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."
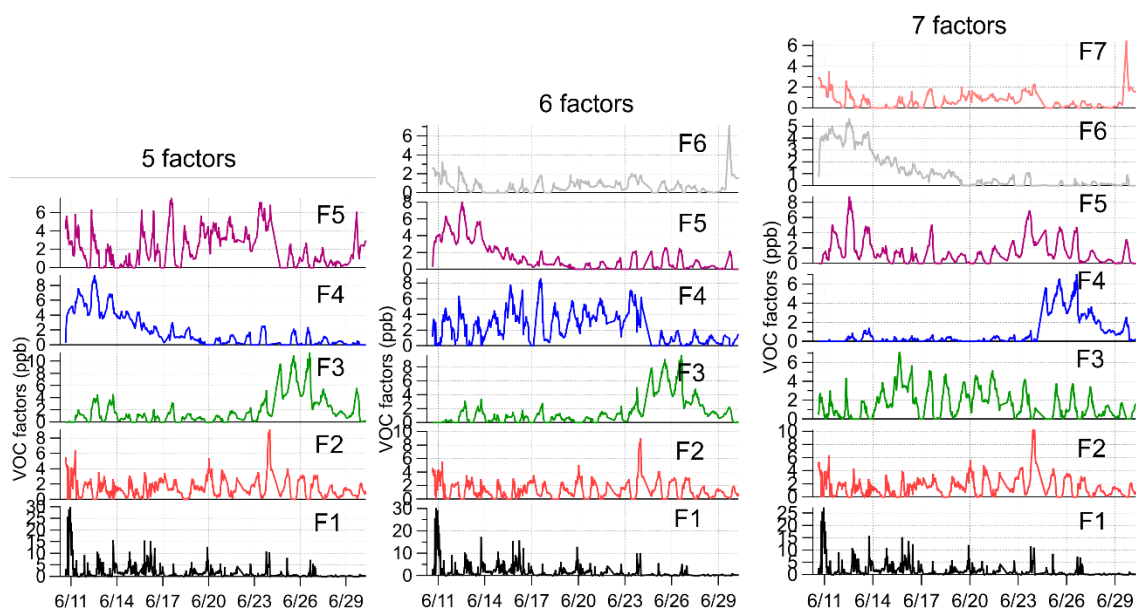
Figure S5. Time series for 5-7 VOC factors resolved from the PMF analysis of Vocus-PTR-ToF-MS data. Compared to the 5-factor solution, a new factor F6 was resolved in the 6-factor solution. Further increasing factor number to 7 only led to the factor splitting, resulting in uninterpretable factor time 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., $C_{10}H_{15}O_{1-3}^{+}$ and $C_{10}H_{17}O_{1-2}^{+}$) 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., $C_{10}H_{17}O_{4-5}^{+}$ and $C_{10}H_{15}O_{4-5}^{+}$) 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".

References

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