

Author response to anonymous reviewers, considering manuscript BG-2013-481: New foliage growth is a significant, unaccounted source for volatiles in boreal evergreen forests,

J. Aalto et al.

Comments for both reviewers conjoined in same document, in order of appearance. The referee comments marked with red colour.

#### General comments, Referee I

The manuscript is well written and describes a very important topic that has been largely overlooked. My main (but little) concerns are that the importance of emissions with regard to aerosol formation should be discussed a bit more careful given the uncertainties that are still connected to the topic. Also, I would like to see the results laid out in a bit more detail (see below). Despite this and the modifications recommended below, I have no major concern about publication.

#### General comments, Referee II

The study by Aalto et al. brings on light an overlooked topic and provides new insights on BVOC emissions. The results indicate that a large fraction of the emitted VOCs originate from the growing process, challenging the empirical models that incorporate temperature and light driven emissions. Nevertheless, no comparison between the measurements and the model is presented. I think that it would have been valuable if the authors enriched their results and arguments with such comparisons. In general, the manuscript is well written and includes a large amount of data collected during three consecutive years. Considering its scientific value and presentation quality it certainly deserves publication but some points should be better addressed beforehand.

We thank both reviewers for positive evaluation and constructive criticism; we feel that the manuscript has vitally improved thanks to your comments.

In this paper, we present only measured data. The purpose of our comparisons between mature and growing foliage is to show 'growth-normalized' emissions; we actually do provide an independent measure for the relative importance of shoot and needle growth processes on emission rates, which can then be implemented to any kind of model. We have now tried to clarify this in the manuscript.

We have tried to implement the specific suggestions as specified below (in the order of appearance in the manuscript, **the comments by referee 1 marked with REF I, and by referee 2 with REF II**):

#### Specific comments

REF I, P1, L23: two orders of magnitude seem to be exaggerated. And while it is said that the higher emissions are particularly found during spring time, it is worth mentioning that in summer the two branches show emissions of similar magnitudes.

**Response:** Regarding most of the compounds and on an average over the year, one order of magnitude difference is correct; however in case of monoterpenes and especially during the spring

time the difference between young and old shoot is indeed two orders of magnitude or even more. Now this is clarified in the abstract by mentioning the details and also the late summer similar emission rates between new and old foliage are specified.

**Previously:** The emission rates of organic vapours (monoterpenes, methyl butenol (MBO), acetone and methanol) from vegetative buds of Scots pine during the dehardening and rapid shoot growth stages were one to two orders of magnitude higher than those from mature foliage.

**Revised:** The emission rates of organic vapours from vegetative buds of Scots pine during the dehardening and rapid shoot growth stages were one (methyl butenol (MBO), acetone and methanol) to two (monoterpenes) orders of magnitude higher than those from mature foliage; this difference decreased and finally disappeared when the new shoot was maturing in late summer.

**REF II, P1, 23:** It would be better if you provide quantitative values with uncertainties.

**Response:** We admit that some quantitative values would be relevant, and have now added certain highlights to the abstract; the details (including uncertainties) are also now included in the results in the form of a table.

**Previously:** The emission rates of organic vapours (monoterpenes, methyl butenol (MBO), acetone and methanol) from vegetative buds of Scots pine during the dehardening and rapid shoot growth stages were one to two orders of magnitude higher than those from mature foliage.

**Revised:** The emission rates of organic vapours from vegetative buds of Scots pine during the dehardening and rapid shoot growth stages were one to two orders of magnitude higher than those from mature foliage; this difference decreased and finally disappeared when the new shoot was maturing in late summer. On average, the springtime monoterpene emission rate of the bud was about 500 times higher than that of the mature needles; during the most intensive needle elongation period the monoterpene emission rate of the growing needles was 3.5 higher than that of the mature needles, and in September the monoterpene emission rate of the same year needles was even lower (50 %) than that of the previous year needles. For other measured compounds (methanol, acetone and MBO) the values were in same order of magnitude, except before bud break in spring, when the emission rates of bud for those compounds were on average about 20-30 times higher than that of mature needles.

