

Reviewer 2

Reviewer comment: The paper makes a useful methodological contribution by incorporating species-specific, bark-beetle-induced BVOC emissions into COSMO-MUSCAT and explicitly separating trunk emissions from standard canopy/needle emissions. The simulations illustrate how an additional stress-related source can alter the diurnal cycle of monoterpenes and affect atmospheric composition. However, the highly idealized infestation scenario should not be interpreted as an estimate of the actual atmospheric effects of the 2021 European bark-beetle outbreak. The main quantitative conclusions depend on several substantial assumptions whose uncertainties have not been adequately evaluated through sensitivity experiments. My comments focus primarily on the emission calculations. I recommend **major revision**. I have a few concerns of the emission calculations

Author response: We thank the reviewer for taking the time and effort spent on our manuscript and for the valuable remarks and suggestions. 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 in italics.

The assumptions made for the modeling study are put further into perspective in order to make clear that we did not intend to simulate the real 2021 bark beetle infestation. Our study analyses the effect on SOA and O₃ of a hypothetical intensive spruce bark beetle infestation. For a realistic representation of the actual bark beetle infestation further model developments as well as detailed available data on bark beetle observations are necessary. The discussion of the limitations was extended accordingly in section 5. In the following we address your comments and recommendations in detail.

Reviewer comment: The emission rates adopted from Jaakkola et al. (2023) represent total bark emissions measured from infested trees, rather than the additional emission above the healthy-bark baseline. Consequently, in Eqs. 3–4, adding the healthy-trunk term to the full infested-trunk emission rate appears to double count constitutive bark emissions for the infested fraction. Moreover, the formulation does not explicitly distinguish the trunk area associated with healthy and infested trees. f_{trunk} has been multiplied by both healthy and induced trunk emissions. I think Equation 4 is conceptually incorrect and should be revised. Although the authors may think that emissions from healthy trunks are much smaller, this does not justify double-counting emissions.

Author response: We agree that the current emission formulation double-counts the healthy trunk emissions. The healthy trunk emissions could be subtracted from the share of the infested spruces. The presented standardized emission rate for monoterpenes in Jaakkola et al. 2023 is 6600 with a standard deviation of 6700 [$\mu\text{g m}^{-2} \text{h}^{-1}$]. The standardized emission rate for healthy stem emissions is 29 with a standard deviation of 51 [$\mu\text{g m}^{-2} \text{h}^{-1}$]. Thus, the healthy emissions are within the standard deviation of the infested. We included a figure and corresponding

discussion in the revised manuscript (lines 513-534, Fig. 10), showing the healthy stem emissions and stressed stem emissions. It becomes visible that the healthy trunk emissions play a minor role in the overall monoterpene emissions. The addition of MTP emissions for healthy stem emissions is in peak less than 1% of the stress-induced emissions. Correcting the presented emission formulation would not change the overall result (SOA: + 55% and O3: ~1-3%).

The response concerning f_{trunk} within this comment is compiled together with the response to the next reviewer's comment.

Reviewer comment: In Eq 4, the derivation of f_{trunk} is insufficiently documented. As described by authors as the ratio between average lateral trunk surface area and the area covered by one tree; converting this ratio cannot get the emission rates to the per ground surface area?. A clarification is needed. Please specify the tree dimensions, geometric assumptions and also the area covered by one tree (crown area or stem area or stand area?)

Author response: We thank the reviewer for raising this point. The land-use input data in COSMO-MUSCAT differentiates the vegetation species and land-use types by using their fraction of grid cell area ($f_{\text{canopy},i}$), rather than concrete areal measures. The stem surface area fraction is calculated from the multiplication of $f_{\text{canopy},i}$ and $f_{\text{trunk},i}$ ($f_{\text{trunk},i}$ = stem area per crown coverage of one tree). The $f_{\text{trunk},i}$ is the ratio calculated from average spruce coverage, height and stem radius, presented in the forest inventory from 2012 (Sachsenforst, 2014). To clarify the meaning of $f_{\text{trunk},i}$, we added a detailed description of the derivation in the appendix and added further explanations within the text.

