

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

A. The manuscript addresses whether the environmental changes causes changes in the emission of biogenic volatile organic compounds (BVOCs) that lead to changes in cloud formation via secondary organic aerosol acting as cloud condensation nuclei. The authors used laboratory chamber experiments to simulate the response of BVOC emissions from a mixture of boreal forest trees for two types of changes: constitutive emissions due to temperature and water availability changes and stressor-induced emissions especially due to insect infestation. The BVOCs emitted were monitored using two complementary analytical instruments: proton-transfer reaction mass spectrometry and gas chromatography/mass spectrometry. SOA and CCN were measured by a scanning mobility particle sizer and a cloud condensation nuclei counter, respectively. The experiments show that changes in temperature and water availability cause differences (but not necessarily statistically significant) in the total VOCs emitted by boreal forest trees, which has a small effect on aerosol hygroscopicity. Insect infestation stresses boreal forest trees differently, the composition of BVOC emissions changes, and for this case, causes enhanced sesquiterpene emission which leads to lower aerosol hygroscopicity.

B. The research is novel because it connects an understudied, yet likely important, climate feedback cycle. This work shows the potential for the environmentally induced and constitutive changes in BVOC emissions to lead to changes in SOA and CCN hygroscopicity, and demonstrates the need to consider these factors in climate models. The latter point is especially well-explained on p. 9 (first full paragraph).

C. The experimental approach is appropriate for the work. One question that the authors should consider is whether the relatively young trees (3-4 years) used in this work would respond similarly to drought, temperature changes, and aphid infestation as mixed-age or mature population (as found in boreal forests) would respond. While it is beyond the scope of this study to study all age ranges and conditions, a comment to address this uncertainty would be helpful.

D. The use of statistics and treatment of uncertainties is appropriate. The manuscript would be improved if the number of replicates used to calculate the standard deviations for the error bars in Figures 2 and 3 (most easily added the captions) were included.

E. The conclusions are valid. The authors explained the limitations of the current work especially in terms of the how temperature and drought impacts this particular boreal forest trees, how the insect infestation used in this work (aphids) is a single example, and that different plants with different insect interactions may create different SOA, which in turn change the hygroscopicity of CCN. This is an important point and it could be strengthened with an example (perhaps review Faaila et al. *Atmos. Chem. Phys.*, 2015, which discussed SOA changes due to simulated herbivory in several plant species)

F. Beyond the suggestions included in this review, the abstract tends to overgeneralize the applicability of this work. It would helpful to acknowledge (in the abstract) the conditionality of this work, i.e. the scope of this work is one group of boreal forest trees and one type of insect infestation.

G. The references are appropriately applied and cited.

H. The text is mostly readable with the exception of a few sentences that are inconsistent with scientific style and the bulk of the text. For example, in paragraph 1, the use of the imperative in "Note that already nowadays environmental stress factors are affect plants" is awkward. In the following sentence "...more than 40% of forest trees in Europe suffer from various stresses in which biotic stresses account for ~40%" of what? The total stresses? Rewording this sentence would improve clarity. Also "nowadays" is a casual and vague term (used also p. 9).

On page 8 the sentence "Climate changes induces both long terms changes and short term changes in the climate parameters" needs to be reworded to improve clarity.

Also on page 8 the following sentence "...more trees will be exposed more often to stresses." may be improved by rewording "...more trees will experience stresses more often."

Reviewer #2 (Remarks to the Author):

Review on "Environmental conditions regulate the impact of plants on cloud formation" by Zhao et al.

This is an interesting paper on a timely, appealing and important topic. However, I think it is in the limit of acceptance for a high profile journal of the Nature family because it does not seem robust, comprehensive, quantitative and novel enough.

See my comments while reading the paper:

Abstract too verbose and with some confusing sentences like for example: "...biotic and abiotic environmental factors that regulate emissions of VOC also modify the CCN activity of the resulting SOA". Does this occur because of a direct effect of those environmental factors or only through the changes they generate on SOA?

