Responses to reviewers' comments on "Sources of organic gases and aerosol particles and their roles in nighttime particle growth at a rural forested site in southwest Germany"

Thanks to all reviewers for their evaluation and constructive comments on the manuscript. We tried to answer all questions and revised our manuscript and supplement accordingly. Point-to-point responses are displayed in blue text and changes in both revised manuscript and supplement are highlighted in red text.

Responses to Reviewer 1:

This paper presents an analysis of VOCs and organic aerosol (OA) that were present at a site in Germany, using online measurements from a variety of mass spectrometric techniques. In particular, online PTR-MS measured VOCs, and CHARON PTR-MS and AMS assessed particle composition. PMF was applied to each of these data sets. A central tenet of the paper is that the additional molecular information provided by PTR enhances the identification of sources relative to what can be learned with the AMS. The relatively weak correlation of some AMS PMF factors with CHARON PMF factors supports this claim. A strength of this paper is that PMF is applied to both gas and particle measurements using PTR-MS. Whereas PMF has been applied to AMS data extensively, there are only a few examples where it has been applied to PTR data, and fewer still (any?) that use PTR to assess both gas and particle composition. A second strength is the clear demonstration of nighttime OA growth via NO3 chemistry (up to a few nm/hour), as seen in the CHARON (and AMS) data sets. This is especially important to illustrate for a mixed use environment, where large populations (and their pollution) are located close to forests. For these reasons I strongly support publication and have only a few critical comments.

Response: We thank the reviewer for the encouraging comments. The point-to-point responses to individual comment are given below.

1. General: Although the title is strictly correct that the measurements are at a forested location, I was surprised to read in the paper that a large city, a very large coal-fired power plant, and a refinery are only a few km away. My recommendation would be to give a new title to the paper because my first impression was that this was going to be a paper on data from a relatively

remote site, dominated by only biogenic emissions. This is clearly not the case with the large, for example, aromatic input, along with the important role of NO3 chemistry.

Response: Geographically, our sampling site is located in the forest center (Figure 1). It is subjected to the input of urban emissions mainly during daytime with southwesterly winds. During nighttime, the wind direction generally shifted to the east and both wind speeds and boundary layer heights are low, thus the impact of urban emission would be minor at night. We demonstrated that the NO_3 chemistry of BVOCs plays an important role for the particle growth and mass increase during nighttime. Therefore, we would like to keep the paper title as "Sources of organic gases and aerosol particles and their roles in nighttime particle growth at a rural forested site in southwest Germany". We can assume that most readers will know that no place in Germany is really remote. To make this clearer we added information on the nearby city in the first sentence of the abstract as follows.

Lines 15-17: The composition, sources and chemical transformation of volatile organic compounds (VOCs) and organic aerosol (OA) particles were investigated during July-August 2021 at a rural forested site in southwest Germany 10 km north of the city of Karlsruhe.

2. Line 223. Small point that PTR does detect alkenes.

Response: Thanks for the comment. We revised this sentence accordingly.

Line 225: Note that PTR-ToF-MS cannot well detect some VOCs such as small alkanes and alkenes with a proton affinity lower than H_2O and/or highly oxidized organic species.

3. Line 299. Delete "with". Otherwise, this paper was very clearly written with almost no typos.

Response: Done.

4. Line 300 and thereabouts. Although the authors mention this, I think more emphasis should be given to the extremely low probability that highly oxygenated species will be detected, both with the heat of the CHARON source and with PTR ionization, i.e., the spectrum is invariably "contaminated" or impacted by oxygenated fragment ions. Does this affect the PMF solutions and interpetation?

Response: We agree that the detection of highly oxidized species by the CHARON-PTR-MS was limited by both CHARON thermo-desorption temperature and PTR ionization. The

temperature of the CHARON thermo-desorption unit was set with 150 ℃ in our study, which was sufficient for the evaporation of semi-volatile organic compounds rather than highly oxidized species. As suggested by previous studies (Riva et al., 2019), PTR-MS with H_3O^+ as the ionization ion are less sensitive to detect highly oxidized species due to fragmentation losses. We emphasize both thermo-desorption temperature and PTR ionization are limited for detecting highly oxygenated species by the CHARON-PTR-MS in the revised manuscript.

Lines 305-308: It is consistent with previous studies that CHARON-PTR-MS is suitable to measure the semi-volatile and less-oxidized organic compounds ($C_xH_yO_{1-3}^+$) in the particle phase. However, the detection of highly oxidized organic species is limited due to the thermo-desorption temperature of CHARON (150 ℃) and the fragmentation during PTR ionization.

As stated above, the contribution of highly oxygenated species to organic aerosol mass measured by CHARON-PTR-MS was minor, thus a negligible effect can be expected on PMF analysis. The CHARON-PMF factors were interpreted with high loadings of high-molecular-weight fingerprint ions (m/z>120), which mostly could be considered as the parent ions of less-oxidized organic compounds (oxygen number <5) or their major fragment ions.

5. The authors identify a primary traffic factor with a strong aromatic character and an aromatic oxygenated factor. Why refer to one as a traffic factor and not the other? In terms of source attribution, they both arise from traffic, only one is more aged than the other.

Response: We characterized a primary traffic VOC factor with high contributions by aromatic hydrocarbons and ethanol. We defined another factor as aromatic-OVOC factor because it had high contributions by oxygenated aromatic VOCs. The oxidation of aromatic hydrocarbons emitted from traffic could produce these oxygenated aromatic-VOCs. However, this factor could be influenced by the transport of urban air containing oxygenated aromatic-VOCs formed from the oxidation of aromatic hydrocarbons emitted from industrial sources (e.g., power plant and refinery) beside traffic emissions. Therefore, we think this factor should be called aromatic-OVOC factor rather than aged traffic factor in this study.

6. I think it is notable that the MOOA factor is so significant, a long way from the coast. We don't hear much about marine input in continental regions, and so I would recommend to emphasize this more, e.g. in the Abstract.

Response: The MOOA factor can significantly contribute to aerosol particle mass when the sampling was influenced by the frequent air masses from Atlantic Ocean, but the $PM_{2.5}$ mass concentrations were generally low. In our previous study (Shen et al., 2019), we investigated the origins of aerosol particles at Linkenheim during summer 2016 by using an AMS and a single-particle mass spectrometer (LAAPTOF), 2 km northwest of our sampling site. Similarly, abundant sodium chloride sea-salt particles measured by LAAPTOF and enhancement of AMS-measured marker ions ($CH_3SO_2^+$) were observed during low-$PM_{2.5}$ mass episode when the site was influenced by air masses originating from the Atlantic ocean (Shen et al., 2019). Since the abstract has a word limit we explained this in the conclusion section with the following sentence:

Lines 642-644: Please note, that the relatively large contribution of the marine factor (MOOA) with 22% ± 16% is not unexpected in central Europe since we detected substantial contributions of sea salt aerosol in a previous study at a location 2 km west (Shen et al., 2019).

7. I am surprised that the cooking factor only shows a peak at lunchtime. This is unlike behavior observed in cities with large evening signals. Why is this? In particular, is this an indication of only a very local source, e.g. from cafeterias on the site? If so, it reduces its importance as likely it will have little regional impact. This may be the reason that the AMS COA factor was not observed. Regardless, it is interesting and valuable to illustrate that it can be observed using palmitic acid as a tracer.

Response: Indeed, there is a canteen located 300 m south of the sampling site, which is only open during lunch time (11:30-14:00) at weekdays. We added this information in Section 2.1 and reformulated Section 3.2.3 accordingly.

Line 115: A canteen is located 300 m south of the sampling site on the KIT campus, which is generally open during lunch time (11:30-14:00) at weekdays.

Lines 465-479: The first OA factor was characterized with high fractions of $C_7H_9^+$, $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$. $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$ can be tentatively assigned as long-chain fatty acids. For example, $C_{16}H_{33}O_2^+$ was tentatively assigned as palmitic acid, which can be used as a tracer for cooking-related OA (Reyes-Villegas et al., 2018). Furthermore, the diurnal variation of this factor showed a peak at lunch time (12:00-14:00), which is in line with the opening hours of the canteen located 300 m south of the sampling site in the campus. Therefore, it is reasonable to assign this factor to cooking emissions. As expected, the time series of COA correlated well

with $C_{16}H_{33}O_2^+$. Note, that we cannot assign particulate $C_7H_9^+$ ion to specific organic compounds, which is likely produced from the fragmentation of oxidized organic compounds of higher mass. In this study, the COA factor was not resolved from the unconstrained AMS-PMF analysis, which is likely due to the lack of specific marker ions of cooking emissions measured by the AMS. On other hand, cooking-related oxidized species may be mixed into HOA resolved from the AMS-PMF analysis, which leads to HOA having a high O:C value (0.26) (Mohr et al., 2012).

8. Line 541 and thereabouts. It is valuable that the NO3 production rate was calculated but why not try to estimate the concentration of NO3, if the major sinks of NO3 are indeed with biogenics (which were measured)? This would be useful for others to be able to frame how likely nighttime biogenic SOA may form in their settings, which may have a different NO3 concentration.

Response: As suggested, we estimated the concentration of NO3 radicals in this study. We provided the information in the supplement and the revised manuscript correspondingly.