Line 224-228:

[‘Considering $E_{\text{trunk},i}$ requires the introduction of the stem surface area fraction ($f_{\text{canopy}} \cdot f_{\text{trunk},i}$) of a tree species i . $f_{\text{trunk},i}$ is the ratio of the stem lateral surface area to the area covered by the crown of one average tree calculated from Norway spruce inventory data. By multiplying $f_{\text{trunk},i}$ with $f_{\text{canopy},i}$, the share of the vegetation within a grid cell is scaled to stem surface. f_{canopy} represents the fraction covered by the area of the crown of a specific tree species within a grid cell, whereby the fraction of stem surface area in a grid cell is calculated.’]

The extent of the bark beetle infestation on the spruces is represented by another ratio (a_{stress}) in equation 6. The stressed emissions are only calculated for the share defined with a_{stress} , which is in our case 0.46. Thus, at peak infestation in August, 46% of the stem area of Norway spruces are infested.

Reviewer comment: Table 2 reports the total monoterpene emission rate from Jaakkola et al. (2023), but provides emission rates and fractions for only selected compounds. It is therefore unclear how the complete measured monoterpene mixture is represented in the model. Are the unlisted compounds omitted or assigned to lumped chemical species? **Please provide a complete mapping from the measured compounds to the model species, demonstrate conservation of total monoterpene emissions, and justify the surrogate reaction rates and SOA yields.** This is particularly important because the conclusions concern oxidation pathways and SOA formation, which depend on emission composition as well as total emission magnitude.

Author response: We appreciate the reviewers point regarding Table 2 and the clarification of lumped BVOC species. The table is indeed not well understandable and required further clarification. We revised the table and its header for better understanding in line XXXX.

Table 2. Applied standardized emission rates (ERS) [$\mu\text{gm}^{-2}\text{h}^{-1}$] for Norway spruce within the stress parametrization adopted from Jaakkola et al. (2023) and emission potentials (EP) applied in the standard MUSCAT set-up (Steinbrecher et al., 2009).

BVOC	ERS healthy trunk [$\mu\text{gm}^{-2}\text{h}^{-1}$]	ERS stress-induced [$\mu\text{gm}^{-2}\text{h}^{-1}$]	EP reference [$\mu\text{gg}_{\text{DW}}^{-1}\text{h}^{-1}$]
Monoterpenes	29±51	6600 ±6700	synthesis 2.1 pool 0.4
Sesquiterpenes	2.1±3.2	53±7	0.1
Isoprene	0.4±0.9	3±7	1.0

In COSMO-MUSCAT, first the total monoterpene emissions are calculated and afterwards the emissions are divided into lumped species. Lumping VOC species in chemical transport models is a state-of-the-art approach, which is among others necessary to limit calculation time. The lumped species in COSMO-MUSCAT are API, LIM, BARO and BHC8. API includes α -pinene, β -pinene, camphene, Δ^3 -carene, α -thujene.

LIM includes limonene, α - and γ -terpinenes, α - and β -phellandrenes, myrcene, trans- and cis-ocimene, and linalool. BARO is equal to p-cymene and BHC8 represents 1,8 cineol (Stockwell et al. 1997, Luttkus et al. 2022). To make the comparison between the scenarios more robust, we did not change the assignment or the fractions. We are aware that this lumping might produce shortcomings when compared to reality as discussed by Luttkus et al. 2024a, 2024b and will be further addressed in future studies.

The description of the lumped BVOC species clarifies also the technical assignment to SOA species. The SOA module was not changed within this study. For a detailed description of relevant gas phase SOA compounds, reaction rates and its partitioning into particle phase, we kindly refer you to the work by Luttkus et al. 2022.

To clarify the treatment of monoterpene species we added in line 278-284:

[‘In the model framework ERS for Norway spruce for total MTP and SQT are adopted from Jaakkola et al. (2023) (see Table 2). For a differentiation of the MTP species, the total MTP emissions are split into lumped species classes, with 83% assigned to API (including α -pinene, β -pinene, camphene, Δ^3 -carene, α -thujene) and 10 % to LIM (including limonene, α - and γ -terpinenes, α - and β -phellandrenes, myrcene, trans- and cis-ocimene, and linalool). The splitting factor is held constant between the reference and the stress scenario for a more robust comparison, although BVOC compositions may differ between healthy and stressed conditions in reality. The lumping is described in section 2.2. Norway spruce specific parameters and empirical constants are presented in Table A1’]

We added the description of the lumped modeled BVOC species in the manuscript in line 175-179 within the description of the SOA module in section 2.2, to also clarify the assignments here.

