Response to referee comments by Michael Staudt

The work by D. Taipale et al. assesses the potential impact of the underestimation of VOC emissions from young Scots Pine foliage on larger scale VOC fluxes as well as on particle formation and growth. Based on data set published by Aalto et al. 2014 the authors extrapolate the seasonal VOC emission potentials to stand and regional levels and compare the outputs with those obtained by the MEGAN modelling approach. They also analyzed the effects of stand age, season and latitude on the potential underestimation of the whole Scots pine tree's foliage emission potential. Furthermore the authors provide a nice literature compilation of available emission data for Scots Pine. The paper is overall written and the topic is interesting and relevant for our scientific discipline and will make a nice paper in Biogeosciences.

We would like to start our reply by sincerely thanking Michael Staudt for taking the time to carefully review our manuscript and for providing constructive and very relevant comments, that when implemented, will notably improve our manuscript.

There is considerable evidence that young developing shoots of coniferous species release larger amounts of terpenes and other VOCs than mature shoots with respect to their needle masses. Not accounting for this may indeed bias emission estimates and assessments of their implications in air chemical processes as suggested by the present study. However, I have some concerns related to uncertainties and the representativeness of the emission data used in the study. The whole modeling exercise bases on a data set from a sole study (Aalto et al. 2014) reporting extremely increased emissions (from needles ?) during shoot growth starting from several hundreds of μg g-1 h-1 at bud burst (?) decreasing progressively later in the season. These data were obtained on few shoots of a single (?) tree in the same population measured by the same methods. A lot of previous studies that measured emissions from Scots pine or other coniferous species at various scales (needles, branch, whole trees or potted plants) reported increased emissions during shoot growth period but as far as I know, none of them observed orders of magnitude higher emissions, but rather percentage to few fold higher emissions (see e.g. Flyckt 1979, Janson 1993, Kim 2001, Komenda & Koppmann 2002, Tarvainen et al., 2005; Hakola et al. 2006; Holzke et al. 2006; Räisänen et al., 2008, 2009; Geron and Arnts 2010. . .). Accordingly, the 2fold higher emission potential applied in the MEGAN model (Guenther et al. 2012) seems not to be so bad. I could not find really convincing arguments in the ms that literature data other than those by Aalto 2014 are not or less valuable and that the assumptions in MEGAN are completely wrong, which altogether questions the representativeness of the Aalto et al. 2014 dataset. Nevertheless it might be okay to use only the Aalto et al. 2014 data and keep the current modelling part as it is for the final paper but then it should be presented as a kind of "worst case scenario" pointing to a large POTENTIAL underestimation of VOC emissions from this type of vegetation. But as long as there are no independent studies (at shoot or needle level) confirming the Aalto et al. 2014 data, the outputs of the presented extrapolations cannot really be taken as granted and must be presented and discussed as such. In other words, I recommend the authors to tone down a bit their statements and conclusions. Speaking "badly" (without intention to offend anyone), the current manuscript version gives a bit the impression of "puffed-up story". This is a pity, because not really necessary.

We thank you for your comments regarding the presentation of our “story”, including concerns related to uncertainties and the representativeness of the data used. As we are sure that you are aware (e.g. judging from your “P.S.”), these concerns were also brought forward by the two anonymous reviewers and thus we refer to our reply to referee #2. In short, we indeed intend to tone down the statements and conclusions and 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.

On L165 we inform that shoot enclosures were used, thus we assumed that it would be evident that the shoot (i.e. both needles and branch) was measured. However, if this is not the case, we can add: “The shoot enclosures enclosed parts of the shoots, i.e. both needles and the woody stem (see Fig. 1 in Aalto et al. (2014)).” on L168 after “2009-2011.”.

The reported emissions of VOCs from new foliage originate from buds in the very beginning of the measurement period. We can point this out in Sec. 2.3.

The information that only one tree was measured is provided on L167. Since referee #2 was also confused about these details (i.e. how many shoots and trees were measured), we suggest to reformulate “...from a ~50 year old Scots pine tree…” to “...from one ~50 year old Scots pine tree…” on L167 and to add the following sentence: “Within one season, one mature shoot and one current year bud/shoot were measured, but during the next growing season, different shoots were chosen for the measurements.”

