## **Response to Referee #1**

We thank the reviewers for their insightful comments and efforts to improve the manuscript. We provide point-by-point response to each comment as follows. In the following text, the reviewers' comments are in black, authors' response are in blue, and changes to the manuscript and supplement information are in dark red.

The study by Song et al. combines real-time measurements of atmospheric volatile organic compound (VOC) and organic aerosol concentrations on a highly complex study site influenced by a biogas power plant, a mixed temperate forest stand, a nearby village and a clear-cut area. The deployed instruments, especially the PTR-TOF-MS coupled with a CHARON particle inlet and the VOCUS-PTR-TOF-MS, are state of the art and allowed the authors to investigate VOC concentrations both in the gas and particle phase during a threeweek field campaign. Additional measurements of trace gases (methane, carbon dioxide, carbon monoxide, ozone), water vapor, particulate matter and black carbon concentrations in the atmosphere, as well as meteorological parameters (temperature, relative humidity, soil moisture, planetary boundary layer height, wind speed and wind direction) built a comprehensive data set that is generally well suited to achieve the objective of the study. This was, as indicated in the title, to characterize the concentrations of biogenic VOCs and their oxidation products at a stressed forest close to a biogas power plant. The sources of the measured VOCs were nicely disentangled on the basis of the wind direction. Investigating the impact of a biogas power plant on atmospheric VOC concentrations in direct contrast to a stressed conifer forest is quite a novelty and also the impact of insect outbreaks in atmospheric chemistry is not well understood yet. Thus, in my opinion, the content of this study fits well with the scope of ACP. However, I have some major concerns regarding the overall presentation of results, as described in detail below.

Briefly, the study is written rather descriptive and substantial conclusions regarding a broader context become not always clear. I would strongly recommend to present the implications of the findings in more detail. Further, there were some inconsistencies regarding the definition of the wind direction sectors with possible implications on the data interpretation. Also, the tree species composition of the investigated forest and the stress status of the same should be characterized with more detail, as no physiological parameters are given in the present version of the manuscript.

Overall, I think the study contributes to a highly relevant topic and should be considered for publication in ACP, but there is still quite some scope of improvement.

**Response:** We appreciate your insightful comments and suggestions, which are helpful for the improvement of our manuscript. Point-by-point response to each comment are given below.

# Specific comments

#### Title

In the title the authors state, that a "stressed pine forest" was investigated. The term "pine forest" is in this context a little confusing or even wrong, as the study was conducted next to a forest composed of *Picea abies* (Norway spruce) and *Fagus sylvatica* (European beech) (L. 86) with no reported occurrence of *Pinus spp.* (Pine). In my understanding, "Pine" should be replaced with "temperate" in L2. Depending on the species composition in the studied

area (which should be described with more detail) the forest type could be further specified as "temperate mixed forest" or "temperate coniferous forest".

**Response:** The Eifel Forest is mainly composed of Norway spruce (Picea abies), so it should be clarified as a temperate coniferous forest. We have changed the title of our manuscript. The updated title is now:

"Characterization of biogenic volatile organic compounds and their oxidation products at a stressed spruce-dominated forest close to a biogas power plant"

In addition, the stress status of the forest is insufficiently documented. With no doubt, the forests in the Eifel were strongly affected by bark beetle outbreaks, heat waves and drought over the last years. However, this regional situation does not explain sufficiently the current status of the investigated forest. For this, further stress indicators like tree mortality, chlorophyll fluorescence, leaf/needle water potential or comparable stress parameters should be included in the study in any case.

**Response:** We have revised the subsection of "2.1 Sampling site" to include more information regarding the stress status of the Eifel Forest during our measurement period. We have also included the leaf area index and soil moisture data for the Eifel Forest during our sampling period, which indicate that the forest was under stress.

## Lines 117-134: "2.1 Sampling site

"In this study, a three-week field campaign was conducted at a site in the northern Eifel Forest (50.72° N, 6.40° E) during June 2020 as a part of the "Heat and Drought 2020" campaign of the Modular Observation Solutions of Earth Systems (MOSES) project of the Helmholtz Association of German Research Centers. The Eifel Forest was suffering from severe droughts, heatwaves and severe bark beetle infestation in the last years (Weber et al., 2022b; Ghimire et al., 2016). Within two years (2018-2020), 14% of the spruce in the Northern Eifel region were removed due to summer droughts and only 28.3% remained in good condition (Montzka et al., 2021). Therefore, the Eifel Forest can serve as an example of a stressed temperate coniferous forest.

