

Reviewer 1

The study "The effect of spruce bark beetle on BVOC, secondary organic aerosol and ozone: Integrating biotic stress in a chemical transport model" investigates the effect of herbivore feeding on Spruce VOC blends and integrates them into a global VOC model. Therefore, this is a very important study to improve our general understanding of the complexities of how biogenic VOCS are produced and how they affect other important pieces of the atmospheric system, such as secondary organic aerosols, clouds and ozone concentration.

Author response: We thank the reviewer for the positive review of our manuscript and for the constructive comments and suggestions to improve it. We have modified the manuscript accordingly as outlined below. In the following, the reviewers' comments are marked in black and our answers in blue. Amendments to the manuscript are highlighted by square brackets and italics writing.

Reviewer comment: General comments: I am not an expert in modelling, but I think that the study is impressive in its ambitions to parametrize not only the emissions from spruce bark, but more importantly adjust them to the biotic stress of beetle herbivory. Truly a great addition to large VOC models, such as COSMO-MUSCAT. Maybe it can be adjusted for other models, one day, such as MEGAN for example?

I find that the manuscript is very well written and mostly ready for publication in Atmospheric Chemistry and Physics.

I have only minor comments for revision.

Author response: Thank you very much for your helpful comments and for sharing additional research on herbivore insect infestations and its effects on VOC and SOA. Your comments helped us to improve the paper, and increase its value for a wider range of readers.

Specific comments:

Reviewer comment: L17: I would recommend saying "an increasing strain" rather than "increasingly strain".

Reviewer comment: L46: I think you mean "threat", instead of "thread". Please correct.

Author response: We included both corrections in the revised manuscript.

Reviewer comment: L68: Hydroperoxyl radicals or hydroxyperoxy radicals? In L 134 it's spelled differently.

Author response: HO₂ radicals are hydroperoxyl radicals, the spelling was corrected.

Reviewer comment: L80: I don't disagree with you here, but please check the new article by Yu et al. 2024 (<https://doi.org/10.1126/science.ado6779>). It might provide some important context here in regards to SOAs formed from BVOCs from evergreen needle trees induced by bark beetle herbivory.

Author response: Thank you for drawing our attention to this very interesting article! We included this new information of SOA also being a stress-signaling ‘tool’ of vegetation in line 24-27:

[‘Further, also the SOA produced from specific BVOC compositions, carries the information of approaching disturbances and can thus be transported over a larger distance (Yu et al. 2024)’].

Reviewer comment: L125: Does hv in R2 stand for solar radiation? If so, maybe you can spell it out in full? There should be enough space. What does M stand for?

Author response: The hv represents the energy of incoming photons i.e. solar radiation which causes then photooxidation. M is usually a molecule like N₂ or O₂ that takes up excess energy of the reacting molecules through collision. A short description for M and hv is added in line 140-138:

[‘Here, M is a molecule absorbing excess energy (e.g. N₂), and hv represents the light intensity of solar radiation’]

Reviewer comment: Equation 4: Have you seen the study by Ghimre et al. (2016; <https://doi.org/10.1016/j.atmosenv.2015.11.049>)? They have measured the emissions of bark VOCs from spruce directly. Does your model match the emissions from that study? I feel like it would be an important study to at least mention in your work, seeing as closely related they are in scope and intention.

Author response: Thank you for pointing at this important study on the emissions of attacked spruces. The development of our parametrization and the applied standardized emission rates follow the findings of Jaakkola et al. 2023, who also compared their results to those of Ghimire et al. 2016. The measurements of Jaakkola et al. are not straightforwardly comparable to those of Ghimire et al. 2016 due to different approaches and insufficient information on the stem temperature. Still, we are aware that the measurements by Jaakkola et al. 2023 are by a manifold higher than those of Ghimire et al. 2016, we therefore included a discussion in the revised manuscript. We are aware of the limitations of including data from a single measurement campaign, based on site-specific conditions.

A short discussion was introduced at line 234-239:

[‘The applied standardized emission rates are 3 to 9 times higher than results from an earlier study by Ghimire et al., 2016 who performed measurements in southern Finland. One source of

uncertainty is the timing of the measurement during the infestation and the age of the entrance holes (Jaakkola et al., 2023). For detailed discussion the reader is referred to Jaakkola et al. 2023. Standardized emission rates derived from measurements at different sites can strongly vary in magnitude, with possible influences by age of trees, genetic variations, general health status and timing within the season of the measurement (Hakola et al., 2023).']

Reviewer comment: L216: Please provide any citation on the claim that isoprene cannot be emitted from or stored in the bark.

Author response: added a citation and further context in line 238-241:

['The contribution of the isoprene from the stem is unclear, as isoprene emission is a light-dependent process and isoprene is typically not stored in a tree trunk (Jaakkola et al. 2023). However, elevated isoprene was found during measurements by Jaakkola et al. 2023 and Ghimire et al. 2016, and is thus included for completion ($ERS_{i, trunk, S}$).']

Reviewer comment: Figure 2: Please spell out the meaning of δSIE in the figure description for an interpretation that is more independent from the main text.

Table 2: Similarly, please spell out the SIE.