We agree with you that it is not justified to suggest a different value for the leaf age factor used in MEGAN by only considering the findings from one study (in this case Aalto et al., 2014). However, findings from only one study can be used to question the current value (so that’s what we did). You are referring to several publications that showed only moderately increased emissions during shoot growth from Scots pines and other coniferous species. Several of these are also cited in our manuscript (Janson 1993, Komenda & Koppmann 2002, Tarvainen et al., 2005; Hakola et al. 2006; Räisänen et al., 2009, see e.g. Fig 4). Flyckt (1979), Kim (2001), and Geron and Arnts (2010) are not cited as they do not deal with Scots pine. Independent of conifers species, it is correct that no other studies have found such a pronounced effect in the emissions from new foliage as Aalto et al. (2014), except Tarvainen et al. (2005) (see e.g. our Fig. 4). This has also been clearly pointed out in Sec. 2.3. HOWEVER, previous studies on Scots pines (like the ones you refer to) have not measured the emissions from buds/growing needles and mature needles separately (as also pointed out in e.g. Table A2). This is a shortcoming, since it might be very very difficult to determine emissions from buds or growing needles, if the majority of needles inside the chamber are mature (as those previous studies also show). Only Räisänen et al. (2009) measured new and one year old needles separately, but measurements of growing needles were only started in the end of July, when the elongation period was almost completed. Their findings are in line with those by Aalto et al. (2014) during the period from which they have measurements, but they didn’t measure during spring. These points are mentioned and discussed e.g. in the introduction and in Sec. 2.3. It should also be said that it is rather likely that other tree species act differently when it comes to emissions originating from growth, thus measurements from other tree species can’t really be used to falsify or verify observations from Scots pines (which has also been pointed out in our conclusion section). In conclusion: since no one has so far utilised the same approach as Aalto et al. (2014) and falsified its representativeness, we have no reasons to not apply them in such a study as this (i.e. this manuscript). Additionally, since our manuscript does not present the measurements themselves, but rather builds on the peer-reviewed paper by Aalto et al. (2014), it seems rather strange that we here should evaluate the validity of the results by Aalto et al. (2014).

In my view the paper would gain impact if the authors discuss more critically the uncertainties and limitations in terms of representativeness of the input data and the reasons why they diverge so much from that of previous studies. Here I offer a few reflections that might be inspiring. One reason for the magnitude higher emissions potentials reported by the Aalto study lies in the measuring scale and the reference unit used. I am convinced that bursting buds and very young expanding shoots still bare of needles release MTs and other VOCs but most of them likely stem from other organs tissues than needles. Also, the (co)-authors published several nice papers showing that VOC emissions from axial organs are important, especially during springtime. Hence relating VOC emissions from buds and very young shoots to a minute amount of needle generates huge and highly variable needle emission potentials that in fact do not exist and could (partly) explain why other studies that measured emission at needle scale as for example Raisänen et al. 2009 found lower emission potentials and lower leaf age effects. In order to see how the emission potentials of Scots pine shoots evolve during the course of the seasons independent of the actual needle mass they wear it would be interesting to express emission rates per whole shoot and/or per whole shoot dry mass.

As stated above, we do not believe that it is our responsibility to evaluate the validity of a peer-reviewed publication (in this case Aalto et al. 2014), however, you are probably very right that we would get more citations if we started to speculate in the differences between studies! So we thank you for your insightful reflections! Thus, we suggest to briefly discuss the uncertainties and limitations of the approach by Aalto et al. (2014) and suggest why the results from Aalto et al. (2014) diverge from other studies by including the following information in Sec. 2.3 (the following is not going to be the final wording, as it is formulated as a reply to Michael Staudt, but it will include the same content): The measurements from developing shoots were done on branches where buds/needles and the woody stem were included. It is thus an aggregate measurement of the whole branch tip. In an elongating bud of Scots pine the stem develops first and growth of needles is very slow during the first ca. 5 weeks of the growth period (in S-Finland conditions). Hence, Michael Staudt is correct in that, during the first weeks, the emissions probably are originating rather from the elongating (green) stem than from the needle primordia. However, due to obvious logistical reasons it is very difficult to quantify the biomass of the stem and needles at a given point of time: when you cut the branch for biomass measurements, then your measurement period for this branch is ending and a new bud or branch has to be set up for measurements which causes a discontinuous dataset. So, even when we fully recognize the potential error source in the reported emission rate per biomass measurement, we still think that there may not be a reasonable way of getting a more accurate estimate. Additionally, most other branch scale measurements have included the stem tissue in the enclosures as well, so this is an error that is prone to all such estimates of emission rates. As also mentioned earlier, we will point out that no previous studies (except Räisänen et al., 2009) have measured the emissions from buds/growing needles and mature needles separately, and that this can be one cause of the observed differences between Aalto et al. (2014) and other studies, since it might be very difficult to determine emissions from buds or growing needles, if the majority of needles inside the chamber are mature. As mentioned earlier (both in this response and in the manuscript), Räisänen et al. (2009) only measured the emissions from growing needles from the end of July onwards and their findings are in line with those by Aalto et al. (2014).