**REF II, P1, L24:** Supporting results are needed.

**Response:** The whole sentence is now deleted because the assertion indeed seems to need some further justification. The basis of the finding is anyway presented in the following sentence: The emission rates of developing shoot didn't always follow either the diurnal cycle in temperature, or the diurnal cycle of the emissions from mature shoot.

REF I, In the end of abstract: I recommend a more cautious formulation because the implications on aerosol formation have not been demonstrated directly.

REF II P2, L1-5: This study does not investigate new aerosol formation events. Therefore, I would suggest transferring this sentence to the introduction or removing it completely

**Response, I&II:** This is now removed from the abstract as no aerosol measurements are presented in this paper. The importance of high emission rates in spring is linked to their coincidence with the frequent new aerosol formation events. Although in this study no aerosol measurements are reported, we still wanted to highlight that our finding potentially has relevance when it comes to certain specific springtime aerosol phenomena. The issue is anyway dealt in conclusions and removing it from the abstract is certainly a good idea.

REF I, P2, L13: The study of Bourtsoukidis et al. (2012) also involves the effect of ozone for the above mentioned sesquiterpenes.

**Response:** We are grateful for the referee for pointing out this reference, and have added some details based on the study of Bourtsoukidis et al. (2012).

**Previously:** The measured atmospheric concentrations of many reactive gases, for example terpenoids (mainly isoprene, mono- and sesquiterpenes), exhibit huge spatial and temporal variations due to factors related to large variation in emission rates and emission composition between or inter-species (Kesselmeier & Staudt 1999, Bäck et al. 2012), to light and temperature-related variations in incident emission rates (Gunther et al. 1993, Komenda & Koppman 2002, Tarvainen et al. 2005) as well as to transport and chemical reactions in the air (Rinne et al. 2007).

**Revised:** The measured atmospheric concentrations of many reactive gases, for example terpenoids (mainly isoprene, mono- and sesquiterpenes), exhibit huge spatial and temporal variations due to factors related to large variation in emission rates and emission composition between or inter-species (Kesselmeier & Staudt 1999, Bäck et al. 2012), to light, temperature and ozone concentration-related variations in incident emission rates (Gunther et al. 1993, Komenda & Koppman 2002, Tarvainen et al. 2005, Bourtsoukidis et al. 2012) as well as to transport and chemical reactions in the air (Rinne et al. 2007).

REF I, P2, L26ff: This is not true. The seasonal isoprenoid model presented by (Zimmer et al., 2003) has been applied to derive seasonal dependencies of monoterpenes emission also for evergreen trees (Grote et al., 2006). More discussion and comparisons with the MEGAN approach can also be found in (Grote & Niinemets, 2008; Monson et al., 2012). The problem however, is that the summer-time emission factor is assumed to represent the maximum emission while enzyme activities related to emission that are specific for leaf expansion are neglected so far.

**Response:** The referee is correct and we have now included some more references and a sentence where the seasonality in process model approach has been discussed. In the previous version we only discussed the empirical models.

**Insertion:** The seasonality has to some extent been implemented in process-based models describing the enzyme activity changes over seasons, such as in SIM-BIM (Grote et al. 2006, see also discussion in Grote & Niinemets 2008, Niinemets et al. 2010 and Monson et al. 2012). However, the phenology-submodel in SIM-BIM suggests that the monoterpene emission onset happens only a few weeks after leaf emergence, and that the maximum emission rates take place in fully grown, current year leaves (Fischbach et al. 2002). Thus, the processes during leaf development and expansion are not taken into account in these models.

Fischbach, R. J., Staudt, M., Zimmer, I., Rambal, S. & Schnitzler J.-P. Seasonal pattern of monoterpene synthase activities in leaves of the evergreen tree *Quercus ilex*. *Physiol. Plant.* 114, 354-360, 2002.

