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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Biotic stress accelerates formation of climate-relevant aerosols in boreal forests" *by* J. Joutsensaari et al.

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We thank the referee for valuable comments. We have considered the comments and have improved accordingly the manuscript (MS). Our detailed responses for the referee's comments are bellow.

Referee's general comment: None of the elements are flawed and the limitations of the techniques are largely recognized appropriately. However, I find the paper is poorly linked, such that it appears as though it is two separate studies. This must be addressed before it is acceptable for ACP, either by making stronger, clearer linkages or by extending the individual aspects so as to make them suitable for separate publications.

Authors' response: The main aim of this study is to investigate the climate change feedback between increased herbivore outbreaks, SOA production, and CCN concentrations from a variety of perspectives to provide a broadened perspective of the potential magnitude of the effect. Consequently, this study summarizes the results from field and laboratory experiments, satellite observations and global scale modeling in order to get wider knowledge of the effects of insect herbivory and outbreaks on SOA formation and the Earth's climate. Not just present results from individual studies. We believe that this integrated interdisciplinary approach that evaluates plant biological, ecological and atmospheric processes concomitantly is novel and warrant. We think that one publication connecting individual aspect together is stronger than separate publications with individual aspects. Therefore, we have clarified the linkages between experimental studies, global scale modeling and satellite observations in order to meet the remarks presented by the referee.

Referee's specific comment: My main concern centres on the lack of linkage between the treatment of SOA formation in the GLOMAP model and the experimental evidence. In only using an enhancement in SOA-precursor emission rates resulting from insect attack, the modelling is isolated from the experiments. It is stated that "for computational affordability, [the authors have] made a simplifying assumption that 13% of [the] oxidation products [of MT] form vapours capable of producing SOA". First, this approach omits any SOA formed from SQT. This may be a reasonable approximation in other GLOMAP applications, but in the context of the current paper where substantial enhancements in SQT are reported from insect attack, this appears over simplistic. The laboratory and field SOA enhancements will undoubtedly include the contributions from such enhancements. The previous demonstration of GLOMAP's ability to respond to increased VOC emissions are based on the MT yield of SOA precursors. Furthermore, scaling the GEIA emissions database of MT emissions to include SQT must assume a constant relationship between SQT emissions and MT and between enhancements of MT and enhancements of SQT. This is clearly not the case from the field measurements reported here, nor from previous studies. Moreover, the omission of stress-responders other than MT or SQT may lead to very significant omission of SOA response. I cannot see a justification for an assumption that a simple scaling of MT emissions will represent a response that can be confidently assumed order of magnitude representative of the measurements.

Authors' response, changes in MS: We have recognized the lack of linkage between the laboratory and field experiments and the GLOMAP modeling. However, at this point in time we do not believe there is sufficient experimental data on SOA yields from plant stress compounds to incorporate the results into a global model in a meaningful way. However, this is a first attempt at global modelling of the phenomenon that serves as a useful illustration of the possible scale of its global consequences. In all likelihood, the results presented here would actually be a conservative lower limit of the potential impacts of herbivore outbreaks because increasing sesquiterpenes would only increase the observed effect. We do agree with the reviewer, however, that a final word on this matter will need much more detailed investigation in the future – this is not, however, in the scope of this study.

The technical reason for omitting sesquiterpene from our simulations was that suitable spatially resolved emission datasets at the global scale were not available when the simulations were run. While such datasets have since emerged, we decided not to rerun the model because 1) of its extensive computational expense, and 2) inclusion of sesquiterpenes would not impact our (rather general) conclusion that the insect herbivory can affect aerosol forcing on a regional scale. This decision still seems valid in the light of recent research findings: For example, D'Andrea et al. (2015) estimated that in the boreal forest region monoterpenes are responsible for up to over 80% of SOA formation, while sesquiterpenes play a much less significant role (always below 20%, but in many areas below 10% contribution to SOA).

However, the reviewer comments make it evident that the original manuscript did a poor job communicating the motivation and aim of the GLOMAP model runs. To remedy this, we have modified the text in the following way:

Section 2.3, first sentence: "An evaluation of the significance of insect damage on atmospheric boreal aerosol was obtained with a global chemical transport model, GLOMAP, to provide a first estimate of the potential scale of the impact.