[‘The modeled MTP species are divided and lumped into four groups. Lumped API includes α -pinene, β -pinene, camphene, δ^3 -carene, α -thujene. LIM lumps limonene, α - and γ -terpinenes, α - and β -phellandrenes, myrcene, trans- and cis-ocimene, and linalool (Stockwell et al., 1997; Luttkus et al., 2022). Two smaller groups are BHC8 (1,8 cineol) and BARO (p-cymene) (Luttkus et al., 2022). Sesquiterpene species are not differentiated in the model and lumped in the SQT class. ’]

Reviewer comment: The reported monoterpene emission rates of 6600 +-6700 ug/m²/h exhibits substantial variability. Applying part of this single mean value to infested spruce throughout Europe does worry me about the representativeness of the outputs. As a minimum, the author should have performed sensitivity analyses covering plausible emission magnitudes and composition and present the results as a range of experiments with different emission magnitudes and clearly state the uncertainty when interpreting the quantitative atmospheric results.

Author response: We agree that the reported monoterpene emission rate by Jaakkola et al. 2023 shows a strong variability. The reported value of 6600 $\mu\text{g m}^{-2} \text{h}^{-1}$ is the median of infested samples in the early infestation stage. The standardized emission rates range from 170 to 13500 $\mu\text{g m}^{-2} \text{h}^{-1}$, and were found to be lower than the measured emission rate. Measurements of BVOC emissions from infested Norway spruce are scarce and lack the feasibility of direct comparison. This was added in line 233-236:

[‘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. 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 and genetic of the tree, general health status and timing within the season of the measurement (Hakola et al., 2023). ’]

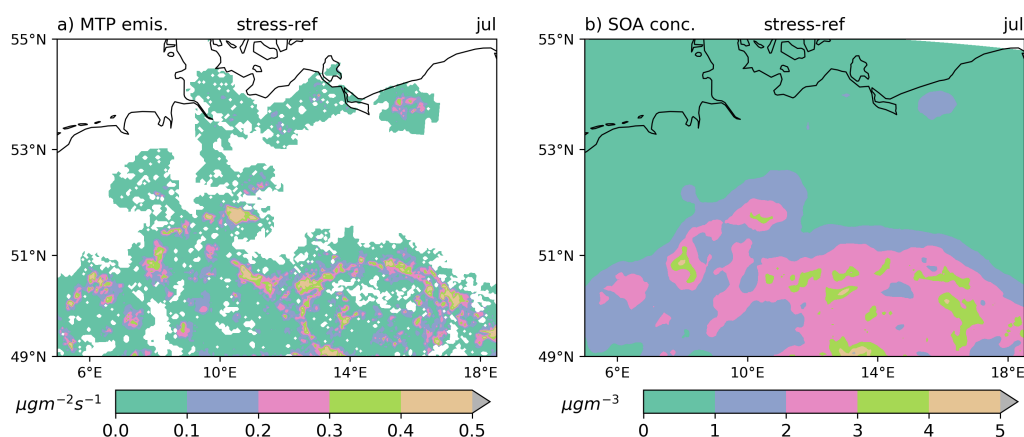
The focus of our study was to investigate the potential effects of additional BVOC emissions associated with bark beetle infestation on atmospheric chemistry. Throughout the manuscript we clarified, that the study is not representative for the real bark beetle infestation situation in 2021. The abstract was revised, clarifying the goal of the study. Further we added clarifications during the description of the parametrization in line 275-278:

[‘At the peak in August, 46~\% of the stem area of spruces is affected within the model. This assumption is likely beyond the real bark beetle infestation in 2021 for whole Europe, and might lead to regional and location-specific overestimation. However, a detailed inventory of the infestation extent and timing was not available. ’]

And in Line 339-342:

[‘Using this high fraction for synchronized spruce infestation leads to regionally overestimated stress effects. The actual spatial extent of the bark beetle infestation in 2021 might differ from our simulated event. Our study investigates the potential impacts on air quality of an upper-bound scenario associated with spruce bark beetle-induced emissions, based on a seasonal development curve of beetle population.’]

