**Reviewer #2**

The study focuses on an interesting and important topic of seasonality of monoterpene emission dynamics, and expands to its importance on secondary aerosol formation. Paper is well written and very easy to read. However, there are some methodological flaws that make the conclusions not well founded.

Most of the results of the paper, if I understood correctly, are based on measurements on one Scots pine shoot. Knowing the intraspecies variability of emissions of VOCs (e.g. Staudt et al., 2001; Bäck et al., 2012), the lack of quantification of variability of the effect of the new foliage is a serious drawback. The data by Aalto et al., (2014) can be taken to show the importance of new needle flush, but its weakness lies in the fact that is shows data from only one shoot. For national level estimate there should be some indication also on the variability of the data and the robustness of the results.

Firstly, we thank the reviewer for taking the time to carefully read and review our manuscript. Thanks for the kind words and valuable suggestions.

The results of this manuscript are based on three years of measurements from one Scots pine tree – not from one shoot only. One page 5, L165-168 in the current version of the manuscript, it currently says: “In brief, the shoot exchange of monoterpenes was measured with an automated gas-exchange enclosure system and analysed by PTR-QMS (Proton Transfer Reaction - Quadrupole Mass Spectrometer) from a ~50 year old Scots pine tree located at the SMEAR II station during 2009-2011.”. All in all, we utilised 6813 observations from 53 weeks obtained during three years (see Table A1 in the appendix of the manuscript). We can alternatively add the information to this sentence that not only one shoot was measured.

We completely agree with the reviewer that the conclusion of this study is limited by the data availability. However, we are in the unfortunate situation that additional suitable data currently does not exist for our analysis. Considering the comments from both reviewers, it is clear that in the manuscript, we need to emphasise that what we try to do is to demonstrate the **potential effects** of monoterpenes from growing pine needles more than providing final definitive answers in this field nor suggesting actual robust emission factors to use in models. We will change the wording, especially in the abstract and conclusion, in order to clarify our aim, and strongly underline the large uncertainties connected to our findings and call for more measurements of new conifers (including Scots pine) foliage. The reviewer requests that for national level estimates there should be some indication on the variability of the data and the robustness of the results. It would for example be possible to add error bars to Fig. 10 which are controlled by the data variability from Fig. 7. Otherwise, it can also be assumed that the lower limit of the emission factor of new foliage compared to that of mature foliage is 2, (because e.g. MEGAN assumes that new foliage has twice as high a potential to emit monoterpenes than mature foliage, however, that estimate is not based on spring time observations), and that estimate is already included in Fig. 10, but a comment on this can be added to the results section.

The authors seem to be in the opinion that the ecosystem emission measurements by micrometeorological techniques are most reliable way to obtain ecosystem scale emission potentials (e.g. page 11). Still the modelling was done based chamber measurement on one shoot. The authors could maybe have somehow scaled the emission potentials they obtained from shoot measurement to fit ecosystem emission measurement. Thus their modeling results would be on a more robust basis.

It is clear that there are different advantages and disadvantages with different measurement – as well as modelling – approaches, and in combination, it must be expected that the best results will be obtained. Thus, this manuscript utilises chamber measurements, ecosystem scale micromet measurements and various modelling efforts. The measurements used are from one tree, not one shoot, see above. We are not completely sure about what the reviewer means when s/he suggests that we could have scaled the emission potentials obtained from shoot measurements to fit ecosystem scale emission measurements. Do you mean that we should have presented the emission factors in the unit of mass/m2/time (e.g. µg m-2 h-1) as it is usually done in micromet measurements, instead of in the unit of mass/foliage mass/time (e.g. µg g-1 h-1) as it is usually done in chamber measurements? We used the latter, because this analysis is indeed based on chamber measurements and because most existing measurements are based on chamber measurements, thus it is easier to compare to other studies when the per-foliage mass unit is used (as e.g. done in Fig. 4). However, the reader is able to unit convert the emission factors to mass/m2/time unit utilising Fig. 1-2. We can remind the reader of this possibility by adding a sentence to the manuscript.

Furthermore, the paper uses only temperature dependent algorithm, even though is extensively cites Taipale et al., (2011) who show that about 40

It is correct that Taipale et al. (2011) found that the ratio of the *de novo* emission factor to the total emission factor varied between 30-46 % and when initialising our analysis, we also discussed among ourselves whether it would be more appropriate to standardise the emission rates using the hybrid algorithm or only temperature. In the end, we decided to standardise using only temperature, since (1) the understanding of light-dependency on emissions from conifers trees is still poor (there exists only very few publications), which was also underlined by Taipale et al. (2011) as they provided large error bars on their results (~10-80%), and (2) existing literature on emissions of monoterpenes from Scots pine is exclusively standardised using only temperature (only exceptions are to our knowledge Taipale et al. (2011) and Rantala et al. (2015)) and these publications do not provide sufficient (if any) information about the light conditions, thus a re-standardisation is not possible and thus a comparison to existing values (as is done in Fig. 4 and Sec. 2.3) would not be possible (see e.g. Langford et al., 2017). The most important point is that our conclusions would not change if a different emissions algorithm was used (see e.g. Fig. 3a in Taipale et al., 2011). We can add our reasoning to Sec. 2.3.

The extrapolation of the importance of new foliage to different age classes as well to northern Finland, based on model, can be very uncertain as we do not know if the VOC emission from new needle flush behaves in the same way as in Hyytiala.

This is completely correct, and we will add clarifying text on the uncertainty that our assumptions can cause both in the method and results sections. We will underline that we present a back-of-the-envelope study with crude extrapolations in order to give an order-of-magnitude estimate of the potential impacts that an exclusion of new foliage can have on model estimates.