Section 3.3, first paragraph: "We used a simplified SOA formation set-up within the GLOMAP model to assess whether the large-scale insect outbreaks could potentially impact Earth's climate. The model set-up is summarized in Fig. 6a: 10 % of the total boreal conifer forest suffered insect herbivory with a 10-fold increase in monoterpene emissions in outbreak areas. Note that the simulations do not include sesquiterpene emissions, as they are not incorporated in the model version used here. Thus the results presented here likely represent a conservative lower-bound of the potential impact as any increases in sesquiterpene emissions would serve to increase SOA yields even more dramatically. Furthermore, D'Andrea et al. (2015) have recently estimated that in the boreal forest region monoterpenes are typically responsible for up to over 80% of SOA formation, while sesquiterpenes play a much less significant role. We do not expect this short-coming to impact our conclusions, which are intended to merely indicate whether insect herbivory could be of regional importance, and thus merit more detailed model studies in the future. A 10-fold increase in monoterpene emissions in 10 % of the total boreal area are conservative estimates — "

Referee's specific comment: There needs to be some clarification of the GLOMAP mechanistic treatment. Is a 13% yield of all oxidation products assumed to be SOA-forming (i.e. from both OH and O3)? If so, what is the justification of the scaling given the enhanced SOA yields were based on O3-initiation only? Unless the yield is from O3-initiation alone, I can only see this as further disconnect between the modelling and measurements.

Authors' response: We are not quite certain what the reviewer means here. The fixed 13% yield of SOA forming compounds from monoterpene oxidation is used both for OH and O3 routes in all areas (i.e. both in insect damaged and background areas). We have based our scheme on the observation that insect damage significantly enhances the monoterpene *emissions*, not on the observation that it enhances SOA yields due to O3 pathway (although that of course is true too). Overall, higher emissions lead to higher monoterpene concentrations, which then can react further both via OH and O3 pathways; therefore, the assumption of higher concentration of SOA forming oxidation products also via the OH route is not unrealistic. We further stress that the yield (13%) stays the same for both oxidation pathways, and in all simulated areas.

Referee's general comment: In summary, I don't see the clear justification for the assumption that the increase in CCN and SOA mass exhibited in GLOMAP for a given increase in MT bears a close relationship to the phenomenon observed in the measurements. Given the disconnect, I would have probably combined the GLOMAP and satellite components into one phenomenological paper (abstract of which is line 16 onwards of the current abstract) and expand the field and lab experiments into a more quantitative mechanistic paper (abstract lines 10-16 of the current abstract).

Authors' response: As mentioned earlier, we believe that one publication including different perspectives to investigate the same question is stronger than separate publications since it evaluates the question across scales - from field and chamber studies to global scale phenomena. We acknowledge that connection between experimental and modeling studies could be stronger. We wish to stress again that the global simulations were not intended as a definitive quantification of the effect of biotic stress on boreal atmospheric aerosol but merely as a first-order approximation to demonstrate that this phenomenon may be of regional importance and thus merits further study.

Referee's specific comment: If all elements of the paper are to remain, the following should be considered: i) the coherence between the model and measurements should include results from OH initiated oxidation in the chamber experiments or eliminate the OH-initiated enhancement in condensable vapour concentration in GLOMAP or make a convincing argument for some correspondence between them.

Authors' response: In our chamber experiments, ozonolysis of terpenes included also

OH induced oxidation because we did not use any OH scavenger which inhibits formation of OH via ozonolysis of VOCs (see Hao et al, 2009). Therefore, we do not believe that chamber experiments with only OH initiated oxidation or the elimination of OH pathways in GLOMAP could change our main conclusions. As stated earlier, GLOMAP model was used to obtain the order-of-magnitude estimate of the potential significance of insect damage on atmospheric boreal aerosol, not to analyze in detail the effect of different SOA formation mechanism.

Referee's specific comment: ii) the extrapolation of chamber yields to models is fraught with danger and it is understandable that the authors have avoided making a linkage. However, some discussion about the relationship between the 13% yield of condensable vapours in the model and the measured SOA yields should be included. This raises a number of points. The authors recognise the effect that walls can have on leading to underestimates. However, the yields from different chambers under different conditions also give wildly differing values, even for single precursors, which need to be mentioned.

Authors' response: We note that extrapolation of chamber yields to models is not straightforward. "SOA yield" as calculated in a chamber experiment is not necessarily meaningful for global modeling applications that are intended to represent an ambient environment. Yields are affected by a complex suite of variables including, but certainly not limited to, precursor concentration (Kang et al., 2011; Kroll et al., 2008; Pfaffenberger et al., 2013; Presto and Donahue, 2006), seed particle composition/loading (Ehn et al., 2014; Hamilton et al., 2011), and mass of absorbing organic aerosol material present in the chamber (Pankow, 1994; Odum et al., 1996). Thus, while it is potentially useful to compare SOA yield curves (yield vs. organic aerosol loading) between different laboratory experiments to gain insight into differences between the SOA formation behavior of the precursor BVOC profiles, it is not meaningful to arbitrarily select a single SOA yield number from those curves to use in a global modeling application. This is why we used a more simplified approach for the global modeling application where an SOA yield value of 13% was used for biogenic emissions as is common practice in global modeling applications. In this study, average SOA mass yields vary between 0.1–3% in control experiments and 5–40% in insect-damage experiments (overall averages 1% and 18%, respectively). The yield used is line with our insect-damage experiments but higher than obtained from control experiments, which might overestimate SOA formation in low organic mass (control) areas. However, this does not change our main conclusions.