The increase in MTP emissions from reference to stress on the regional D0 domain is 44% in July (see Table C1 in Appendix), at the same time SOA increases on the domain average by 44% (see Table C2 in Appendix). Changing the emission potential by a certain factor would reduce or increase the effect on SOA in a similar range for larger regions. This can be seen in the conducted simulations already, where there are regions with differently strong increases in BVOC emissions (due to different spruce abundance), but similarly strong increases in SOA. The figure below shows this for the regional domain. Due to transportation, the distribution of the SOA is more homogenous, compared to the monoterpene emissions.



We further discussed the likely overestimation due to assumptions in the extent of the bark beetle infestation and the applied emission rates.

Line 612- 614:

[‘On the regional domain average, an increase of MTP emissions by 44 % increases SOA by the same percentage (44 %) in July. Locally, in spruce-abundant areas as Frydlant and Kosetice, SOA more than doubles, with doubled MTP emissions. As discussed before, the bark beetle scenario likely overestimates effects regionally and inherits uncertainties concerning the temperature dependency, however, a substantial increase in the BVOC emissions is nearly translated into the same increase in SOA concentrations.’]

We are aware that the current scenario represents an upper bound scenario on the regional / continental scale, but it is representative on the local scale with a strong infestation, although the exact locations of such infestation events are not available for all of Europe.

Line 495- 499

[‘An upper-bound scenario was simulated to estimate the potential effects of bark beetle infestation on atmospheric chemistry. The simulation does not replicate real bark beetle infestation in 2021, which requires further constraining by real infestation data. The current scenario likely overestimates on regional and continental scale, however, it is representative for locations with a strong infestation, although the exact locations of such infestations are not available for whole Europe.’]

and in the conclusion in line 652- 654:

[‘The assumed synchronized infestation peak by 46% likely overestimates the effect at the regional and continental scale, however, is likely representative for highly infested locations. Detailed information on infested forest sites was not available for whole Europe.’]

Reviewer comment: Although the author explained that the stem temperature is unavailable, I don’t think it is justifiable to assume that emissions are temperature-independent. Given that enhanced nighttime monoterpene concentrations and SOA formation are central findings, sensitivity tests using plausible temperature responses are needed to determine how strongly these results depend on the assumed temperature-independent source.

Author response: We appreciate the reviewer’s point and understand the concern. We do not claim that the emissions caused by bark beetle infestation are temperature independent. Indeed, higher stem temperature accelerates emissions from infested trees, as also shown in Jaakkola et al. 2023. At the same time, it is also reported that the measured stem emissions were not found to have a distinct diurnal cycle. Including a temperature dependency as widely applied with the correction factor $\gamma = \exp(\beta(T - T_0))$ (Guenther et al. 1993, 2012) includes a diurnal cycle of pool emissions and would not account for accelerated emissions. Developing a new temperature dependency estimate was beyond this study. Furthermore, the mechanisms underlying the enhanced BVOC emissions are not fully resolved. The increase may reflect a combination of increased bark beetle activity and release of stored resin, enhanced volatilization, and induced tree defense responses. Due to these uncertainties and the ones arising from measurements, as discussed before we did not take a temperature-dependency for the stem emissions into account so far. We added this in line 242-245:

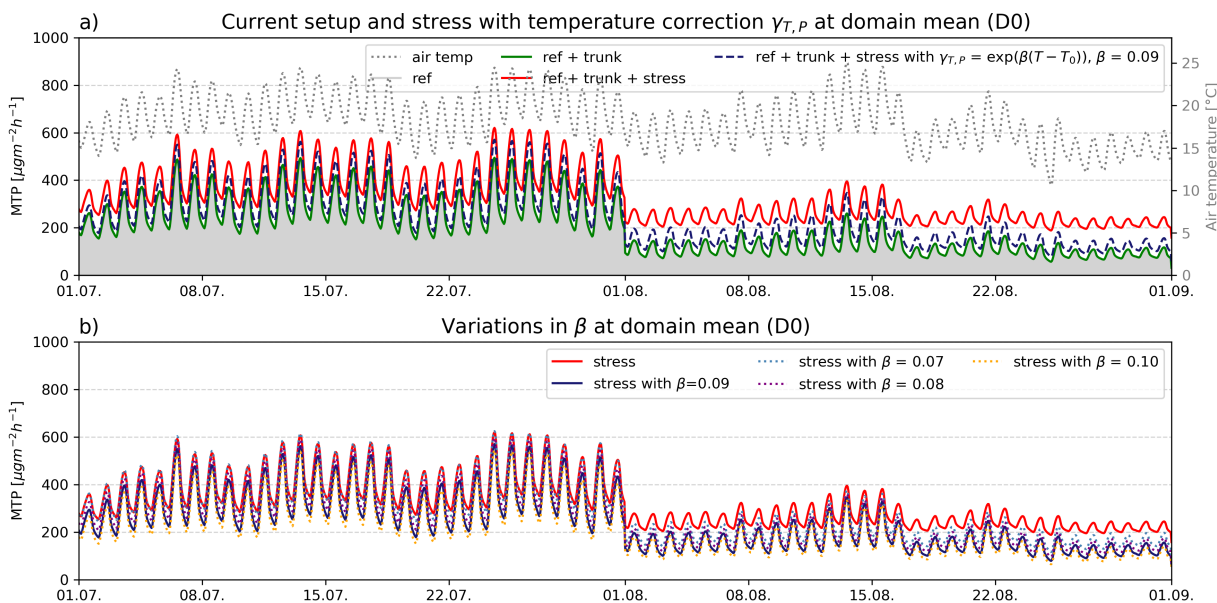
[‘Jaakkola et al. (2023) did not find a distinct diurnal cycle in stem emissions; however, they found an accelerated temperature dependency of the emissions. Neither effect can be adequately reflected by using the standard temperature correction factor ($\gamma_{T,P}$), which introduces a diurnal

cycle of emissions as a result of the dependency on air temperature and does not capture the temperature response of observed stem emissions. ’]

The parametrization adds an additional amount of BVOC into the model system, in order to analyze potential effects on SOA and O₃. To address the reviewers concerns we calculated the monoterpene emissions offline using a temperature correction factor according to the above formula with different assumptions on β without coupling it to the chemistry mechanism in MUSCAT. As outlined above, the relative change in BVOC emissions roughly translates into a similar relative change in modeled SOA concentration. With the temperature correction, the emissions of *Picea abies* (Norway spruces) at Norunda (14% spruces) follow a strong diurnal cycle, with peak emissions for warmer temperatures. As expected, especially nighttime emissions are reduced due to lower nighttime temperatures. Overall, the emissions for July and August decrease from 486.24 to 278.30 [$\mu\text{g m}^{-2} \text{h}^{-1}$]. The reduced emission increase of 30 % (previously 54 %) compared to the reference simulation would roughly result in a SOA increase of 30 % (previously 54 %). On the regional domain average (D0 domain, 6% spruces) the average MTP emissions without temperature correction are 326.31 [$\mu\text{g m}^{-2} \text{h}^{-1}$] and reduce with temperature correction to 245.29 [$\mu\text{g m}^{-2} \text{h}^{-1}$].

As the emission increase of MTP in the stress scenario of 44 % on D0 domain mean resulted in 44 % increase in modeled SOA concentration, this means that in a stem temperature-corrected stress scenario MTP emission would increase by 33% (relative to the air temperature-corrected reference scenario with an approximate increase in SOA by about a similar percentage.

We included an additional figure (Figure 10) and further discussions concerning the temperature dependencies into the manuscript’s discussion.