Lines 103-114 (Supplement): According to previous studies (Xu et al., 2015; Yu et al., 2019), the steady-state NO₃ radicals concentration [NO₃·] can roughly be estimated by the following equation:

$$[NO_3 \cdot] = \frac{k_1[NO_2][O_3]}{j_{NO_3} + k_2[NO] + \sum k_i[VOC_i]}$$

where j_{NO_3} is the NO₃ photolysis rates, k_1 and k_2 are the rate coefficients with 3.5 x 10^{-17} cm^3 molecules^{-1} s^{-1} and 2.7 x 10^{-11} cm^3 molecules^{-1} s^{-1} at 298 K, respectively, k_i is the rate constant of NO₃ reacting with VOC species. In this study, we focused on the nighttime NO₃ chemistry, thus the j_{NO_3} was assumed as zero. The data of NO and NO₂ were obtained from the Eggenstein air quality monitoring station (state environmental agency, LUBW) located ~2.5 km southwest of the sampling site. We estimated the sink of NO₃ radicals as dominated by reactions with isoprene, monoterpenes, sesquiterpenes, benzene, toluene, C₈-and C₉-aromatic hydrocarbons. We added the time series and diurnal variations of steady-state NO₃ radical concentrations in the revised supplement (Fig. S19). We observed rapid decreases of steady-state NO₃ radical levels during early nighttime, and remaining at low concentrations during the night, which is mainly due reactions with monoterpenes.

Lines 592-594: The concentrations of steady state NO$_3$ radicals were also roughly estimated in supplement Text S3. We observed rapid decreases of steady-state NO$_3$ radicals during early nighttime, and stayed at low concentrations at night, which was mainly due to the sink of terpene oxidation.
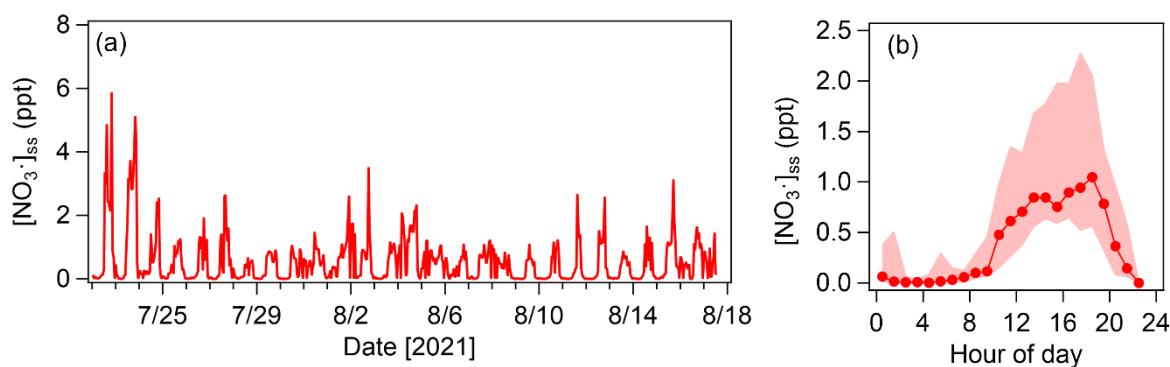
Figure S19. (a) Time series of estimated steady-state [NO$_3$·] concentrations and (b) diurnal variations of [NO$_3$·].

9. How do the authors handle a size-dependent enrichment factor (Figure S1) when calculating the total aerosol mass observed by CHARON?

Response: Based on the SMPS measurements, the geometric particle size was varying in the range of 20-112 nm (average: 48 ± 12 nm) during the entire campaign. Therefore, we used an average enrichment factor of 6 for the particles with sizes <150 nm to calculate the total aerosol mass measured by the CHARON-PTR-MS.

10. It appears that the authors are attributing all of C5H9+ to isoprene. How good an assumption is this?

Response: Yes, we attributed all of C$_5$H$_9$$^+$ signals to isoprene in this study. Previous studies have found that the fragmentation of 2-methyl-3-buten-2-ol (C$_5$H$_{11}$O$^+$, MBO) emitted from biogenic sources inside PTR instruments can significantly contribute to the C$_5$H$_9$$^+$ signals (Karl et al., 2012). In this study, the time series of C$_5$H$_9$$^+$ was correlated with that of C$_5$H$_{11}$O$^+$ (r= 0.69) as shown in Fig. S17 in the revised supplement, suggesting that the fragmentation of MBO could contribute to the signals of C$_5$H$_9$$^+$. However, the concentrations of C$_5$H$_{11}$O$^+$ were much lower than those of C$_5$H$_9$$^+$, thus the contributions from C$_5$H$_{11}$O$^+$ fragmentation to C$_5$H$_9$$^+$ were much lower than those from the isoprene parent ion. In addition, Coggon et al., (2024) recently reported that PTR-measured C$_5$H$_9$$^+$ signals can be significantly contributed by the

fragmentation of cycloalkanes and long-chain aldehydes (e.g., octanal and nonanal) emitted from anthropogenic sources such as cooking emissions especially in urban regions. These long-chain aldehydes were not measured in this study, thus their fragmentation to $C_5H_9^+$ were expected to be minor. Therefore, it is reasonable to assume that $C_5H_9^+$ were mainly related to isoprene at this location.
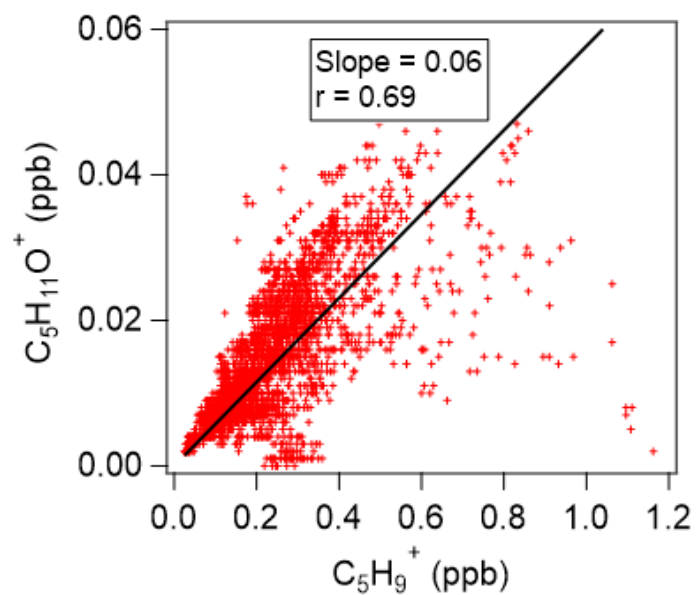
Figure S18. Scatter plot of $C_5H_9^+$ and $C_5H_{11}O^+$ in the gas phase during the entire campaign.

Response to Reviewer 2

The manuscript by Song et al. investigates the chemical composition and sources of VOC and OA, as well as contribution of OVOC and CHON to nighttime OA mass and particle growth events in a rural forested area in Germany in summer, by the deployment of advanced mass spectrometry techniques, very extensive source apportionments (for several datasets for both gas and particle phase) and air mass back trajectory analysis. In this study they have found that the CHARON-measured OA mass accounted on average for 63 ± 18% of the total OA mass (4.2 ± 2.8 μg m-3) concurrently measured by an aerosol mass spectrometer (AMS). After the source apportionment done by the PMF they also have found three factors of oxygenated VOC (OVOC), namely aromatic-OVOCs, biogenic-OVOCs and aged-OVOCs contributed on average 11% ± 9%, 37% ± 29%, 29% ±21% of total VOC concentrations, respectively. Finally, they have also observed regularly nighttime particle growth, which they attributed to the semi-volatile organic compounds and organic nitrates formed from the oxidation of biogenic VOC. This is a very interesting phenomenon. The study is well designed, the manuscript is well written, and the data are well presented with very impressive efforts. I would therefore recommend its publication on ACP.

Response: Thanks for the constructive evaluation on our manuscript. The point-to-point responses to individual comment are given below.

Below few comments for the authors.

Specific:

Line 220-222. What's the fraction of these 98 VOC ions to the total VOC signal/concentration detected by PTR-MS? Is the total VOC shown in Figure 2d the sum of these 98 VOC ions or all detected VOC ions including excluded ones? Would be nice to show the time series of total 98 VOC ions and total VOC ions in SI.

Response: In Figure 2d, we showed these 98 VOC ions as the total VOCs. We excluded some low-molecular weight species like formaldehyde and methanol. We also removed less abundant VOC species with many missing data points less than the method detection limit especially for high-molecular weight species (m/z>150). The average fraction of the 98 VOC ions to the total VOC signals was 85% ±3% during the entire measurement period (Figure R1). We revised this sentence accordingly.

Line 223: The average fraction of the 98 VOC ions to the total VOC signals was 85% ±3% during the entire measurement period.

Line 265: The total mixing ratios of measured VOCs (98 VOC ions) ranged from 7.6 to 88.9 ppb with an average of 31.2 ± 13.4 ppb.
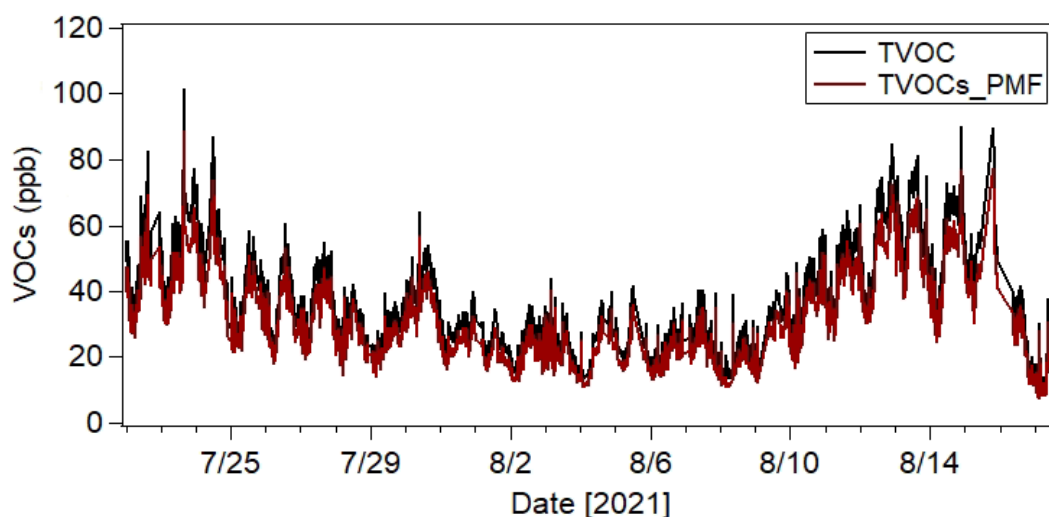
Figure R1. Time series of TVOCs measured by the PTR-MS and total concentrations 98 VOC ions for the PMF analysis (TVOCs_PMF).