The annual average of emission potentiai, as shown in Figures 9 and 10, is not really a good metric. Even large emission potential in spring does not necessarily lead to large annual emission as the temperatures in spring are lower than in summer.

As such we agree with the reviewer that it is maybe not the best metric. The reason for including a seasonal average is that even after 31st of May (which we have defined as the end of spring in our division) we still predict that new Scots pine foliage contributes with the majority of the emissions of monoterpenes from the canopy for a couple of months (see Fig. 8 and Aalto et al., 2014). Additionally, one should recognise that models usually only use one value for the emission potential throughout the season. However, we are willing to exclude the values provided from the full season from Fig. 9 and 10.

Detailed comments:

Page 1, line 15: “. . .assume that the contribution of BVOCs from new conifer needles is minor to negligible.” This statement isounds wrong. The models assume that the contribution of new needles to be equal to mature ones.

Yes, the statement sounds wrong. We will reformulate the sentence to: “Models to predict the emissions of biogenic volatile organic compounds (BVOCs) from terrestrial vegetation largely use standardised emission potentials derived from shoot enclosure measurements of mature foliage. In these models, the potential of new foliage to emit BVOCs is assumed to be similar (or twice as high) to that of mature foliage, and thus new foliage is predicted to have a negligible to minor contribution to canopy BVOC emissions during spring time due to the small foliage mass of emerging and growing needles.”

Page 6, lines 220-221: “. . .(when data based on Aalto et al. (2014) is not considered).” Why is this part in parenthesis? It seems to me to be an integral part of the sentence, without which it would be misunderstood.

Sure, we will remove the parentheses (without the content) and thus write: “A few points range up to ~6 μg g-1 h-1 , while only one measurement point results in a potential of ~15 μg g-1 h-1 when data based on Aalto et al. (2014) is not considered.”.

Page 10, lines 364-365: “The spring time differences in emission potentials lead to uncertainties in predictions of monoterpene emissions that are much greater than what has been estimated by Lamb et al. (1987) and Guenther et al. (2012).” The Lamb et al., (1987) paper dates before a lot of BVOC emission modelling activities and measurements that today form the body of the literature were conducted. I wonder how relevant their estimates are today.

Though Lamb et al. (1987) is a rather old paper, we refer to it, because it is one of very few papers that provides a quantitative assessment of the uncertainty used for projections of BVOC emissions. Due to its age, we were also hesitant to cite it, but in the MEGAN v.2.1 model description paper (Guenther et al., 2012) the authors state that these first quantitative uncertainty estimates by Lamb et al. (1987) provide a general guidance on the accuracy of BVOC emission estimates. Thus we concluded that they are still valid, and hence we also referred to Lamb et al. (1987). If the reviewer disagrees with today’s relevance of Lamb et al. (1987), we can omit reference to that paper.

Figure 7: You could add “south” and “north” above the top left and right panels, respectively, to indicate the whole column of panels. Similarly, you could indicate the rows, by e.g. having a label on right side for 10, 25 and >50.

Thank you for the great suggestions. We will change the figure accordingly.

Figure 8 is very confusing. It could be better to divide it to smaller figures, or better indicate what is what. Not including the April data of Taipale et al., (2011) and Rantala et al., (2015) to panels c, I and o seems cherry picking, to make the fit between data look better than it is.

Perhaps we could also here utilise your great suggestion from above: “south” and “north” could be added above subfigure a and d respectively and we could add the age label on the right side of the figures. Alternatively, within the subfigures, we could write “(a) 10y, south”, “(b) 25y, north” and so on instead of just “(a)” and “(b)” and so on. We could also add “zoom of g-l” above subfigure m.

Taipale et al. (2011) does not include any data from April (only from May-August). April data from Rantala et al. (2015) was excluded as it represents the measured flux during the entire month, while new Scots pine foliage only emerges halfway through the month as also pointed out on page 9, L327-329. It is naturally possible to include the April data point from Rantala et al. (2015) in Fig. 8 and in Sec. 3.2 clarify that the April data is not directly comparable to our estimations.

Table 1: Please indicate what is the total emission, so that comparison of “additional” emission has a reference point.

This is in principle a fine idea, but we are hesitant to do so, as we fear that that number can later on be misused by hasty readers as an estimate for total monoterpenes emissions from Finland. Since the largest uncertainty in our study is caused by the emission rates of new foliage (due to inter-species variability and limited data availability), and since our study can therefore only aim at providing order-of-magnitude estimates, rather crude assumptions (provided in Sec. 4.2) were used for calculating the additional emissions in Table 1. However, such a crude treatment is not fit for providing robust national level estimates of total emissions of monoterpenes.

Table 3: 1e7 etc is not proper way to indicate powers of ten. It should be 107.

Of course. We will correct this.

Please check the number of digits on any numerical results given. Giving results this uncertain with three significant digits (implying uncertainty in the order of percents) is excessive.

Thank you for pointing this out. We will naturally correct it.

Additional references

Bäck, J., Aalto, J., Henriksson, M., Hakola, H., He, Q., and Boy, M.: Chemodiversity of a Scots pine stand and implications for terpene air concentrations, Biogeosciences, 9, 689–702, doi:10.5194/bg-9-689-2012, 2012.

Ghirardo, A., Koch, K., Taipale, R., Zimmer, I., Schnitzler, J.-P., and Rinne, J.: Determination of de novo and pool emissions of terpenes from four common boreal/alpine trees by 13CO2 labelling and PTR-MS analysis, Plant Cell Environ., 33, 781–792, 2010.

Staudt, M, Mandl, N, Joffre, R, Rambal, S, 2001: Intraspecific variability of monoterpene composition emitted by Quercus ilex leaves.