonic data was recorded continuously (even during the time of the profile measurements) this could avoid the problem of the time delay between fluxes and profiles.

>> Thanks for the suggestion. The approach using von Karman constant (with z as measurement height or considering displacement such as z-d) is known to fail in the roughness sub-layer (1.5 – 3 times of canopy height), and in a tall canopy environment it is challenging to determine displacement.

3) Another potential problem of the K-theory is that it might have problems in convective conditions, i.e. during noon when the fluxes are highest. Furthermore, in my opinion you probably should not use the level at 9.5m (L3) but L2 at 13.6m instead as L3 is close above the canopy (8.8m from Fig 1) and you most likely don't have a well developed turbulence there (but patterns of large scale eddies penetrating the canopy layer). This

would not work well with the K-theory. However, the BVOC gradients get smaller as well and might approach the measurement uncertainties…

>> It is known that K-theory based on an aerodynamic approach often fails in the roughness sub-layer. However, with real measurement for BVOCs, our study result clearly shows very similar K values among 3 dominant compounds at the site, even if their source/sink strength and chemical life times are varied. This suggests that use of K-theory by direct BVOC flux and vertical gradient measurement is reliable for estimation of fluxes of other compounds not measured using EC. In order to test the reliability of this conclusion, we applied the method to historical data from the same site, and we confirmed that the fluxes estimated by K-theory using K_{univ} were similar in magnitude to those discussed in this manuscript.

For the heights we selected for vertical gradient data, we refer to our response to Reviewer #1's specific comment (6).

4) The site description (p25085) mentions a clear day-night pattern in wind direction, which might be a circulation system causing advective flows. Can you comment on this? Furthermore, you mention a new tower was placed north of the old one, but only 10m away. You state that the wind direction during nights is predominantly from 30 - 60^o, and during day from 220 – 280^o, so there is a risk that the towers might influence the measurements. Did you check this?

>> Advection normally affects background concentration change, equally affecting concentration at all measurement levels, thus the gradient (dC) should not be changed. The axis between the old and new towers runs roughly perpendicular to typical daytime and nighttime wind direction, thus influence of the towers upon each other should be minimal. Also, advection flow influence is expected to be small since the footprint of the forest is homogeneous over a very large area.

5) You should briefly mention the measurement levels for the meteorological data (p25085).

>> Yes, we now mention all the levels for south tower measurements (i.e. at 12.5m, 8.75m, 4.9m, 3.0m, and 1.2m).

6) Instrument setup: the mismatch of the sampling times for EC-fluxes and K-theory might cause some problems, especially during times when conditions change rapidly, e.g. during morning and evening hours. Could that be the outliers in Fig 8?

>> We agree and have considered this point. To minimize the impact of the time interleaving between EC and gradient measurements, we interpolated concentration data between gradient measurements, calculating K's to match the EC-flux measurement time. Resulting fluxes measured by EC vs. K_{univ} for m/z 33, 69, and 81 showed no significant difference (within 10% in slope and R-square > 0.87).

The outliers in Fig 8, are scattered in time-of-day, and do not occur in specific time periods such as morning or evening.

7) Was the BVOC inlet heated to avoid condensation in the inlet line?

>> No, the inlets were not heated. However, we didn't find any condensation in the inlet during the campaign. A high flow rate was maintained through the sampling line in order to minimize memory effects in the tubing and air stagnation that might promote condensation.

8) Section 2.2.2; calibration: Your calibration routines (p25087) remain unclear to me. I assume the first mentioned calibration (twice daily) is a two point calibration (zero and one concentration but several masses), the second one performed irregularly during instrument service is a 5 (6)-point calibration but only for d-limonene? Could you clarify this?

>> Multipoint calibration was conducted before/after every instrument power off/on using the same standard cylinder used for daily calibration. The multipoint calibration measured all of the same masses that were measured in the twice-daily two-point calibrations. In the text we showed d-limonene as an example of the concentration levels used. To clarify this, we removed this example, and added a statement 'using the same standard cylinder used for regular base calibration'.

