

**RC: The manuscript aim to relate BVOC emissions to ambient mixing ratios thereof. That is to my opinion a valuable and needed topic to address because many models yet explain bare emissions and it has to be further estimated what ambient mixing ratio or concentration that will be. However, that ambient values are more crucial for a proper air chemistry and to assess the emission impact on the atmosphere as well as immediate feedbacks to the within the atmosphere-biosphere system. One very positive topic in this manuscript is therefore the "feedback" possibility by applying ozone, deposition and dilution losses to the emission algorithm.**

**The major problem of the manuscript is, to my opinion, that the authors get lost in a large set of possibilities the data allows to discuss. That lead to a broad but in many places speculative arguing. An example is the diel variation in emissions, without physiological parameters like photosynthesis, evapotranspiration, light, temperature and humidity given on a sub daily scale the special shape of such emissions can't be discussed. Also, the correlation analysis between oxygenated and terpenoid compounds (representing rather different biophysical and biochemical processes) would need further information and qualify a separate publication. Here, that disturbs a clear statement how the "translation" from emitted VOC to their ambient mixing ratios is conducted as given by the title.**

**AC: We thank anonymous reviewer for the positive evaluation of our attempt to link the emission rates with the ambient mixing ratios. We agree that our data provides a large set of possibilities to be investigated. We have tried to formulate the manuscript such that it presents the most important trends observed, discussing in detail the strengths and weaknesses of each point. For example, since we do not have physiological parameters such as evapotranspiration and photosynthesis measured, we have calculated the absolute humidity values as an indication of the leaf to vapor pressure deficit, which is the driving force for transpiration and stomatal behavior. In the revised manuscript the diel variations in emissions will be improved with similar diel patterns of temperature, light and humidity but their discussion section will be deleted as suggested**

**We believe that our correlation analysis between the oxygenated and terpenoid compounds fits within the scope of the manuscript and does not require a separate publication. Our intention was to investigate how (similar to emission rates in Fig. 6) these correlations are changing in outbound conditions. Within this context we agree, that the lack of additional physiological measurements restricts our potential discussion of the biophysical and biochemical processes that occur. But our discussions, even if they are speculative, may help to point out the potential feedback mechanism to enhance further discussions and measurements.**

**How the emissions are used to estimate local ambient mixing ratios will be explained in expanded form in the new manuscript.**

**RC: The measurement system used is an open chamber approach that would need a proper set mass balance (and by that box model) equation to assess the dynamics of the measurements and then apply that as corrections to the emission algorithm used. Currently, it remains a small miracle how the equations have been formulated.**

**AC: The equation has been formulated without miracles but with the use of current published knowledge, obtained by previous studies (eg Ruuskanen et al., 2005; Tarvainen et al., 2005; Hakola et al., 2006; Ortega et al., 2008; Rottenberger et al., 2008; Aaltonen et al. 2011; Mochizuki et al. 2011; Kolari et al., 2012):**

$$E = F \frac{C_2 - C_1}{m}$$

The only difference (which is actually a development) is that  $C_2$  has been corrected for the mentioned effects of dilution, deposition and chemical reactions.

Aaltonen, H., Pumpanen, J., Pihlatie, M., Hakola, H., Hellén, H., Kulmala, L., Vesala, T., and Bäck, J.: Boreal pine forest floor biogenic volatile organic compound emissions peak in early summer and autumn, *Agr. Forest Meteorol.*, 151, 682–691, 2011.

Bourtsoukidis, E., Bonn, B. and Noe, S. M.: On-line field measurements of BVOC emissions from Norway spruce (*Picea abies*) at the hemi-boreal SMEAR Estonia site, under autumn conditions, *Bor.Res. Environ.*, 19, in press, 2014 (<http://www.borenv.net/BER/pdfs/preprints/Bourtsoukidis.pdf>)

Hakola, H., Tarvainen, V., Bäck, J., Ranta, H., Bonn, B., Rinne, J., and Kulmala, M.: Seasonal variation of mono- and sesquiterpene emission rates of Scots pine, *Biogeosciences*, 3, 93-101, doi:10.5194/bg-3-93-2006, 2006.