Or the sentence starting with "Our findings" better state which are they? It is not clear.

Or in introduction: For example it is not only "nowadays" that environmental stress factors are affecting plants....this has occurred always.

Or the sentence "...This implies a strong feedback between plants' emissions and climate ". This is still not demonstrated at least its actual quantitative importance.

Or the sentence " effects on VOCs emission of plants are much less investigated than those on net CO₂ exchange..." It is true but avoid giving the wrong impression that there is not much on this, since in fact, there are multitude of studies in this area.

The statistics are not clear nor robust either. For example, there is no variance data presented for monoterpene/sesquiterpene ratio or no "n" in the figure 2 caption

The experimental design: tested hygroscopicity of SOA are in a quite narrow range. One wonders what would happen in a wider range.

Different compounds have different responses to temperature, so I wonder how this was not appreciated with increasing temperatures tested. How there was no change in emission pattern for constitutive and even more for induced emissions? Any explanation for that?

Figure 3. The caption statement on significance of changes in k does not seem to be appreciated in the figure. Unclear.

Water shortage (drought) appear in page 8 as a surprise not presented, nor treated, before. By the way, this paragraph on drought needs more work and detail.

In the following paragraph, more than modeled values the reader is interested in actual measurements ...not possible?

Next paragraph: I agree this is an important study with important and interesting information such the one on biotic stresses likely having significant influence on the CCN concentration in areas where biogenic SOA components dominate particle formation..... but this, by itself, is not completely new (this same research group itself has studied this for long), not very robust, not very extensive, not wholly convincing, not with real measurements, and specially with not much quantitative information, at least for a paper in a high profile journal like this one. It seems instead very adequate for a good atmospheric journal.

Figure 4 caption....why modeled and not measured values?

Experimental design-Extended figure 1. Why not using a single chamber where plants are in contact with ozone as it in fact occurs in nature?

Try to explain more clearly the caption of figure extended data figure 2.

Extended data figure 4: I am still surprised of this absence of change with temperature, and still would very much recommend having the same graph for the different monoterpene compounds and the different sesquiterpene and others compounds since they have different physic-chemical properties and temperature sensitivities

Extended data figure 6. As in most parts of the text I miss statistics here. How significant are the changes? They do not seem very strong or significant.

7 *Reviewers' comments:*

8 *Reviewer #1 (Remarks to the Author):*

9 *A. The manuscript addresses whether the environmental changes causes changes in the*
10 *emission of biogenic volatile organic compounds (BVOCs) that lead to changes in cloud*
11 *formation via secondary organic aerosol acting as cloud condensation nuclei. The authors*
12 *used laboratory chamber experiments to simulate the response of BVOC emissions from a*
13 *mixture of boreal forest trees for two types of changes: constitutive emissions due to*
14 *temperature and water availability changes and stressor-induced emissions especially due to*
15 *insect infestation. The BVOCs emitted were monitored using two complementary analytical*
16 *instruments: proton-transfer reaction mass spectrometry and gas chromatography/mass*
17 *spectrometry. SOA and CCN were measured by a scanning mobility particle sizer and a*
18 *cloud condensation nuclei counter, respectively. The experiments show that changes in*
19 *temperature and water availability cause differences (but not necessarily statistically*
20 *significant) in the total VOCs emitted by boreal forest trees, which has a small effect on*
21 *aerosol hygroscopicity. Insect infestation stresses boreal forest trees differently, the*
22 *composition of BVOC emissions changes, and for this case, causes enhanced sesquiterpene*
23 *emission which leads to lower aerosol hygroscopicity.*

24 *B. The research is novel because it connects an understudied, yet likely important, climate*
25 *feedback cycle. This work shows the potential for the environmentally induced and*
26 *constitutive changes in BVOC emissions to lead to changes in SOA and CCN hygroscopicity,*
27 *and demonstrates the need to consider these factors in climate models. The latter point is*
28 *especially well-explained on p. 9 (first full paragraph).*

