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 three-week 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 ~2.5 ± 0.02 m^2 m^{-2} based on the ERA5 reanalysis data. The soil moisture was measured to be 0.3 ± 0.04 m^3 m^{-3} at a station located ~150 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 m^3 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 set-ups 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 ± 0.5 µg m^{-3}, consisting mainly of semi-volatile monoterpene oxidation products. The average mixing ratios of isoprene (0.58 ± 0.54 ppb) and monoterpenes (2.5 ± 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 0-120º, 120-240º, 240-310º and 310-330º, respectively. Almost no winds were coming from the sector of 330-360º during the measurement period.

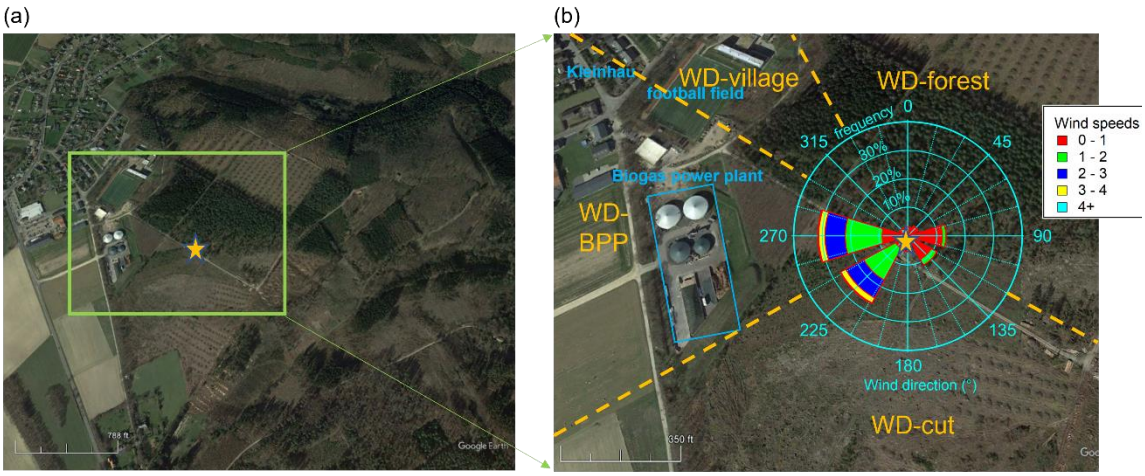

Figure 1. (a) Location of the sampling site (orange star) (©Google Earth); (b) a close look at sampling site with the centered wind rose for the entire measurement period. The orange dash lines are shown for distinguishing different sectors of wind direction (WD). The WD-forest of 0-120º is influenced by an intact forest area, the WD-cut of 120-240º is influenced by a clear-cut area, the WD-BPP of 240-300º is influenced by a biogas power plant (blue rectangle) and the WD-village of 300-330º 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 10th 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 long-term 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 (5th-30th June 2020) at our measurement container, the soil moisture values were relatively low with an average of 0.3 ± 0.03 m^3 m^{-3}. 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 4th June 2020, the soil moisture values in most areas in the Eifel Forest were lower than 0.3 m^3 m^{-3} (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.

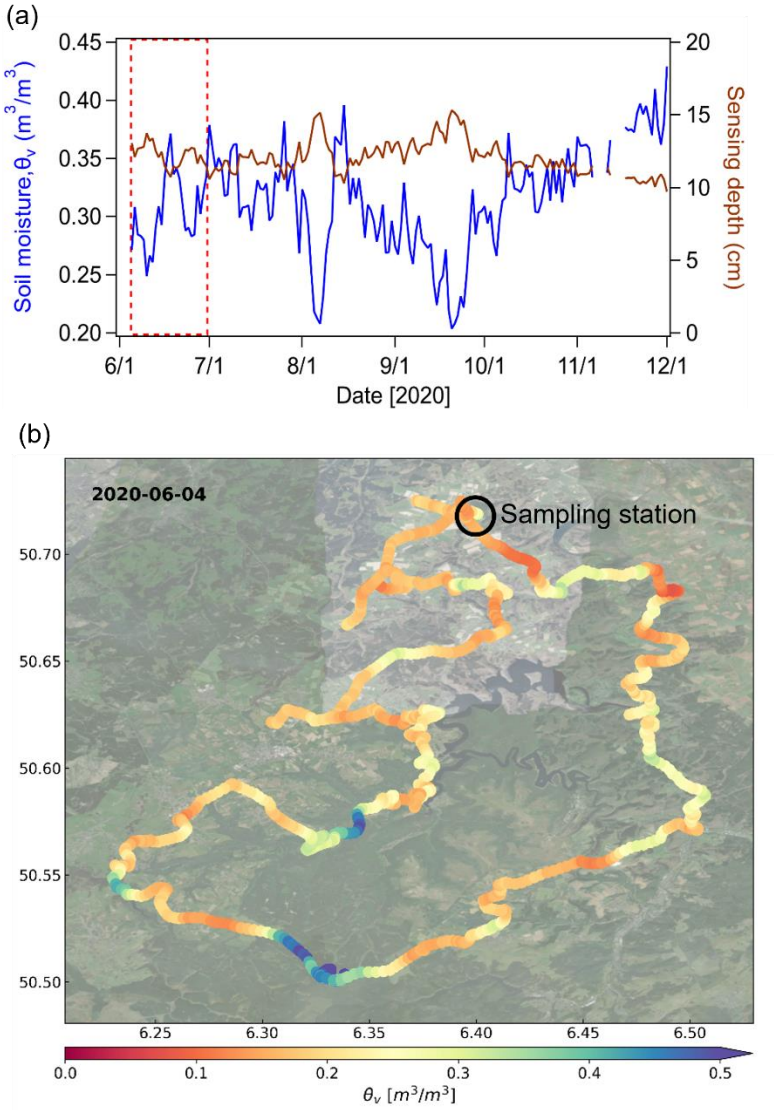

Figure S7. (a) Time series of daily soil moisture (θ_v) and sensing depth measured by a cosmic ray neutron sensor (CRNS) which was located ~ 150 m southwest of our sampling site. The red dashed box shows the concurrent sampling period at our measurement

container during 5th-30th June 2020; (b) Spatial distribution of soil moisture in the northern Eifel derived from the measurement by a CRNS rover during 4th 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 ºC for three consecutive measurement days. During the high-T episode, the daily maximum temperature exceeded 25 ºC for three consecutive measurement days.

Lines 297-302: "During the entire measurement campaign, we observed two characteristic episodes, Episode 1 (0:00 9th of June to 0:00 12th of June) and Episode 2 (12:00 23rd of June to12:00 26th of June), for different meteorological conditions. During

Episode 1, the daily maximum temperature remained below 20 ºC for three consecutive measurement days. During Episode 2, the daily maximum temperature exceeded 25 ºC 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 ± 0.54 ppb, slightly higher than that previously reported in a Norway spruce-dominated forest (0.32 ± 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 ± 5.3 ppb) in this study was also higher than that reported in a Norway spruce-dominated forest (0.50 ± 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 0-120º for the intact forest, 120-240º for the clear-cut area, 240-310º for the biogas power plant and 310-330º for the residential area of the village. No winds were coming from the sectors of 330-360º 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_3 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:

$$F_p = \frac{C_{p,i}}{C_{g,i}+C_{p,i}} \quad (3)$$

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 α-pinene and β-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% α-pinene and β-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 α-pinene and β-pinene. We have revised this sentence as follows.