Line 294-296. The authors attributed the low mass range of detected ions mainly to the fragmentation of larger masses, instead of other potential reasons like low oxygenated VOC there. Probably the mass spectra of FIGAERO-CIMS could help to support this attribution if the FIGAERO mass spectra show higher mass range ions.

Response: Previous studies have shown that the fragmentation of higher masses during the PTR ionization contributed to the lower masses significantly (Gkatzelis et al., 2018; Peng et al., 2023; Leglise et al., 2019). Here we selected $C_9H_{14}O_4$ and $C_7H_8O_4$ measured by the FIGAERO-CIMS for the comparison to the parent and/or fragment ions measured by the CHARON-PTR-MS in Figure R2. $C_9H_{14}O_4$ and $C_7H_8O_4$ can be assigned as the marker ions for monoterpene oxidation products (e.g., norpinonic acid and isomers) and toluene oxidation products (e.g., tetrahydroxybenzene and isomers), respectively. The fragmentation of $C_9H_{15}O_4^+$ and $C_7H_9O_4^+$ in the CHARON-PTR-MS could produce $C_9H_{13}O_3^+$ and $C_7H_7O_3^+$, respectively by losing one H_2O. Good correlations were observed for $C_9H_{14}O_4$ measured by FIGAERO-CIMS vs. $C_9H_{13}O_3^+$ measured by CHARON-PTR-MS, and $C_7H_8O_4$ vs. the sum of $C_7H_9O_4^+$ and $C_7H_7O_3^+$. It indicates that the ions at lower mass range were related to the fragmentation of higher mass in the CHARON-PTR-MS. Please note that $C_9H_{15}O_4^+$ was not measured by the CHARON-PTR-MS, which was probably fragmented. In addition, we adopted an assumed maximum sensitivity (~22 cps/ppt) for FIGAERO-CIMS to quantify the concentrations of oxygenated organic compounds, which may lead to the underestimation.
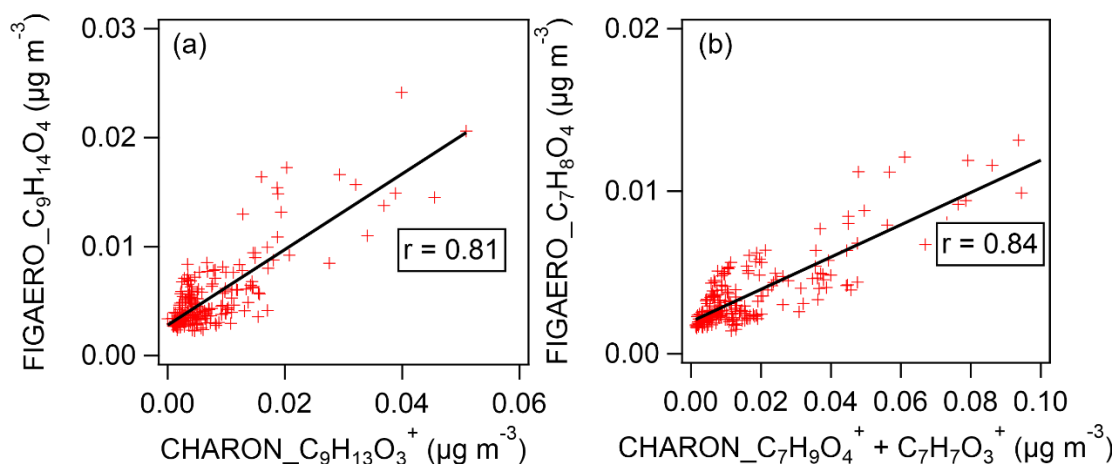
Figure R2. Scatter plots between (a) $C_9H_{14}O_4$ measured by FIGAERO-CIMS vs. $C_9H_{13}O_3^+$ measured by CHARON-PTR-MS, (b) $C_7H_8O_4$ measured by FIGAERO-CIMS vs. the sum of $C_7H_9O_4^+$ and $C_7H_7O_3^+$ measured by CHARON-PTR-MS.

Line 299 and 482. It seems the dominating ion for $CxHyO2+$ group is $C3H5O2+$ (methylglyoxal/acrylic acid) from Table S2 and S3. Why is it so high? It seems to be present both in aged-OVOC factor in gas phase and F5/6 factor in particle phase from CHARON-PTR-MS PMF results. This ion seems to be related to the long range transported Atlantic air masses as discussed in Line 379? Does the F5/6 factor correlate with MOOA factor or MSA (similar to the MOOA factor with MSA fragment ion ($CH3SO2+$) from AMS-PMF in Fig 5b)?

Response: We believe that $C_3H_5O_2^+$ was significantly produced from the fragmentation of less-volatile organic compounds. Figure S12 shows the evolution of $C_3H_5O_2^+$, $C_4H_9O^+$, $C_4H_7O^+$ during an individual CHARON-PTR-MS measurement cycle. Unlike $C_4H_9O^+$ and $C_4H_7O^+$ the $C_3H_5O_2^+$ needs more time to reach its plateau at each CHARON measurement. This suggests that particulate $C_3H_5O_2^+$ could be contributed by the fragmentation of less-volatile organic compounds. We also added the time series of particulate $C_3H_5O_2^+$, sum of F5 and F6, and MOOA resolved from AMS-PMF analysis (Fig. S14) in the revised supplement. It can be seen that the time series of sum of F5+F6 correlates with MOOA (Fig. S14). In addition, we found that the time series of F6 correlated better with MOOA (r = 0.40) compared to that of F5 (r = 0.1), thus F6 was more related to long-range transported Atlantic air masses.
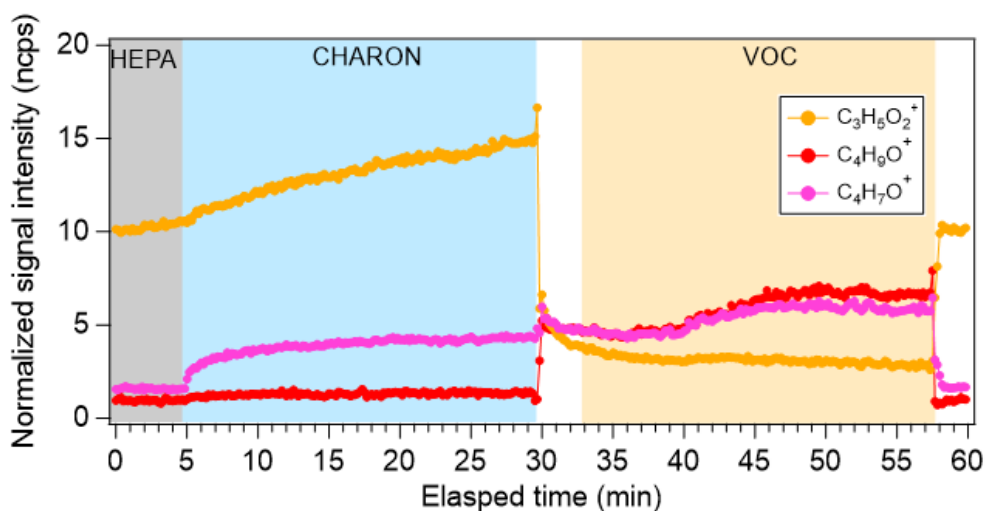
Figure S12. Time series of selected ions including $C_3H_5O_2^+$, $C_4H_9O^+$, $C_4H_7O^+$ during an individual CHARON-PTR-MS measurement cycle.
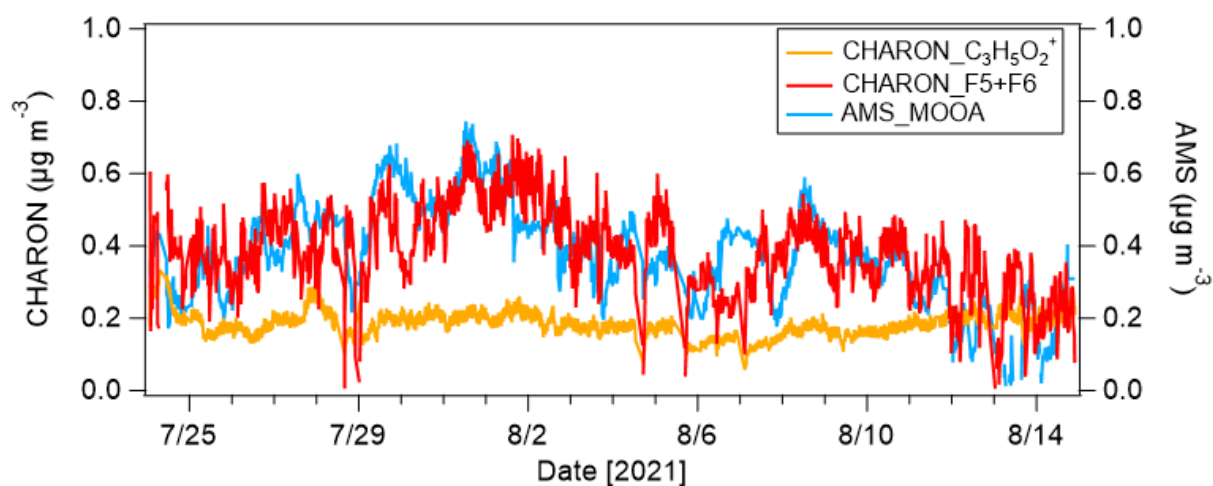
Figure S14. Time series of particulate $C_3H_5O_2^+$, sum of unassigned factor 5 and 6 (F5 + F6) form the CHARON and MOOA resolved from the AMS-PMF analysis.