29 *C. The experimental approach is appropriate for the work. One question that the authors*
30 *should consider is whether the relatively young trees (3-4 years) used in this work would*
31 *respond similarly to drought, temperature changes, and aphid infestation as mixed-age or*
32 *mature population (as found in boreal forests) would respond. While it is beyond the scope of*

33 *this study to study all age ranges and conditions, a comment to address this uncertainty*
34 *would be helpful.*

35 **Response:**

36 We thank the reviewer for the supporting remarks.

37 We have accepted the suggestion and made a remark on the use of young rather than more
38 mature trees. Indeed, the effect of age on how trees respond to stresses is understudied and
39 largely unknown. We agree with the reviewer that it is outside the scope of this paper. We
40 therefore added the following remark to better indicate the limitations of our statements in the
41 revised manuscript (now page 9):

42 “We investigated young boreal tree species. It is possible that different plant species with
43 different ages may exhibit different responses in VOC emissions to stresses and thus further
44 change SOA composition and properties.”

45 *D. The use of statistics and treatment of uncertainties is appropriate. The manuscript would*
46 *be improved if the number of replicates used to calculate the standard deviations for the*
47 *error bars in Figures 2 and 3 (most easily added the captions) were included.*

48 **Response:**

49 We accepted this comment. In the revised manuscript, we have added the number of
50 measurements used to calculate the standard deviations in the captions of Figure 2 and
51 separately in two newly added tables for Figure 2 and Figure 3 (now Extended Data Table. 1
52 and Table 2).

53 *E. The conclusions are valid. The authors explained the limitations of the current work*
54 *especially in terms of the how temperature and drought impacts this particular boreal forest*
55 *trees, how the insect infestation used in this work (aphids) is a single example, and that*
56 *different plants with different insect interactions may create different SOA, which in turn*
57 *change the hygroscopicity of CCN. This is an important point and it could be strengthened*
58 *with an example (perhaps review Faoula et al. Atmos. Chem. Phys., 2015, which discussed*
59 *SOA changes due to simulated herbivory in several plant species).*

60 **Response:**

61 We thank the reviewer for the supporting remarks and have accepted the suggestion. In the
62 revised manuscript, we have added an example to strengthen our discussion as follows.

63 “We investigated young boreal tree species. It is possible that different plant species with
64 different ages may exhibit different responses in VOC emissions to stresses and thus further
65 change SOA composition and properties. For example, simulated herbivory on different tree
66 species has been shown to cause different responses in VOC emissions, which alters the SOA
67 composition with some variability^{33,34}.”

68 *F. Beyond the suggestions included in this review, the abstract tends to overgeneralize the*
69 *applicability of this work. It would helpful to acknowledge (in the abstract) the conditionality*
70 *of this work, i.e. the scope of this work is one group of boreal forest trees and one type of*
71 *insect infestation.*

72 **Response:**

73 In the revised manuscript we have modified the abstract to fix the overgeneralization. We
74 clearly state that the study was based on one group of boreal forest trees and one type of
75 insect infestation (aphid here).

76 *G. The references are appropriately applied and cited.*

77 *H. The text is mostly readable with the exception of a few sentences that are inconsistent*
78 *with scientific style and the bulk of the text. For example, in paragraph 1, the use of the*
79 *imperative in "Note that already nowadays environmental stress factors are affect plants" is*
80 *awkward. In the following sentence "...more than 40% of forest trees in Europe suffer from*
81 *various stresses in which biotic stresses account for ~40%" of what? The total stresses?*
82 *Rewording this sentence would improve clarity. Also "nowadays" is a casual and vague term*
83 *(used also p. 9).*

84 *On page 8 the sentence "Climate changes induces both long terms changes and short term*
85 *changes in the climate parameters" needs to be reworded to improve clarity.*

86 *Also on page 8 the following sentence "...more trees will be exposed more often to stresses."*
87 *may be improved by rewording "...more trees will experience stresses more often."*

88 **Response:**

89 We thank the reviewer for the helpful comments pointing out our unclear sentences.

90 In the revised manuscript, we have accepted the suggestions and modified these sentences to
91 make them clear.

92 The sentence "Note that already nowadays environmental stress factors are affect plants" now
93 reads:

94 “Already at present, environmental stress factors strongly affect plants^{14, 15}.”