9) P25089, L9: did you apply any filtering to remove data with too low turbulence, e.g. based on u*?

>> Filtering by u* might be important particularly for night time data when turbulence is lower. However, we didn't use this criterion to filter the data. The cutoff value for u* is controversial (0.1 m/s – 0.2 m/s), and we think it doesn't significantly matter for our calculation for K's since night time K's are almost all around 0 (very small flux at nighttime). Even if we use this filter, our data used in calculating K's are only few points.

10) P25091, L4pp: can you comment on the storage term calculations? Methanol and MT were emitted during night as well under low turbulence, m69 is light-dependent and should be emitted when conditions start getting turbulent. Would you have an example of a reactive compound that should be emitted under low-light conditions, but show a weak storage?

>> In response, we added two references (Rummel et al.,2007 and Wolfe et al., 2009) for some readers who may be interested in the storage term.

As briefly explained in the manuscript, when the thermal stratification of the atmosphere is stable or turbulent mixing is weak, BVOCs emitted from leaves and the soil may not be measured by EC flux. Under such conditions the storage term becomes non-zero, so it must be added to the eddy covariance flux measurement if we expect to obtain a measure of BVOCs flowing out of the soil and vegetation. The storage term can be calculated by Eq (3) in the manuscript as the integration function on the right hand side. The storage term at night for BVOCs is negligible, because the concentration decreases by the early morning due to nighttime oxidation processes, along with decreasing emission from the plant. This results in little accumulation within the canopy. However, in the morning after sunrise (hour 6-8) BVOC concentration suddenly increases below the EC flux measurement height, with stable (or neutral) air conditions, and this substantially affects total flux. In addition, with PTR-MS measurement alone it is nearly impossible to determine which specific monoterpene and sesquiterpene compounds are being directly emitted from plants. Thus, it is hard to which reactive compounds having weak storage in the canopy are emitted under low-light conditions.

11) P25092, L27: from fig 4b, I'd say that this is a rather pronounced vertical gradient for m69 and you present high fluxes for m69 as well, so this statement is unclear to me.

>> Even though *m/z* 69 includes MBO and isoprene in concentration, flux of *m/z* 69 is mostly contributed by MBO since isoprene emission at this site is minor as mentioned at the last

paragraph of Section 3.2. So, isoprene concentration within m/z 69 does not contribute much to the flux, but it changes the background concentration of m/z 69.

12) P25095, L15: was Kuniv even calculated when not all 3 K-values could be calculated, i.e. when Km69 was missing? How many K-values did you use for Kuniv at least?

>> We used at least one K value to determined K_{univ} when others were not available. We rephrased this in the text: 'At least one to three K values (K_{m33}, K_{m69}, and/or K_{m81}) were calculated for every hour, and averaged to a K_{univ} of which was then applied to estimate fluxes for all compounds with measured vertical gradients.'

13) P25095, L24pp: I suppose you try to avoid comparing K with K taken from the inverted equation. Either you for example use another way to get K (see comment above), or you take only Km33 and Km81 but leave Km69 for validation as anyway only 51% of Km69 were available. However, m69 is light-dependent, m33 and m81 are not.

>> We added a table with slope and R-square to compare three K's each other, and all these are in good agreement. Also we found, K_{univ} is more relevant to use since more averaging reduces uncertainty. The reason that K_{m69} was less available is the nighttime m/z 69 gradient is too small to be well resolved, since it is a light-dependent species. Flux by K-theory does not consider light and/or temperature dependence of a species directly, but the vertical flux is determined by calculating the constant K against the vertical gradient, whose strength may or may not be impacted by light and/or temperature. For example, no m69 emission at nighttime due to light-dependence results in no gradient and vertical flux, while strong vertical gradient of m33 and m81 (not light dependent) leads to fluxes even at nighttime. However, K values at nighttime are very small due to low turbulence of the air, and thus, a tiny flux even of compounds having a strong vertical gradient.

14) P25097, L1pp; and fig3: are the mentioned fluxes EC-data, or data calculated by the K-theory?