Lines 670-672: "It is reasonable to assume that these monoterpenes are mainly α-pinene and β-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.

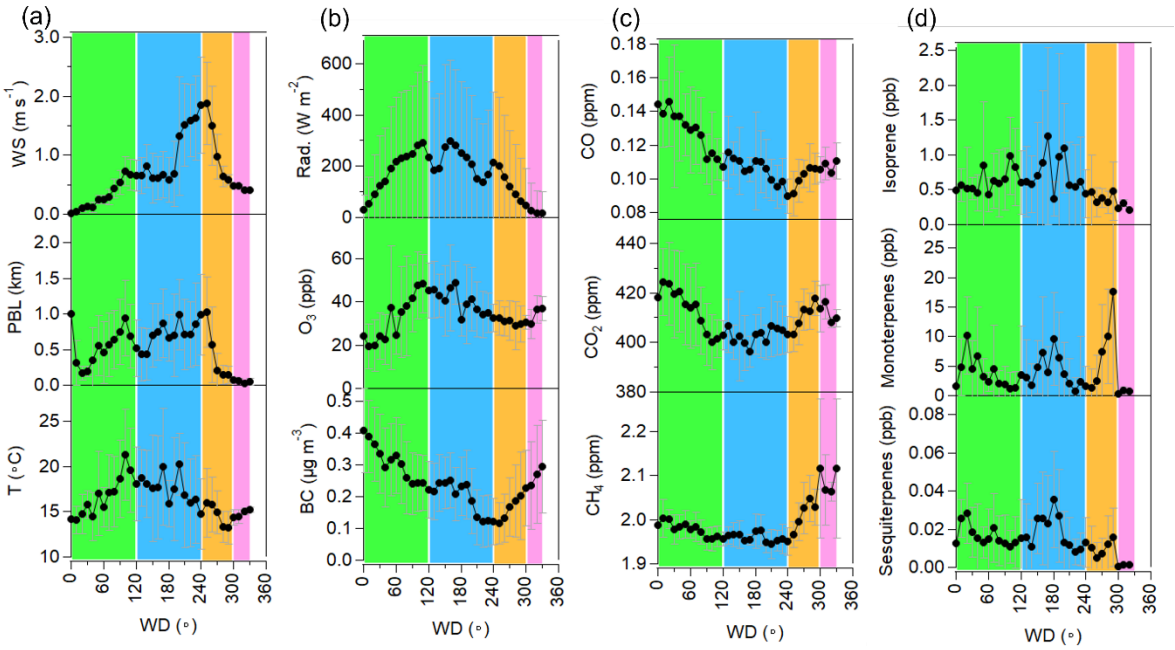

Figure 5. Variations of (a) wind speed (WS), planetary boundary layer (PBL) and ambient temperature; (b) global radiation, O_3 and BC mass concentrations; (c) mixing ratios of CO, CO_2 and CH_4 and (d) mixing ratios of isoprene, monoterpenes and sesquiterpenes as a function of wind direction (WD). The black dots and whiskers represent the mean values and standard deviations in each WD bin of 10º. Data within the WD1 of 0-120º is influenced by an intact forest area (light green), the WD2 of 120-240º is influenced by a clear-cut area (light blue), the WD3 of 240-300º is influenced by a biogas power plant (yellow) and the WD4 of 300-330º 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.

Response to Referee #2

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**.

General Comments

This study presents a detailed investigation into the real-time measurements of biogenic volatile organic compounds (BVOCs) and their oxidation products in both gas and particle phases in a stressed pine forest near a biogas power plant. The authors performed comprehensive measurements using two advanced mass spectrometers and analyzed the influence of various factors including meteorology, local emissions, and chemical transformation processes. This study provides valuable information, but it has a more limited scope than typical research articles. Several major and minor comments need to be addressed before the manuscript can be considered for publication.

Major Comments

The authors emphasize in the title and discussion that this forest is stressed. However, there are no details about the nature of this stress (e.g., when it occurred, to what extent, any dead trees, etc.). A discussion about potential changes in emissions due to this stress would be beneficial.

Response: We have provided more details about the stress status of the Eifel Forest in the subsection of "2.1 sampling site". Moreover, we have included the discussion on meteorological and physical (e.g., leaf area and soil moisture) conditions during the sampling period, emphasizing that the Eifel Forest was under stress during our measurement period.

Lines 116-134: "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 ~2.5 ± 0.02 m^2 m^{-2} based on the ERA5 reanalysis data. The soil moisture was measured to be 0.3 ± 0.04 m^3 m^{-3} at a station located ~150 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 m^3 m^{-3}) in most areas covering our sampling site (Fig. S7). Therefore, the Eifel Forest was under relatively dry condition during our measurement period."

The authors identified two organic acid factors using PMF based on VOCUS-PTR data. However, these could be fragments of larger parent ions and not necessarily acids. Additionally, the choice of identifying 6 factors instead of 5, 7 or more needs justification. Figure S6d suggests that 6 factors may not fully explain the measured signals. For source apportionment, the correlation analysis between the factor and its dominating species seems unnecessary and does not support source identification convincingly.

Response: We have provided the time series of 5-7-factors from the PMF analysis in the revised supplement. Compared to the 5-factor solution, a new factor F6 was resolved in the 6-factor solution. However, with 7 or more factors, we were unable to separate the terpene-dominated factor into two distinct sources related to biogenic emissions and BPP emissions. Instead, the 7-factor or higher-factor solutions led to factor splitting, resulting in additional uninterpretable factors. Therefore, we have retained the 6-factor solution as the most interpretable result.