Line 308. The authors attributed the correlation of CxHy+ with CxHyO1-2+ mainly to the fragmentation of larger masses. Li et al. (2020) have also reported similar observations that CxHyO1-2+ species followed more the CxHy+ trends. How about the role of nighttime chemistry and decreased PBL here, considering higher monoterpenes/sesquiterpenes (Line 266 and Fig S5b) and SV-OOA1 factor (Line 404 and Fig 5c) at night as well as SV-OOA1 factor correlates with VOC terpene factor (Line 403) and nighttime-BSOA factor (Line 469)?

Response: We attributed to the ions of $C_xH_y^+$ in the particle phase as the fragment of higher masses as they had good correlations with particulate $C_xH_yO_{1-2}^+$. In the gas phase, $C_xH_y^+$ ions are mainly contributed by aromatic hydrocarbons and monoterpenes and/or their fragmentation. However, these hydrocarbon species generally have high volatilities, thus they cannot exist in the particle phase. The nighttime chemistry and decreased PBL can increase the concentrations of $C_xH_yO_{1-2}^+$ and their fragment $C_xH_y^+$ ions mainly formed from the oxidation of terpenes, contributing to SV-OOA or nighttime-BSOA factor at night.

Line 328-330. Ethanol is a relatively long-lived compound and could have contributions from regional transport or lab solvent usage. Could the authors exclude this contribution e.g. via air mass trajectory results? If not, the reviewer may suggest to remove these two sentences and make the point directly about correlations between traffic VOC with ethanol.

Response: The contribution of solvent usage to ethanol was minor. We deleted these two sentences to make the point clear.

Line 367-373. The biogenic OVOC factor with smaller OVOC. Does it correlate with monoterpene or sesquiterpene SOA markers detected by CHARON or FIGAERO? Probably this could be utilized to support the factor attribution.

Response: We observed no significant correlation between the biogenic OVOC factor with monoterpene or sesquiterpene SOA markers measured by CHARON-PTR-MS or FIGAERO-CIMS, which can be explained by two reasons. Firstly, frequent rainfall can scavenge the aerosol particle species and influence their temporal variations, while it has less effect on the trends of OVOCs. Secondly, as indicated by the strong correlations with O_x (r = 0.82), the temporal variations of biogenic OVOC factor were mainly influenced by the gas phase chemistry and the atmospheric oxidation capacity. However, the formation mechanisms of SOA may be different with those for smaller biogenic OVOCs.

Line 379-382. Considering the high fractions of this factor from the marine air mass clusters, have the authors tried to correlate this factor with MOOA factor or MSA (similar to the MOOA factor with MSA fragment ion (CH3SO2+) from AMS-PMF in Fig 5b)?

Response: No significant correlations were found between the factor of aged-OVOCs and MOOA or the MSA fragment ion. As mentioned above, frequent rainfall events were observed during the days with the marine air mass clusters (Fig. 2), which can strongly scavenge the aerosol particles. However, the temporal variations of OVOCs were less affected by rainfall but were mainly related to the variations of wind directions.

Line 395-397. Maybe also partially due to the PBL dilution as shown in Fig 5c?

Response: Yes. We revised this sentence accordingly.

Line 411: No HOA peak was found in the afternoon, which is likely associated with strong photochemical oxidation, high wind speeds and expanded PBL in the afternoon leading to the decrease of HOA mass.

Line 443. This factor also contains quite high C7H9+ in addition to the fatty acid peaks. Is it from toluene or fragment ion from monoterpenes/sesquiterpenes? Have the authors tried to correlate this factor with MOOA factor or MSA (similar to the MOOA factor with MSA fragment ion (CH3SO2+) from AMS-PMF in Fig 5b), or check the air mass trajectories? Fatty acids like palmitic acid can also have marine sources (Mashayekhy Rad et al., 2018). Also higher fractions of biogenic-OVOC from long range transported marine air masses was shown in Line 492.

Response: The charcoal denuder used in the CHARON inlet can strip off aromatic hydrocarbons and monoterpenes efficiently (>99.999%). Therefore, it is unlikely that particulate $C_7H_9^+$ was contributed by toluene or the fragmentation of monoterpenes or sesquiterpenes. Particulate $C_7H_9^+$ had a good correlation with particulate $C_7H_{11}O^+$ (r = 0.92, Figure R3), therefore $C_7H_9^+$ could be derived from the fragmentation of $C_7H_{11}O^+$. $C_7H_{11}O^+$ can be tentatively assigned as 2,4-heptadienal in the gas phase, which is formed from the cleavage of fatty acid hydroperoxides in heated cooking oils reported by previous studies (Fullana et al., 2004; Takhar et al., 2023). In addition, we cannot fully exclude that the fragmentation of other particulate oxygenated species contributing to $C_7H_9^+$ in the particle phase. Therefore, it is more reliable to use long-chain fatty acids to characterize this factor related to cooking emissions. There is no significant correlation between AMS-MOOA factor and CHARON-COA factor or palmitic acid, therefore the contributions of marine sources to fatty acids were rather minor. We reformulated this paragraph in the revised manuscript.
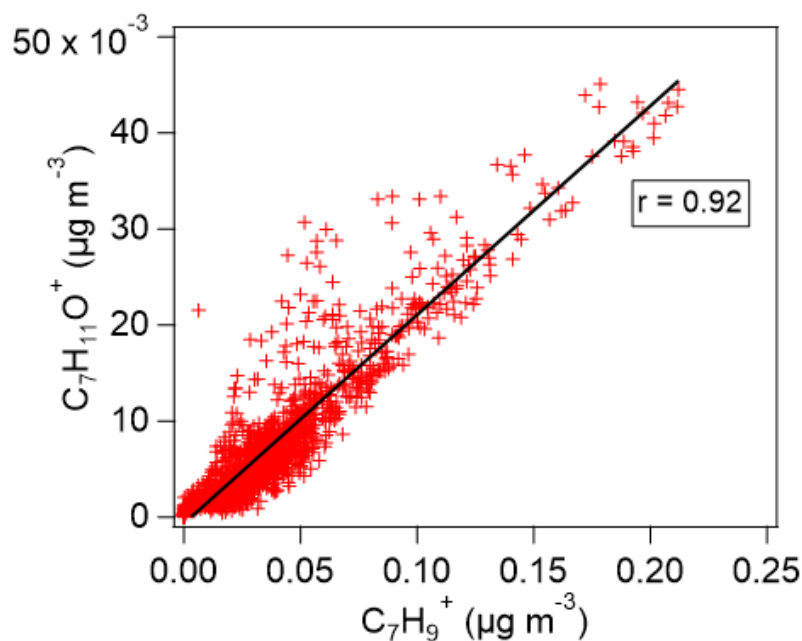
Figure R3. Scatter plot between $C_7H_9^+$ and $C_7H_{11}O^+$ ions measured by the CHARON-PTR-MS.

Lines 465-479: The first OA factor was characterized with high fractions of C_7H9^+, $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$. $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$ can be tentatively assigned as long-chain fatty acids. For example, $C_{16}H_{33}O_2^+$ was tentatively assigned as palmitic acid, which is a fatty acid mainly released from the cooking emissions. Palmitic acid can be used as a tracer for cooking-related OA (Reyes-Villegas et al., 2018). Furthermore, the diurnal variation of this factor showed a peak at lunch time (12:00-14:00), which is in line with the opening hours of the canteen located 300 m south of the sampling site in the campus. Therefore, it is reasonable to assign this factor associated with cooking emissions. As expected, the time series of COA is well correlated with $C_{16}H_{33}O_2^+$. Note that we cannot assign particulate $C_7H_9^+$ ions to specific organic compounds, which is likely produced from the fragmentation of oxidized organic compounds of higher masses. In this study, the COA factor was not resolved from the unconstrained AMS-PMF analysis. However, the AMS-PMF yielded the HOA factor with a high O:C value (0.26), which might be associated with the mixing of cooking-related oxidized species into HOA (Mohr et al., 2012).

Lower fractions of biogenic-OVOC were observed for long-range marine air masses (C4-C5) compared to continental air masses (C1-C2). We reformulated the sentence to avoid any confusion in Line 492.

Lines 526-529: Both continental air mass clusters C1 and C2 showed highest fractions of biogenic-OVOCs to total VOC mixing ratios (40% and 55%), indicating the importance of biogenic VOC oxidation to OVOC contribution. Comparatively lower fractions of biogenic-OVOCs (30% and 35%) were observed for long-range transported air masses from Atlantic Ocean (C4-C5).

Minor technical details:

Line 107. Use either "western" or "central".

Response: Revised.

Line 145. Add "respectively" after "100 °C".

Response: Done.

Line 150. Add "Th" after "398".

Response: Done.

Line 287. The percentage is slightly different from the values in the abstract in Line 21. Please double check.

Response: Revised.

Line 416. Please keep consistent for "R" or "r" in the text, also in all plots if it means the same such as Fig S6.

Response: Done.

Line 536. Typo for SV-OOA1?

Response: Done.

Fig 9 and 10. Would be nicer to change to horizontal plot for better visualization. Also type of "ug -3" in Fig 10.

Response: Done.

Fig S5. Please change the axis color of wind speeds or BLH to be consistent with their marker color.

Response: We updated this plot accordingly.