>> We presented EC-flux data in both figures for methanol (m33), MBO+isoprene (m69), and monoterpenes (m81). So, we added 'EC' in front of 'flux' if we used EC flux data to clarify between flux by EC and K-theory.

15) I actually like the discussion with 'older' data and analyses from the same site (back to 1999). There are not many sites where this is possible!

>> Thank you!

Response to Referee #3 (Dr. Ruuskanen)

We thank Dr. Ruuskanen for the insightful review.

Specfic Comments:

-The authors use the term flux for eddy covariance flux as well as for flux in general. Please correct 'flux' to 'EC flux' at least on page 25088, lines 14 and 17 and page 25095, line 5.

>> Thank you for pointing this out. As suggested we corrected the term 'flux' with EC flux if we used EC flux data in the text.

Introduction:

-P25083, line 13. Presently also the use of time of flight mass spectrometers enable direct EC flux determination of unlimited number of VOCs, e.g. Ruuskanen, Müller, Schnitzhofer, Karl, Graus, Bamberger, Hörtnagl, Brilli, Wohlfahrt, Hansel, Eddy covariance VOC emission and deposition fluxes above grassland using PTR-TOF, Atmos. Chem. Phys., 11, 611–625, 2011. Please correct the text accordingly.

>> We added the following sentence with references after stating the limitations of PTR-MS in EC flux.

"Recently, use of the new time-of-flight mass spectrometer (PTR-TOF-MS) allows measurement of full mass spectra at high mass resolution fast enough to measure fluxes for all masses simultaneously (Ruuskanen et al., 2011, Park et al., 2013), yet it is more expensive than the PTR-MS."

-P25083, line 20: Flux gradient method was not initially introduced by Goldstein et al., the only one referenced here. Many comparisons of indirect gradient fluxes with the fluxes from the direct eddy covariance method have been done in the 1980's. Please also reference them, one possibility is an overview by Baldocchi, Hincks, Meyers, Measuring Biosphere-Atmosphere Exchanges of Biologically Related Gases with Micrometeorological …, Ecological Society of America, 69: 1331–1340. Please add reference.

>> We added a reference as suggested, though VOC flux estimation by flux gradient method has only been reported in a few studies previously and Goldstein et al was the first to do this with in-situ VOC measurements.

-P25084, line 1: In addition to different source profiles, the fluxes and thus gradient K values are influenced by chemical reactions mainly leading to loss of reactive VOCs in during the transport from emission to the measurement point, e.g. Rinne et al., Effect of chemical degradation on fluxes of reactive compounds – a study with a stochastic Lagrangian transport model, Atmos. Chem. Phys., 12, 4843-4854, 2012.

>> We thank you for the suggestion and excellent reference. However, per our response to reviewer #2's comment 2), we have decided not to mention source profiles of different scalars.

Methods:

-Page 25085, line 25. Please clarify to text: Were the moments contaminated by the generator removed from the dataset? If not, why?

>> Yes, we removed a few data points that were affected by the generator exhaust, particularly for benzene (m/z 79). We added the following sentence to mention this in the text.

‘we removed generator contaminated spikes such as benzene (*m/z* 79).’

-Page 25090, line 27: correct spelling ‘mesaurements’ should be ‘measurements’

>> Thank you.

-Page 25091, line 5: Please clarify how was the storage affect taken into account? Where the gradient flux measurements during the morning hours 6-8 o’clock corrected for the storage that was over 50% of the EC flux for methanol and monoterpenes? Please add how much the storage accounted for in the night. Please add: What criteria were used to filter values due to noise from real flux from the gradient flux values?

>> We refer to our response to comment 10) from reviewer #2 above. As requested, we added a mention of the nighttime storage term by stating ‘were extremely small or negligible (less than 0.7%) during the daytime (08:00 – 18:00 PST) and at nighttime (20:00 – 06:00 PST).’.