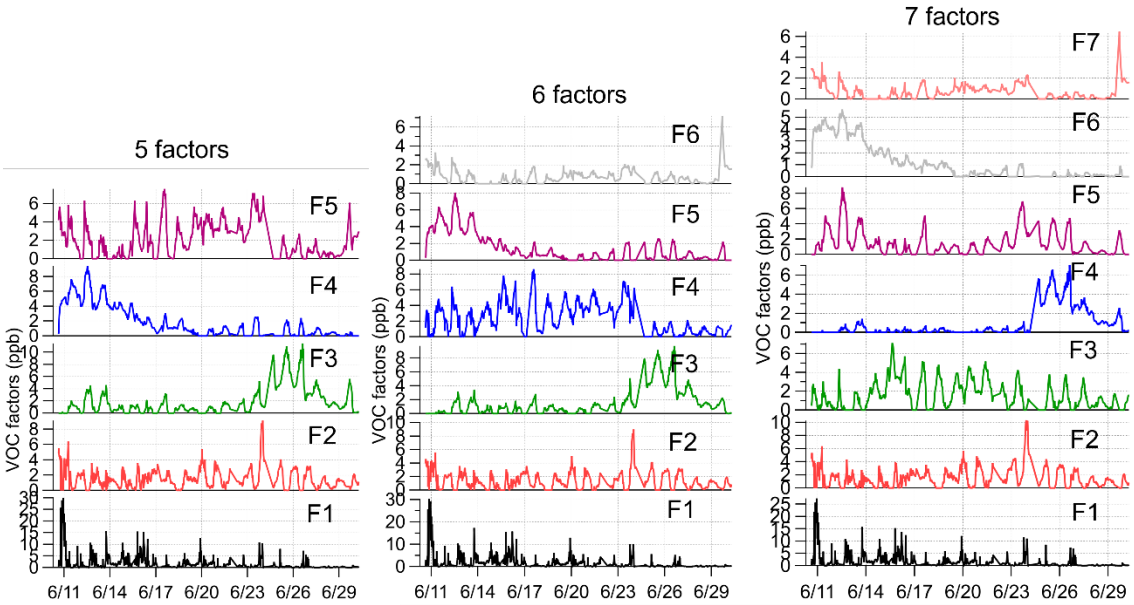

Figure S5. Time series for 5-7 VOC factors resolved from the PMF analysis of Vocus-PTR-ToF-MS data. Compared to the 5-factor solution, a new factor F6 was resolved in the 6-factor solution. Further increasing factor number to 7 only led to the factor splitting, resulting in uninterpretable factor time series.

It is important to avoid assigning factors solely based on correlation analysis between the factor and its dominating species. To ensure a robust and interpretable result, we have rephrased the source apportionment of VOCs, identifying the factors based on their profiles and temporal variations. We have revised these two original factors related to organic acid to two factors related to BVOC oxidation during daytime (day-SecVOC2 and day-SecVOC3).

Lines 535-570: "In this study, we also resolved three factors related to the oxidation of BVOCs during daytime denoted as day-SecVOC1, day-SecVOC2 and day-SecVOC3. The factor profile of day-SecVOC1 was characterized with high fractions of acetic acid ($C_2H_5O_2^+$) as well as isoprene and its oxidation products (e.g., $C_5H_9^+$, $C_4H_7O_{1\text{-}4}^+$ and $C_5H_9O_{2\text{-}4}^+$). This factor was also dominated by stronger oxidized products of monoterpenes with oxygen atom numbers >3 (e.g., $C_{10}H_{17}O_{4\text{-}5}^+$) compared to other factors in the higher mass range. The diurnal variations of day-SecVOC1 factor showed high concentrations during daytime. Therefore, the day-SecVOC1 factor can be mainly attributed to the photochemical oxidation of isoprene and monoterpenes during daytime. Li et al., (2021) resolved one factor representing isoprene and its oxidation products and another factor representing stronger oxidized products of monoterpenes from the binPMF analysis for a low-mass (*mz*50-200) and a high-mass range (*mz*201-320), respectively, for two European forest sites. They found that these two factors had a similar diurnal pattern with high daytime concentrations. In our study, we performed the PMF analysis for the full mass range (m/z40-220) of the major VOC ions and resolved the day-SecVOC1 factor containing high fractions of oxidized products of isoprene and monoterpenes. This suggests that isoprene oxidation products and higher oxidized products of monoterpenes were mainly related to the daytime oxidation processes. The factor profiles of both day-SecVOC2 and day-SecVOC3 were characterized with high fractions of acetone ($C_3H_7O^+$) and acetic acid ($C_2H_5O_2^+$).

Acetone and acetic acid could be contributed by biogenic and anthropogenic secondary sources (Khare et al., 1999; Jacob et al., 2002). The factor profile of day-SecVOC2 also had high fractions of $C_2H_7O_3^+$ (acetic acid water cluster), $C_3H_7O_2^+$ (propionic acid), $C_4H_9O_2^+$ (butyric acid) and $C_3H_9O_2^+$ (e.g., propylene glycol). These gaseous organic acids could be formed from the oxidation of BVOCs like monoterpenes (Friedman and Farmer, 2018). In addition, the factor profile of day-SecVOC3 showed higher fractions of $C_4H_7O_4^+$ (e.g., succinic acid) compared to other factors. The time series of day-SecVOC3 showed the highest correlations with $C_2H_7O_3^+$ (acetic acid water cluster, r = 0.79), $C_2H_5O_2^+$ (acetic acid, r = 0.63) and $C_3H_9O_3^+$ (propionic acid water cluster) compared to other factors. The time series of day-SecVOC3 factor also showed strong correlations with $C_4H_6O^+$ (r = 0.90, **Fig. S10**) and $C_2H_5O_3^+$ (r = 0.89), which can be assigned as the isoprene oxidation product as deprotonated $C_4H_7O^+$ (MVK+MACR) and glycolic acid, respectively. In addition, O_3 was only weakly correlated with day-SecVOC2 (r = 0.27), but much better correlated with day-SecVOC3 (r = 0.57). Moreover, a better correlation was found between O_3 and the sum of these two factors (r = 0.70). The diurnal variations of both factors showed higher concentrations during daytime. Based on these results, we identified day-SecVOC2 and day-SecVOC3 as representing low-molecular weight oxygenated organic compounds produced from the daytime photooxidation of BVOCs."

The conclusion about the impact of relative humidity (RH) on gas-to-particle phase partitioning should consider the influence of temperature changes (about 10-15°C difference), which could not be excluded here.

Response: We agree. The ambient temperature was anticorrelated with RH. Therefore, increasing RH and decreasing temperature promoted the gas-to-particle partitioning of these weakly oxidized monoterpene products. We have revised this conclusion in the abstract correspondingly.

Lines 39-41: "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."

Minor Comments

More details about the tree species are needed.

Response: We have added the information of tree species.

Line 98: "Norway spruce (*Picea abies* (L.) Karst.)"

Lines 54-55 & 390-394: Discuss the temperature and light dependency of monoterpene emissions. Are they emitted in higher amounts during the daytime? The authors should discuss the different synthesis, storage, and emission mechanisms of isoprene (de-novo) and monoterpenes (mainly pool emissions from boreal pines).

Response: Thank you for the comment. Yes, the emissions of monoterpenes are higher for Norway spruce-dominated forest during daytime due to higher temperatures and radiation. We have included the temperature and light dependency of monoterpene emissions in the introduction. Although we cannot demonstrate the impact of different emission mechanism on isoprene and monoterpenes in this study, we have mentioned this possibility in the revised manuscript.

Lines 64-73: "The diurnal pattern of isoprene concentrations in forests shows typically higher values during daytime (Yáñez-Serrano et al., 2021; Li et al., 2020; Hakola et al., 2012), since isoprene emissions increase with temperature and sunlight intensity as

result of increased de-novo production and direct release. In contrast, monoterpenes are mainly released from storage pools of boreal pines. The emissions and composition 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). It has been reported that these stresses can alter the emissions of BVOCs, especially of terpenoids (Ghimire et al., 2016; Jaakkola et al., 2023; Byron et al., 2022)."

Line 133: Add the diameter of the sampling tube (also for other relevant sections).

Response: We have added the diameter for the sampling tube in the revised manuscript.