As shown in Fig. 1, the measurement site is situated directly next to a stand of Norway spruce with a few shrubs and blueberry plants also surrounding the area. To the south and southeast of the measurements site, there were some clear-cut areas due to bark beetle infestation in the years of 2018-2020. Additionally, the measurement site was located ~400 m southeast of a football field in the small village Kleinhau belonging to the municipality of Hürtgenwald, Germany (population about 9000) and ~250 m east of a BPP (BioEnergie Kleinhau GmbH). The biomass substrate used for the biogas production in this BPP consisted mainly of crop waste (e.g., corn stover). The measurement site was affected by the BPP emissions especially for westerly wind directions."

**Lines 290-296:** "The leaf area index of the Eifel Forest during our measurement period was determined to be  $\sim 2.5 \pm 0.02 \text{ m}^2 \text{ m}^{-2}$  based on the ERA5 reanalysis data. The soil moisture was measured to be  $0.3 \pm 0.04 \text{ m}^3 \text{ m}^{-3}$  at a station located  $\sim 150 \text{ m}$  southwest of the sampling site. In addition, the spatial distribution of soil moisture in the northern Eifel Forest also showed low values ( $< 0.3 \text{ m}^3 \text{ m}^{-3}$ ) in most areas covering our sampling site (Fig. S7). Therefore, the Eifel Forest was under relatively dry condition during our measurement period."

#### Abstract

The abstract could be substantially improved by adding a few sentences at the beginning about the general topic of the study, the research gap and the specific research questions.

: "In the WD-forest group [...] biogenic emissions of isoprene, monoterpenes and sesquiterpenes [...] exceeded the photochemical consumption" – is this surprising? I think this is exactly what we would expect for a temperate forest, especially, when it is stressed.

**Response:** In the revised manuscript, we divided the measurement data of BVOCs into two groups with one mainly influenced by biogenic emissions from an intact forest and a clear-cut area (biogenic-group) and another one by the anthropogenic emissions from a BPP and a village (anthropogenic-group). In the biogenic group, we observed that the diurnal variations of isoprene, monoterpenes and sesquiterpenes showed higher mixing ratios during daytime even when atmospheric oxidants like  $O_3$  and OH radicals had high concentrations. It is expected that higher temperature would enhance the emissions of BVOCs for a temperate forest. In addition to biogenic emissions, the ambient concentrations of BVOCs are also affected by the levels of atmospheric oxidants. Therefore, we emphasize that the increase of these BVOCs during daytime were driven by higher temperatures, which exceeded their photochemical consumption.

We added the following sentences to the abstract to introduce to the subject.

**Lines 19-22:** "Biogenic volatile organic compounds (BVOCs) are key components of the atmosphere, playing a significant role in the formation of organic aerosols (OA). However, only few studies have simultaneously examined the characteristics of BVOCs and OA in the forest under the impact of consecutive droughts and extensive bark beetle infestations."

I would recommend the authors to have a closer look on studies, that were conducted at the "Stations for Measuring Ecosystem-Atmosphere Relations" (SMEAR) in Estonia and Finland (SMEARII), because there are quite some similarities between the experimental setups and ecosystems studied (eg. Bourtsoukidis, E., Bonn, B., & Noe, S. M. (2014). On-line field measurements of BVOC emissions from Norway spruce (Picea abies) at the hemiboreal SMEAR-Estonia site under autumn conditions. *Boreal environment research*, *19*(3), 153."

: Here, the authors limit the scope of their conclusions to their specific study site. I would strongly recommend to highlight aspects of the study that are relevant for a broader context and/or more generalizable.