The reliability of the b=0.09 normalization procedure could be more discussed and tested. If I understood correctly the authors used this normalization to compare the Aalto et al data with literature data. On the other hand, only the Aalto emission data were used in the extrapolation and apparently this normalization was unable to explain the observed emissions variation over brief periods or even within a day. As a result the seasonal evolution of the thus calculated emission potential might be an overestimation. Wouldn't be more appropriate to apply another normalization, which explains better diel emission variation, for example those suggested by Aalto et al. 2015, or a fitted beta-value on diel emission variations?

Again, thank you for your valid suggestion. This comment is a bit in line with that of referee #2 where s/he wondered why we only used the temperature dependent algorithm and not e.g. the hybrid algorithm, and thus we generally refer to the reply we made to referee #2. It is naturally possible to do as you suggest, but it would ruin our chances of comparing with other studies (which you have emphasised in your review that we should do more), and it introduces more uncertainty, since the temperature dependency/sensitivity is very sensitive to a low number of data points and any noise in the emission rate measurements. Considering the fact that the ratio of the emission rates of new and mature foliage (Aalto et al., 2014) follows the same pattern as that of the emission potentials shown in our manuscript, it is not well justified to change the method of standardisation. Since referee #2 also commented on this, it is clear that a better justification for the used algorithm is needed in the manuscript. We will add such (summarising our reply to both referee #2 and Michael Staudt) on L170.

The Aalto et al. 2015 paper also specifically describes monoterpene emissions bursts from 1-year and 2-years old Scots pine shoots (hence with mature needles) that happen especially during the spring period. I guess it is impossible to predict and quantify these temporary episodic bursts and hence could not be included in the present extrapolation/upscaling study. However, if these bursts exist as described in the Aalto et al. 2015 paper, they will reduce the relative contribution of young growing needles to the whole tree emissions during spring and may also - together with peak emissions from stems, partly explain the higher particle formation observed during this period.

Yes, you are completely correct, and in fact those bursts are partly (see below for further comment) also observed in the data presented in Aalto et al. (2014), and thus they are included in our study too. Fig. 5c shows the monoterpene emission potential of mature foliage that we used in our study. Since it is based on weekly calculated emission potentials, it is naturally not possible to observe the dynamics of those individual bursts, but anyhow they contribute to the emission potential we calculated and used, and from Fig. 5c it is also possible to see that the emission potential of mature foliage is higher during spring than later in the season. With that said, the emission bursts presented in Aalto et al. (2015) mainly take place before growth onset, and thus before the period that our manuscript targets. In fact, these monoterpene emission bursts start to be over when growth onsets. This is probably also the reason why they do not impact our emission potential of mature foliage much (see Fig. 5c).

As also responded to referee #1, we do not claim that the discrepancy between observed and predicted spring time NPF can solelemly be explained by VOC emissions from new foliage. Instead we just calculate how much higher the formation and growth rates would be if we account for the enhanced emissions from new foliage. It seems that we need to clarify this in the manuscript (in Sec. 4.3) and when doing so, we can also mention the possibility that excluded emissions from stems and emission bursts from mature foliage earlier in the season (i.e. before there is any buds or new needles) could make the numbers (i.e. formation and growth rates) even higher.

Another point of discussion I missed in this as well as in the studies by Aalto et al. is resin exudation. Pine shoots can exudate resin in micro droplets that are hardly visible but contribute well to boost emissions. For example Eller et al 2013 (http://dx.doi.org/10.1016/j.atmosenv.2013.05.028) reported that small amounts of resin is exuded from healthy, undamaged Ponderosa pine tissues, in particular from young growing needles and branches.