Kolari, P., Bäck, J., Taipale, R., Ruuskanen, T. M., Kajos, M. K., Rinne, J., Kulmala, M., and Hari, P.: Evaluation of accuracy in measurements of VOC emissions with dynamic chamber system, *Atmos. Environ.*, 62, 344–351, doi:10.1016/j.atmosenv.2012.08.054, 2012

Ortega, J. and Helmig, D.: Approaches for quantifying reactive and low-volatility biogenic organic compound emissions by vegetation enclosure techniques – Part A, *Chemosphere*, 72, 343–364, 2008.

Rottenberger, S., Kleiss, B., Kuhn, U., Wolf, A., Piedade, M. T. F., Junk, W., and Kesselmeier, J.: The effect of flooding on the exchange of the volatile  $C_2$ -compounds ethanol, acetaldehyde and acetic acid between leaves of Amazonian floodplain tree species and the atmosphere, *Biogeosciences*, 5, 1085-1100, doi:10.5194/bg-5-1085-2008, 2008.

Tarvainen, V., Hakola, H., Hellén, H., Bäck, J., Hari, P., and Kulmala, M.: Temperature and light dependence of the VOC emissions of Scots pine, *Atmos. Chem. Phys.*, 5, 989-998, doi:10.5194/acp-5-989-2005, 2005.

In the revised version we will present the equation in a more detailed manner in order to avoid similar confusions:

“Emission rates of the measured VOCs were calculated by using the mass balance equation (eg. Ruuskanen et al., 2005):

$$E = F \frac{C'_2 - C_1}{m} \quad (1)$$

$F$  is the total flow rate through the chamber,  $C_1$  is the concentration at the last measurement interval before the cuvette closes and  $C_2$  is the last measurement conducted with closed cuvette. The final value is corrected for dilution ( $k_{dil}$ ), ozone reaction losses ( $k_{VOC}[O_3]$ ) and dry deposition on the cuvette walls ( $k_{dep}$ ):

$$\text{Where } C'_2 = C_2 \cdot \exp((k_{dil} + k_{chem} + k_{dep}) \cdot t)$$

Time  $t$  is the closing period ( $t=3\text{min}$ ), while  $m$  is the total dry needle biomass of the enclosed branch. Hereafter, biogenic emissions were quantified in  $\text{ng g(dw)}^{-1} \text{h}^{-1}$ . More details on the correction terms applied can be found in Bourtsoukidis et al. (2012) “

**RC: I do not challenge the choice of the emission algorithm chosen, but everything should be discussed carefully taking into account that the temperature dependent equation has a rather small explanatory power (see r2 in table 2). Especially if there are obvious other emission driving factors discussed.**

**AC:** This paper presents the variability of temperature dependencies under different environmental conditions. We will further expand our discussions on this topic (see specific comments below) but one should also notice the large amount of data that were used for the fittings ( $4391 < N < 16200$ ) and keep in mind that the measurements have been conducted in the field and not inside a laboratory with controlled conditions.

**RC: I would skip the correlation analysis and take the diel variations as they are without discussing them as there are lacking informations on the physiological state of the branch.**

**AC:** We prefer to retain the diel correlations but will improve the plot in response to the reviewers critique by adding diel variations in temperature, O<sub>3</sub>, radiation and absolute humidity. Although biophysical parameters were not directly measured the biological proxies would provide valuable information to the manuscript. The detected abiotic parameters are commonly used because of their accessibility with respect to measurements and in models and thus kept as valuable information for the reader.

**RC: A focus could be drawn on the change in the emission composition (Fig. 4) and it's possible consequence for the seasonality in emissions and further in context to Fig. 8 where the emissions are mapped to ambient mixing ratios. Here, the non-linearity should be discussed, at least for oxVOCs.**

**AC:** We appreciate the suggestion and we will revise the manuscript accordingly. For specific revisions please see the specific comments.