**Response:** We have reviewed relevant studies previously published and compared our results with these findings. We have expanded our research scope and revised the abstract accordingly. The updated abstract is as follows:

Lines 19-45: "Biogenic volatile organic compounds (BVOCs) are key components of the atmosphere, playing a significant role in the formation of organic aerosols (OA). However, only few studies have simultaneously examined the characteristics of BVOCs and OA in the forest under the impact of consecutive droughts and extensive bark beetle infestations. Here we present real-time measurements of OA and BVOCs at a stressed Norway spruce-dominated forest near a biogas power plant (BPP) in western Germany during June 2020. A proton-transfer-reaction time-of-flight mass spectrometer coupled with a particle inlet (CHARON-PTR-ToF-MS) and a Vocus-PTR-ToF-MS were used to measure OA and BVOCs. The average mass concentration of OA was  $0.8 \pm 0.5 \ \mu g \ m^{-3}$ , consisting mainly of semi-volatile monoterpene oxidation products. The average mixing ratios of isoprene (0.58  $\pm$  0.54 ppb) and monoterpenes (2.5  $\pm$  5.3 ppb) were higher than the values previously measured in both German temperate forests and boreal forests. Based on wind direction analysis, BVOC data were categorized into two groups with one

mainly influenced by the biogenic emissions from an intact forest and a clear-cut area (biogenic-group) and another one by the anthropogenic emissions from a BPP and a village (anthropogenic-group). High mixing ratios of monoterpenes were observed in the anthropogenic-group, indicating a significant contribution of BPP emissions. In the biogenic-group, the variations of BVOC mixing ratios were driven by the interplay between meteorology, biogenic emissions and their photochemical consumption. Positive matrix factorization analysis of VOCs revealed substantial contributions of oxygenated organic compounds from the photochemical oxidation of BVOCs during daytime, while monoterpenes and their weakly oxidized products dominated at night. Furthermore, increasing relative humidity and decreasing temperatures promoted the gas-to-particle partitioning of these weakly oxidized monoterpene products, leading to an increase in nighttime OA mass. The results demonstrate the variations of BVOCs are influenced not only by meteorological conditions and biogenic emissions but also by local BPP emissions and subsequent chemical transformation processes. This study highlights the need to investigate the changes of biogenic emissions in European stressed forests."

## Introduction

L40-42: There are several earlier publications that should be cited here as a primary source, eg. Rasmussen & Went 1964 (10.1073/pnas.53.1.215) or Trainer et al. 1987 (https://doi.org/10.1038/329705a0)

**Response:** We have cited these earlier studies accordingly.

### Methods

Subsection "2.1 Sampling Site": Information about the species composition, as well as about the stress status of the forest stand should be added to this subsection (see comment above). Furthermore, the clear-cutting areas mentioned in L112 seem to cover large areas around measurement location (Fig. 1a). There also seem to be some afforested areas in the close vicinity of the measurement location, which potentially influenced the measurements. The authors should indicate clearly in figure 1a which areas are covered with intact forest, and which areas are affected by clear-cutting or afforestation. Adding a colored layer to the satellite image might be suitable for this purpose.

**Response:** As mentioned before, we have included more information regarding the stress status of the Eifel Forest in subsection 2.1 "Sampling site". We have also revised Figure 1 to show different wind sectors in detail. The measurement site was affected by an intact forest, a clear-cut area, a BPP and the residential area of Kleinhau in the sectors of 0-120°, 120-240°, 240-310° and 310-330°, respectively. Almost no winds were coming from the sector of 330-360° during the measurement period.



**Figure 1.** (a) Location of the sampling site (orange star) (©Google Earth); (b) a close look at sampling site with the centered wind rose for the entire measurement period. The orange dash lines are shown for distinguishing different sectors of wind direction (WD). The WD-forest of 0-120° is influenced by an intact forest area, the WD-cut of 120-240° is influenced by a clear-cut area, the WD-BPP of 240-300° is influenced by a biogas power plant (blue rectangle) and the WD-village of 300-330° is influenced by the residential areas of Kleinhau.

L138-142: Here, different temperature settings of the drift tube of the CHARON-PTR-TOF-MS are described. In L161-167 is stated, that measurements with the drift tube temperature set to 80°C were discarded – please join this two paragraphs.

**Response:** We have joined these two subparagraphs to explain the different temperature setting of the drift tube of the CHARON-PTR-ToF-MS during the two measurement stages.

Lines 166-176: "Finally, the electric field (E/N) of the CHARON-PTR-TOF-MS was kept at ~97 Td and ~57 Td for the gas and particle phase measurement modes respectively during the second measurement stage. Please note that during the first measurement stage the actual temperature of the drift tube fluctuated and was lower than the intended temperature of 120 °C (Fig. S1). This made it difficult to quantify organic compounds in the particle phase measured by the CHARON-PTR-ToF-MS. For the gas phase measurements, we corrected the major VOC data from the first measurement stage based on the gas calibration and the cross-comparison with Vocus-PTR-ToF-MS measurements. Consequently, we can present the major VOC species measured by the CHARON-PTR-ToF-MS for the entire campaign, while the particle phase data for first measurement stage were excluded for further analysis in this study."