Response to Reviewer 3

The manuscript proposed by Junwei Song et al. entitled "*Sources of organic gases and aerosol particles and their roles in nighttime particle growth at a rural forested site in southwest Germany* " deals with the chemical characterisation of Volatile Organic Compounds (VOCs) and organic aerosol (OA) at the molecular level, and the relation with particle growth events at a rural forested site in southwest Germany during summer 2021. The study used a combination of state-of-the-art measurement and advanced statistical tools (i.e. PMF) to evidence sources contribution to VOCs and OA. The authors interestingly show an important contribution of different biogenic SOA factors to OA, while traditional AMS-PMF approach was not able to distinguish the different processes contributing to SOA, highlighting the need for molecular level characterization. They hypothesise that BVOC oxidation with ozone and NO3 radicals produced semi and low volatile compounds that participate to nighttime particle growth events. The paper is of excellent quality and well written, it reads quite well. However I have some points that are worth to be clarified, even if it does not question the quality of the study. The paper is of great interest for the scientific community, and consequently I recommend the publication of the paper after the authors address the following points:

Response: We thank the reviewer for the detailed evaluation on our manuscript. We provide the point-to-point response to each question as below and revised our manuscript based on the reviewers' constructive comments.

Main comments

L. 184: Does the 18.4 enrichment factor used as a constant value during the entire campaign? The authors have particle size distribution measurements (SMPS), why not using these data to calculate an enrichment factor for each time point of the field campaign based on size distribution (in mass), and the enrichment factor response as a function of particle diameter? This maybe can help to increase the agreement between AMS and Charon.

Response: There are some limitations for the adaptation of the CHARON enrichment factors at each time point based on the SMPS measurements. Firstly, CHARON-PTR-MS and SMPS have different measurement time resolution (10 s for CHARON-PTR-MS and 6 min for SMPS). Besides, the enrichment factor of CHARON is not only related to the particle size but also to some extent to the particle composition (Peng et al., 2023). In our study, the SMPS measurements showed the geometric particle sizes ranging between 20-112 nm (average: 48 ± 12 nm) during the entire campaign. Therefore, we adopted an average enrichment factor of 6 for the particles with the sizes <150 nm to calculate the total aerosol mass measured by the CHARON-PTR-MS.

Lines 37-40 (Supplement): Based on the SMPS measurements, the geometric particle sizes ranged between 20-112 nm (average: 48 ± 12 nm) during the entire campaign. Therefore, we

adopted an average enrichment factor of 6 for the particles with the sizes <150 nm to calculate the total aerosol mass measured by the CHARON-PTR-MS.

L. 142-144 and text S1 It is a good approach to remove the first 290 s of each CHARON measurement, but I am wondering why such a specific timing (why not 5 min / 300 sec?). Then, the authors did not exclude the same 290 sec in gas phase mode, which is questioning. Indeed, some sticky semi volatile species that are highly concentrated in particle phase (or, at least, more concentrated in gas phase concentrated in particle than in gas phase) will take time to re equilibrate to gas phase or background concentration. This could lead to an overestimated gas phase concentration of these compounds. Is there an obvious reason for not applying such an exclusion on gas phase? Maybe the authors can show in the supplement one or two time series as examples of gas/particle (and particle gas) transition of sticky compounds to support their approach.

Response: Thanks for the comment. We also excluded the last 10 s data of each CHARON measurement to avoid the interference during the shift from CHARON particle measurement mode to gas phase measurement mode, so we totally excluded 300 s data for each CHARON measurement. We set three minutes for the transition from CHARON measurement mode to gas-phase measurement mode, which allows the equilibrium of gas species in the sampling line before starting each VOC measurement. Therefore, we only excluded the first 10 s and last 10 s at each VOC measurement mode to avoid any inferences due to the switching between different measurement modes. We provided an example of the measurement cycle of the CHARON-PTR-MS as shown in Fig. S13 of the revised manuscript. We modified the description of CHARON-PTR-MS data processing in the supplement accordingly.
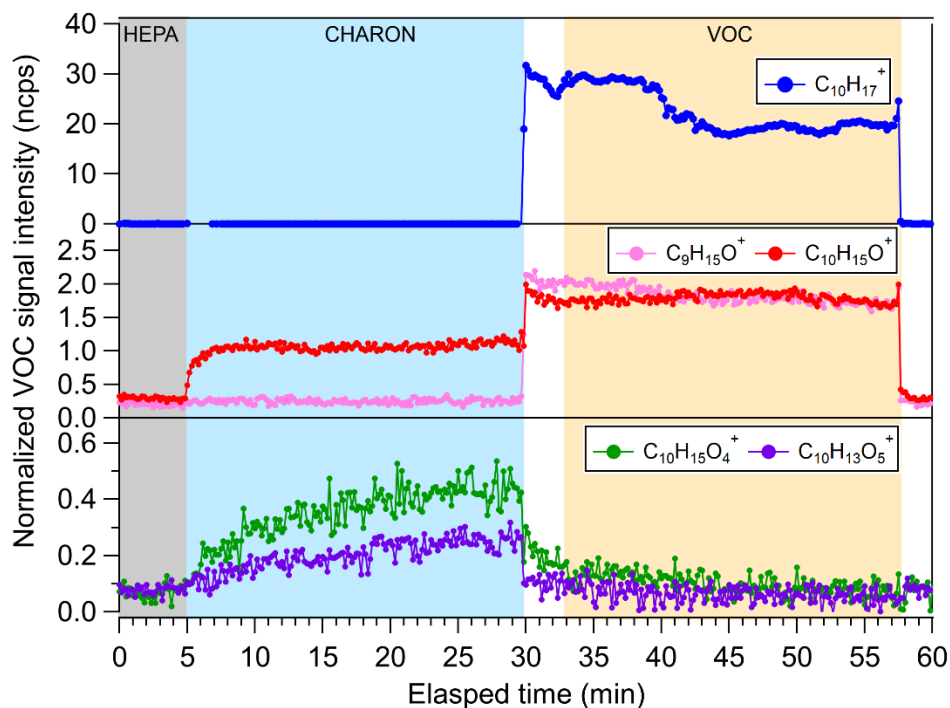
Figure S13. An example of the alternating measurement cycle of the CHARON-PTR-MS including 5-min HEPA mode, 25 min CHARON mode, 3 min transition time, 25 min VOC mode and 2 min transition mode on 13th August 2021. Time series of selected ions are shown including $C_{10}H_{17}^+$, $C_9H_{15}O^+$, $C_{10}H_{15}O^+$, $C_{10}H_{15}O_4^+$ and $C_{10}H_{13}O_5^+$.

Lines 28-44 (Supplement): As shown in Fig. S13, the signals of particulate more oxidized species ($C_{10}H_{15}O_4^+$ and $C_{10}H_{13}O_5^+$) measured by the CHARON slowly reached plateau values compared to less oxidized species ($C_{10}H_{15}O^+$). The PTR-MS shows slower responses to some organic species especially more oxidized species in the particle phase (Piel et al., 2021). Thus, the initial 290 s particle-phase data at each CHARON measurement cycle were excluded. We also excluded the last 10 s particle-phase data of each CHARON measurement to avoid any inferences due to the switching from particle-phase measurement to gas-phase measurement. Thus, in total 300 s data of each CHARON measurement were excluded. Then the processed particle data were corrected by the interpolate subtraction of HEPA filter background. Note, that the transition time was set to 3 minutes for the switching from CHARON measurement mode to VOC measurement mode before starting the gas-phase measurements. In addition, for the gas phase data, we only excluded the first 10 s and the last 10 s data at each VOC measurement cycle to avoid any inferences due to the switching between different measurement modes.

L. 287 290: Are AMS and Charon really comparable in this study? The authors wrote the AMS measured NR.PM2.5, while the ADL in the CHARON only extracts PM1. In addition, only masses above 60 are included in the CHARON analysis, while some compounds might significantly fragment at masses below this threshold (m/ 41 or 43 for example can be important fragments, see Leglise et al., (2019), cited in the manuscript).

Response: The AMS and the CHARON measurements can be compared in this study. Firstly, both instruments are connected to the same $PM_{2.5}$ inlet for measuring the particle composition. Secondly, our OPC measurements show that the $PM_{2.5}$ mass was mainly comprised of PM_1 (~93%, Figure. S6). Thus, the limitation by the CHARON ADL should be minor for the comparison between AMS and CHARON.

Indeed, some organic compounds measured by the CHARON-PTR-MS could fragment to lower masses like m/z41 ($C_3H_5^+$) and m/z43 ($C_3H_7^+$). These ions have lower molecular weights, so their mass concentrations calculated by the IDA analysis software were relatively low. The contributions of these smaller ions to total OA mass measured by the CHAON were low (<10%). In this study, we excluded these small ions for the PMF analysis because they can originate from the fragmentation of different organic compounds and cannot provide useful source information.