Lines 154-155: "perfluoroalkoxy tube (1/4 inch inner diameter)"

Lines 227 & 302-304: Clarify how many compounds were detected and identified by both instruments, and how many were excluded from further analysis due to low signal. Explain what is meant by "missing".

Response: With the Vocus-PTR-ToF-MS measurement, 287 VOC ion peaks within the mass range of m/z 40–445 were quantified after background correction. Following selection, 157 VOC ion peaks with assigned chemical formulas (primarily mainly $C_xH_y^+$ and $C_xH_yO_z^+$) were used for the PMF analysis. For the CHARON-PTR-ToF-MS measurement, 939 ion peaks were automatically identified using IDA software, and 388 of these were assigned chemical formulas (mainly $C_xH_y^+$ and $C_xH_yO_z^+$). After background correction, 112 VOC ions measured by the CHARON-PTR-ToF-MS were considered for comparison with those simultaneously measured by the Vocus-PTR-ToF-MS.

We have provided this information in the revised supplement **S2, Lines 56-57:** "With the Vocus-PTR-ToF-MS measurement, 287 VOC ion peaks within the mass range of m/z 40–445 were quantified after background correction. Following selection, 157 VOC ion peaks with assigned chemical formulas (primarily mainly $C_xH_y^+$ and $C_xH_yO_z^+$) were used for PMF analysis. For the CHARON-PTR-ToF-MS measurement, 939 ion peaks were automatically identified using the IDA software, and 388 of these ions were assigned with chemical formulas (mainly $C_xH_y^+$ and $C_xH_yO_z^+$). After background correction, 112 VOC ions measured by the CHARON-PTR-ToF-MS were considered for comparison with those simultaneously measured by the Vocus-PTR-ToF-MS."

Missing data is misleading since we mean the data below the limit of detection. We have provided the detailed calculation method of data uncertainties in the revised manuscript.

Lines 251-256: The uncertainties were calculated with the following equations:

$$Unc. = \begin{cases} LOD \times \frac{5}{6} & conc \leq LOD \quad (1) \\ \sqrt{LOD^2 + (Error\ fraction \times conc.)^2} & conc. > LOD \quad (2) \end{cases}$$

where the concentrations of a VOC ion below the limit of detection (LOD) were replaced with half of the LOD and the associated uncertainties were set to 5/6 of the LOD using the Equation 1. The uncertainties of a VOC ion above the LOD were calculated using the Equation 2, assuming an error fraction of 10%.

Line 229: The explanation for excluding C4H9+ is unconvincing. Figure S5 shows a high contribution of C4H9+ from 6/11 to 6/14, but a low contribution outside this period despite

relatively stable TVOCs signal (by CHARON-PTR). Was this due to instrument performance?

Response: Yes. The significant variation of $C_4H_9^+$ was attributed to the instrument performance of the Vocus-PTR-ToF-MS. We only observed the large contribution of $C_4H_9^+$ to the VOCs measured by the Vocus-PTR-ToF-MS during the measurement period of 6/11 to 6/14. During other measurement periods, the values of $C_4H_9^+$ were mostly below the detection limit. Therefore, we excluded this signal from the PMF analysis.

Lines 292-300: Discuss the potential impacts of fragmentation on the mass and O:C and H:C ratios detected by CHARON-PTR.

Response: A previous study has reported that the fragmentation of organic compounds in the CHARON-PTR-ToF-MS can result in a negative bias in the determination of bulk organic aerosol parameters. Therefore, the average O:C ratio of bulk OA measured by the CHARON-PTR-ToF-MS was lower than that measured by the AMS. We have added one sentence to clarify this point.

Lines 356-358: "Please note that the fragmentation of organic compounds in the CHARON-PTR-ToF-MS may result in lower average O:C values of bulk OA compared to those measured by the AMS (Leglise et al., 2019)."

Lines 310-312: It is also common to see C5-C8 compounds in boreal forest environments as oxidation products from monoterpenes. Compare these results with more field observations using CIMS with other reagent ions that cause less fragmentation and provide some conclusions.

Response: We agree that the oxidation of monoterpenes can produce C5-C8 compounds. Unfortunately, this study lacks concurrent measurements with a CIMS using a soft ionization source (e.g., iodide). To the best of our knowledge, no measurements with the CHARON-PTR-ToF-MS or other CIMS have been available for specific C5-C8 compounds in the boreal forest environment. Therefore, we are unable to make such comparisons. In the future, we plan to make such comparisons by concurrently measuring of BVOC oxidation products with the CHARON-PTR-ToF-MS and the FIGAERO-iodide-CIMS in field observations.

Line 316: Have you done any correlation analysis between the parent ions and their potential fragment ions? How strong are these correlations?

Response: Yes, we have performed correlation analysis between parent ions and their potential fragment ions for several organic compounds (e.g., $C_8H_{13}O_4^+$, $C_9H_{15}O_3^+$ and $C_{10}H_{17}O_3^+$) in the particle phase measured by the CHARON-PTR-ToF-MS. Good correlations were observed between $C_8H_{13}O_4^+$ (norpinic acid) and $C_8H_{11}O_3^+$ ($r = 0.97$) as well as between $C_9H_{15}O_3^+$ (norpinonic acid) and $C_9H_{13}O_2^+$ ($r = 0.73$). However, no correlation was found between $C_{10}H_{17}O_3^+$ (cis-pinonic acid) with $C_{10}H_{15}O_2^+$, likely due to the strong fragmentation of cis-pinonic acid, resulting in low concentrations of the parent ion $C_{10}H_{17}O_3^+$. In addition, we found a good correlation for $C_{10}H_{15}O_2^+$ and $C_4H_7O^+$ ($r = 0.72$), both of which were likely produced from the fragmentation of cis-pinonic acid.

We have added this figure in the supplement (now Figure S9).