L170: Please explain, why another time period than 2020/06/05-2020/06/30 was chosen for the measurements with the Vocus-PTR-TOF-MS. The reasons are currently not clear.

**Response:** Due to a malfunction of the Vocus-PTR-ToF-MS, it was not available for measurement at the beginning of the campaign. We started the concurrent VOC measurements of the two PTR-ToF-MS on 2020/06/10 when the Vocus-PTR-ToF-MS was working properly. We have added one sentence to avoid any confusion.

**Lines 193-194:** "The Vocus-PTR-ToF-MS was not available for measurements before 10<sup>th</sup> June 2020 due to a technical problem."

L205: Soil moisture has an extremely high local variability. In this study only one soil moisture probe was used – the authors should be aware that the soil moisture data are not very reliable and should be transparent about this in the manuscript.

**Response:** We agree that soil moisture has a high local variability. In this study, the longterm soil moisture was measured by a cosmic ray neutron sensor (CRNS) at a station which was located ~150 m southwest of our sampling site. The CRNS was calibrated properly in this study, thus it can provide reliable soil moisture data. During the concurrent sampling period (5<sup>th</sup>-30<sup>th</sup> June 2020) at our measurement container, the soil moisture values were relatively low with an average of  $0.3 \pm 0.03$  m<sup>3</sup> m<sup>-3</sup>. This indicates that these measurement days were already very dry at our sampling site. In addition, the spatial distribution of soil moisture in the northern Eifel Forest was determined by mobile CRNS measurements with a rover. For example, during 4<sup>th</sup> June 2020, the soil moisture values in most areas in the Eifel Forest were lower than 0.3 m<sup>3</sup> m<sup>-3</sup> (Fig. S7), indicating that the forest was under dry conditions. To clarify the status of stress for the forest, we have provided additional information in the methods section.



**Figure S7.** (a) Time series of daily soil moisture  $(\theta_v)$  and sensing depth measured by a cosmic ray neutron sensor (CRNS) which was located ~ 150 m southwest of our sampling site. The red dashed box shows the concurrent sampling period at our measurement

container during 5<sup>th</sup>-30<sup>th</sup> June 2020; (b) Spatial distribution of soil moisture in the northern Eifel derived from the measurement by a CRNS rover during 4<sup>th</sup> June 2020. The black circle shows the location of the sampling container.

#### **Results and discussion**

The authors make intensive use of the supplement and present in total 23 (!) Figures and Tables (10 in the main manuscript and 13 in the supplement). This makes it sometimes difficult to follow the overall line of argumentation throughout the manuscript. I would highly recommend to opt for fewer Figures and make a selection based on relevance to support the main conclusions.

**Response:** We have reorganized the order of Figures and Tables in the main manuscript and supplement. We have selected the most important and relevant ones to support our conclusions.

Throughout the manuscript many abbreviations are used. Some of them are common and make the text easier to understand (eg. PTR-TOF-MS, VOC, OA, SOA, LOD, PM2.5, PM10). However, some abbreviations are introduced, but never used again and should, in my opinion, be removed from the text (e.g. TDU in L.124 or FIMR in L171). Further, there are some abbreviations for short terms, like black carbon and planetary boundary layer, where the terms could be written out in full in order to reduce the total number of abbreviations and make the text easier to follow. In any case, the entire manuscript should be carefully checked to ensure that all abbreviations are introduced the first time they are used in the text (see technical comments). Further, nested abbreviations (explaining one abbreviation with another one) like in "semi-volatile oxygenated OA" (L293) should be avoided. Instead, please write out e.g. "semi-volatile oxygenated organic aerosols"

**Response:** We completely agree with this comment. We have checked the abbreviations throughout the manuscript. We have introduced the abbreviations correctly in the revised manuscript.

The authors should reconsider, whether giving averages over the entire campaign is the best way to present their data. Especially, for parameters with diurnal variations, like ambient temperature (see L.249) or BVOC concentrations, it would be more informative to report day and night averages, and for temperature, additionally, daily maximum and minimum values. This might not be necessary for more constant parameters, like soil moisture.