Thanks for pointing out this feature. Resin exudation from the buds is indeed a phenomenon that affects the emissions, and is observed in the Scots pine branches we have measured as well. This is probably a natural defense that protects the developing buds from feeding insects, and occurs before the buds start elongating. Naturally exposed resin on developing cones, buds and the bases of needles may contribute up to 10% of the total ecosystem monoterpene flux while the resin is fresh (Eller et al. 2013). We will mention this in Sec. 2.3 when elaborating on the way the measurements were carried out.

Some specific comments

L33: remove “ecological” since a by-product is not formed for ecological reasons
OK

L53: I suggest removing “still”
OK

L93: “static needleleaf development” is an unclear awkward wording, please change

You are right, it's not a very good sentence. We will reformulate the sentence from "If a model utilises rather static needleleaf development combined with only slightly higher emission potentials of new than mature needles, the influence of new coniferous foliage to canopy BVOC emissions is predicted to be minor (Guenther et al. 2012)" to "If a model assumes that the emission potential of new needles is only slightly higher than that of mature foliage, then the influence of new coniferous foliage to canopy BVOC emissions is predicted to be very minor, since the mass of emerging and growing needles is very small during spring time (Guenther et al. 2012)."

L97: suggest replacing "complete" by "better"
OK

Chapter 2.3: Even though I appreciated much the literature compilation done by the authors, I found this M&M chapter rather unconvincing and the ideas behind unclear.
OK. The reason behind this literature compilation (which is also given on L187 onwards in our manuscript) is that there exists large variability in emission rates, not only between species, but also within species, which results in large uncertainties in the emission potentials used in models. Since our work is only based on measurements from one publication, we naturally need to compare our emission values with those obtained in other studies. Since all 3 referees have pointed out in their reviews (and as we have pointed out in our manuscript, but obviously not clearly enough), that the conclusion of this study is limited by the data availability, it is evident that our literature compilation needs to stay in the manuscript, and we even need to expand the discussion of it as suggested by Michael Staudt, but that the motivation behind it needs to be clarified. So we will clarify the motivation in Sec. 2.3.

L179 ff: "normalization", see my comments above
See our reply above (where you commented on this).

L197-198 ". . .hence is able to generate significant seasonal variations (Hellén et al., 2018)". The reasoning behind this statement is unclear to me.
Here we are actually referring to the same problem you raised a bit earlier on in your review, namely the fact that beta is in reality not a constant, and when treated as a constant, the seasonal evolution of the calculated emission potentials might not be completely correct. We suggest to reformulate the sentence "If the emission was not already standardised, a value of β = 0.09 °C-1 was used as this is the most commonly used value in the literature for monoterpenes, though β is known to vary during the season and can be different for individual monoterpene isomers (Hakola et al., 2006; Hellén et al., 2018), and hence is able to generate significant seasonal variations (Hellén et al., 2018)." to "If the emission was not already standardised, a value of β = 0.09 °C-1 was used as this is the most commonly used value in the literature for monoterpenes. However, β is in reality known to vary during the season and can be different for individual monoterpene isomers (Hakola et al., 2006; Hellén et al., 2018), and hence can cause significant seasonal variations in the calculated emission potential which are not necessarily true (Hellén et al., 2018)."

L213 ”Raisanen et al. (2009), who. . .” This study was conducted on needles not whole Scots pine shoots and the difference in emission potentials was only significant on a needle dry weight basis, not on a needle surface basis. There is another study by these authors on whole Scots pine trees in OTCs, which could be considered (Raisanen et al. 2008; https://www.sciencedirect.com/science/article/abs/pii/S1352231008000496)

This is correct, and in the manuscript, we have also not claimed that they did shoot measurements. We can add the additional information about the measurements to the sentence you refer to by reformulating: “Räisänen et al. (2009), who provide emission potentials of new and mature needles, individually, show that the potential of new needles to emit monoterpenes is twice as high as that of mature needles. This is based on measurements from August-September, and is in accordance with findings by Aalto et al. (2014), who show that the difference in the potentials of the two needle age classes is about a factor of two in August (Fig. 3f).” to “Räisänen et al. (2009), who measured the emissions from new and mature needles, individually, and without contributions from the woody parts of the branches, show that the potential of new needles to emit monoterpenes is twice as high as that of mature needles when calculated based on the dry mass of the needles. This is based on measurements from August-September, and is in accordance with findings by Aalto et al. (2014), who show that the difference in the potentials of the two needle age classes is about a factor of two in August (Fig. 3f). However, when Räisänen et al. (2009) determined their emission potentials based on needle surface, instead of needle dry mass, the authors did not find a significant difference in the emission potentials.”. Räisänen et al. (2008) is a very interesting paper, but unfortunately we cannot include it in Fig. 4 and the related discussion in Sec 2.3 as the values are standardised using the hybrid algorithm and it is not possible to re-standardise the emissions with the information presented in the paper.