We also note that differences in chambers, conditions, precursors, etc. have an effect on yields. However, we think that yields determined form the emissions of real boreal forest trees are more representative than from single component (e.g. a-pinene) experiments. We have included discussion on the relationship between model and experimental yields in the revived version of MS (Chapter 3.2).

We have already discussed (page 10868) the effect of total organic mass loading and wall losses on SOA mass yields as well as compared previous studies on boreal trees. To revised versions of manuscript, we added a general note on the variation of yields depending on chambers, conditions and precursors (Chapter 3.2).

Authors' changes in MS: We have added the following discussion at the end of paragraph on SOA mass yields (Chapter 3.2): "Furthermore, the SOA mass yields determined from different chambers under different conditions can vary widely (Lee et al., 2006; Shilling et al., 2008; Mentel et al., 2013; Hao et al., 2011) and therefore it is not straightforward to assess a suitable yield values for model calculations. In this study, we have used a fixed 13 % yield (all cases) in the GLOMAP modeling as is common established practice for regional to global scale modeling applications. This value is also consistent with our insect-damage experiments (overall averages 18%). This fixed yield value might overestimate SOA formation in control areas with lower VOC and SOA mass concentrations, so the potential effect observed here would be a lower estimate of the potential effect."

Referee's specific comment: Ozonolysis experiments are very susceptible to the relationship between the position and timing of introduction of ozone into chamber and relationship to the measurement point. A description of the mixing of the plant emissions with the ozone rich air and the methodology for SOA sampling in the chamber should be included.

Authors' response, changes in MS: The schematic of ozonolysis experiments is shown in Fig. 1 and described on page 10859. At the inlet of the reactor, air flow from the seedling headspace (2 L/min) was mixed (in T-fitting) with an ozone-enriched air flow (total flow 17 l/min). O3 concentrations were measured from main air flow at the inlet and outlet of the reactor. Aerosol samples were collected from the main air flow at the outlet of reactor (see Fig. 1). We have clarified those points in the revised MS (Ch. 2.2.1 and 2.2.3). We have also chanced O3,inlet values in Table 1, now the dilution due to plant chamber air flow was taken into account (values now ca. 10 % lower).

Referee's specific comment: Chamber experiments using mixtures are more complicated than using single precursors. Since the timescales for oxidation of MT and SQT will be very different, the concept of yield for mixtures is problematic and the way that dilution of a mixture in the real atmosphere occurs will mean that yields do not combine in any way similar to the way they combine in a chamber (and in neither case will they be linear). The method for calculating yield (as plotted in Figure 5) should be described. In reality it will be a combination of sequential yields, the most reactive first. Since ozonolysis of SQT gives significant OH yields, the subsequent SOA formation of less reactive precursors will include some OH-initiation (helping the linkage between the modelling and experiments a little, but not sufficiently, since it will be so different to the atmosphere) and depend on the relative reaction rates of the remaining mixture with different oxidants.

Authors' response: We note that experiments with VOC mixtures are more complicated (e.g. complicated reaction chemistry) than pure compound experiments. Although, timescales for oxidation of different compound will be very different and subsequent reactions occur, we believe that SOA mass yields determined form real plant emissions are more representative than those derived from single component (e.g. a-pinene) experiments since multi component system in closer atmospheric situations than one component system. The method to calculate yield is described on the page 10861.

Referee's general comment: I may have missed the point of the paper, but the above queries should be addressed before it is suitable for publication in ACP. Alternatively, the point of linking the experimental and modelling elements of the paper should be much more clearly made.

Authors' response: Briefly, the main point of the papers is that more frequent insect outbreaks in a warming climate could result in substantial increase in biogenic SOA formation in the boreal zone and, thus, affect aerosol forcing of climate at regional scales. This should be considered in future climate predictions.

Authors' general remarks: To study comprehensively the effect of insect outbreaks on global scale, very time consuming and intensive studies in different areas are needed, e.g., laboratory and field experiments with different tree species to evaluate effects of biotic and abiotic stress on VOC emissions and SOA formation, updating VOC emission models in global aerosol and climate models (e.g. MEGAN) based on field and laboratory results, aerosol and global scale modeling, etc. We think that this would require a several-year-long project to concern all aspect in detail. Ultimately, this manuscript demonstrates that across a range of scales (laboratory experiments to regional modeling) and across a variety of investigation methods (laboratory and field measurements, GLOMAP modeling, satellite data products) the potential impact of herbivore outbreaks on SOA formation is significant even when the assumptions used would arguably result in a conservative, lower-bound estimate of the effect. This study provides a strong rationale prompting further investigation, and highlights the importance of research on this biosphere-atmosphere climate change feedback.

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