**Response:** Thank you for this suggestion. We have added the daily maximum and minimum values for ambient temperatures and BVOC mixing ratios accordingly.

L252 ff.: Please, add some kind of systematic definition of the two episodes. A systematic definition could be: If the temperature of single days was > the 50% quantile of the temperature of the entire measurement campaign for a number of x consecutive days, then these days were defined as high-T episodes.

**Response:** We have provided a systematic approach to define these two episodes. During the low-T episode, the daily maximum temperature remained below 20 °C for three consecutive measurement days. During the high-T episode, the daily maximum temperature exceeded 25 °C for three consecutive measurement days.

**Lines 297-302:** "During the entire measurement campaign, we observed two characteristic episodes, Episode 1 (0:00 9<sup>th</sup> of June to 0:00 12<sup>th</sup> of June) and Episode 2 (12:00 23<sup>rd</sup> of June to 12:00 26<sup>th</sup> of June), for different meteorological conditions. During

Episode 1, the daily maximum temperature remained below 20 °C for three consecutive measurement days. During Episode 2, the daily maximum temperature exceeded 25 °C for three consecutive measurement days. Therefore, hereafter we define these two episodes as low-T and high-T episodes, respectively."

Please, explain and quantify what "good agreement" means in L. 266.

**Response:** We have revised "good agreement" to "good to fair correlations (r = 0.92 and 0.59 for isoprene and monoterpenes respectively)" in this sentence.

**Lines 312-314:** "Isoprene and monoterpenes were quantitatively measured by the CHARON-PTR-ToF-MS and Vocus-PTR-ToF-MS with good to fair correlations (r = 0.92 and 0.59 for isoprene and monoterpenes, respectively)."

Throughout the entire section "Results and discussion" the French Landes forest is used as one of the main references to compare the results of this study with (eg. L 268, L269, L271, L280, and more). I have some doubts, whether the Landes forest, which is (other than the forest investigated in the present study) a pine forest with oceanic climate, the best choice to compare the results with to this extend. I would recommend to check the literature carefully for studies that were conducted in forests dominated by Norway Spruce and incorporate them into the discussion. One relevant study might be Petersen et al. 2023 (10.5194/acp-23-7839-2023) published in this same journal.

**Response:** Thank you for this valuable comment. We have extended the discussion to include the comparison of BVOCs with different types of forest ecosystems.

**Lines 314-327:** "During the entire campaign, the average mixing ratios of isoprene was  $0.58 \pm 0.54$  ppb, slightly higher than that previously reported in a Norway sprucedominated forest ( $0.32 \pm 0.17$  ppb) in central Germany (Bourtsoukidis et al., 2014) and a mixed-conifer forest (max. 0.25 ppb) with Norway spruce and Scots pine (*Pinus sylvestris* L.) in Sweden (Petersen et al., 2023). The level of isoprene in this study was comparable to that (~0.6 ppb) observed in French Landes forest dominated by maritime pine trees (*Pinus pinaster* Aiton) during summer time (Li et al., 2020), but higher than those (0.01-0.2 ppb) reported for the boreal forests in Finland dominated by Scots pine (Li et al., 2021a; Hellén et al., 2018). The average mixing ratios of monoterpenes ( $2.5 \pm 5.3$  ppb) in this study was also higher than that reported in a Norway spruce-dominated forest (0.50  $\pm 0.21$  ppb) in central Germany (Bourtsoukidis et al., 2014), but lower than that observed in the French Landes forest (~6 ppb) (Li et al., 2020). Relatively low mixing ratios of monoterpenes were reported previously for the boreal forests in Finland (~0.8 ppb) during summertime (Li et al., 2020; Mermet et al., 2021)."

L284: Without direct calibration the measured sesquiterpene concentrations are probably not only lower than the actual concentrations due to fragmentation, but also due to the typically relatively low transmission rate of sesquiterpenes during the proton transfer reaction in the PTR-TOF-MS.

**Response:** This is correct. We have added one more sentence to explain the limitation of sesquiterpene quantification.

**Lines 336-337:** "In addition, sesquiterpenes may experience wall losses inside the inlet tubing and the instrument, and have low transmissions (Li et al., 2020)."

