Reply to editor's comment

We appreciate the Editor's view and share his wish to provide a fruitful interaction between the authors and the referee. Our previous response to referee #1 (posted on 6th of March) was not intended as a final response, but more meant to serve as part of an interactive discussion and correct potential misunderstandings. This is also the reason why we tried to post it as soon as possible, did not follow the appropriate structure of a final reply, and why we did not include the specific suggestions for changes to our manuscript. Those specific suggestions are presented here together with other "new" text in red, while parts of our original reply is included in blue when deemed needed.

Reviewer #1

The authors consider the implications of failing to account for springtime monoterpene emission bursts from new needles when modelling biogenic emissions and subsequent aerosol formation and growth. The pronounced seasonality of the emissions of some biogenic volatile organic compounds (bVOCs) from some species of vegetation has been reported previously and has to some extent been included in the current generation of bVOC emissions models. While the magnitude of the change in monoterpene emission potential included in the leaf age activity factor in MEGANv2.1 (the most widely used bVOC model) is substantially smaller than that reported for Scots pine at the SMEAR II field station, emission potentials in MEGAN are for ecosystem / plant functional types rather than individual species.

While I very much appreciate the concept that the authors are attempting to demonstrate and agree that this could have significant implications for biogenic emissions and atmospheric composition, air quality and climate at the local scale I do not feel that the work presented here is sufficiently conclusive.

Firstly, we would naturally like to thank the referee for his/hers valuable comments.

There exists a large variety of important biosphere-atmosphere models of different scales that have different purposes and aims, but that all try to answer questions within the categories of air quality, climate, atmospheric composition and more. Not all models are global. It is correct that global models utilise emission potentials for plant functional types, but many of the models that are not global models don't, even though they still contain e.g. the full version of MEGAN or only the emission response algorithms from e.g. MEGAN. Independently, when modelling boreal forests, one should be aware of the discrepancy that an exclusion of the enhanced emissions from new Scots pine foliage can result in. As mentioned in our previous reply, it would be very difficult to suggest a better value for the coefficients in the expression for the leaf age emission activity factor (as also pointed out in Sec 4.1), since no other boreal species than Scots pine was studied, since it is not transparent how models attain the emission potentials of their plant functional types, and since the emission measurements of new foliage from other boreal needle evergreen trees do not exist. Reflecting upon the referee's comment, it is clear that we need to specify and clarify these items in the introduction.

In the introduction, we will try to even stronger underline the motivation of this study, which arises from the fact that Aalto et al. (2014) have falsified that emerging and mature Scots pine foliage have the same potential to emit BVOCs. Such evidence naturally calls for a quantification of its potential atmospheric impacts.

In effect, this study is based on 3 years of measurements of monoterpene emissions from a single Scots pine tree at a single site, extrapolated to assume that all species at this one site behave in the same way and that the same behaviour would be observed at all boreal forests in Finland (although the magnitude of the effect would differ according to length of growing season). The authors point to previous work that also reports elevated emissions from new needles (up to a factor of two according to Räisänen et al, 2009) BUT ignore the fact that the same authors observed similar differences between emissions measured from mature needles (Räisänen et al, 2005) and fail to acknowledge that extensive measurements of bVOC (mostly isoprene) emissions in Estonia by Noe et al and Niinemets et al found substantial differences in emissions between trees, between locations but also within the same tree. i.e. the community are well aware that the extrapolation of emissions potentials from a limited number of measurements to the ecosystem, regional or even global scale must result in highly uncertain emission estimates.

We are thankful to the referee for pointing this out and wish to highlight that we have acknowledged the large intraspecies variability as well as variability in individual trees' emission rates of monoterpenes in Sec. 2.3. However, nowhere in the manuscript do we claim, nor assume, that other boreal forest species – or all tree species in Finnish forests - behave in a similar way as Scots pine. This is of course also the reason why we did not compare our emission potentials to those used in models as specified on e.g. page 6, L198-203 and on page 8, L307-309. Since our entire manuscript only deals with Scots pine, it seems at best needless, and at worse confusing, to point out that no other species were considered. To turn it around; if such a thing had been done, shouldn't it then have been pointed out in the manuscript?

Concerning the reference to Räisänen et al, we refer to our earlier reply and suggest that we do not comment on the difference in the emissions from mature needles in the manuscript.