95 The sentence "...more than 40% of forest trees in Europe suffer from various stresses, in
96 which biotic stresses account for ~40%." now reads:

97 “...more than 40% of forest trees in Europe suffer from various stresses where biotic stresses
98 account for ~40% of the total stresses^{14,16}.”

99 The word “nowadays” in page 9 has been changed to “at present”.

100 The sentence "Climate changes induces both long terms changes and short term changes in
101 the climate parameters" have been reworded to improve its clarity. Now it reads:

102 “Climate change induces both long term, slow changes in the climate parameters such as
103 global mean temperature change and short term episodic changes, such as heat and water
104 shortage extremes.”

105 The sentence "...more trees will be exposed more often to stresses" on page 8 has been
106 modified to "...more trees will experience stresses more often" in the revised manuscript.

107 *Reviewer #2 (Remarks to the Author):*

108 *Review on "Environmental conditions regulate the impact of plants on cloud formation" by*
109 *Zhao et al.*

110 *This is an interesting paper on a timely, appealing and important topic. However, I think it is*
111 *in the limit of acceptance for a high profile journal of the Nature family because it does not*
112 *seem robust, comprehensive, quantitative and novel enough.*

113 **Response:**

114 We thank the reviewer for carefully reviewing our manuscript and giving constructive
115 comments. Based on these comments, we have substantially modified our manuscript. All the
116 comments have been addressed and we believe that these revisions have substantially
117 improved the manuscript. In the following, we provide the one-by-one responses to the
118 comments and the corresponding changes to the manuscript. The original comments are
119 shown in italics.

120 *See my comments while reading the paper:*

121 *Abstract too verbose and with some confusing sentences like for example: "...biotic and*
122 *abiotic environmental factors that regulate emissions of VOC also modify the CCN activity of*

123 *the resulting SOA". Does this occur because of a direct effect of those environmental factors*
124 *or only through the changes they generate on SOA?*

125 **Response:**

126 In the revised manuscript, we have omitted this sentence by condensing the entire abstract.

127 *Or the sentence starting with "Our findings" better state which are they? It is not clear.*

128 **Response:**

129 In the revised manuscript, we have modified this sentence to make it clear. Now it reads:

130 "The coupling of plant stresses, VOC composition and CCN activity points to an important
131 impact of induced plant emissions on cloud formation and climate."

132 *Or in introduction: For example it is not only "nowadays" that environmental stress factors*
133 *are affecting plants....this has occurred always.*

134 **Response:**

135 In the revised manuscript, we have modified this sentence. Now it reads:

136 "Already at present, environmental stress factors strongly affect plants^{14,15}."

137 *Or the sentence "...This implies a strong feedback between plants' emissions and climate".*
138 *This is still not demonstrated at least its actual quantitative importance.*

139 **Response:**

140 In the revised manuscript, we have changed this sentence to make is more precise. Now it
141 reads:

142 "This implies a potential important feedback between plants' emissions and climate."

143 *Or the sentence "effects on VOCs emission of plants are much less investigated than those on*
144 *net CO2 exchange..." It is true but avoid giving the wrong impression that there is not much*
145 *on this, since in fact, there are multitude of studies in this area.*

146 **Response:**

147 We agree. In the revised manuscript, we have modified this sentence to avoid potential
148 misunderstanding. Now it reads:

149 "The effects of environmental factors on VOC emissions of plants have been investigated by
150 a number of studies, but less intensive than their effects on the net CO₂ exchange of

151 plants^{8,9,17}. Only few studies address effects of environmental factors on induced VOC
152 emissions and SOA formation^{13, 19}.”