Author response: The table 2 was revised substantially in response to the second reviewer. In this context also abbreviations were spelled out to facilitate its readability.

Reviewer comment: L260: Please replace "Scandinavia" with "Fennoscandia", as Finland is clearly included in the regional D0 domain. Also applies to rest of the text, for example

Author response: Thank you for pointing this out to me, I changed Scandinavia to Fennoscandia as it has clearly high shares of spruces and is represented on the continental N0 domain.

Reviewer comment: Figure 3: It's a nice figure, but if you are presenting % in the right-hand Y-axis, I think it could be actual percentages, rather than the decimal numbers. Because technically it's showing now between 0 and 1%, rather than from 0 - 100% (which I assume was your intention, judging by the density of evergreen forests in northern Europe). Please correct this.

Author response: Thank you for pointing at this mistake, the colorbar has been revised and the borders have been removed (see next comment).

Reviewer comment: Figure 3, 4, etc: Please redraw all maps in your manuscript without country borders. This is a clear instruction from the journal guidelines - "Please adhere to United Nations naming conventions for maps used in your manuscript. In order to depoliticize scientific articles, authors should avoid the drawing of borders or use of contested topographical names."

Author response: Thank you for bringing this to our attention. We removed the borders on all maps.

Reviewer comment: L300: All at the same time or not all at the same time? I'm a little bit confused here.

Author response: The study assumes that 46% of the spruce trees across the European continent are infested all at the same time, which is not representing local differences in bark beetle infestation. The intention of the study is to show the theoretical impact of a large scale herbivore infestation with a focus on the atmospheric chemistry i.e. SOA and ozone.

Adapted passage starting in line 339-342:

[‘The assumed peak emission induced by bark beetle stress occurs in August and affects 46 % of the spruces across Europe all at the same time. The share of 46 % results from the German forest inventory report and represents the share of all significantly defoliated spruces in 2021, despite the origin of stress. Using this high fraction for synchronized spruce infestation leads to regionally overestimated stress effects. The actual spatial extent of bark beetle infestation in 2021 might differ from our simulated event.’]

Reviewer comment: L323: Heights? Do you mean elevation?

Author response: The sentence is brought into more context line 366-367:

[‘The biogenic module of COSMO-MUSCAT does not account for different heights of trees or shrubs as BVOC emission sources.’]

Reviewer comment: Figure 5: Please spell out MTPs here as well.

Author response: was adopted

Reviewer comment: L339: You might be interested in how the bark-beetle induced VOC emission from evergreen trees actually responds to outdoor environments with elevated levels of NO_x or O₃. Does it compare to your predictions? (<https://doi.org/10.1038/s43247-025-03175-3>)

Author response: Thank you for sharing this really interesting study. Our aim was to investigate the effect of biotic stress induced emissions on air quality. So far, our model framework does not contain the impact of pollutants on VOC emissions, however, this is part of future studies.

Reviewer comment: Figure 7: Could you please explain again here what do N0 and D0 domain mean, so that the reader does not need to go back to the main text to see?

Author response: Descriptions of the grid resolution of the domains was now included in the respective figure description.

Reviewer comment: L398: Can you please provide a reference for different SOA formation from BVOCs and AVOCs?

Author response: The sentence was written in a misunderstandable way. The intention here was not to differentiate between reaction pathways of anthropogenic and biogenic VOC, but rather all VOC species follow different reaction pathways.

The passage was rewritten in order to clarify the motivation at line 448-452:

['Different VOCs from anthropogenic and biogenic sources exhibit varying oxidation rates by O_3 , OH, and NO_3 , resulting in the formation of different lower-volatility products that partition into the particle phase. Therefore, the resulting SOA composition depends on the VOC precursor composition and the prevailing oxidant regime. ']

Reviewer comment: L434: Recent findings show that SOAs are also an effective communication tool for plants, especially for evergreen trees (<https://doi.org/10.1126/science.ado6779>). I feel like this is important to consider and acknowledge.

Author response: The passage was adopted in line 490-492 as follows:

['The SOA formed during stressed conditions can transport information of herbivore infestation and approaching disturbances to more distant trees throughout atmosphere, which leads to BVOC composition adaptations (Yu et al. 2024). ']

Reviewer comment: L463: One thing to consider here is that SQTs can act as repellents for bark beetles, it affects bark palatability (<https://doi.org/10.1038/s43247-025-03175-3>). So maybe this is an important aspect that is not being modelled in COSMO-MUSCAT and that's why you see the difference. SQT emission has been shown to be negatively correlated with beetle damage. I think that SQT emission is intrinsically tied to the biotic stress factor.

Authors response: In our model framework the SQT emissions are modeled similar as the MTP emissions to increase with infestation. Indeed, it is an interesting finding that sesquiterpenes are an effective repellent against bark beetles and can lead to reduced tree damage.

Included in line 560-562:

['Despite their low emission rate, SQT are an important factor in plant protection processes (Simin et al. 2026) and atmospheric chemistry (Bourtsoukidis et al. 2012, Bourtsoukidis et al. 2018). ']

In regard to the parametrization of the VOCs, it is known that COSMO-MUSCAT has shortcomings in the sesquiterpene representation. This would have exceeded this study and is planned to be part of future studies.