### Specific Comments

**RC: Page 30189, line off: The sentences in the end of the abstract are very unclear. What do you want to tell here? Do you apply only temperature dependent emissions or as it should be light and temperature dependent emissions? Spruce emit both ways, see Ghirardo et al. 2010 Plant Cell & Environment doi:10.1111/j.1365-3040.2009.02104.x for a recent reference.**

**AC:** The intention was to emphasize the deviations between the algorithm and the monoterpene emissions as they can be seen in Fig. 9. The last sentence is revised to

*"Finally, we evaluate the temperature dependent algorithm that describes the temperature dependent emissions by grouping the data in ten different temperature regimes. Highest deviations between the algorithm and the measurements were observed for monoterpenes. The observed discrepancy was attributed to the additional light dependency of monoterpene emissions that was observed"*

**RC: Page 30190, line 9: I guess, you want to tell northern hemisphere vegetation instead of "north hemisphere..." here.**

**AC:** Correct. This will be changed in the revised version.

**RC: Page 30191, line 1: "... they grow in" instead of "grow at"**

**AC:** Will be corrected

**RC: Page 30193, line 9: What is "leak tight"? Given your reference to Ruuskanen, that mean the cuvette is open, not tight. It should be clear as you later need this fact in your equation to describe the situation when measuring.**

AC: Thank you for this point. It was meant to be "not leak tight". We will reformulate the sentence as follows:

*"During the closing period, ambient air was allowed to enter the enclosure by a small hole at the back of the chamber in order to avoid pressure differences (Ruuskanen et al., 2005) while an inbuilt fan ensured homogenous mixing."*

**RC: Page 30195, line 14ff: I would not name equation 1 a "mass balance" equation, that is a source term for an emission. A mass balance should be  $dy/dt = \Sigma k_1 y - \Sigma k_2 y$  with a set of rate parameters  $k_1$  that represent source terms and a set of rate parameters  $k_2$  that represent the sink terms operating on the vector of relevant state variables  $y$ . Equation 1 can be the solution of such a mass balance equation as a function of time. I further assume you mean  $k_{VOC} [O_3] = k_{chem}$ . This should be written or defined here.**

AC: Yes, we agree that equation 1 is the solution of a mass balance equation. Detailed information on this equation has been provided in the general comments. We will also further clarify as suggested by stating

*" $k_{VOC} [O_3] = k_{chem}$ "*

**RC: Page 30195, line 23: I guess you mean 3 minutes, not seconds here. At least the cited papers used that time interval for closing the chamber.**

AC: You are right, we meant 3minutes.

**RC: Page 30196, line 2: What are "nearby tree emissions"? Do you mean the tree itself you measure (i.e branches outside the cuvette) or the emissions from trees in the surrounding? That is very unclear. As you want to refer to ambient concentrations  $C_a$ , that need to be well defined. Given your sentence here,  $C_a$  is all outside your cuvette! This may vary for several places (inside or above) canopy or near to a forest edge etc but still would stay just a  $C_a$  for your situation. Otherwise you need to define some  $C_{a0}$  that is ambient concentration far away from sources or so.**

AC: We mean the mixing ratios that would have been measured if the cuvette was not there and we had just measured the mixing ratios with the PTRMS inlet. We appreciate the suggestion of naming the  $C_a$  into  $C_{a0}$  as the surrounding ambient mixing ratios of the measurement point.

**What do you further mean with "steady state of the measured monoterpene signal"? Is that you have been waiting long enough to have constant chamber concentration reached? Or, a constant emission?**

**AC:** Emission rates cannot be dramatically altered in a 3 minute period, unless a driving force is drastically changed. Therefore, we mean the monoterpene mixing ratios as they are monitored by the PTR-MS.