Grote, R., Mayrhofer, S., Fischbach, R. J., Steinbrecher, R., Staudt, M. & Schnitzler, J.-P. Process-based modelling of isoprenoid emissions from evergreen leaves of *Quercus ilex* (L.) *Atmos. Environ.* 40, 152-165, 2006.

Groter, R. & Niinemets, Ü. Modeling volatile isoprenoid emissions – a story with split ends. *Plant Biol.* 10, 8-28, 2008.

Niinemets, Ü, Monson, R. K., Arneth, A., Ciccioli, P., Kesselmeier, J., Kuhn, U., Noe, S., Peñuelas, J. & Staudt., M. The leaf-level emission factor of volatile isoprenoids: caveats, model algorithms, response shapes and scaling. *Biogeosciences* 7, 1809-1832, 2010.

Monson, R. K., Grote, R., Niinemets, Ü. & Schnitzler, J.-P. Modeling the isoprene emission rate from leaves. *New Phytol.* 195, 541-559, 2012.

**REF II, P3, L1-2: Please provide a reference.**

**Response:** This indeed seems to need a clarifying reference. Rinne et al. 2009 is now added as a reference.

Rinne, J., Bäck, J. & Hakola, H.: Biogenic volatile organic compound emissions from the Eurasian taiga: current knowledge and future directions. *Boreal Environ. Res.*, 14, 807-826, 2009.

**REF II, P4, L16-18: Why did you choose to close the cuvette with this rate? In other studies (eg Ruuskanen et al., 2005; Bourtsoukidis et al., 2012) the authors operated their systems by closing the cuvette 3 times per hour and therefore they gained better resolution. Do you have any indications that such frequency might alter the emission responses?**

**Response:** It's true that the temporal resolution in our results is not ideal. This is due to our measurement set-up. The results were obtained using the custom-made gas exchange measurement system which includes the PTR-MS as one of the analysers. In addition to the gas exchange measurements, the same PTR-MS is also used for recording the atmospheric VOC concentrations, scheduled for 1 hour in every 3 hours. Zero air is measured 15 minutes per every 3 hours. That means that our enclosure measurements use the PTR-MS only for 1 hour and 45 minutes per every 3 hours. Within this time frame we measure emission rates from ten separate shoot, stem and soil chambers, giving typically 4 measurement points (closures) for the shoot chambers per three hours.

Ruuskanen et al. 2005 used the earlier version of the same measurement system where the temporal resolution for the VOC emission rate measurement was better. However, we believe that with this temporal resolution we are able to capture the main dynamic features with sufficient accuracy, and without compromising the other measurement targets.

**REF II, P4, L20: What was the retention time? Which were the losses for the compounds measured?**

**Response:** The total retention time was about 40 seconds in 2009 and 55 seconds in 2010-2011. Those details are now added to the methods section. Kolari et al. (2012) (already cited elsewhere in the manuscript) have studied the losses in our measurement system and found that they are in the range of 5-30 %. We are confident that those losses do not affect the comparisons between the emissions of mature and developing shoot using similar measurement setups for both.

P5, L10: A sentence added: The total retention time of the sample tubing was about 40 seconds in 2009 and 55 seconds in 2010-2011.

**REF II, P4, L25: Briefly report averaged variations on the parameters measured during one closure.**

**Response:** Details of the typical changes in parameters during closure are now added to manuscript as follows:

During the closure the temperature inside the enclosure tended to increase slightly, especially daytime when the increase in temperature during the closure was typically 2-4 °K. The PPFD measurements were conducted outside the chamber and followed ambient variations.

**REF II, P4, L29-31: what about possible effects of removing the buds? (eg. Hakola et al., 2006).**

**Response:** We are grateful for addressing this issue because majority of findings by Hakola et al. (2006) match quite nice with our findings. First, we are confident that bud removal should not have any major impact on the springtime (April) emissions, because the buds were removed weeks or months before the study period whereas Hakola et al. found that the direct impact of bud removal on terpenoid emissions lasted for one week, and gradually diminished thereafter. Secondly, in the study of Hakola et al. the growing shoot emitted more monoterpenes and MBO than the shoot that was not growing (matches to our results). Thus, we did the following revisions:

P4, starting from L29:

**Previously:** All buds of the mature shoot and all auxiliary buds of the developing shoot were gently removed about one month before the installation to avoid contamination with fresh resin flowing from the scars.