For me, it doesn't always become clear whether a statement refers to results of the authors, or rather to a cited study. As an example in L.324: "The fragmentation pattern of oxidized

organic compounds in the CHARON-PTR-TOF-MS varied depending on the instrument settings (Leglise et al. 2019). What is the meaning of the reference in this case?

**Response:** This statement refers to an example for the variation of fragmentation pattern of oxidized organic compounds the study cited. We have revised this sentence to avoid any confusion.

Lines 385-387: "Previous studies have shown that the fragmentation pattern of oxidized organic compounds in the CHARON-PTR-ToF-MS varied depending on the instrument settings (Leglise et al., 2019; Gkatzelis et al., 2018)."

In my opinion a drawback of the study is, that the wind sectors are not defined uniformly and, that the clear-cut sites are not represented adequately in the sector definition. While in L334ff there are three wind sectors defined (0-240° forest, 240-300° biogas power plant, 300-330° village), there are only two sectors defined in L372 (0-240° forest, 240-330° biogas power plant). Based on Figure 1a it seems like there was intact forest from ~0-90°, afforested or clear-cut areas from ~90-240°, the biogas power plant from ~240-300 and a zone influenced by forest emissions and anthropogenic emissions of the village from ~300°-360°. I would kindly ask the authors to check the definition of the sectors and indicate the land use with a colored layer in figure 1 (see comment above).

**Response:** We have checked the definition of different sectors carefully. Based on the wind rose plot and geographical conditions around our sampling site, we define four major wind sectors including 0-120° for the intact forest, 120-240° for the clear-cut area, 240-310° for the biogas power plant and 310-330° for the residential area of the village. No winds were coming from the sectors of 330-360° during the entire measurement period. We have revised Figure 1 as mentioned before. We have also revised the discussion on how the different meteorological conditions influence the variations of BVOCs and particles in section 3.2.

L401: Keep in mind, that plants also emit less VOCs during nighttime (see eg. Holzke et al. 2006 for European Beech, doi.org/10.1007/s10874-006-9027-9, Fig. 3a; and Ghirardo et al. 2010 doi.org/10.1111/j.1365-3040.2009.02104.x Fig. 1b; Meischner et al. 2024 doi.org/10.1093/treephys/tpae059, Fig. 4 for Norway Spruce).

**Response:** Indeed, the plants typically emit less VOCs at night due to lower temperature and the absence of sunlight. However, reactive VOC like monoterpenes and sesquiterpenes are typically also depleted faster during daytime. This can influence the concentrations observed depending on the photochemical activity and the corresponding rate coefficients. During nighttime, the remaining oxidants like ozone or NO<sub>3</sub> radicals may also reduce the concentrations.

L564: I would avoid statements about the concentration of sesquiterpenes, since measurements were not calibrated, as described in L. 282ff.

**Response:** We agree and have revised this sentence accordingly.

Lines 693-695: "The average mixing ratios of isoprene and monoterpenes were higher than the values previously measured in both German temperate forests and boreal forests during summertime (Mermet et al., 2021; Li et al., 2021a; Hellén et al., 2018; Bourtsoukidis et al., 2014)."

L577: Formulas and calculations should be defined and explained in the material and method section.

**Response:** We have moved these calculations to the method section.

#### Lines 271-276: 2.4 Calculation of particle-phase fraction of organic compounds

To estimate the gas-to-particle partitioning processes, we calculated the particulate mass fraction  $(F_p)$  of organic compounds by the Equation 3:

$$F_p = \frac{c_{p,i}}{c_{g,i} + c_{p,i}} \tag{3}$$

where  $C_{p,i}$  and  $C_{g,i}$  are the particle and gas phase concentrations of the individual organic compound measured by CHARON-PTR-ToF-MS and Vocus-PTR-ToF-MS, respectively.

L598: I have some difficulties to follow the argumentation why European beech should have emitted mainly  $\alpha$ -pinene and  $\beta$ -pinene. For Norway spruce this might be correct, however, there is strong evidence, that European beech emits mainly sabinene (>30 % of total monoterpene emissions) and only <10%  $\alpha$ -pinene and  $\beta$ -pinene (Holzke et al. 2006, Table 2, 10.1007/s10874-006-9027-9)

**Response:** Indeed, European beech emits mainly sabinene. Our sampling site is mainly surrounded by Norway spruce, mainly emitting  $\alpha$ -pinene and  $\beta$ -pinene. We have revised this sentence as follows.