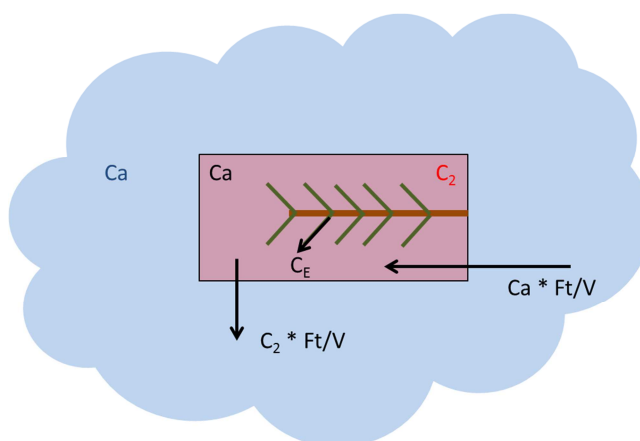
**Equation 2 does not make real sense as it tells that  $C_a$  the ambient concentration equals some concentration  $C_2$  minus kind a difference drawn away by a flow and kind a dilution because that is replaced by the ambient concentration with the same flow. To my opinion, here should be the chamber's concentration that equals these source/sink terms. In general, I would recommend to present a real mass balance equation (this actually also defines a box model) with all its sources and sinks relevant. From that, it is possible to derive all the solutions that describe the dynamics of the system presented here.**

**What is the unit of  $C_a$  if solved equation 3 replacing the variables by their appropriate units?**

**Another problem according to units, you state in the description of equation 1 that  $C_1$  and  $C_2$  are concentrations, therefore something given as mass/volume as usual in atmospheric sciences. Equation 3 seems to end up in pptv which is a mixing ratio. Here it is needed to clarify the units and possible conversions to explain the equations you used for estimating the ambient mixing ratios.**

**AC:** We are very grateful for this comment, since through it we have realized that there was a term missing in our balance approach. This additional term corresponds to complete mass balance approach proposed by the reviewer as it takes into account the mass loss through measurement from the cuvette. The new results when this term is included do not change substantially but a small increase in the calculated ambient mixing ratios is observed. Therefore, Fig.1, table 3 and Fig. 8 will be revised accordingly.

The detailed logic on formulating eq.2 is illustrated below:



Taking into account the dynamics of the system, the measured concentration  $C_2$  will be:

$$C_2 = C_a + C_E + C_a \frac{Ft}{V} - C_2 \frac{Ft}{V}$$

$C_1$ ,  $C_2$ ,  $C_E$  and  $C_a$  are concentrations ( $\text{mg}/\text{m}^3$ ). We chose to present the difference (line 16) between the measurements and the model in  $\text{ppb}_v$  since it is easier for the reader and since we wanted to present the rest of our results in volume mixing ratios. This will be clarified in the revised version.

The remaining comments are embodied in our revised text below:

“When the enclosure is open, it can be assumed that ambient concentrations ( $C_{a0}$ ) are identical to the concentrations measured by the PTR-MS when the cuvette is open ( $C_1$ ). Here, by ambient concentrations we define the concentrations that would have been measured at the same point but without the chamber present. Nevertheless, the measurements were conducted inside a chamber with an enclosed branch and our inlet was located at the inner side of the chamber. By using a box model when the chamber is closed and by assuming steady state of the measured monoterpene signal, we can derive an equation that calculates the ambient concentrations:

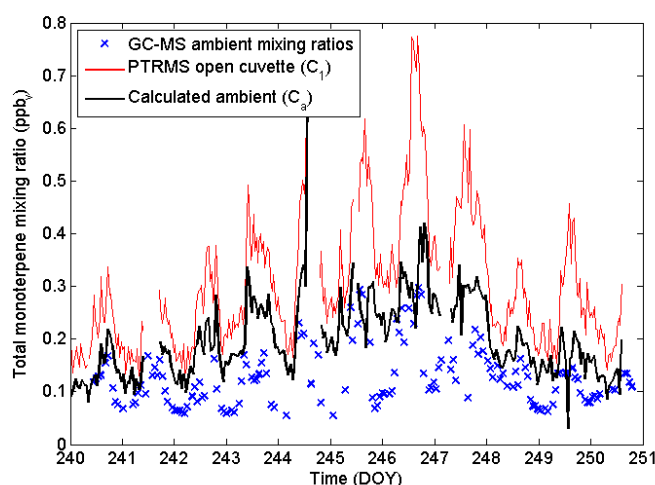
$$C_2 = C_{a0} + C_E + C_{a0} \frac{Ft}{V} - C_2 \frac{Ft}{V} \quad (2)$$

Where  $V$  is the volume of the chamber and  $C_E = \frac{E \cdot m \cdot t}{V}$  is the mass entering the system as emissions. Solving the above equation for  $C_a$  we can derive the ambient mixing ratios:

$$C_{a0} = C_2 - \frac{\frac{E \cdot m \cdot t}{V}}{1 + \frac{Ft}{V}} \quad (3)$$

Equation 3 has been evaluated by comparing the derived results with the sum of ambient monoterpene mixing ratios, as they were measured by Gas Chromatography–Mass Spectrometry (GC-MS) techniques (Nölscher et al., 2012) during the PARADE campaign (Fig. 1). The GC-MS measurements have been performed 100m away and therefore further dilution effects are expected. Despite some discrepancies, averaged values for the reported period indicate a minor difference between the model and ambient mixing ratios ( $0.056 \pm 0.021$  ppb<sub>v</sub>). In contrast, raw data with the open chamber are almost double compared with ambient mixing ratios quantified by GC-MS measurements. Therefore, we assume the above equation derives representative ambient mixing ratios without the influence of the tree in the immediate vicinity, as this is supported by the comparison of these measurements. Here, we note that while Eq. 3 is in concentration, we express the difference (and the subsequent results) in ppb<sub>v</sub> for ease of comparison with other studies. Under flow-through enclosures, a second empty chamber is used as reference and ambient mixing ratios can be directly monitored. Since we have used a dynamic method and our inlet was in the inner side of the chamber and hence it always affected the monitored mixing ratios, we used Eq. (3) to derive all the ambient mixing ratios presented.”

Revised Fig. 1:



**RC: Page 30198, line 11ff: Are these ppb values (NOx) also ppbv?**

AC: yes, we will correct in new version

**RC: Page 30199, line 5: The negative fluxes may also originate from the problem of formulating the proper place in space where the mass balance holds. Is the model located inside the chamber? That would make everything in relation to the Ca as measured before closure and negative values might occur for weak emission and stronger losses as example.**

AC: Maybe there is a small misunderstanding here. The negative fluxes refer to the emission equation, as described by eq. 1 and it is different from the box model that describes the ambient mixing ratios. When during one closure there is decay on the measured VOC mixing ratio (even if the last point was corrected accordingly for chemical reaction, dilution and deposition) the sign of eq. 1 will be negative. It is not the first time that negative fluxes are observed. It means that the loss processes are stronger than the emissions.

**RC: Page 30201, line 6: "... For the rest months" I would say here "For the later months...". In line 14, same page you may define the correlation coefficient as CC because you use that later on.**

AC: It will be revised as suggested. Also the cc is defined in detail.

**RC: Page 30202, line 13ff: The sentence "In all cases..." I do not understand it's message? That reads like a sentence missing some parts. To what was monoterpenes "highest", how does the acetone's temperature dependency links here?**

AC: The intention was to comment on the results presented in Table 2. In all cases means spring, summer, autumn, day, night, i.e. complete dataset. Acetone temperature dependency fits in the way that we emphasize which was the strongest emission potential and which was the strongest temperature dependency. In order to make it more clear we have revised the sentence as following:

*"In Table 2 we present the results of regression analysis performed between the enclosure temperature and the emission rates, separated for different seasons and light abundance. Monoterpene emission potential was the highest calculated for every season and light abundance conditions. From seasonal point of view...."*

**RC: Line 18ff: What do you like to express here? That is not understandable. Your data say that E30 in summer is about 1/3 of the spring value and E30 in autumn is 1/3 of the summer value. Your relation here is kind of opposite expressed.**

AC: we added a semi-colon to make it more clear

*" $E_{30,MT(summer)} \approx 1/3 E_{30,MT(spring)}$ ;  $E_{30,MT(autumn)} \approx 1/3 E_{30,MT(summer)}$ "*

**RC: Line 24: Figure 1 shows monoterpenes according to the axes description! Not sesquiterpenes. You need to be precise here what is shown and described.**

**AC:** In page 30196, line 3 we say “assuming steady state of the measured monoterpene signal”. Additionally in line 10 we clearly state “The latest equation has been evaluated by comparing the derived results with the sum of ambient monoterpene mixing ratios, as they were measured by Gas Chromatography–Mass Spectrometry (GC-MS) techniques...”. Nevertheless, we acknowledge possible confusion because of page 30202, line 24. We will correct this typo.

**RC: Page 30203, Line 1ff:** It is bit puzzling as you employ only temperature dependent algorithms but here you clearly tell that there has to be a light dependency as well for monoterpenes. If you look to your table 2, it is also clear that the temperature bound algorithm is only explaining a mere of 40% of your data’s variation in general and during night time that diminishes to just 10% in the case of monoterpenes. Well, given that, the algorithm just can’t explain the measurements. You may trust it a bit more in the case of sesquiterpenes, that rises once to mere 70%.

**AC:** We employ only temperature algorithms since we do not have continuous PAR measurements. We clearly state throughout the manuscript (abstract, results and conclusions) that there is a light dependency based also on your comments. Additionally, I would like to repeat that the amount of ambient data used in these correlations is rather large. When analyzing the data in temperature regimes (Fig.9) there is only 20% of MT emissions missing. In the latter case, the observed deviation may be attributed to additional light induced emissions.

**RC: Page 30204, line 9:** I would rather say the metabolic pathways are known (not "now known") because this was clear rather long time. Maybe the interplay and the dependencies between several relevant pathways get more and more known and will be ongoing studied I guess.

**AC:** we have deleted the “now”

**RC: Line 18ff:** What is LAh? That whole section remains a bit puzzling to me. As it is well known, that also conifers emit light and temperature dependent and the presented correlations support this finding. Why you try to avoid getting into that (already in the section 3.3 before) and why not apply a mixed algorithm in the case of monoterpenes. As light and temperature are not independent from each other the temperature related algorithm seems to work most times kind of well enough which does not mean it has to be true. It might not be of importance for the major topic here, the explanation of ambient mixing ratios, but try to avoid strange reasonings like that given in line 20, "Despite ...". The monoterpene emissions will originate from both, pools and recent fixed carbon additionally complicated by non-specific storages (see eg. Niinemets et al., Trends in Plant Science Vol.9 No.4, 2004) that occur in plant leaves due to physicochemical properties of the compounds. I am a bit in doubt about correlating such weak predicted emissions to each other and draw conclusions from that, especially as the emission algorithm used just covers temperature as dependent variable and you have here any kind of other impacting factors (ozone, humidity, light).

**AC:** In the text we say that we have used the lower and the uppermost 25% of absolute humidity data (p30203, lines 12-13) ( $LAh < 6.1 \text{ gm}^{-3}$ ;  $Hah > 13.2 \text{ gm}^{-3}$ ). Therefore, LAh is the lowest 25% ( $LAh < 6.1 \text{ gm}^{-3}$ ) of our dataset.



We do not avoid getting into the light dependency of monoterpenes. We do mention it at every part of the manuscript (abstract, results, discussion, conclusions). As already mentioned above no PAR data are available and therefore complete light and temperature algorithms could not be tested.