**Revised:** All buds of the mature shoot and all auxiliary buds of the developing shoot were gently removed about one month before the installation to avoid contamination with fresh resin flowing from the scars (Hakola et al. 2006).

P8, starting from L23:

**Previously:** Remarkably, our measurements show that in addition to methanol, the initial phases of new biomass development in buds influence dramatically also the MBO and monoterpene emissions.

**Revised:** In addition to methanol, the initial phases of new biomass development influence dramatically also the MBO and monoterpene emissions, which is in line with the results of Hakola et al. (2006).

REF II, P5, L2: what was the temperature of the heated line?

**Response:** The heating doesn't use constant temperature; instead it has constant power ( $5 \text{ W m}^{-1}$ ). This way the heating reduces surface effects roughly in same manner along wide temperature range.

**Previously:** During 2010 and 2011, heated fluorinated ethylene propylene (FEP) tubing (length 64 m , internal diameter 4 mm) was leading the sample air towards the  $\text{CO}_2$  and  $\text{H}_2\text{O}$  analyzers, and the VOC subsample was taken from that line before the other analyzers.

**Revised:** During 2010 and 2011, heated ( $5 \text{ W m}^{-1}$ ) fluorinated ethylene propylene (FEP) tubing (length 64 m , internal diameter 4 mm) was leading the sample air towards the  $\text{CO}_2$  and  $\text{H}_2\text{O}$  analyzers, and the VOC subsample was taken from that line before the other analyzers.

REF II, P5, L11: Counts per second or mixing ratios instead of concentrations would have been more accurate.

**Response:** VMR truly is more accurate way to present this. The sentence is modified accordingly.

**Previously:** The PTR-QMS measures the total concentration of all compounds having equal mass with a resolution of 1 amu (atomic mass unit) and was operated here with 12.5 s measurement interval and integration time of  $1 \text{ s amu}^{-1}$ .

**Revised:** The PTR-QMS measures the volume mixing ratio of all compounds having equal mass with a resolution of 1 amu (atomic mass unit) and was operated here with 12.5 s measurement interval and integration time of  $1 \text{ s amu}^{-1}$ .

REF II, P5, L32-33: Can you present a ratio? Did you conduct any GC-MS measurements to ensure that there was no isoprene emitted? Additionally and according to the literature (Fall et al., 2001 and Warneke et al., 2003) other aldehydes and ketones can be also found in this mass and it worth mentioning.

**Response:** Emissions from the same trees have been measured previously using GC-MS and in these measurements only a very small isoprene emission was seen (Hakola et al. 2006, Tarvainen et al. 2005). In Hakola et al (2006) we have discussed that the observed small isoprene emissions are

actually potentially artefacts, as MBO can be dehydrated in the adsorbent tubes to isoprene which is then detected in analysis. Obviously aldehydes and ketones should be mentioned here.

**Previously:** Based on this, we assume that in this case the emission at 69 amu is mostly composed of the MBO fragment.

**Revised:** Based on this result, and the previous GC-MS measurements from same trees conducted by Tarvainen et al. (2005) and Hakola et al. (2006), we assume that in this case the measured emission at 69 amu is mainly composed of the MBO fragment. However, as PTR-MS is mass-specific, we cannot exclude the potential traces of isoprene and certain aldehydes and ketones in the M69 (Fall et al. 2001, Warneke et al. 2003), if they are emitted from Scots pine shoots.

Fall, R., Karl, T., Jordan, A. & Lindinger, W.: Biogenic C5 VOCs: release from leaves after freeze-thaw wounding and occurrence in air at a high mountain observatory, *Atmos. Environ.*, 35, 3905–3916, 2001.