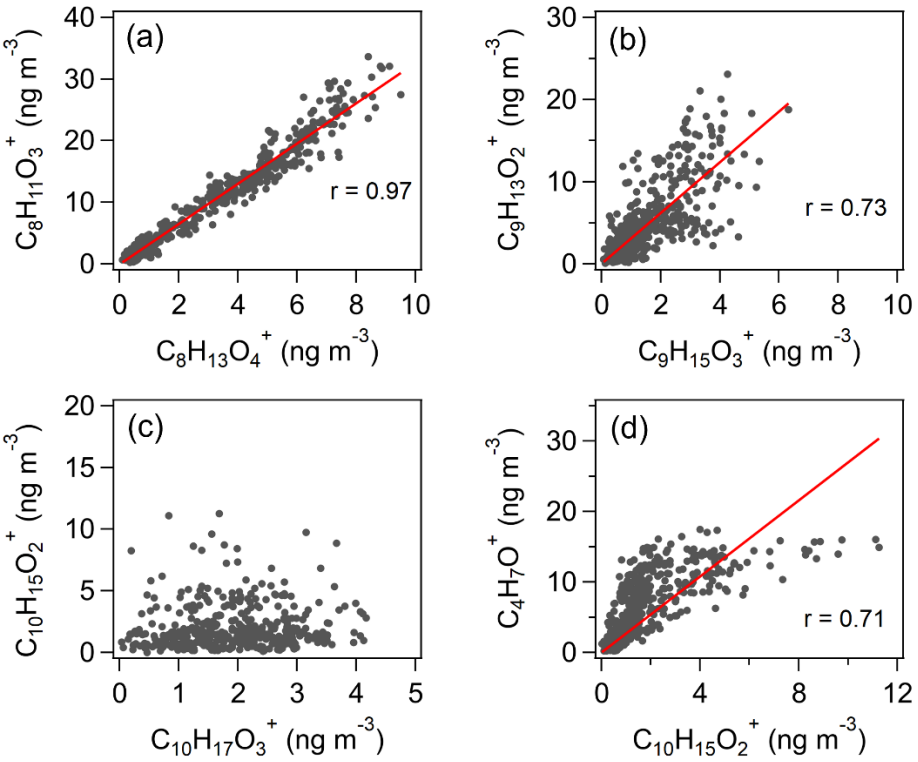

Figure S9. Scatter plots of parent ions and their potential fragment ions: (a) $C_8H_{13}O_4^+$ (norpinic acid and its isomers) vs. $C_8H_{11}O_3^+$; (b) $C_9H_{15}O_3^+$ (norpinonic acid and its isomers) vs. $C_9H_{13}O_2^+$ and (c-d) $C_{10}H_{17}O_3^+$ (cis-pinonic acid and its isomers) vs. $C_{10}H_{15}O_2^+$ and $C_{10}H_{15}O_2^+$ vs. $C_4H_7O^+$.

Lines 598-599: Are there also spruce and beech trees at the sampling site?

Response: Yes. Our sampling site was located adjacent to an intact forest stand dominated by Norway spruce.

Technical Corrections

Line 391: Add a space between "values" and "during".

Response: Added.

SI

Figure S9, panel (b): Confirm if the isoprene signal was also multiplied by 10, as in panel (a). Double-check the legend.

Response: The legends are correct in both panels. The scale of y axis in panel (a) is larger than that in panel (b), so the isoprene signal was scaled by a factor of 10 in panel (a) for a better visualization.

Line 155: Correct to "pink and grey" instead of "grey and pink".

Response: This figure is removed.

Response to Referee #3

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**.

General comment

The study by Song et al. presents atmospheric observations of VOCs and aerosol composition at an interesting site in Germany, influenced by a nearby biogas power plant (BPP), a temperate forest, and a local village. This setting offers a unique opportunity to disentangle the contributions of these sources to ambient VOC composition and to assess their impact on local organic aerosol loading and atmospheric chemistry processes. The authors employed state-of-the-art analytical instrumentation for VOCs (VOCUS-PTR-ToF-MS) and aerosol composition (Ionicon PTR-ToF-MS coupled with CHARON), alongside a comprehensive array of gaseous, particulate, and meteorological measurements. Additionally, they conducted PMF analysis on 157 VOCs, creating a robust framework for both source identification and atmospheric impact assessment. Despite the evident efforts behind this study, its scientific conclusions are hindered by generalized statements that fall short in communicating a clear and novel message. However, the technical aspects of the paper are exceptionally well-articulated and valuable for future users of the equipment. Given the technical strengths and the uniqueness of the site, this study is valuable for the literature, but the following comments should be addressed before it is considered for publication.

Major Comments

1. The scientific focus of the study needs substantial improvement, and a clear conclusion should be articulated. While the results section is rich in information, it lacks flow and a cohesive scientific message that ties the observations together.

Response: In this study, we characterize BVOCs and their oxidation products in a stressed Norway spruce-dominated forest using online mass spectrometry, focusing on how meteorological conditions, source emissions, and chemical oxidation influence their temporal variations. In the revised manuscript, we present our observations in a coherent flow. The first section provides a general overview of our measurements. The second section examines the impacts of meteorological conditions, biogenic and BPP emissions on BVOC variations in detail. In the third section, we conduct a PMF analysis to distinguish the contributions of different sources and oxidation processes to BVOCs. In the last section, we discuss the temporal variation of BVOC oxidation products in both the gas and particle phases, as well as the role of meteorological conditions in their partitioning processes. We have improved the conclusions to highlight the most important findings from this study.

2. The PMF analysis requires further consideration. The study appears to be designed to distinguish the chemical fingerprints of the BPP from the forest and other sources, such as anthropogenic emissions from the nearby village. However, the identification of a factor labeled 'terpenes' suggests that the two dominant sources at this location were not successfully separated. Figure S6a-b indicates that alternative solutions were possible, and the choice of the 6-factor solution may not be as robust as implied. Moreover, additional analyses should be conducted; it is common practice to correlate factors with

external variables, yet only vague correlation values (r) are provided here. I recommend that the authors reconsider their PMF solution and potentially re-run the analysis with a more robust setup, such as applying stricter criteria for data inclusion (e.g., considering thresholds greater than 20% for missing values (L227)).

Response: We performed the PMF analysis not only to distinguish the sources but also identify different chemical oxidation processes contributing to the VOCs. We were unable to separate a factor dominated by the terpenes into two distinct sources of biogenic emissions and BPP emissions. The source profile of BPP emissions might resemble that of terpenes from biogenic emissions. In the revised manuscript, we have defined this factor as terpene-dominated. Furthermore, we combined the meteorological data to demonstrate the relative importance of biogenic emissions and BPP emissions for different measurement periods. We applied strict criteria to select the most abundant species, ensuring a robust PMF analysis setup. We have provided the time series of 5- to 7-factors from the PMF analysis in the revised supplement. Compared to the 5-factor solution, a new factor F6 was resolved in the 6-factor solution. However, with 7 or even more factors, we were unable to separate the terpene-dominated factor into two sources related to biogenic emissions and BPP emissions respectively. Instead, 7-factor or more-factor solutions led to factor splitting, resulting in additional uninterpretable factors. Therefore, we have retained the 6-factor solutions as the most interpretable result. As suggested, we also attempted to include more VOC species in the PMF analysis, considering thresholds of more than 20% for missing values. However, this did not enhance our ability to distinguish the factors or provide additional insights into the sources and chemical processes contributing to the VOCs.

Lines 494-507: "The first factor profile was dominated by the monoterpene parent ion ($C_{10}H_{17}^+$) and its fragment ion ($C_6H_9^+$) (Tani et al., 2003; Kari et al., 2018). Furthermore, this factor was characterized with higher fraction of monoterpenoids such as $C_{10}H_{17}O^+$ (camphor or monoterpene oxide) and $C_{10}H_{19}O^+$ (linalool or monoterpene water cluster) in high mass range (*m/z*140-230) (Li et al., 2020). These monoterpenoids can be emitted by leaves and flowers directly (Joó et al., 2010). Therefore, we define this factor as a terpene-dominated factor. As discussed before, the variations of monoterpene concentrations were influenced by the BPP emissions and biogenic emissions depending on the wind directions. In this study, PMF analysis could not separate the relative contribution of biogenic emissions and BPP emissions to monoterpenes directly. This is probably due to the source profile of BPP emissions dominated by monoterpenes that is similar to that of biogenic emissions. Based on WD analyses, this factor was expected to be mainly associated with the biogenic emissions when the winds were coming from the forest. In contrast, when the winds were coming from the BPP, this factor was significantly contributed by the BPP emissions."