The referred sentence (“Despite”) will be revised as suggested:

*“Monoterpene emissions originate from both pools and recent fixed carbon additionally complicated by non-specific storages (Ninements et al., 2004)”*

**RC: Page 30207, line 2: Either "...and their importance should..." or "...and the importance of them should..."**

**AC:** We thank the reviewer for the correction. It will be revised as

“and their importance should”

**RC: Line 13: What is the "time maximum"? Do you mean the seasonal emissions here or the time of the daily maximum emission?**

**RC: Line 22ff: The sentence "This might be linked..." needs revision. It is unclear what you want to state here.**

**RC: Page 30208, until line 15: Generally, the whole section 4.1 would need some additional information from photosynthetic status and the evapotranspiration of the branch enclosed. Many things, like the time of maximal emission are bound to diel cycles and the state of the leaf/branch. Water soluble compounds (like methanol or acetone) will rely a lot on the capability of the tree to evaporate. High humidity impacts here a lot on the possible emission activity. As long as there is no information on photosynthesis or light and temperature given it can't be assessed why as example monoterpenes emit more in the afternoon. The cuvette was placed at the side of the tree, when the sunlight reached there? Was it all day? Any shading occurred? There can be many reasons for the actual shape of the diel variation in emission and without additional data/information all that argumentation here remains speculative**

**AC:** The reviewer is correct. Since we did not measure photosynthesis, we will delete the complete discussion on the diurnal patterns.

**RC: Page 30209, line 1: What competition? Between the factors presented here or between the species, that is not clear at this point..**

**AC:** we mean between factors and we have added that in the sentence

*“chemical mechanism elicited by temperature, light, ozone or competition complicating their emission investigation between the aforementioned parameters”*

**RC: Later, after line 5, you say that the storage pools run empty? Do you have a citation for that? Are you sure that this happens? As told before, here you would need kind of information on the physiological state of the branch**

**AC:** We do not have any citation and we are not sure that this happens. This is why this sentence is located at the discussion part and formulated in a very careful manner (“it is tempting to assume”).

**RC: Line 14: You argue about the humidity as a driving factor, well, yes as this controls the actual evapotranspiration and therefore how the plant is able to do photosynthesis. Further, it is one force that will draw out water soluble compounds. So, clearly it should "drive" emissions but do you have a citation of the water film protection?**

**AC:** We do agree that humidity can be a driving factor by controlling the evapotranspiration. As citation for the water film protection one can refer to figure 9 of Bourtsoukidis et al. (2012). However driving factors are used differently in different communities.

**RC: At the end of that page, you come back to the light dependent monoterpene emissions. You do not use the light dependency, because equation 5 is not light dependent it is clear that such features will not be mapped adequately. To my opinion, it is anyway a benefit to include the possible ozone losses if the sample system is large and the time inside the chamber is long according to the typical atmospheric reaction rates. Furthermore, your system was open and there happens a replacement of the air drawn to the PTRMS including ozone in the ambient air. I would rather try to concentrate on that topic here and not discuss too much about processes you can not prove.**

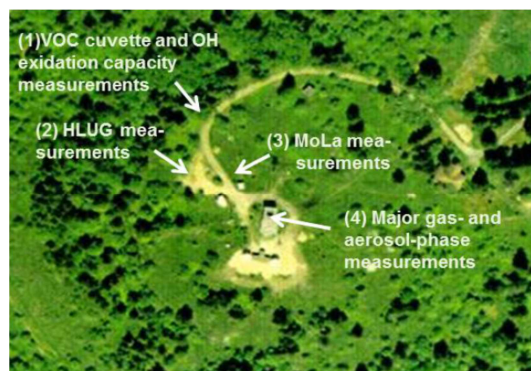
**AC:** The ozone losses, the reaction rates obtained, the sampling system, the dilution effect and the general characterization of the system have been provided in the material and methods section as addition to Bourtsoukidis et al 2012. As stated we consider both times, i.e. during the closure and during the opening of the cuvette for revealing the ambient close to the branch concentration of a compound and exactly this is what extends the measurements and links emissions and ambient concentrations or mixing ratios.