Line 517-530:

[‘The additional bark beetle induced emissions in the stress scenario are based on the parametrization using emission rates standardized at 30°C without a temperature correction due to insufficient information on the temperature dependency of the emissions from bark beetle infested trees. Furthermore, Jaakkola et al. (2023) reported no diurnal cycle of the stem emissions. To test the influence of a temperature correction factor, we calculated MTP emissions offline with the respective functions. The offline calculation uses the air temperature and radiation from the MUSCAT simulations; however, air temperature can only be seen as an approximation for bark temperature.

We applied the widely used temperature correction factor ($\gamma_{T,P} = \exp(\beta \cdot (T - T_0))$) of the pool canopy emissions also on the trunk emissions (see also Appendix A). Figure 10a shows the MTP emissions of spruces for the regional domain (D0) average. The $\gamma_{T,P}$ leads to a clear diurnal cycle, reducing especially nighttime emissions due to the lower air temperatures. On average, the stress-induced MTP emissions decrease by 25 % when including the temperature correction. Thus, the increase from the reference to the stress-induced case with temperature correction is approximately 33 %, and thus SOA would also likely increase by about the same percentage. The temperature within a tree trunk is influenced by the wounding of bark beetles, as this affects the sap flow. Further research is needed to constrain this for a suitable parametrization.’]

In the offline calculation we further added variations with different beta values, which control the temperature dependency in models. It shows that already slight variations in beta influence the peak emissions.

We agree that further research is needed to refine the temperature dependency of BVOC emissions in models, however, this is beyond the scope of this study.

Reviewer comment: More generally, the equations are difficult to follow because the definitions and interpretations of some symbols are incomplete. Please review all equations and accompanying text to ensure that every symbol, unit, and parameter value is clearly defined when first introduced.

Author response: Abbreviations and symbols were reviewed and supplemented with further descriptions.

Reviewer comment: Abstract: Please briefly describe the stress parameterization and its key assumptions, particularly the prescribed infestation extent and the treatment of bark-emission temperature dependence. The conditional nature of the quantitative results should be clear.

Author response: The abstract was revised and the simulation setup was clarified.

Line 4-10:

[‘Based on a seasonal bell-shaped spruce bark beetle population growth and measurements of healthy and infested spruces from Jaakkola et al. (2023), a parametrization for stress-induced BVOC emissions was developed and included in the chemical transport model COSMO-MUSCAT (Consortium for Small scale MOdelling - MUltiScale Chemistry Aerosol Transport). This study investigates an upper-bound scenario of intensive Europe-wide spruce bark beetle infestation, with an assumed peak extent in August affecting 46 % of spruces. The stress parametrization elevates the BVOC emissions from spruces under the assumption of diurnally constant stress-induced emissions, which leads to increased nighttime monoterpene concentrations.’]

Reviewer comment: L10: suggest changing to “The enhanced BVOC emissions”

Author response: The abstract was revised as outlined above.

Reviewer comment: L53-54, bark beetles often occur after storm as well.

Author response: added storms

Reviewer comment: L112: specify which anthropogenic emissions

Author response: We added a clearer description by naming the anthropogenic emissions in line 118-122:

[‘The prescribed anthropogenic emissions are carbon monoxide (CO), carbon dioxide (CO₂), ammonia (NH₃), NO_x, black carbon (BC), sulfur dioxide (SO₂), non-methane VOC (NMVOC), fine particulate matter smaller than 10µm (PM₁₀), fine particulate matter smaller than 2.5µm (PM_{2.5}), methane (CH₄), and dinitrogen oxide (N₂O).’]

Reviewer comment: L189: Explain what biomass density is. Aboveground or leaf biomass? If it is aboveground biomass, the author needs to explain why use the aboveground biomass instead of leaf biomass and provide further supports

Author response: We clarified this by adding *foliar* biomass density

Reviewer comment: L220-224, Please state explicitly that the bark-emission rates standardized to 30 °C are applied without an actual-temperature correction, if this is the intended implementation. The current explanation leaves this unclear.

Author response: The reason to not implement a temperature dependency for the stem emissions is discussed in the response to a previous comment and further throughout the manuscript. In line 249-250 the following sentence is extended:

[‘For those reasons, the parametrization is simplified by using the standardized emission rates, without a temperature correction factor.’]