153 *The statistics are not clear nor robust either. For example, there is no variance data*
154 *presented for monoterpene/sesquiterpene ratio or no "n" in the figure 2 caption.*

155 **Response:**

156 In the revised manuscript, we improved the statistical analysis. We have added error bars of
157 the ratio of monoterpene to sesquiterpene. We also now specify the number of measurements
158 used to calculate standard deviations in the captions of Figure 2 and in two newly added
159 tables for Figure 2 and Figure 3 (now Extended Data Table. 1 and Table. 2).

160 *The experimental design: tested hygroscopicity of SOA are in a quite narrow range. One*
161 *wonders what would happen in a wider range.*

162 **Response:**

163 The hygroscopicity parameter (κ) of SOA from different biogenic organic precursors and
164 under different reaction conditions, converges to a narrow range, and most studies reported κ
165 in the range of approximately 0.05-0.2, roughly varying around 0.1 (see references (Prenni et
166 al., 2007; Frosch et al., 2011; Lambe et al., 2011) and the references in Extended Data Table
167 4). We guess that this is what the reviewer meant by “quite narrow range”. The κ values of
168 SOA found in our study are consistent with this range. In our experimental design, we
169 systematically changed the environmental factors (biotically stressed versus non-stressed;
170 different temperatures and water content), and we did not intentionally narrow or widen the
171 range of hygroscopicity of SOA.

172 *Different compounds have different responses to temperature, so I wonder how this was not*
173 *appreciated with increasing temperatures tested. How there was no change in emission*
174 *pattern for constitutive and even more for induced emissions? Any explanation for that?*

175 **Response:**

176 The “emission pattern” in this study refers to “the relative contributions of different classes of
177 VOC” as stated in our manuscript. Different individual compounds are lumped into three
178 classes: monoterpenes, sesquiterpenes and others. We agree that the emissions of different
179 compounds may respond differently to changes in temperature. As a matter of fact, we
180 observed that within each class (e.g. sesquiterpenes or monoterpenes), the relative
181 contributions of some individual compounds can change with temperature (see newly added

182 Extended Data Fig. 5). However, the relative ratios of other compounds did not change much
183 with temperature (see newly added Extended Data Fig. 5). Overall the relative contributions
184 of total monoterpenes, sesquiterpenes and others remained largely stable as shown in
185 Extended Data Fig. 6. An explanation for the stability of the relative contributions may be
186 that all three classes of volatiles contain stress-induced emissions. For stress induced
187 emissions, temperature dependence can be very different from those for constitutive
188 emissions, because it is not only the temperature dependence of the enzymatic system
189 synthesizing the VOC that determines the temperature dependence but also the temperature
190 dependence of the biosynthetic pathway eliciting the stress induced emissions. Hence we
191 cannot provide a general explanation for our observation that the contributions of
192 monoterpenes, sesquiterpenes and other emissions did not change substantially with
193 temperature.

194 Our concept is to use plants as holistic VOC sources to overcome biases by synthetic VOC
195 mixtures in many previous laboratory studies. We do not intend to present a biological
196 explanation for such changes. The focus of our investigations is how real and exemplary
197 constitutive and induced VOC emissions and their changes due to stresses affect SOA
198 properties such as κ and finally affect cloud formation. κ changes clearly with the change of
199 the MT/SQT ratio and this result holds even when we do not have a clear answer to the
200 question why the MT/SQT ratio was not substantially changed by temperature but clearly
201 changed by insect infestation.

202 In the revised manuscript, we have added the following explanation to clarify this point and
203 added an additional figure in the supplement (newly added Extended Data Fig. 5). In
204 addition, in order to avoid potential misunderstanding, we have changed the wording “VOC
205 emission pattern” to “VOC emission composition” throughout our manuscript.

206 “In contrast to biotic factors, abiotic factors such as mild heat (up to 35 °C) did *not*
207 significantly change the relative contributions of different VOC classes (monoterpenes,
208 sesquiterpenes and others, as described in Method) for both constitutive emissions (Extended
209 Data Fig. 3) and induced emissions (Extended Data Fig. 4). *Within* each class, the
210 contribution of some individual compounds changed, most distinct in the “others” class,
211 because specific compounds respond to temperature changes differently (c.f. Extended Data
212 Fig. 5).”