L.320: Is traffic the right name for this factor or anthropogenic primary emission would more appropriate? The diurnal cycle of BTEX (figure S5) is more pronounced based on a night/day basis rather than peaking during rush hours (e.g. benzene and toluene peaked at nighttime)

Response: We defined this VOC factor as traffic based on the factor profile with high contributions of BTEX and also ethanol. The diurnal variations of BTEX showed no significant peak at morning rush hours (7:00-9:00), but that of ethanol showed the peak. We compared the diurnal variations of other traffic-related species like NO_2 and BC (Figure R4), which also showed the peaks at morning rush hours (6:00-8:00). The data of NO_2 were obtained from the Eggenstein air quality monitoring station near a main road with high traffic emissions especially at morning and evening rush hours.
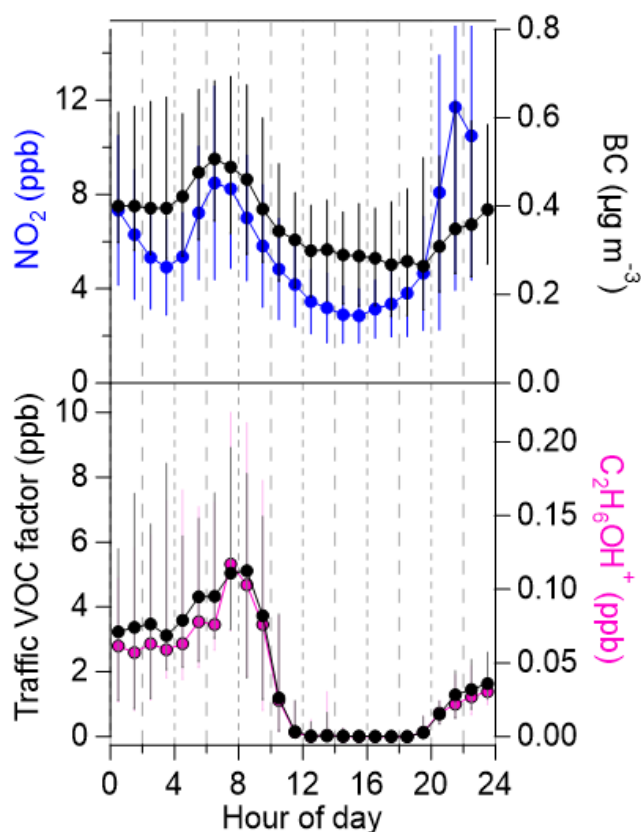
Figure R4. Diurnal variations of NO$_2$, BC, traffic VOC factor and ethanol (C$_2$H$_7$O$^+$) during the entire campaign.

L. 337: Why does C6H9+ ion is attributed to monoterpene fragment rather than isoprene, while it is explained that a correction factor has been applied to this ion accounting for isoprene fragmentation? I do not understand the logic here.

Response: We attributed C$_6$H$_9^+$ to the major fragment ion of monoterpenes. The parent ion of isoprene is C$_5$H$_9^+$. We reported the concentrations of isoprene and monoterpenes based on the concentrations of C$_5$H$_9^+$ and C$_{10}$H$_{17}^+$ with the correction on the fragmentation correspondingly. For the input of PMF analysis, no correction was made for the VOC ions.

L. 374 382: I agree with the fifth VOC factor is long range transport and it should be noted that O/C ratio of this factor is higher than others, supporting this hypothesis.

Response: We calculated the O/C ratios of five VOC factors. The O/C ratio of the fifth factor as aged-OVOC factor was 0.39, which was close to that of biogenic-OVOC factor (0.39) but higher than the other three VOC factors (0.07-0.31). This indicates that the fifth factor was related to the long-range transport.

Lines 388: The O/C ratio of this factor (r = 0.39) was higher than that of the other VOC factors like terpenes and traffic VOC (0.07-0.31).

3.2.1: I think the analysis in this section could be supported by polar plot analysis (concentration as function of wind speed and direction; see for example the fig 7 in the paper of Languille et al., 2019). All factors should have a preferential wind direction (biogenic from east, anthropogenic southwest, etc.)

Response: Thanks for the suggestion. We performed a polar plot analysis of VOC factors and OA factors resolved from the AMS-PMF analysis. High concentrations of traffic-related species including HOA, BC, traffic VOC factor were observed at the northeast sector with low wind speeds. Meanwhile, we found SV-OOA2 and aromatic-OVOC factors showed high concentrations at the center of polar plots, suggesting that they were associated with lower wind speeds. Note that slightly mass increase of HOA, SV-OOA2 and aromatic-OVOC factor were found at the south/southwest winds. This is consistent with the results from the air-mass back-trajectory analysis that the transport of urban air can also contribute to the concentrations of OA and VOC. In contrast, high concentrations of LV-OOA and biogenic-OVOC factor were found for the sectors with east and/or southeast wind directions, indicating that they were influenced by the oxidation of biogenic emissions. We added these plots as Figure S9 in the revised supplement. We also provided additional discussions on the origins of VOCs and OA in the revised manuscript (Section 3.2.1 and 3.2.2).

Lines 335-338: A shown in Fig. S9, the bivariate polar plot analysis showed that higher concentrations of traffic VOC factor were observed at the wind sector of northeast with low wind speeds (0-1.5 m s-1), suggesting that it was related to local traffic emissions. Lines 374-376: Moreover, the bivariate polar plot analysis showed that high concentrations of this factor were observed at the wind sectors of east and southeast, suggesting that it was mainly related to the biogenic emissions. Lines 389-391: The bivariate polar plot analysis showed that high concentrations of this factor were observed at west or southwest wind sectors with high wind speeds.

Lines 413-415: In addition, the bivariate polar plot analysis also showed that HOA was associated with the northeast sector with low wind speeds (0-1.5 m s-1), suggesting that HOA was mainly attributed to local traffic emissions. Lines 438-440: The bivariate polar plot analysis showed that high concentrations of LV-OOA were observed at the wind sectors of east and/or

southeast, suggesting that it was mainly associated with the oxidation of biogenic emissions. Lines 451-452: The bivariate polar plot analysis also showed that high concentrations of MOOA were observed at the wind sectors of west with high wind speeds.
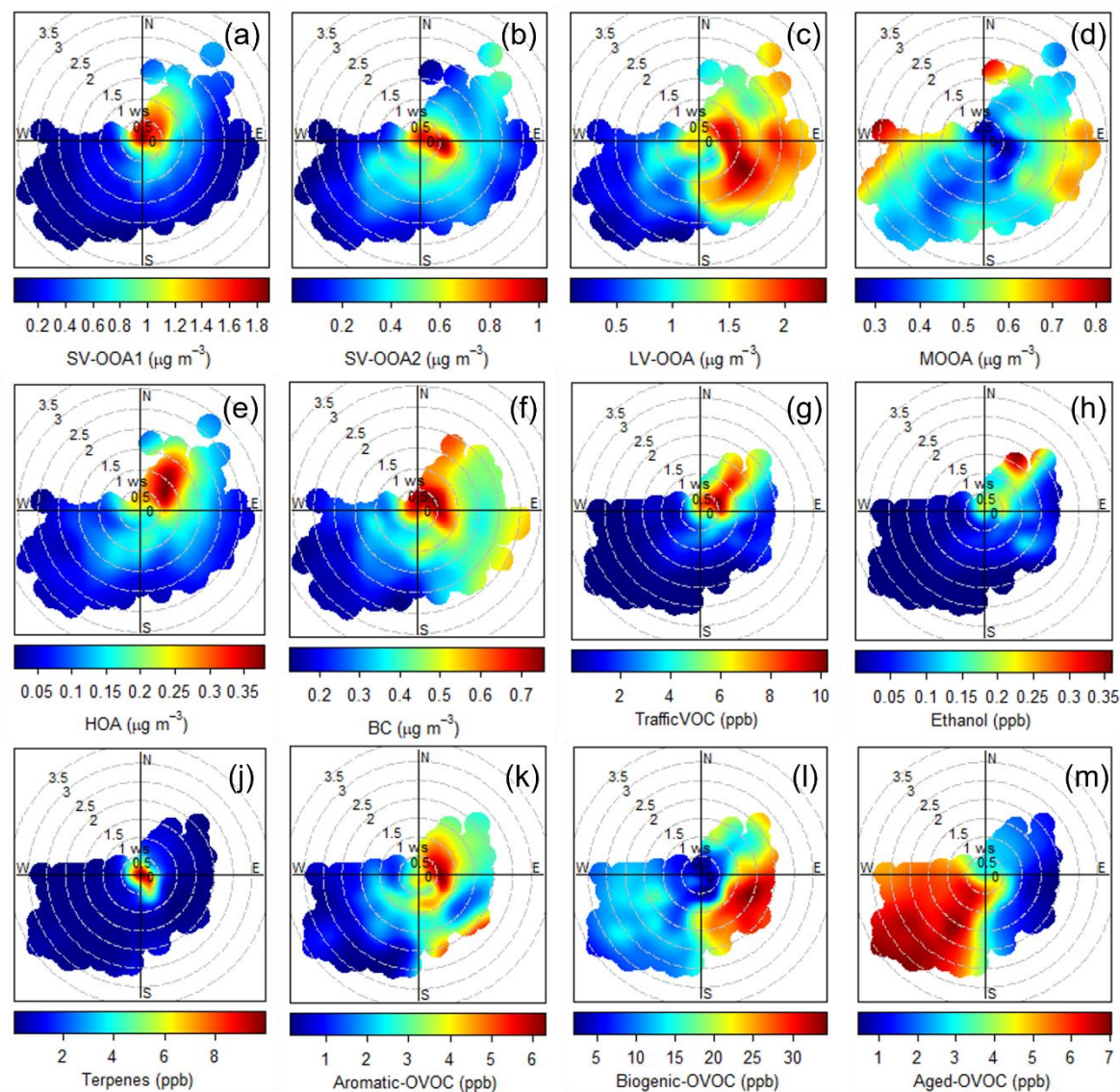
Figure. S9 Bivariate polar plots of (a-f) BC and OA factors including HOA, SV-OOA1, SV-OOA2, LV-OOA and MOOA resolved from the AMS-PMF analysis and (g-h) ethanol and VOC factors including traffic VOC, terpenes, aromatic-OVOC, biogenic-OVOC and aged OVOC.

L390-397: As for VOCs, not sure HOA is only traffic, considering the moderate correlation to BC. And that diurnal cycle is not mainly driven by rush hours. In addition, refineries and coal fire plant might contribute to this factor. In general, it would be good to check whether these industrial sources at source west affect both VOCs and OA.