In light of both reviewers' comments about the large intraspecies variability of VOC emissions, it is very clear that we need to clarify the aim and also underline the uncertainties associated with this study, especially in the abstract and conclusion sections. For further elaboration, we refer to our reply to referee #2.

The important question here then is whether the difference in total emissions and potential impact on atmospheric oxidation is sufficiently large to warrant the inclusion (or rather increase) of leaf-age based differences in monoterpene emission potentials in a global modelling framework such as MEGAN. And to my mind, while I accept that it could well be of significance locally, the authors do not demonstrate that its importance extends beyond Finland.

As specified earlier and above, our aim does not include to directly change the age dependent emission factors in MEGAN or in any other model, which is also clearly stated in Sec. 4.1, e.g. on page 10, L391-392. However, since evergreen trees are dominating in many ecosystems, we believe it is warranted to discuss the impact of such observation on the modelled emission patterns. This needs to be underlined at least in the introduction.

(1) While Scots pine is the dominant species in Finnish conifer forests, it is not the only one, and to scale the effect up at the very least the authors should consider the full mix of species in these ecosystems. At least one previous study has reported the difference in emissions factors for all of the major tree species at these sites.

In our manuscript, national level estimates only consider the area of Scots pine trees growing on forest land, and thus exclude the area of Scots pines growing on poorly productive forest land and the areas where other species grow. This has been clearly stated in e.g. both Sec. 2.4 and 3.4, thus no further elaboration would be assumed needed.

To our knowledge, there do not exist BVOC measurements of *new* foliage from other boreal species, thus it would not be possible to consider their contribution, and even if such data would be available, it is not within our aim. As also replied to reviewer #2, we will add a call for measurements of new foliage from Scots pine as well as from other tree species.

(2) Although the authors state that SMEAR II is representative of forests in southern Finland they do not explain how they have concluded this, and similar for SMEAR I in northern Finland. They are thus already extrapolating from, at best, 2 sites to an entire country even before trying to argue that it is of global importance.

Thank you for pointing out this potential misunderstanding in our manuscript. We do not believe that we have explicitly stated that the SMEAR II station is representative of forests in southern Finland nor that the SMEAR I station is representative of forests in northern Finland (not to say that they are not). L243 on page 7, might be the cause of this confusion, and thus we suggest to reformulate “The premise is that this is representative for southern Finland” to “In order to conduct our analysis, we have to assume that this is representative for southern Finland” or similar. Observations from SMEAR I and II are naturally utilised due to data availability and in order to provide estimates across a latitudinal gradient.

In the introduction, on page 2, L67-71, we have tried to argue for the potential global importance, highlighting the fact that Scots pine is the most widely distributed pine species in the world, and that it is one of the most dominant evergreen tree species. Also the fraction of Finland which Scots pine covers is pointed out.

The most up-to-date Finnish forest statistics lists the volumes (10^6 m3) of main tree species in south FI and north FI (https://stat.luke.fi/en/forest-resources-region_en-2):

	south	%	north	%
pine	713	43.90	530	62.21
spruce	572	35.22	168	19.72
birch	269	16.56	142	16.67

This data shows that pine forests cover from 44 to 62% of the total volume of forests in the south and north, respectively. Hence, it is warranted to say that SMEAR II and I forests, both dominated by Scots pine, are at least among the most abundant in Finland.

(3) Aerosol formation and growth depends on more than just monoterpene emissions and the fact that models currently under predict new particle formation at SMEAR II during the spring does not conclusively demonstrate that this discrepancy is entirely due to an under-estimation of total monoterpene emissions. Aerosol formation potential differs widely between different monoterpenes and actual aerosol yield has also been shown to depend on the mix of bVOCs emitted not just the quantity and broad type (e.g. Kiendler-Scharr et al., 2009; McFiggans et al., 2019). A PTR does not distinguish individual monoterpenes. While the authors are able to show that aerosol formation would be increased if there was indeed a spring burst in emissions at SMEAR II it is not clear what oxidation pathways are included in there model and hence it is hard to be certain that it is emissions alone that are incorrectly modelled.