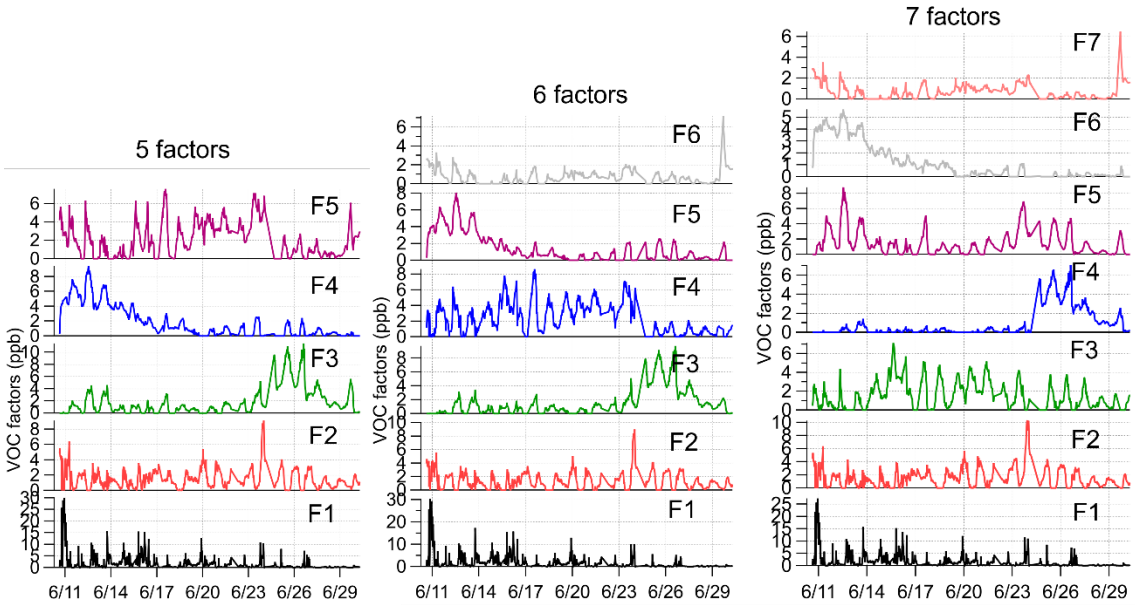

Figure S5. Time series for 5-7 VOC factors resolved from the PMF analysis of Vocus-PTR-ToF-MS data. Compared to the 5-factor solution, a new factor F6 was resolved in the 6-factor solution. Further increasing factor number to 7 only led to the factor splitting, resulting in uninterpretable factor time series.

Specific Comments

L1. The current title does not accurately reflect the content of the paper. The authors assume a stressed forest (L90), but there is no evidence provided to support that the forest ecosystem was under stress during the measurement period. While droughts, heatwaves, and beetle infestations are known to occur in most temperate forests, claiming 'stress' in this context is an unsupported assumption. This claim should be corrected throughout the paper.

Response: The Eifel Forest is dominated by Norway spruce (*Picea abies*), thus it should be regard as a temperate forest. Correspondingly, we have also changed the title of the manuscript.

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

In the revised manuscript, we have also provided additional information to support the claim that the Eifel Forest was stressed during our measurement period.

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 ~2.5 ± 0.02 m^2 m^{-2} based on the ERA5 reanalysis data. The soil moisture was measured to be 0.3 ± 0.04 m^3 m^{-3} at a station located ~150 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 m^3 m^{-3}) in most areas covering our sampling site (Fig. S7). Therefore, the Eifel Forest was under relatively dry condition during our measurement period."

L35-37. This conclusion is rather weak, especially considering the extensive use of highly sophisticated analytical equipment in this study. Please strengthen this conclusion in line with the major comments provided above.

Response: We have strengthened the discussion and conclusions accordingly and have revised the abstract to highlight the major findings of this study. The revised abstract is now:

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 ± 0.5 µg m^{-3}, consisting mainly of semi-volatile monoterpene oxidation products. The average mixing ratios of isoprene (0.58 ± 0.54 ppb) and monoterpenes (2.5 ± 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."

L53. You may consider citing two recently published, relevant papers: Weber et al., 2023 (https://www.nature.com/articles/s41467-022-34944-9) and Bourtsoukidis et al., 2024 (https://www.nature.com/articles/s43247-023-01175-9).

Response: We have cited these two recently published and relevant papers.

L74-75. The study by Penuelas and Staudt (2010; https://doi.org/10.1016/j.tplants.2009.12.005) is more appropriate for citation at this point.

Response: This study has been cited correctly in the revised manuscript.

L90, L100, etc. Please remove all comments on stress.

Response: Removed as suggested.

L213. You may consider adding the following relevant studies that deal with PMF analysis on PTR data: Desservetazz et al. (2023; https://doi.org/10.1016/j.scitotenv.2023.166592) and Jain et al. (2023; https://doi.org/10.5194/acp-23-3383-2023).

Response: We have cited these two recently published and relevant papers.

L263. How do the PMF factors relate to these distinct events?

Response: The terpene-dominated factor also increased during events with CH_4 spikes. In contrast, the other PMF factors, which were mainly associated with the secondary oxidation of anthropogenic and biogenic VOCs, showed no significant changes during these distinct events.

L268, L280. Please also compare the findings to other German forests.

Response: We have also compared the BVOC concentrations to other studies in different forests including a Norway spruce-dominated forest in central Germany.

Lines 314-327: "During the entire campaign, the average mixing ratios of isoprene was 0.58 ± 0.54 ppb, slightly higher than that previously reported in a Norway spruce-dominated forest (0.32 ± 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 ± 5.3 ppb) in this study was also higher than that reported in a Norway spruce-dominated forest (0.50 ± 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)."

L282-286. Consider moving this part to the Methods section.

Response: We consider that it is important to discuss the limitation of sesquiterpene quantification by the PTR-ToF-MS in this study. Therefore, we still keep this part with some modifications as suggested by another reviewer.

Lines 334-340: "It should be noted that the quantification of sesquiterpenes is affected by the degree of sesquiterpene fragmentation inside the PTR-ToF-MS, which can vary significantly depending on the instrument setting (Kim et al., 2009; Kari et al., 2018). In addition, sesquiterpenes may experience wall losses inside the inlet tubing and the instrument, and have low transmissions (Li et al., 2020). Due to a lack of a dedicated sesquiterpene calibration in this study, the quantification of sesquiterpenes measured by the PTR-ToF-MS can be regarded as the lower limit without the consideration of fragmentation."

L340. The term "PBL" appears for the first time here, so it needs to be defined. Additionally, the connection between wind direction and boundary layer height is unclear, leading to a weakly supported statement in L341-342.

Response: PBL should be defined as the abbreviation of planetary boundary layer. We have defined it correctly in the revised manuscript. Lower wind speeds and PBL heights contribute to less dilution of the CH_4 emitted from the biogas power plant.