Response: The correlation of HOA with BC could be deteriorated due to the different emission ratios of HOA/BC of different vehicles and fuel types of the cars passing the sampling site. Generally, gasoline vehicles have higher emission ratios of HOA/BC (0.9-1.7) than diesel vehicles (0.03-0.61) (Dewitt et al., 2015). In this study, the average HOA/BC was 0.38 ± 0.31, suggesting dominant contributions of diesel vehicles. We resolved HOA from the unconstrained AMS-PMF analysis in this study, which may mix with other sources like cooking emissions to some extent. As mentioned above, we performed the polar plot analysis of VOC factors and OA factors from the AMS-PMF. The impact of industrial sources including refineries and coal-fired power plant on HOA should be minor. We reformulated this paragraph in the revised manuscript as follows.

Lines 407-415: The correlation of HOA and BC could be deteriorated due to different emission ratios of HOA/BC of different vehicles and fuel types of the cars passing the sampling site. Generally, gasoline vehicles have higher emission ratios of HOA/BC (0.9-1.7) than diesel vehicles (0.03-0.61) (Dewitt et al., 2015). In this study, the average HOA/BC was 0.38 ± 0.31, suggesting dominant contributions of diesel vehicles. The diurnal cycle of HOA showed a peak at the morning rush hours (8:00-9:00). No HOA peak was found in the afternoon, which is likely associated with photochemical oxidation, high wind speeds and an expanded PBL in the afternoon leading to the decrease of HOA mass. In addition, the bivariate polar plot analysis showed that HOA was associated with the northeast sector with low wind speeds (0-1.5 m s-1), suggesting that HOA was mainly related to local traffic emissions.

3.2.3 The reasons explaining why a 6 factors solution has been selected should be clarified (why for CHARON-PMF there is not the same plots (fig S8 and S9) as for AMS and VOCs in the supplementary?). From what I see, factors 5 and 6 seems to be anticorrelated with a nearly similar mass spectrum. So the combination of both will probably result in rather flat signal, that can potentially be attributed to some kind of background or something else (it seems like one factor that has been split in half)? Both factors are mainly attributed to C3H5O2+ and altogether they explain 42 % of the OA, which is quite high (I guess this is the highest ion at m/z 73 on fig S7, that is really high based on table S3)! Can this be due to a contamination in the set up (filter, lines material, etc.)? Another option would be to exclude this ion from the analysis, if this can be a contamination or if it biases the analysis? In that sense, a five factors solution summing the 2 last factors should be suitable too.

Response: We added the key diagnostic plots for the 6-factor PMF solution of CHARON-measured OA in the revised manuscript. Compared to the 6-factor PMF solution, we cannot separate the factor of COA with a 5-factor PMF solution in this study. We found that the F6 showed a better correlation with MOOA resolved from the AMS-PMF analysis (r = 0.40) compared to F5 (r = 0.09). We also sumed the fifth and sixth factors (F5 + F6) and fund that the time series of the summed F5+F6 better correlated with that of MOOA (r = 0.59), while no good correlation was found between particulate $C_3H_5O_2^+$ measured by the CHARON and MOOA (r = 0.08). We speculate that F6 is likely associated with long-range transported marine air masses. The effect of contamination should be minor because we correct the CHARON data with the interpolate subtraction of HEPA filter background. In addition, we looked at the evolution of $C_3H_5O_2^+$ during an individual CHARON measurement. The $C_3H_5O_2^+$ signals during CHARON measurements were higher than those measured during HEPA background measurements. However, unlike other low-molecular weight ions $C_4H_7O^+$ and $C_4H_9O^+$, the $C_3H_5O_2^+$ signals need more time to reach stable values during CHARON measurement. Therefore, $C_3H_5O_2^+$ is likely associated with the fragmentation of less-volatile organic compounds in the particle phase.
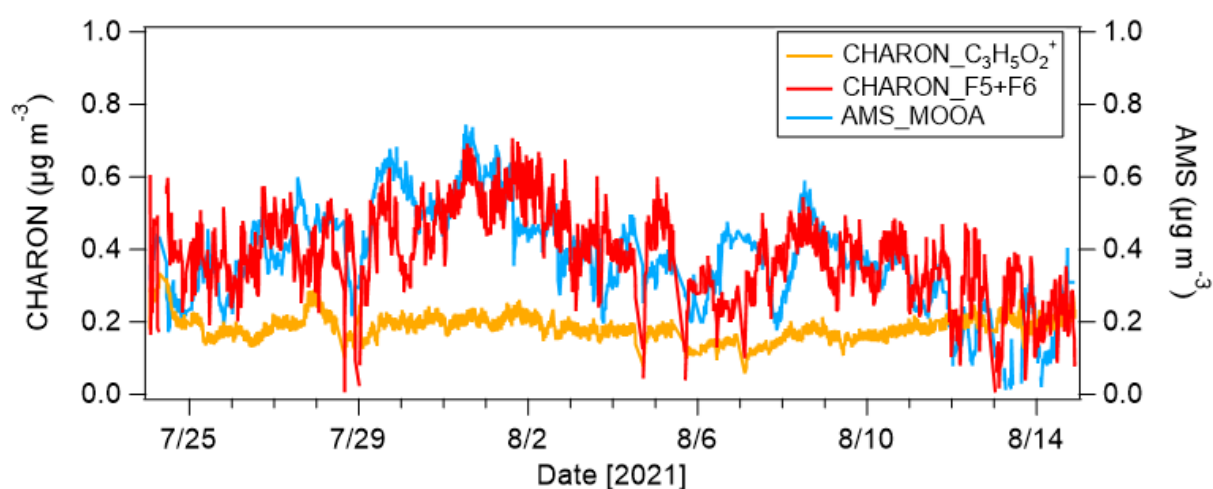
Figure S14. Time series of particulate $C_3H_5O_2^+$, sum of unassigned factor 5 and 6 (F5 + F6) and MOOA resolved from the AMS-PMF analysis.
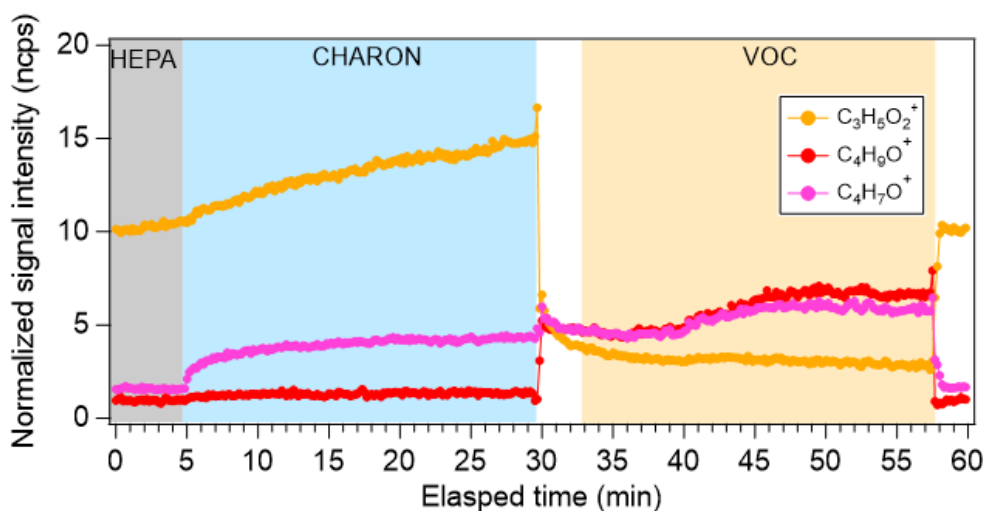
Figure S12. Time series of selected ions including $C_3H_5O_2^+$, $C_4H_9O^+$, $C_4H_7O^+$ during an individual CHARON-PTR-MS alternatingly measurement cycle.

L. 443. I fully agree with the assignment of factor 1 CHARON OA to cooking OA, but I have 2 remarks. First the ion C16H33O2 is not the only significant one, there is another important one that can be a long chain fatty acid C16H35O3+. The second one is the C7H9+ peak, commonly attributed to toluene in gas phase, which is for sure not the case in OA. Where does this ion may come from (also seen in factor 2).

Response: We attributed the first factor as cooking OA due to high loadings of long chain fatty acids including $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$. $C_7H_9^+$ was not attributed to toluene since the gas denuder of CHARON inlet can strip off toluene efficiently (99.999%). $C_7H_9^+$ was likely contributed by the fragmentation of different oxygenated species of higher masses, which cannot be used as a marker ion for cooking emissions. We reformulated this paragraph in the revised manuscript as follows.

Lines 465-479: The first OA factor shows high fractions of $C_7H_9^+$, $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$. $C_{16}H_{33}O_2^+$ and $C_{16}H_{35}O_3^+$ can be tentatively assigned as long-chain fatty acids. For example, $C_{16}H_{33}O_2^+$ was tentatively assigned as palmitic acid, which is a fatty acid mainly released from the cooking emissions. Palmitic acid can be used as a tracer for cooking-related OA (Reyes-Villegas et al., 2018). Furthermore, the diurnal variation of this factor showed a peak at lunch time (12:00-14:00), which is in line with the opening hours of the canteen located 300 m south of the sampling site on the KIT campus. Therefore, it is reasonable to assign this factor with cooking emissions. As expected, the time series of COA well correlated with $C_{16}H_{33}O_2^+$. Note,

that we cannot assign the particulate $C_7H_9^+$ ion to specific organic compounds, since it is likely produced by fragmentation of different oxidized organic compounds of higher masses. In this study, the COA factor was not resolved from the unconstrained AMS-PMF analysis. However, the AMS-PMF yielded the HOA factor with a high O:C value (0.26), which might be associated with the mixing of cooking-related oxidized species into HOA (Mohr et al., 2012).

It is also interesting to note that COA contributes to 9 % of OA according to CHARON PMF, but it is not reported in AMS-PMF while it has been already identified in urban AMS-PMF studies. How can that be explained?