**Lines 670-672:** "It is reasonable to assume that these monoterpenes are mainly  $\alpha$ -pinene and  $\beta$ -pinene because our sampling site was in a forest dominated by Norway spruce known to emit mainly pinenes (Christensen et al., 2000; Hakola et al., 2017)."

# Conclusion

618: This is inconsistent with L.282 where it says, that sesquiterpenes could not be quantified due to missing calibration standards. Hence, sesquiterpene measurements should only be used to calculate correlations with other parameters or interpretation of temporal variations.

Response: We have modified this sentence to avoid any inconsistence.

Lines 693-695: The average mixing ratios of isoprene and monoterpenes were higher than the values previously measured in both German temperate forests and boreal forests during summertime (Mermet et al., 2021; Li et al., 2021a; Hellén et al., 2018; Bourtsoukidis et al., 2014).

#### Figures

Figure 1b - RH: Please, change the color of the x-axis to black. The pink color could be interpreted as constantly low precipitation rates.

**Response**: We have separated the original Figure 1b to Figure 2 in the revised manuscript. This figure has been revised accordingly.

Figure 4: I really like the highlighted areas that indicate the wind direction from the biogas power plant. Why not adding shaded areas for the other sectors, too?

**Response:** Thank you for the suggestion. We have revised the original Figure 4 to Figure 5 and added the shaded areas for different wind sectors accordingly.



**Figure 5.** Variations of (a) wind speed (WS), planetary boundary layer (PBL) and ambient temperature; (b) global radiation,  $O_3$  and BC mass concentrations; (c) mixing ratios of CO, CO<sub>2</sub> and CH<sub>4</sub> and (d) mixing ratios of isoprene, monoterpenes and sesquiterpenes as a function of wind direction (WD). The black dots and whiskers represent the mean values and standard deviations in each WD bin of 10°. Data within the WD1 of 0-120° is influenced by an intact forest area (light green), the WD2 of 120-240° is influenced by a clear-cut area (light blue), the WD3 of 240-300° is influenced by a biogas power plant (yellow) and the WD4 of 300-330° is influenced by the village (pink).

Technical corrections

L26: Please, introduce the abbreviation for wind direction (WD)

Response: corrected.

L50: Please, change "forests" to "forest ecosystems"

Response: changed.

L55: Please, change "showed" to "shows"

Response: changed.

L57: Please, change "sunlight" to "sunlight intensity"

Response: changed.

L58-59: Please, assign cited studies to specific stress types, since not all of the cited studies in L59 addressed the effect of high temperature, drought AND herbivory attack on BVOC emissions from trees

**Response:** We have changed the cited studies to each specific stress types in the revised manuscript.

Lines 68-71: "The emissions and compositions of BVOCs from trees varies with abiotic and biotic stresses such as high temperature (Teskey et al., 2015; Kleist et al., 2012), drought (Peron et al., 2021; Bonn et al., 2019) and herbivore attack (Jaakkola et al., 2023; Kari et al., 2019; Faiola and Taipale, 2020)."

L60: This is optional, but may be the sentence becomes clearer if "significantly" is exchanged with "especially". In this way the role of terpenoids in the stress response of trees is highlighted and the sentence is less redundant with the previous one.

**Response:** We have revised this sentence accordingly.

Lines 71-73: "It has been reported that these stresses can alter the emissions of BVOCs especially terpenoids (Ghimire et al., 2016; Jaakkola et al., 2023; Byron et al., 2022)."

L64: Please, change "showed" to "shows"

Response: changed.

L64ff: "...lower values during daytime", compared to what?

**Response:** we have revised this sentence to make it clear.

**Lines 75-78:** "The diurnal variation of monoterpene concentrations shows lower values during daytime than nighttime in the boreal forests, which were attributed to the rapid photochemical consumption and expanded boundary layer heights (Hellén et al., 2018; Hakola et al., 2012)."

L83/L84: A connecting sentence would make the text easier to follow

**Response:** We have added the sentences to connect these two paragraphs.

**Lines: 92-95:** "However, our understanding of the interplay between gas and particle phases of BVOC oxidation products in real forest atmosphere, particularly in stressed forest, remains limited. Addressing these gaps is crucial for assessing the impact of various environmental factors on BVOC emissions and their subsequent transformation (Faiola and Taipale, 2020)."