Warneke, C., de Gouw, J. A., Kuster, W. C., Goldan, P. D. & Fall, R.: Validation of atmospheric VOC measurements by proton-transfer-reaction mass spectrometry using a gas-chromatographic pre-separation method, *Environ. Sci. Technol.*, 37, 2494–2501, 2003.

**REF I, P6, L27:** Could you please give a reference for the assumption that all needle age classes have the same biomass? It is somehow against intuition. Also literature that I am aware of indicates that at least needle ages classes of three years and older are considerably diminished (Niinemets & Lukjanova, 2003; Xiao & Ceulemans, 2004).

**Response:** The assumption of equal needle mass between needle age classes is based on the constant needle mass on stand scale (Vose et al 1994). This is now clarified in the text. The number of living needle age classes is mentioned on line 32; and is based on our field measurements, e.g. Ilvesniemi et al. (2009). Besides of assuming that in Scots pine forest there are three living needle age classes, we assume that the older needle age classes have fallen down. We are aware that this is a simplification, but it is necessary for being able to estimate the role of new foliage to the total canopy emissions. The uncertainty caused by differences between the biomasses of needle age classes is now estimated by conducting two modified calculations: One where the proportion of growing needle age class has been increased by 10 %, and one where it has been decreased by 10 %. The result of this calculation is presented in the new table 2 (presented somewhat later), and the effect is mostly lower than 10 %.

**Previously:** To estimate the contribution of the developing foliage compared to the total foliage emissions, three simplifying assumptions were applied:

**Revised:** To estimate the stand level contribution of the emissions from developing foliage compared to the emissions from whole canopy, three simplifying assumptions were applied:

**Previously:**

- i) All remaining needle age classes have the same mass per land area in the autumn. As the canopy closure has taken place several decades ago, the stand needle mass is rather constant; all needle age classes have same mass.

**Revised:**

- i) All remaining needle age classes have the same mass per land area in the autumn. This assumption is based on the fact that the canopy closure has taken place several decades ago and therefore the stand needle mass is rather constant (Vose et al. 1994); thus there is no major difference in yearly new foliage production.

Vose, J. M., Dougherty, P. M., Long, J. N., Smith, F. W., Gholz, H. L. & Curran, P. J. Factors influencing the amount and distribution of leaf area of pine stands. *Ecol Bull.* 43, 102-114, 1994.

REF I, P7, L6: I think you assume everything identical, not almost identical, right?

**Response:** Yes, we assume exactly identical in the calculation, and hypothesize that the environmental conditions of different needle age classes are anyway almost identical. This is now clarified so that the difference between the assumption and the justification are clearer.

**Previously:**

- iii) All needle age classes at the same canopy height face about identical (light and temperature) environmental conditions. The canopy structure and growth mode of Scots pine produce almost even spatial distribution of all living needle age classes. The spatial distribution is to some extent skew so that the newest needle age class is shading the older ones, but because Scots pine canopy is fairly sparse the light and thermal conditions of all needle age classes are assumed to be almost identical.

**Revised:**

- iii) All needle age classes at the same canopy height face identical (light and temperature) environmental conditions. The canopy structure and growth mode of Scots pine produce almost even spatial distribution of all living needle age classes. The spatial distribution is to some extent skew so that the newest needle age class is shading the older ones, but because Scots pine canopy is fairly sparse the light and thermal conditions of all needle age classes are almost identical.

REF I: The results are presented biomass-based. However, as also noted by the authors, emissions are often expressed on a leaf area basis. For comparison reasons, I would therefore recommend to calculate the emissions also based on leaf area. This could also been used for a tree-level upscaling exercise. Additionally, I would like to see a table with summed up emission values per year, otherwise the numbers presented in the text are difficult to digest.

**Response:** We admit that the per leaf area emissions would be valuable for comparison reasons. The specific leaf areas for the monitored shoots were now added to the method section. The emission rates are expressed only in figure 2; the other results would end up to same results despite of the unit used in calculation because they are using relative units based on comparing the difference between the mature and developing shoot.