For the data filtering, we did not filter out any flux data below flux detection limit because we think very small flux is also real flux if we have a good measure of both wind speed and BVOC mixing ratio. However, we removed the vertical gradient data if the gradient is too small (i.e. not well resolved) in calculating K values as stated in the section 4.1.

Results:

-Page 25092, line 10: Please rewrite: ‘Diurnal patterns of… … peak in the morning…’ Consider changing to ‘Mixing rations of … …were highest in the morning…’

>> We rewrote as suggested.

-Page 25093, line 10 and Figure 5: are the measurements above the noise level? Please add detection limits of each m/z to methods or Table 1.

>>Now the detection limits for each *m/z* are in Table 1.

-Page 25094, line 10: Please consider editing: starting the section where ‘Fluxes of the additional 14 measured BVOC species were determined..’ (Page 25096, line 3) by stating that ‘Fluxes of the other 14 species selected for vertical gradients (should be vertical gradient measurements) were not measured…’

>> Yes, we changed this sentence. Thank you

-Page 25094: The authors derive K from methanol, isoprene-MBO and monoterpene measurements so that it would fit the sources and sinks of also the other 14 VOCs better

than CO2 flux. However, the evaluation of sinks e.g. chemical loss is discarded. More importantly the authors show that methanol has a different source profile than the isoprenoids (Fig 4), and appears to be emitted from the forest floor. Please add discussion on: Why is the universal K derived from the three different VOCs and how much does it differ from the K derived from CO2 and water vapor?

>> As mentioned above, we omitted the sentence discussing different source profiles between BVOCs and CO_2/H_2O in both the introduction and analysis sections. In addition, we did not measure CO_2 and H_2O vertical flux and gradient data at the same location, so it is hard to argue if there is significant discrepancy between those K values.

-Page 25096: Please add to text information on the reliability of the gradient fluxes, all values derived might not be due to a flux, but due to noise in a low signal: Did the small positive and negative fluxes of m/z 71, 113, 155 pass the quality controls and were the mixing ratios also above detection limit?

>> Yes, we removed mixing ratio data below detection limit. In addition, we should be very careful about filtering low fluxes under quality control if any compound has bi-directional flux and gradient is small then flux should be also small. So, flux detection limit cannot be defined particularly for bi-directional flux species.

-Page 25099: Degeneration of organic matter e.g. soil litter is also a well known source of methanol release in the understory. Could the sources of methanol emissions be dominated by release from plants trough stoma (T and PAR driven release) during the day and litter decay (T driven release) in the night?

>> Yes, it is possible. Our comparison between the T algorithm and T+L algorithm was more focused on daytime data. We inserted this information.

-Page 25100, line 4: Add references.

>> We added a reference. (Bouvier-Brown et al., 2012) which briefly described understory shrub removal in 1999.

-Page 25101, line 18: please remove the circular statement of 'excellent agreement' of EC and gradient fluxes derived from EC measurements.

>> We omitted the sentence as suggested

-Fig. 7: Please edit the figure colors, so that the Kuniv line is darker also in legend than the shaded area in a B&W print. Please change std to percentiles e.g. quartiles to present the variation of the K values. Use of std requires normally distributed data, and the K values clearly are not

>> We newly created figure 7 with all individual K's to show the range of K's and used 10-90th percentile for K_{univ}.

-Fig. 9: Please check, do the vertical bars denote standard error or deviation from the mean? Please change std to percentiles (see comment on Fig. 7.).

>> We changed the error bars with 25-75th percentile.

-Fig, 10: Are the fluxes derived from gradient method or both methods? Please clarify to the caption.

>> We clarified this in the figure caption that indicates the EC flux data were used for MBO+isoprene, monoterpenes, methanol, and the others are estimated by flux-gradient similarity method.

-Fig. 11: Please check the legend. Change to consistent form e.g. 'Compound (m/z)', also in the other figures. The total hourly mean BVOC diurnal flux in Fig. 10 is valuable, hopefully in the future the authors will continue on the determination deposition of oxidation products and consider devoting measurement time on the for the direct eddy covariance method.

>> We changed it. We also hope to study OVOC deposition in more detail.

Thank you.