Lines 400-402: "We observed that the mixing ratios of CH_4 increased significantly in the WD-BPP along with the decrease of wind speeds and PBL heights and corresponding weaker dilution"

L343-349. This entire section needs to be rewritten for improved clarity. Several statements are vague and weakly supported by the data. For example, while it is suggested that temperature is not the main driver, there is no discussion of other environmental factors within the forest that could be influencing the results. A more thorough examination of these potential drivers is necessary.

Response: Based on the wind and geographical conditions around the sampling site, we divided the entire measurement period into four wind direction (WD) sectors. Correspondingly, we have rewritten this section to provide a detailed discussion of the factors driving BVOC variations within each WD sector.

Lines 393-440: "We firstly analyzed the variations in the mixing ratios of gas species as a function of wind direction (WD) with a bin of 10° (**Fig. 5**). According to the wind and geographical conditions around the sampling site (**Fig. 1b**), we divided the entire measurement period into four WD sectors including WD-forest (0-120°)), WD-cut (120-240°), WD-BPP (240-300°) and WD-village (300-330°). Within the sectors of WD-forest and WD-cut, the sampling site was influenced by an intact forest dominated by Norway spruce and a clear-cut area, respectively. In contrast, the sampling site was influenced by the winds coming from the BPP and the village residential areas within the sectors of WD-BPP and WD-village, respectively. We observed that the mixing ratios of CH_4 increased significantly in the WD-BPP along with the decrease of wind speeds and PBL heights and corresponding weaker dilution. In contrast, constantly low mixing ratios of CH_4 were observed in the WD-forest and WD-cut even when both wind speeds and PBL decreased significantly. The results indicate that the enhancement in CH_4 mixing ratios in the WD-BPP was

mainly attributed to the BPP emissions. In addition, CH_4 mixing ratios remained higher for WD-village, which was likely associated with the anthropogenic emissions from the village residential areas. We also observed a significant increase of monoterpene mixing ratios in the WD-BPP along with lower ambient temperature (~15 °C) and decreasing radiation. This suggests that the increase of monoterpene mixing ratios in the WD-BPP was due to BPP emissions rather than biogenic emissions. In contrast to CH_4, monoterpenes showed very low values in the WD-village, suggesting a minor contribution of anthropogenic emissions from the village residential areas to monoterpenes.

We also observed significant variations in the mixing ratios of isoprene, monoterpene and sesquiterpene in the WD-forest and WD-cut, likely associated with changes in meteorological conditions, biogenic emissions and/or chemical oxidation processes. Specifically, the mixing ratios of monoterpenes and sesquiterpenes increased in the WD-forest of 0-30° but isoprene showed no significant change. The meteorological condition in the WD-forest of 0-30° was characterized by low ambient temperature, low wind speed and shallow PBL during nighttime. Unlike isoprene, monoterpenes and sesquiterpenes can still be released from the Norway spruce in the dark (Van Meeningen et al., 2017). Monoterpenes and sesquiterpenes could accumulate during nighttime in the WD-forest of 0-30° as a result of low concentrations of atmospheric oxidants like O_3. Besides, we observed an increase of isoprene mixing ratios in the WD-forest of 60-120° which was coincided with the increases of wind speed, PBL, ambient temperature and radiation during daytime. In contrast, monoterpenes and sesquiterpenes showed low mixing ratios of in the WD-forest of 60-120°. It is expected that higher temperature and radiation enhanced biogenic emissions, resulting in the increase of isoprene mixing ratios. However, lower mixing ratios of monoterpenes and sesquiterpenes were likely attributed to the photochemical oxidation exceeding their biogenic emissions. The strong photochemical oxidation processes were characterized by higher radiation and O_3 mixing ratios in the WD-forest of 60-120°. In the WD-cut of 120-180°, we observed simultaneous increase of isoprene, monoterpenes and sesquiterpenes, which were associated with enhanced biogenic emissions induced by higher temperature. Conversely, simultaneous decrease of isoprene, monoterpenes and sesquiterpenes mixing ratios were observed in the WD-cut of 180-240° along with high ambient temperature. Note that the sampling site were more influenced by the distant Norway Spruce trees in the WD-cut of 120-180° compared to the WD-cut of 180-240° (**Fig. 1a**). In addition, the wind speeds were significantly higher in the WD-cut of 180-240°. Therefore, the decreases in isoprene, monoterpenes and sesquiterpenes mixing ratios in the WD-cut of 180-240° were attributed to the reduced biogenic emissions of fewer Norway spruce and the dilution effect caused by higher wind speeds. The dilution effect was supported by the lowest CO mixing ratios and BC mass concentrations observed in the WD-cut of 180-240°."

L368-369. How can isoprene, monoterpenes and sesquiterpenes originate from chemical transformations?

Response: The mixing ratios of isoprene, monoterpenes and sesquiterpenes are affected by atmospheric oxidation processes.

L389. Please compare with temperate forests.

Response: We have compared the diurnal variations of isoprene with those previously observed in temperate forests dominated by the Norway spruce.

Lines 450-453: "As expected, isoprene showed higher concentrations during daytime in the biogenic-group, which is similar to the diurnal behavior of isoprene emission rate in previous observations in Norway-spruce dominated forests (Bourtsoukidis et al., 2014; Juráň et al., 2017)."

L411 and in general. It appears that you are using O_3 mixing ratios to attribute the atmospheric degradation of terpenes. What about the opposite, i.e., O_3 formation? Local terpene emissions contribute to O_3 formation, which is particularly relevant for emissions upwind of the measurement site and in relation to some of the PMF factors. This aspect should be addressed.

Response: The reaction rates of monoterpenes with O_3 are 4-5 orders of magnitude lower than those with OH radicals (Atkinson and Arey, 2003). Therefore, the daytime oxidation of terpenes is generally dominated by OH radicals rather than O_3. Higher solar radiation and higher mixing ratios of O_3 indicate a stronger photochemical oxidation during daytime of the high-T episode. We have revised this sentence accordingly.

Lines 470-474: "Meanwhile, higher radiation and constantly high mixing ratios of O_3 (40-60 ppb) were observed during daytime of the high-T episode. The photochemical O_3 production is supported by also by higher BVOC mixing ratios. However, the increasing biogenic emissions due to higher temperatures and solar radiation obviously exceeded the photochemical consumptions."

L426 & L436. A factor named 'terpenes' indicates that the entire scope of the PMF did not achieve its purpose. This may actually be considered the weakest point of the study.

Response: We were unable to distinguish the relative contributions of biogenic emissions and BPP emissions to the terpenes from the PMF analysis. This difficulty likely arises because the source profile of BPP emissions resembles that of terpene-dominated biogenic emissions. We also attempted to increase the number of factors and include additional VOC species in the PMF analysis, but these results did not help in separating these two sources contributing to the monoterpenes. We don't think this is a weak point of our study but demonstrates the limitations of PMF analysis. However, we were able to distinguish the sources not only by using the wind directions but also the temperature dependence to illustrate the relative importance of biogenic emissions and BPP emissions for the monoterpenes or this terpene-dominated factor.

L448. An R^2 value of 0.46 does not truly indicate 'well-correlated' parameters.