We also agree that annual cumulative emissions would be very useful, however there are many caveats that make the accurate summing very difficult. First, we have some missing data points (due to calibrations, technical problems etc.), and second, we do not have proper gap filling procedures for emission measurements. The only applicable way to produce the annual cumulative emissions would be to use quite complicated and strong averaging and/or emission models as a tool. In our case, the results are not relying on any particular averaging method or model, and only describe the relative significance of growing foliage to the total emissions at each point in time. The next step would be to make a model parameterized with the measured emissions, however this is a major work, and therefore we think that it cannot be done here.

**Insertion to P5, L26:** The specific leaf area for year 2009 was  $0.11 \text{ m}^2 \text{ g}^{-1}$ , and for years 2010-2011  $0.9 \text{ m}^2 \text{ g}^{-1}$ .

**REF II, P8, L5:** Report here (ii) the days of elongation for each year

**Response:** Only days having needle elongation rate higher than  $0.5 \text{ mm d}^{-1}$  were chosen for comparison to distinguish as clear effect of needle elongation on VOC emissions as possible. The total length of the needle elongation period is now added to the text as well as the starting and ending days of the most intensive needle elongation period.

**Insertion to P5, L26:** Inside the enclosure, the most intensive needle elongation days (needle elongation rate  $\geq 0.5 \text{ mm d}^{-1}$ ) took place between the 1<sup>st</sup> of June and the 25<sup>th</sup> of July. The total needle elongation period was 104 days in 2009 and 2010, and 102 days in 2011.

**REF II, P8, L26:** In addition to the figures, a table presenting the quantitative differences would be useful to the reader. The following sentence can be revised, including discussion from the table.

**Response:** We agree with the referee that such a table is useful and illustrative. Therefore it is now added to the results.

**Insertion:**

Table 1. The ratio between VOC emission rate of the growing needles to the emission rate of mature needles (unitless), obtained by dividing the daily average emission rate of growing needles with the daily average emission rate of mature needles. The medians represent data from all three measurement years, and the periods cover April, the most intensive needle elongation days (needle elongation rate  $\geq 0.5 \text{ mm d}^{-1}$ , in June and July), and September.

|                                    |                | Methanol   | Acetone    | MBO        | Monoterpenes |
|------------------------------------|----------------|------------|------------|------------|--------------|
| <b>April</b>                       | Upper quartile | 39         | 42         | 23         | 770          |
|                                    | <b>Median</b>  | <b>25</b>  | <b>29</b>  | <b>18</b>  | <b>520</b>   |
|                                    | Lower quartile | 11         | 19         | 10         | 260          |
| <b>Intensive needle elongation</b> | Upper quartile | 4.2        | 3.3        | 6.5        | 30           |
|                                    | <b>Median</b>  | <b>2.4</b> | <b>1.8</b> | <b>3.5</b> | <b>3.5</b>   |
|                                    | Lower quartile | 1.3        | 1.1        | 2.8        | 0.3          |

|                  |                |            |            |            |            |
|------------------|----------------|------------|------------|------------|------------|
|                  | Upper quartile | 0.8        | 1.0        | 0.5        | 1.2        |
| <b>September</b> | <b>Median</b>  | <b>0.5</b> | <b>0.8</b> | <b>0.4</b> | <b>0.5</b> |
|                  | Lower quartile | 0.2        | 0.6        | 0.2        | 0.2        |

REF II, P10, L3-5: I think you should be more careful when speaking about “total emissions” since you were only monitoring few VOCs. It would be best if you wrote “total measured emissions” (also in other parts). Moreover, a table with the contributions for each of the three periods would very helpful and informative.