L86: Please, complete common species with Latin species names eg. "Norway spruce (*Picea abies* (L.) H. Karst.)"

Response: Added (Line 98).

L92: Please, quantify the increase in BPPs, if possible

**Response:** We added one sentence to describe the quantity of biogas power plants in Europe.

Lines 104-105: "Europe is the world leader in biogas electricity production with more than 18,000 BPPs (Brémond et al., 2021)."

L124: The term "thermo-desorption unit" appears only once in the entire text, so the abbreviation can be deleted (same for PEEK in L135)

Response: deleted.

L133: Please, introduce the abbreviation "PFA"

**Response:** This has been corrected.

L145: Please be consistent with the abbreviation "TOF" or "ToF" throughout the whole text **Response:** We have checked and corrected the typos throughout the manuscript.

L201: Is "malfunction" instead of "multifunction" meant? Please, check.

**Response:** This has been corrected.

L202: Please, introduce abbreviations for "relative humidity" and "planetary boundary layer". Check consistency with other parts of the manuscript, eg. in L 202 is says "boundary layer" and in L976 "planetary boundary layer"

**Response:** We have introduced the abbreviation correctly throughout the revised manuscript.

L208: Please, avoid introducing abbreviations in the title.

**Response:** We have removed the abbreviations in the title.

L226: Please, change "into" to "from"

Response: Changed.

L252-254: Please, change punctuation. E.g. Two characteristic episodes, [..], were observed [...].

Response: Changed.

L350: In my opinion, it is not ideal to start a new paragraph with a reference to the supplement.

**Response:** We have reorganized this paragraph in the revised manuscript.

L395: Please, add ",respectively," after "<0.01 ppb"

Response: added.

## References

- Bonn, B., Magh, R. K., Rombach, J., and Kreuzwieser, J.: Biogenic isoprenoid emissions under drought stress: different responses for isoprene and terpenes, Biogeosciences, 16, 4627-4645, 10.5194/bg-16-4627-2019, 2019.
- Faiola, C. and Taipale, D.: Impact of insect herbivory on plant stress volatile emissions from trees: A synthesis of quantitative measurements and recommendations for future research, Atmospheric Environment: X, 5, 100060, https://doi.org/10.1016/j.aeaoa.2019.100060, 2020.
- Jaakkola, E., Gärtner, A., Jönsson, A. M., Ljung, K., Olsson, P. O., and Holst, T.: Spruce bark beetles (Ips typographus) cause up to 700 times higher bark BVOC emission rates compared to healthy Norway spruce (Picea abies), Biogeosciences, 20, 803-826, 10.5194/bg-20-803-2023, 2023.
- Kari, E., Faiola, C. L., Isokääntä, S., Miettinen, P., Yli-Pirilä, P., Buchholz, A., Kivimäenpää, M., Mikkonen, S., Holopainen, J. K., and Virtanen, A.: Time-resolved characterization of biotic stress emissions from Scots pines being fed upon by pine weevil by means of PTR-ToF-MS, 2019.
- Kleist, E., Mentel, T. F., Andres, S., Bohne, A., Folkers, A., Kiendler-Scharr, A., Rudich, Y., Springer, M., Tillmann, R., and Wildt, J.: Irreversible impacts of heat on the emissions of monoterpenes, sesquiterpenes, phenolic BVOC and green leaf volatiles from several tree species, Biogeosciences, 9, 5111-5123, 10.5194/bg-9-5111-2012, 2012.
- Peron, A., Kaser, L., Fitzky, A. C., Graus, M., Halbwirth, H., Greiner, J., Wohlfahrt, G., Rewald, B., Sandén, H., and Karl, T.: Combined effects of ozone and drought stress on the emission of biogenic volatile organic compounds from Quercus robur L, Biogeosciences, 18, 535-556, 10.5194/bg-18-535-2021, 2021.

Teskey, R., Wertin, T., Bauweraerts, I., Ameye, M., McGuire, M. A., and Steppe, K.: Responses of tree species to heat waves and extreme heat events, Plant, Cell & Environment, 38, 1699-1712, <u>https://doi.org/10.1111/pce.12417</u>, 2015.