We completely agree with the referee that the influence of BVOCs on aerosol processes is far from simple. Our calculations of the potential impacts on the predictions of NPF and growth are order-of-magnitude calculations, too rough to even consider the different potentials of individual monoterpenes to participate in aerosol processes. The complete set of equations used for these calculations are provided in Sec. 4.3 and from there it is possible to do the re-calculations using the input values provided in the manuscript and get the same results as listed in Table 3. In the manuscript, we could possibly further underline that the calculation is connected with large uncertainty and that no other equations than those presented in Sec. 4.3 were used.

(4) For the sake of argument, let's assume that the authors are correct in their assumptions that all tree species in Finnish forests show the same enhancement in monoterpene emissions during the spring as observed at SMEAR II. At most, the authors state that emissions increase from Finnish forests by 25% but taking into account the effect of latitude they estimate the actual increase in monoterpene emissions from Finland to be of the order of 27 Gg y-1 (i.e. 0.027 Tg y-1).

Again, we want to stress that we have not assumed or claimed that other tree species show the same enhancement in monoterpenes emissions during spring. As mentioned earlier, if we had done so, we should have stated it, probably not the other way round.

(a) bVOC emissions are dominated by emissions from tropical forests (and the same holds true for evergreen ecosystems). Using the total emissions from each plant functional type in Guenther et al (2012) as a baseline, an increase of 0.027 Tg y-1 of monoterpenes equates to an increase of 0.4% in monoterpene emissions from boreal evergreen needleleaf trees or a 0.12% increase in total bVOC emissions from this ecosystem category. In a global context, this would be an increase of <0.02% in total global monoterpene emissions.

Here we refer to our earlier reply. In the manuscript, we will speculate that the emission trait is probably not specific to Finnish Scots pine trees (nor is it sure that all Finnish Scots pines emit like this), and thus our findings could be of importance in simulations of all places where Scots pines make up a major fraction of the land cover. However, due to lack of measurements, we will naturally underline that we cannot be sure of this, and that more measurements are required.

With this manuscript, we are able to show the potential consequences (on modelling of canopy emissions and aerosol processes) of new Scots pine foliage as a very strong emitter of monoterpenes. Our work should motivate readers to investigate if such a phenomenon is also a trait in other evergreen species, which is naturally possible. Since many evergreen trees, especially in tropical regions, have several needle cohorts flushing annually, the impact of new needle emissions might be even larger than what is observed in northern forests. This motivation will be clarified in the manuscript.

(b) Assuming instead that all boreal evergreen ecosystems exhibit the same pattern of emissions and that there we are currently underestimating monoterpene emissions from these high-latitude forests by 25%. This would amount to a 1% increase in global monoterpene emissions or a 0.15% increase in total bVOC emissions

Here we also refer to our earlier reply. In the introduction and/or Sec. 4.3 we can further clarify the importance of BVOCs to the formation of new particles in the boreal region vs. e.g. the tropics.

(5) Given the limited magnitude of the increase when viewed in terms of global annual emissions, what is probably of greater importance then is the impact that these additional emissions would have on springtime atmospheric chemistry. And here, the authors demonstrate that it does make a difference for these two specific sites BUT (as noted above) do not give sufficient detail of the assumptions made in deducing aerosol formation and growth from lumped monoterpene emissions and do not put it into a global context. Is the effect substantial enough to make a difference to local or global climate or local or regional air quality? Or just an interesting phenomena in boreal conifer forests?

Yes, the reviewer is correct, timing is here the key word. As replied earlier, all equations used for our aerosol calculations are provided in Sec. 4.3. We further refer to our earlier reply concerning this point and as mentioned above, we can further clarify the importance of BVOCs to the formation of new particles in the boreal region vs. e.g. the tropics.

What we are therefore left with is a review of existing measurements from SMEAR II (and to a lesser extent SMEAR I), a comparison against other observed monoterpene emission potentials in similar forests and a statement that the effect is substantial enough to warrant inclusion in global bVOC emission models beyond what is already accounted for. In my opinion the authors need do far more to justify their conclusion. At the very least they need to account for the full range of tree species in a Finnish coniferous forest but to really make a case for publication I feel they must show that such a substantial burst is seen in all evergreen needleleaf ecosystems, and to fully model this (i.e. at a global scale) to show the impact on total emissions and on total aerosol production.

Please see our replies above.

This is a particular shame as SMEAR II is an incredibly rich dataset that deserves constant re-visiting and re-evaluation. Were the authors able to conduct the extended analysis required to support their conclusions it would be a welcome addition to the literature.