Response: The factor of nighttime-biogenic OVOC showed better correlations with $C_{10}H_{15}O^+$ ($r = 0.68$) and $C_{10}H_{17}O_2^+$ ($r = 0.65$) compared to other factors. We have identified the factors based on their factor profile and temporal variations rather than their correlations with the dominating species. We have deleted this sentence in the revised manuscript.

L570. Please provide a more detailed explanation of why you calculated the OA/ΔCO ratios.

Response: Since CO is relatively long-lived, normalizing the observed OA to the concurrent background-corrected CO helps minimize the impacts of uncertainties in

boundary layer dynamics. We have added more explanation on the calculation of OA/ΔCO ratios.

Lines 646-648: "CO is relatively long-lived, normalizing the observed OA mass concentrations to the background-corrected CO helps to minimize the impacts of boundary layer dynamics (De Gouw and Jimenez, 2009)."

L599. Needs citations.

Response: We have provided the citations to support this statement.

Lines 670-672: "It is reasonable to assume that these monoterpenes are mainly α-pinene and β-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)."

L616-617. As mentioned earlier, you are not dealing with a stressed forest here. However, it is worth noting that June 2020 coincided with the COVID-19 lockdowns in Germany. Can you provide any insights into the potential influence of this on your dataset? For example, was the BPP operating as usual or at a reduced capacity?

Response: Our sampling site is far from urban regions and only close to a small village of Kleinhau (population about 9000). Moreover, the wind was mainly coming from the forest or the BPP rather than the residential areas of Kleinhau during the entire measurement period. The BPP was operated as usual during our sampling period. Therefore, the potential changes of anthropogenic emissions induced by the COVID-19 lockdown should have negligible influence on our dataset.

L628-631. This statement is confusing and seems to imply that BVOC emissions are larger than their chemical sinks, which is a rather generalized comment. Please consider removing it.

Response: We have removed this generalized statement.

L640-642. This is another generalized sentence that simply states, "high temperatures and radiation will enhance BVOC emissions, which will be oxidized in the atmosphere." While this is accurate, it represents textbook knowledge and highlights the need for clearer, more specific scientific conclusions rather than relying on well-known statements.

Response: In fact, our study concludes that the diurnal variations of BVOC oxidation products were influenced by the interplay between biogenic emissions and chemical oxidation processes, both of which are enhanced by the temperature and radiation. We have rephrased the conclusions to emphasize the key findings from this study.

Lines 687-727: "4 Conclusions

In this study, we investigated the characteristics of VOCs and OA particles simultaneously measured by a CHARON-PTR-ToF-MS and a Vocus-PTR-ToF-MS at a Norway-spruce-dominated forest stressed by bark beetles and droughts close to a BPP in western Germany during June 2020. The average mass concentration of OA particles detected by the CHARON-PTR-ToF-MS was 0.8 ± 0.5 μg m^{-3}. The chemical composition of OA ions ranged between C_2 and C_{10} with oxygen atom numbers of 0-5, which were mainly attributed to the semi-volatile organic compounds formed from monoterpene oxidation. 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., 2021; Hellén et al., 2018; Bourtsoukidis et al., 2014) which may be due to stressed trees with long

lasting droughts and bark beetle infestation and differences in the meteorological conditions. Based on the WD analyses, BVOC data were categorized into two groups to distinguish the impacts of biogenic emissions from an intact forest and a clear cut (biogenic-group) and anthropogenic emissions from a BPP and a village (anthropogenic-group). The mixing ratios of CH_4 and monoterpenes showed significantly higher values in the anthropogenic-group. This was expected for CH_4, and it is also known that BPP can release high concentrations of monoterpenes during biowaste storage and fermentation processes (Salazar Gómez et al., 2016; Papurello et al., 2012). In the biogenic-group, the variations in mixing ratios of isoprene, monoterpenes and sesquiterpenes were driven by the interplay between meteorological conditions, biogenic emissions and subsequent chemical oxidation processes. Based on the PMF analysis of VOCs measured by the Vocus-PTR-ToF-MS, six factors were resolved, representing the major sources and/or products of chemical transformation processes. During the entire measurement period, TVOCs were largely composed of oxygenated organic compounds formed from the photochemical oxidation of BVOCs during daytime. However, monoterpenes and their weakly-oxidized products (e.g., $C_{10}H_{15}O_{1\text{-}3}^+$ and $C_{10}H_{17}O_{1\text{-}2}^+$) dominated the TVOCs during nighttime. These weakly-oxidized monoterpene products in the particle phase also showed higher mixing ratios during nighttime. In contrast, more-oxidized monoterpene products (e.g., $C_{10}H_{17}O_{4\text{-}5}^+$ and $C_{10}H_{15}O_{4\text{-}5}^+$) in both gas and particle phases were more abundant during daytime. By combining the gas and particle data measured by the CHARON-PTR-ToF-MS and the Vocus-PTR-ToF-MS, we found that increasing RH and decreasing temperature led to an increase in the particulate fraction of weakly-oxidized monoterpene products, consistent with the findings from recent simulation chamber studies (Surdu et al., 2023; Luo et al., 2024). Overall, this study demonstrates that the variations of BVOCs are influenced not only by meteorology and biogenic emissions but also by local anthropogenic emissions (e.g., from a BPP), and subsequent chemical transformation processes in a typical stressed European coniferous forest. The impact of soil moisture, tree species composition and tree health conditions on the variations of BVOC concentrations could not be fully addressed due to the relative short observation period. Future long-term field measurements including seasonality and detailed tree characterization are necessary to assess the impacts of droughts and bark beetle outbreaks on BVOC emissions and subsequent formation of SOA."

L649. Please specify this 'minor role'.

Response: We cannot demonstrate the impact of soil moisture in the variations of BVOC concentrations probably due to the relatively short observation period. We have rephrased this sentence to avoid any confusion.

Lines 722-724: "The impact of soil moisture, tree species composition and tree health conditions on the variations of BVOC concentrations could not be fully addressed due to the relative short observation period."

L665. While no relationship between soil moisture and BVOC emissions was demonstrated here, the data were still used for analysis. Therefore, unless there are other reasons behind this decision, I would recommend including Heye Bogena in the author list.

Response: We asked Heye Bogena to become a co-author but he suggested to be acknowledged for his contributions in the Acknowledgements section.

Technical Comments

L23 and in numerous other parts. Technically, the values reported in ppb are volume mixing ratios, not concentrations. Volume mixing ratios (such as ppb) indicate the number of molecules of a substance relative to the total number of air molecules and are independent of temperature and pressure. In contrast, concentrations refer to the mass or number of molecules per unit volume of air, which can vary with changes in temperature and pressure. Please correct this accordingly.

Response: We fully agree. We have replaced concentrations by the volume mixing ratios for the gas species in ppb throughout the revised manuscript.

L115. Please homogenize the temperature units.

Response: Corrected.

L212. The term "non-methane VOCs" is uncommon and might cause confusion. The term "non-methane hydrocarbons (NMHC)" is typically used to describe lighter compounds so in this context, it's clearer and more appropriate to simply refer to them as VOCs. I recommend removing "non-methane" and referring to these compounds as VOCs throughout the paper.

Response: We have corrected all "non-methane VOCs" to "VOCs".

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