Reviewer comment: L235, figure 2. we cannot call this function as “exponential population growth” as it is rather bell-shaped seasonal function.

Author response: We thank the reviewer, for pointing us towards this incorrect description. We changed it to:

[‘Seasonal bark beetle population growth...’]

Reviewer comment: Section 4.1

Norunda is a mixed forest, and I think it is mainly dominated by Scots pine. It will be interesting to show contributions from spruce, pine, healthy trunks, and stress separately. The shortcomings in capturing baseline emissions might be confounded with missing stress emissions. Then, the improved nighttime pattern could be influenced by the non-temperature-dependent sources rather than accurately representing infestation. A sensitivity experiment is clearly needed to justify that the improvements come from infestation.

Author response: We are aware that the measurement site of Norunda is a spruce-pine mixed forest. It needs to be noted, that the simulated emissions represent the whole grid cell of Norunda with a size of 14 km x 14 km, of which only 14 % is covered by spruce (*Picea abies*). The other land use types in the grid cell are mainly agricultural land (56.7%), pine (*Pinus sylvestris*) (11.4%), birch (5%), wetland (3%), artificial surface (8%) and minor contribution from other tree species e.g. oaks.

We included this information in line 353-356:

*[‘Compared are the diurnal cycle for July and August for MTP and SQT for the grid cell including Norunda with a grid cell size of 14 km x 14 km. The land-use distribution within the grid cell is 56.7 % agriculture, 14 % spruce (*Picea abies*), 11.4 % pine (*Pinus sylvestris*), 5 % birches, 8 % artificial surfaces, 3 % wetland and the remaining is attributed to other tree species.’]*

In our model experiment, we adapted only the emissions for *Picea abies* (Norway spruce), as stem emissions of both healthy and infested trees were available from Jaakkola et al., 2023. Thus, the simulated emissions from pine trees are without stem emissions and still in the constitutive mode. We only considered the spruce bark beetle, and no pine beetles. Thus, the presented emission increases are solely due to infested Norway spruce (with the assumed infestation rate of 46 %). Accounting for further tree-species specific stem emissions and herbivore infestation is open for future research.

The measurements used for comparison are from a non-infested forest. Therefore, the comparison between the modeled stress-induced emissions and the measurements does not allow

to draw conclusions on model improvements from the implementation of the stress parametrization. The comparison to the measurements was meant to qualitatively evaluate the order of magnitude of the emissions of the reference and stress scenario. The increase in MTP emissions from the reference to the stress simulation in July is one order of magnitude during nighttime, and a factor of two throughout the day. Compared with emissions measurements in the healthy forest as shown in Fig. 5, stress-induced emissions are higher by a factor of 2.

The changed diurnal cycle of the concentrations under higher BVOC emissions from the stress case merely shows that the constitutive BVOC emission parametrization in COSMO-MUSCAT likely underestimates nighttime emissions and concentrations. However, this should not be drawn back to a better model performance due to the integration of the stress parametrization.

We clarified this in line 376-384:

[‘The diurnal cycle under the stress-induced emissions seems to be in closer agreement with the measurements, however this should not be interpreted as improved model performance. The stress-related MTP concentration is clearly higher than the measurements from the healthy forest, however, close to the 95th percentile of measured MTP concentration. From the comparison with the healthy forest³⁸⁰ measurements we can merely conclude qualitatively that the magnitude of the stress-induced emissions is within a realistic range and that the standard BVOC emissions are potentially too low. The standard parametrization together with the chemistry mechanism in COSMO-MUSCAT need to be further developed to better represent BVOC. The conclusions we draw from the comparison are discussed in Section 5. ’]

And line 514-517:

[‘With the stress parametrization MTP emissions increase substantially compared to the reference scenario, in particular throughout the night. Although this does not translate into improved model performance, it highlights the need to include additional emission sources, adjust existing emission potentials and refine temperature dependencies. ’]

And line 568-571:

[‘As the Norunda measurements represent a healthy forest without significant bark beetle disturbance, we can conclude that the standard COSMO-MUSCAT biogenic module is missing emission sources, however, in that specific case the missing emissions cannot be directly attributed to spruce bark beetle infestation. ’]