213 *Figure 3. The caption statement on significance of changes in k does not seem to be*
214 *appreciated in the figure. Unclear.*

215 **Response:**

216 In the revised manuscript, we have added p values and number of measurements for
217 calculating standard deviations (newly added Extended Data Table. 2) for the statistics result
218 in order to make it clear. Now it reads:

219 “The change of κ is not statistically significant in the constitutive emission case (t-test,
220 $p=0.09$, for the case of 25 °C and 35°C) and is only significant from 22 °C to 34 °C in the
221 induced emission case (t-test, $p=0.017$). The error bars represent the standard deviations of
222 the measurement (with symmetric positive and negative values, and the detailed number of
223 measurements included in Extended Data Table.2).”

224 *Water shortage (drought) appear in page 8 as a surprise not presented, nor treated, before.*
225 *By the way, this paragraph on drought needs more work and detail.*

226 **Response:**

227 In the revised manuscript, we have introduced the effect of drought early in the manuscript
228 (page 2, lines 60-62).

229 “We investigated the effect of aphid infestation as an example of biotic stresses and the effect
230 of heat and drought as examples of abiotic stresses for both constitutive emissions and
231 induced emissions.”

232 And in the revised manuscript, we have modified this paragraph further by adding more
233 details including one additional figure panel (now Extended Data Fig. 6a). Now it reads:

234 “In addition to heat, we also investigated the effect of water shortage (drought) on
235 plant VOC emissions. Similar to the effect of heat, we found that with monoterpenes
236 dominating the total emissions (>80%), the general emission composition of a pine did not
237 change much with drought (see Extended Data Fig. 6a). However, drought decreased the total
238 amount of emissions (Extended Data Fig. 6b). The magnitude of the response of VOC
239 emissions to drought was relatively insensitive compared to heat. We conclude that
240 decreasing VOC emissions by drought should affect the CCN activity of SOA in the opposite
241 way as heat, and the overall effect would be a smaller activated fraction and less cloud
242 droplets.”

243 *In the following paragraph, more than modeled values the reader is interested in actual*
244 *measurements ...not possible?*

245 **Response:**

246 We guess that the reviewer referred to the paragraph discussing Fig. 4. The aim here is to
247 assess how much the changes of hygroscopicity (κ) of biogenic SOA due to biotic stresses
248 found in our laboratory study can impact the CCN number concentration in the ambient -
249 from the microphysical point of view. Since besides hygroscopicity, also particle size affects
250 the CCN number concentration, the size distribution needs to be kept constant in order to
251 demonstrate the impact of changes of hygroscopicity (κ). Therefore, we derived the CCN
252 number concentration from a given constant size distribution using different κ values
253 representing different scenarios.

254 In field measurements, it is difficult to apportion the changes of particle size to specific
255 sources, e.g. induced emissions. One reason is that pre-existing aerosols in the field
256 measurement which also contribute to CCN number concentration can also vary, making it
257 difficult to specifically extract the effect of various stresses.

258 Admittedly, both the field measurement and model studies on CCN activity and number
259 concentration are important to understand the impact of stresses of plant on cloud formation
260 ability of biogenic SOA.

261 In the revised manuscript, we have added a brief discussion on this.

262 “Currently, there are no direct field measurements reporting the effects of biotic stress on the
263 CCN activity of biogenic SOA and CCN number concentrations, to our knowledge. Future
264 field measurements of CCN in periods when biotic stresses induced emissions are dominant
265 will help to assess the impact of biotic stresses on CCN activity and concentration.”