**Response:** We admit that the term ‘total emissions’ is potentially misleading. In this context we mean emissions from the whole foliage (whole canopy), not emissions of all VOCs. It is obvious that the word ‘total’ can be deleted from several sentences without losing any significant information (the message is perhaps even more clear after that), and in some cases we have to formulate the term more carefully. Following reformulations were conducted:

P6, L21, title:

**Previously:** Estimation of the contribution of developing needle age class to the total canopy emission

**Revised:** Estimation of the contribution of developing needle age class to the whole canopy emission

P6, starting from L23:

**Previously:** To estimate the contribution of the developing foliage compared to the total foliage emissions, three simplifying assumptions were applied:

**Revised:** To estimate the stand level contribution of the emissions from developing foliage compared to the emissions from whole canopy, three simplifying assumptions were applied:

P7, starting from L7:

**Previously:** Based on the above assumptions, the contribution of the developing needle age class to the total emissions ( $C_D$ , %) is:

**Revised:** Based on the above assumptions, the contribution of the developing needle age class to the whole canopy emissions ( $C_D$ , %) is:

P10, starting from L3:

**Previously:** Our results show that although the new shoots make up only a small biomass in the beginning of the growth period (Fig. 3 a), their contribution to the total foliage emissions is large (Fig. 3 c-f).

**Revised:** Our results show that although the new shoots make up only a small biomass in the beginning of the growth period (Fig. 3 a), their contribution to the emissions of whole foliage is large (Fig. 3 c-f).

P10, L12:

**Previously:** ... contribute about half or one third of the total emissions from Scots pine foliage.

**Revised:** ... contribute about half or one third of the emissions from Scots pine foliage.

P10, L14:

**Previously:** ... needles to the total emissions from the foliage is approximately equal with mature shoots.

**Revised:** ...needles to the emissions from the whole foliage is approximately equal with mature shoots (table 1).

Figure 3 text:

**Previously:** Figure 3. The growth of needles and the relative contribution of developing shoot to the total foliage emissions. The points represent one week averages in three consecutive years (2009-2011). a. length of growing needles; b. elongation rate of growing needles. The contribution of developing needle age class to total emissions of c. monoterpenes, d. MBO, e. methanol, and f. acetone.

**Revised:** Figure 3. The growth of needles and the relative contribution of developing shoot to the emissions of whole foliage. The points represent one week averages in three consecutive years (2009-2011). a. length of growing needles; b. elongation rate of growing needles. The contribution of developing needle age class to whole canopy emissions of c. monoterpenes, d. MBO, e. methanol, and f. acetone.

REF I, P10, L6: 'using a model of needle and shoot' is a bit puzzling. Please refer to the method section where this has been explained and to what is the essence of the model.

**Response:** The whole sentence was deleted because it was not needed here.

REF II, P10, L21: This is an important finding and you may consider adding it to the abstract. Please add the uncertainties in the values given.

**Response:** We agree on the importance of this finding, and a sentence about that is now added to the abstract.

**Insertion to Abstract:** During spring and early summer the buds and growing shoots are a strong source of several VOCs, and if they are not accounted in emission modeling a significant proportion of the emissions –from few percents to even half of the annual cumulative emissions – will remain concealed.

**Insertion to P10, L26,** table 2 added to this connection:

Table 2. The proportion (%) of the concealed whole canopy emissions if the effect of growing Scots pine needle biomass on VOC emissions is not taken into account. 0 % indicates no effect of the growing biomass; 100 % indicates an equal emission from growing and mature needles; and >100% a greater proportion of growing biomass than the mature foliage to whole canopy emissions. Average+10% and average-10% represent modified calculations where the proportion of growing needles has been increased or decreased by 10%, respectively. The periods represent the first half of and the whole thermal growing season (daily mean temperature permanently > 5°C).

|                                       |                | Methanol  | Acetone   | MBO       | Monoterpenes |
|---------------------------------------|----------------|-----------|-----------|-----------|--------------|
| <b>April-<br/>midsummer</b>           | Max            | 31        | 60        | 176       | 368          |
|                                       | Average+10%    | 31        | 46        | 52        | 211          |
|                                       | <b>Average</b> | <b>29</b> | <b>45</b> | <b>50</b> | <b>200</b>   |
|                                       | Average-10%    | 25        | 43        | 46        | 182          |
|                                       | Min            | 22        | 32        | 22        | 149          |
| <b>Thermal<br/>growing<br/>season</b> | Max            | 16        | 60        | 43        | 158          |
|                                       | Average+10%    | 12        | 23        | 30        | 103          |
|                                       | <b>Average</b> | <b>11</b> | <b>21</b> | <b>28</b> | <b>98</b>    |
|                                       | Average-10%    | 8         | 19        | 24        | 87           |
|                                       | Min            | 2         | 18        | 22        | 78           |