**AC: Page 30210, line 17: You refer to the daily maximum mixing ratios that occur at the middle of the day on figure 1, that can not be seen on the figure as it's not clear where the day of year tick mark is located (start, end or middle of the day?).**

**Further, the finding that is opposite to other findings might be a bit more discussed. What is about that "constant homogenous mixing"? What about the forest edge? Referring to unpublished data is a bit weak reasoning to explain a controverse result.**

**RC:** Even if figure 1 clearly illustrates the daily maxima (the tick is located at the beginning of the day, ie 00:00am), the reader can always see in Fig. 5, where the diel patters are displayed and presented in detail.

In the revised version we will add the complete reference which also includes a photo of the location the measurements took place (Bonn et al., 2013)



Bonn, B., Bourtsoukidis, E., Sun, T. S., Bingemer, H., Rondo, L., Javed, U., Li, J., Axinte, R., Li, X., Brauers, T., Sonderfeld, H., Koppmann, R., Sogachev, A., Jacobi, S., and Spracklen, D. V.: The link between atmospheric radicals and newly formed particles at a spruce forest site in Germany, *Atmos. Chem. Phys. Discuss.*, 13, 27501-27560, doi:10.5194/acpd-13-27501-2013, 2013.

**RC: Line 17: Is the time in "Independent of time..." the daily or seasonal time interval? Later the same line, I guess it's "European conditions" not "Europe conditions".**

**AC:** Changed as : "*Independent of the season*" and "*European conditions*"

**RC: Page 30212, end and next page: I can not understand how you come to this conclusion. You speak here of the "induced" emissions? But your model can rather weakly describe the emission's temperature relation. You also stated before that all data that where very high after some injury etc have been excluded, to my understanding your data are more or less thought to cover the constitutive emissions?**

**AC:** We came to this conclusion from the results illustrated in Fig. 9. As we explained before, in this figure we grouped our data in 10 regimes with the same probability between the limits. The goal was to diminish temporary variabilities and minimize the uncertainty which derives from outliers.

The data have been filtered in such a way so there are no unnatural phenomena included. This is why we have excluded all the data a. after installation, b. during injury, c. during length measurements as have already described in the text.

**RC: Figure 7: What correlation coefficient you have been using (Pearson)? That is not noted in the text.**

**AC:** It was Pearson's and we will add this

**RC: Figure 8: To my opinion, for the water soluble compounds, most prominently for methanol and acetaldehyde, a bit less clear for acetone, the linear relationship does not hold. Is there any reason to have it linear? They seem to follow some logarithmic or power law better. For isoprene and monoterpenes the situation is not as clear but may also qualify better a non-linear behavior. The only one that seem to qualify is the sesquiterpene emission to ambient mixing ratio. A sure way to assess if a model qualifies is a residual analysis, whenever the residuals are not normal distributed and random the model can not (fully) describe the data. What are the colors in the graph?**

**AC:** No, there is no need for linear relationship. We choose to present it this way in order to present how emissions are mapped in ambient mixing ratios. In case of no other sources (e.g. SQT) the linear relationship is evident. Nevertheless, in the revised manuscript we will not apply any kind of relationship and we will just map the emissions with the ambient mixing ratios.

The colors in the graph are monthly means (green=spring, red=summer, blue=autumn). We thank the reviewer for noticing. The legend will appear in the revised version.

**Final authors comment:** We are grateful for the detailed, fruitful and in-depth review provided by anonymous reviewer #1. His/her comments resulted in the revision and more detailed approach of the

mass balance equation that was used for calculating the ambient mixing ratios. Additionally his/her comments resulted in substantial improvement on the weak points of the manuscript. This contribution we will be additionally acknowledged in the acknowledgments section at the end of the manuscript.