266 *Next paragraph: I agree this is an important study with important and interesting*
267 *information such the one on biotic stresses likely having significant influence on the CCN*
268 *concentration in areas where biogenic SOA components dominate particle formation..... but*
269 *this, by itself, is not completely new (this same research group itself has studied this for*
270 *long), not very robust, not very extensive, not wholly convincing, not with real measurements,*
271 *and specially with not much quantitative information, at least for a paper in a high profile*
272 *journal like this one. It seems instead very adequate for a good atmospheric journal.*

273 **Response:**

274 We would like to emphasize the novelty of this study with the background of previous
275 studies. Although there are multitude of studies on the effect of various stresses on VOC
276 emissions, there are only very few studies investigating the particle formation and SOA
277 properties formed from real plant VOC emissions including the studies of our group.
278 Importantly, no studies have directly investigated the CCN activation, the important
279 microphysical properties for climate, as a function of various stresses, to the best of our
280 knowledge.

281 Moreover, we guess the reviewer referred to field studies with the statement “not with real
282 measurements”. Our study is a laboratory study, which investigates the quantitative impacts
283 of various stresses under well-controlled conditions using real plants as representative,
284 holistic VOC sources. Our concept of laboratory studies is complementary to field studies as
285 it allows repeatedly investigating VOC mixtures from complex sources under well-controlled
286 physical conditions. (It is also complementary to classical laboratory studies as we use
287 complex real VOC sources rather than using single VOC or simple, artificial mixtures). We
288 studied a key quantity which is one prerequisite for vegetation-climate interactions: the cloud
289 droplet activation of the resulting SOA. In this sense we understand our manuscript - in its
290 inherent limitations- as also a trigger to promote more studies in the area including field
291 studies - a scope of Nature Communications. Our studies are necessary and helpful to direct
292 field studies, where many environmental factors (temperature, solar radiation intensity,
293 ambient oxidants concentrations, background aerosol type and concentrations etc.) affect the
294 VOC emission and CCN activation and the system is even more underdetermined.
295 Underdetermination and limited reproducibility make it extremely complicated to extract
296 quantitative effects of certain stresses on CCN activity and number concentrations. In
297 contrast, in well-controlled laboratory studies, we investigated the effects of certain stresses
298 systematically and quantitatively without interferences from other factors. We agree with the
299 reviewer in that we cannot and therefore did not, directly extrapolate our results to general
300 ambient cases. We did point out the principle importance based on the measurements, though.
301 This study clearly shows that environmental factors that affect plant emissions eventually
302 propagate all the way to CCN activity of SOA formed from these plant emissions and once
303 more highlights the importance of environmental conditions in the potential impact of
304 terrestrial vegetation on cloud formation and climate. An important conclusion from this
305 study is hence, that such effects should be included in climate models and are worth further

306 studies. We therefore debate that this makes our manuscript unsuited for publication in
307 Nature communications, especially as we now address the well taken major critics.

308 In addition, regarding “quantitative information”, we directly quantified for the first time
309 those parameters: changes of hygroscopicity (κ) of SOA due to insect infestation and heat.
310 These are the complete data set achievable in laboratories studies. And we further quantified
311 the effects of the biotic stresses on the CCN number concentrations in boreal forest areas
312 dominated by biogenic SOA. Admittedly, due to the complexity of the various environmental
313 stresses, “the overall impacts are complex and cannot be assessed quantitatively here” (as we
314 stated) or by any single study.

315 In the revised version, we clarified the manuscript, made the statistics more robust and
316 described our findings more quantitatively (see the changes based on each comments). We
317 wish that - given the clarifications and additional data - now the reviewer can kindly accept a
318 certain inherent lack of immediate generalization. This indeed will need field measurements,
319 which are beyond the scope of this study. Still our study represents an important step to better
320 understand these impacts.

321 *Figure 4 caption....why modeled and not measured values?*

322 **Response:**

323 Please see our responses to the similar question above (page 9, lines 243-244). The particle
324 size distribution was taken from our observations within the PEGASOS campaign near
325 Hyytiälä, Finland, in order to demonstrate the effect of our findings regarding size and κ to a
326 realistic situation. During this field campaign no CCN measurements were done.

