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Comment

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

Anonymous Referee #2

Received and published: 31 May 2015

This manuscript presents an extension and consolidation of previous work by the authors and builds on other recent efforts by others to demonstrate the potentially significant contribution of SOA formed from the enhancement of VOCs resulting from biotic stressing of boreal tree species. The authors use a combination of laboratory, field and modelling studies to corroborate their findings and make an argument that this aspect of VOC-mediated biogeochemical feedback warrants substantial further investigation. The implications in terms of future climate scenarios with increased incidence and changing geographical distribution of insect outbreaks are hitherto poorly understood.

I have few comments to make about the detailed methodology of the chamber experiments, the plant treatment, VOC measurements, the generality of the GLOMAP modelling or the satellite retrieval of AOD beyond the relatively trivial minor points below.

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None of the elements are flawed and the limitations of the techniques are largely recognised 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.

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.

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

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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.

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).

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.

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. 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. 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

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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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10853, 2015.

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