L 225-228. “The reported emission potentials of Scots pine seedlings . . . than plants growing in the field.” Please add references.

Yes, references are always good! However, to our knowledge, there does not exist any publications that specifically prove that plants grown in the laboratorium emit VOCs differently than plants growing in the field. It is therefore challenging to add one or a few references to this sentence, which is also the reason why we didn’t do it in the first place. However, if one studies the reported VOC emissions from individual plant species and compares them to each other, the limited evidence available indicates that plant VOC emissions differ greatly between locations (like laboratory, research garden and forest) as e.g. also concluded by Faiola and Taipale (2020). Niinemets (2010) additionally made a nice review that illustrates that it is unlikely that trees grown under optimal conditions should exist in nature. And since the emissions of VOCs depend on many more environmental variables than temperature and light, our comment/speculation in the manuscript cannot be viewed as very controversial. In lack of better, one option would therefore be to add citation to Niinemets (2010) and Faiola and Taipale (2020) at this point in the manuscript.

L324: “Please be aware that the measured canopy, within an area. . .” long sentence; consider rephrasing

Indeed the sentence is long, and we simply suggest to split it from originally to “Please be aware that the measured canopy, within an area with a radius of 200 m, is only covered by ~75% Scots pine (and ~25% other tree species). Thus our results cannot be directly compared to Taipale et al. (2011) and Rantala et al. (2015), but these two studies provide the most suitable observations for validation of our results.”

L345 “The underestimation. . .” Here and elsewhere in the text as well in the Figure legends I suggest to add “potential” or “estimated” to read “the estimated underestimation”, because the outputs resulting from the presented extrapolation and modelling study should be considered as a case study.
Yes, this is a good idea that better captures what we actually show, and thus we will change the manuscript accordingly.

Figure 3 legend is insufficient. The origin of the data should be mentioned; measurements made on how much shoots and trees, normalized how. . .?
OK, we will extend the figure caption by adding: “The emission potentials are calculated based on the measurements presented by Aalto et al. (2014). Emission rates were obtained from one ~50 year old Scots pine tree at the SMEAR II station. Within one season, one mature shoot and one current year bud/shoot were measured, but during the next growing season, different shoots were chosen for the measurements. The emission potentials were standardised by Eq. (5) in Guenther et al. (1993) (T_s = 30 °C, β = 0.09 °C $^{-1}$). See Sec. 2.3 for more details.”

Figure 8 is very dense and hard to read; showing only the left column graphs (a-f) in a bigger size might be sufficient.
True, and this point was also raised by referee #2. One option is that we follow our suggestion as replied to referee #2, however, as you point out, it might actually be a better idea to only show a-f and then enlarge them, since, in their current stages, it is already very difficult to distinguish between “MEGAN style” and “Mature needles”. Subfigures g-r could alternatively be added to the appendix.

Michael Staudt

PS: Please note that the comments above were written as a review at an earler state of the submission, which I did not finish in time and therefore was temporary excluded from the reviewing process (I aplogize for the delay). Meanwhile the authors have already responded to several of my comments since these were also addressed by the other referees. Nevertheless I hope that they will keep the discussion running.
No worries, and thanks a lot for taking the time to review our manuscript! Sincerely speaking, your comments were very constructive and valuable and will improve our manuscript!

References:

Faiola, C. and Taipale, D.: Impact of insect herbivory on plant stress volatile emissions from trees: A synthesis of quantitative measurements and recommendations for future research, Atmos. Environ. X., 5, 100060, https://doi.org/10.1016/j.aeaoa.2019.100060, 2020.

Niinemets, U.: Mild versus severe stress and BVOCs: thresholds, priming and consequences, Trends Plant Sci., 15, 145–153, doi:10.1016/j.tplants.2009.11.008, 2010.