327 *Experimental design-Extended figure 1. Why not using a single chamber where plants are in*
328 *contact with ozone as it in fact occurs in nature?*

329 **Response:**

330 There are three main reasons to use one plant chamber and one reaction chamber.

331 First, we need to determine VOC emissions from plants and initial concentrations of VOC for
332 SOA formation in the photochemical reaction. The initial VOC concentrations are essential to
333 determine the SOA yield (SOA mass formed per mass of reacted VOC). If there were only a
334 single chamber with plants and oxidants such as O_3 and OH together, neither real VOC
335 emissions nor the initial VOC concentrations in the reaction can be determined due to the
336 reaction loss with OH and O_3 .

337 Second, the UV light used in the laboratory setup to simulate photochemical reactions and
338 SOA formation can damage plants, which can confound the effects of the studied conditions
339 on plants.

340 Third, the plant chamber itself cannot provide the enough residence (reaction) time that is
341 needed for SOA formation and particle growth from small to large. In order to avoid water
342 vapor condensing caused by transpiration in the plant chamber, the flow rate needs to be
343 high. This reduces the residence time since residence time is inversely proportional to the
344 flow rates.

345 *Try to explain more clearly the caption of figure extended data figure 2.*

346 **Response:**

347 We have improved the caption to explain the caption more clearly. We have also added
348 another panel to explain the critical activation diameter and split previous panel b into two
349 panels to make it more clear.

350 *Extended data figure 4: I am still surprised of this absence of change with temperature, and
351 still would very much recommend having the same graph for the different monoterpene
352 compounds and the different sesquiterpene and others compounds since they have different
353 physic-chemical properties and temperature sensitivities.*

354 **Response:**

355 Please see our response to the similar question above (page 6, lines 172-174).

356 In the revised manuscript, we have added the explanation of this point and have accepted the
357 reviewer's suggestion. We have added one figure in the Methods part to show relative
358 fractions of individual compounds of monoterpenes, sesquiterpenes and others. While relative
359 ratios of certain compounds can change with temperature, those for other compounds remain
360 largely invariant with temperature. The overall VOC emission composition regarding the
361 relative contributions of monoterpenes, sesquiterpenes and others did not change much with
362 temperature.

363 *Extended data figure 6. As in most parts of the text I miss statistics here. How significant are
364 the changes? They do not seem very strong or significant.*

365 **Response:**

366 The detailed effects of biotic stress, heat and drought shown in this figure was calculated
367 using the results found in our laboratory study. Therefore, there are no error bars on it. We
368 apologize for not clearly explaining this. In the revised manuscript, we have clearly described
369 it.

370 This figure mainly works as one example of the typical effects of various stresses on CCN
371 number concentration and the exact values are not our focus since they may depend on many
372 parameters and hypothesis used. But the changes on CCN number concentration (indicated
373 by the color bar) shown here are strong. For the constitutive emissions, the heat or drought
374 cause a 27% increase or a 37% decrease in the CCN number concentration, respectively. The
375 biotic stress alone causes a 47% increase while biotic stress plus heat causes a 93% increase
376 compared to the reference case of constitutive emissions at room temperature (right lower
377 circle).

378 In the revised manuscript, we have also added the numbers of changes in the caption.

379

380 **References**

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The revisions to the manuscript were appropriate for the comments and suggestions that I (reviewer 1) made for the initial review.

Reviewer #2 (Remarks to the Author):

Although I am still surprised by the absence of changes in BVOC composition with warming and drought, I think the authors have satisfactorily answered our questions and solved our concerns. I think, as already said for the first version, this is an interesting paper showing how constitutive and induced BVOC emissions, and their changes due to stresses, affect SOA properties such as κ , and how they finally affect cloud formation. The authors have also interestingly concluded that decreasing VOC emissions by drought should affect the CCN activity of SOA in the opposite way as heat, resulting in less cloud droplets. Although the authors cannot directly extrapolate their results to general ambient cases, I agree their results are helpful to direct future field studies. They moreover highlight the importance of environmental conditions in the potential impact of terrestrial vegetation on cloud formation and climate and therefore the interest and need of such effects being included in climate models. I am happy to now recommend acceptance.

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Response:

We thank the two reviewers for carefully reviewing our manuscript again and giving the supportive remarks.