REF I, P10, L28: 100 days – reference? Seems to be a bit high (reference?), i.e. compared to (Jach & Ceulemans, 1999)

REF II, P10, L28: the “about 100days” should be changed in something more accurate

**Response:** ‘About 100 days’ is based on the actual needle elongation measurements conducted within this study. However, the sentence is now modified so that the generalization only qualitative. Since we want to discuss and compare the new foliage growth of different tree species and the potential effect of that on the VOC emissions, we feel that it is enough to describe the new foliage growth of boreal evergreen conifers in a qualitative manner.

**Previously:** In boreal evergreen conifers with a single flush per year, such as in Scots pine, the new foliage growth lasts about 100 days, and is characterized by a rather short and intensive shoot and needle elongation period (fig. 4).

**Revised** In boreal evergreen conifers with a single flush per year, such as in Scots pine, the new foliage growth is characterized by a rather short and intensive shoot and needle elongation period (fig. 4).

REF I, P10, L31: I wonder why poplars are missing here.

**Response:** Poplars are included in the revised version of the manuscript. Thank you for pointing this out.

**Previously:** However, in many other ecosystems, both evergreen and deciduous tree species can have several flushes (e.g. loblolly pine, longleaf pine and red oak), or are freely flushing (e.g. many birches, elms),...

**Revised:** However, in many other ecosystems, both evergreen and deciduous tree species can have several flushes (e.g. loblolly pine, longleaf pine, red oak), or are freely flushing (e.g. many birches, elms and poplars),...

REF I, P12, L20: also light dependent isoprenoid and MBO production respond on temperature and not 'light only'.

**Response:** That's true, and the sentence is now corrected. Thank you for pointing this out.

**Previously:** The less water-soluble isoprenoids are not as much controlled by stomatal actions, and their emission rates follow light only (MBO) or light and temperature (monoterpenes),...

**Revised:** The less water-soluble isoprenoids are not as much controlled by stomatal actions, and their emission rates follow light and temperature,...

REF II, P13, L12: The errorbars at Fig 7 do not allow speaking about "significantly different diurnal patterns"

**Response:** Response to P13, L12: The error bars indeed indicate quite high variation, but the statistical test still showed some significant differences between the diurnal cycles of emissions from mature and growing shoot. The referee is correct in the opinion that the diurnal cycles were not overall different. The sentence is now formulated more carefully.

**Previously:** The observed significantly different diurnal pattern in emissions in growing and mature needles suggests that during the period of high metabolic activity, the biosynthesis of methanol, MBO and monoterpenes is indeed linked to the up-regulation of the respective pathways, and a potential circadian regulation of biosynthesis has been suggested (Laothawornkitkul et al. 2009).

**Revised:** The observed differences in diurnal patterns of emissions from growing and mature needles suggest that during the period of high metabolic activity, the biosynthesis of methanol, MBO and monoterpenes is indeed linked to the up-regulation of the respective pathways, and a potential circadian regulation of biosynthesis has been suggested (Laothawornkitkul et al. 2009).

REF II, P13, L29: The word evidently suggests that such comparisons have been performed. Please add a figure in which you will demonstrate the differences between the model and measurements or reformulate the sentence.

**Response:** The sentence is now reformulated.

**Previously:** Evidently the existing model approaches using only light and temperature as direct drivers for emissions are not able to capture the effect of growing foliar biomass on emissions.

**Revised:** This relationship and the divergence between the diurnal cycles in emissions from mature and growing needles indicate that existing model approaches using only light and temperature as direct drivers for emissions should be modified to capture the effect of growing foliar biomass on emissions.

REF I Technical Comments:

- P1, L25: double 'from'

Corrected

- P2, L13: Guenther instead Gunther

Corrected