Response: COA was not resolved from the unconstrained AMS-PMF analysis in this study. The COA was probably mixed into the HOA because the factor profile of COA resemble that of HOA both with high contributions of hydrocarbon ions ($C_xH_y^+$). Compared to AMS-PMF studies in urban regions, the strength of cooking emissions is usually weak in our rural sampling site. We can identify the COA from CHARON-PMF analysis since the CHARON can measure the long-chain fatty acids (e.g., $C_{16}H_{33}O_2^+$) as the markers of cooking emissions.

L. 497: High contribution of traffic VOC factor is observed for cluster C3. But Figure 1 showed that it is more long-range transport rather than local cluster. The VOC traffic factor is thus probably not only pure traffic.

Response: As stated before, the traffic VOC factor was more related to local traffic emissions as indicated by the polar plot analysis. The air mass cluster C3 is related to long-range transport but has less effect on local traffic emissions.

L.525: Please details what are the calculations made here.

Response: We rephrase this sentence to avoid any confusion.

Line 567: Furthermore, we calculated the diurnal variations for the mass fractions of SV-OOA1 and SV-OOA2 in total OA mass to normalize for the impact of meteorology.

L. 541-543: The NO3 production term should include temperature dependant rate constant (kO3.NO2 = 1.4 × 10−13 exp.(−2470/T) cm3 molecule−1 s−1 (Vrekoussis et al., (2004)). A rough evaluation of the sink for NO3 due to NO should be evaluated to see if N-oxidation products are less concentrated the night NO concentration was higher.

Response: Thanks for the comment. Following the same calculation method we used in our previous study (Huang et al., 2019), we adopted the rate coefficient at 298 K to calculate the production rate of NO_3 radicals without accounting for the temperature dependence. This allows for the comparison of NO_3 radical production rates with previous studies.

We calculated the diurnal variations of NO and NO_2 that were measured at the Eggenstein air quality monitoring station located ~2.5 km southwest of the sampling site (Figure R5). During nighttime, the concentrations of NO were relatively low (~1 ppb) compared to those of monoterpenes (~3 ppb). Therefore, the sink of NO_3 radicals due to reaction with NO was minor.
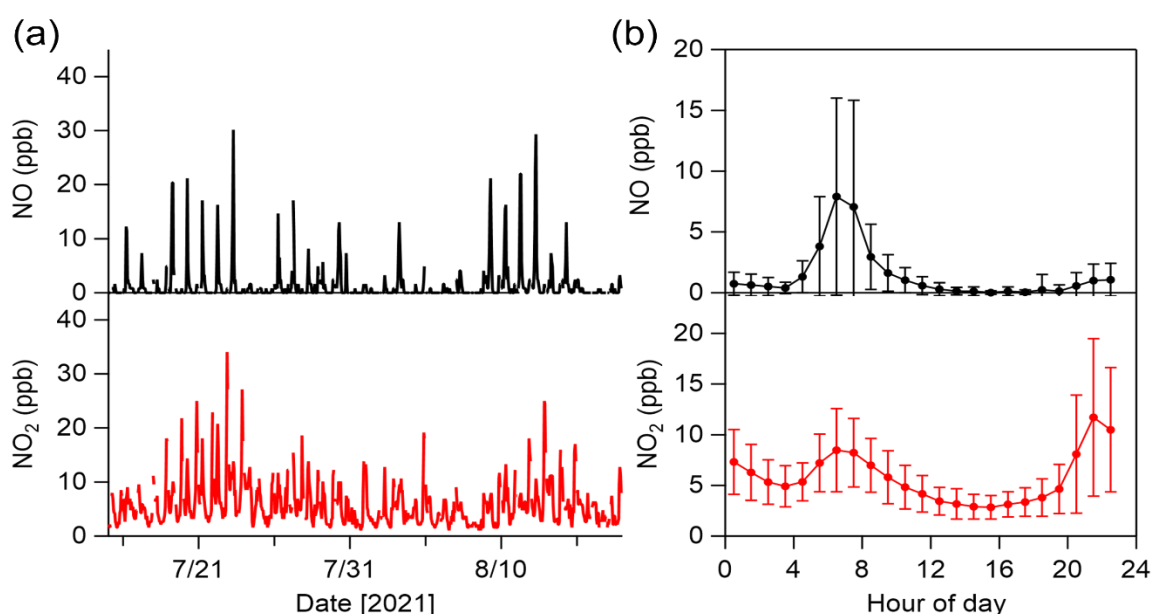
Figure R5. (a) Time series of NO and NO_2 concentrations measured at Eggenstein air quality monitor station located ~2.5 km southwest of the sampling site, (b) diurnal variations of NO and NO_2.

L.557- 590: I am in line with the authors hypothesis of nighttime contribution of BVOC oxidation to SOA and growth. But I see here two limits in the approach, the first one being that most of aerosol shown in the particle size distribution that depict a growth were not detected neither by AMS nor by the CHARON due to their low diameter. Making the link between these growth events and the aerosol chemical composition should be done with high caution, as this is not a direct link. The second limit is that nights when no growth were observed, the terpenes and related oxidation products should be lower. This case is not shown by the author but could strengthen their hypothesis if verified (maybe include an additional figure in the supplement).

Response: We agree the reviewer's comment. Due to the limitation of instruments, it is difficult to make the direct link between aerosol particle growth and aerosol chemical composition in this study. However, we observed the aerosol particle mass increase along with these particle size growth processes simultaneously. Furthermore, the meteorological conditions were stagnant with low wind speeds and low PBL heights during these particle growth events. The oxidation of terpenes by O_3 and NO_3 radicals produced SV-OOA and organic nitrates, contributing to the increases of OA mass and thus particle growth. We updated Figure 10 with adding the time series of wind speeds and boundary layer heights. As suggested, we provided Figure S17 in the supplement to show that lower concentrations of terpenes and SV-OOA were observed during non-particle growth events.
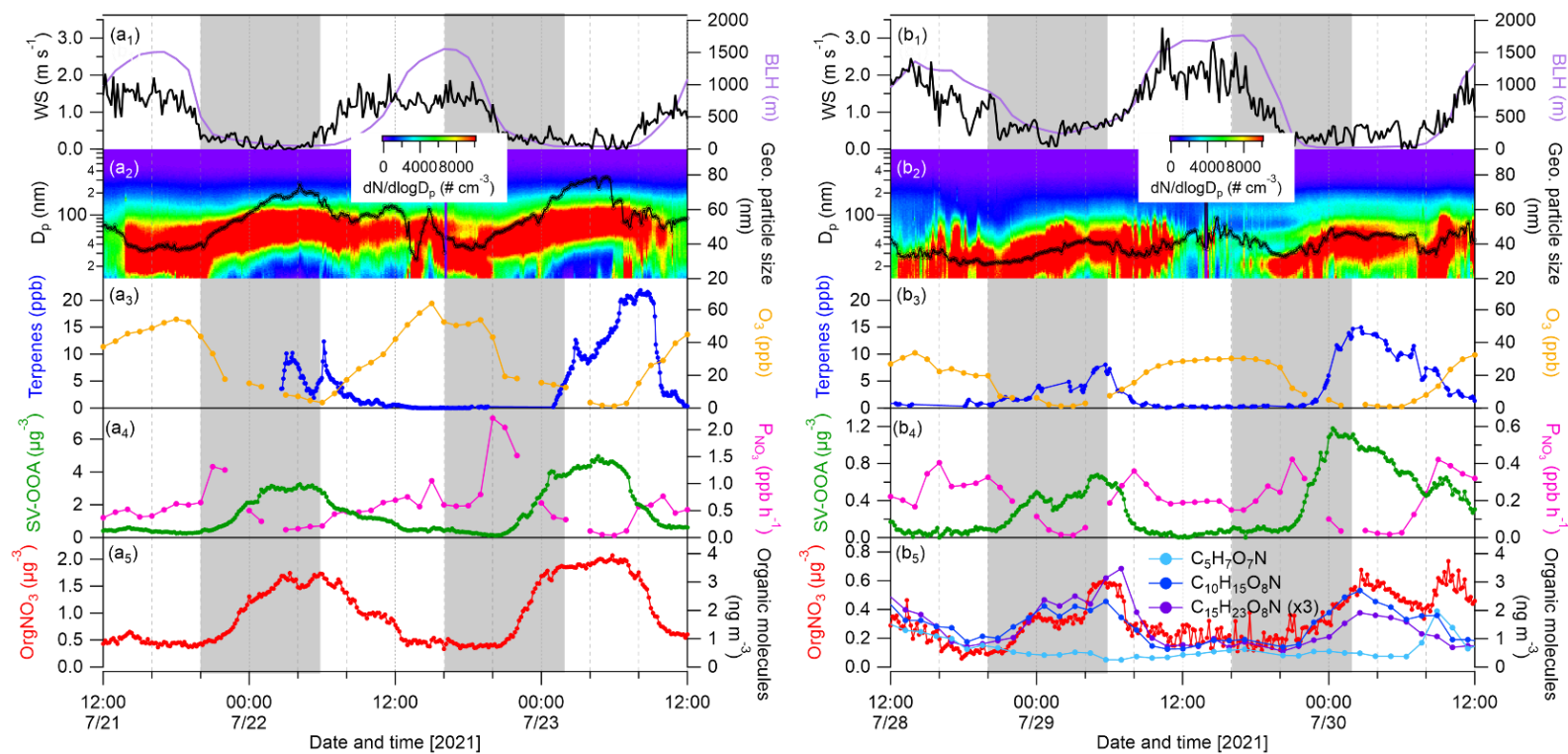
Figure 10. Cases showing the nighttime particle growth events as marked in gray shaded areas. Time series of wind speed, boundary layer height, particle number size distributions and geometric mean particle size, and mixing ratios of terpenes factor and O_3, production rate of nitrate radicals (P_{NO_3}), and mass concentrations of SV-OOA1 and organic nitrate calculated from the AMS during 21st-23rd (a1-a5) and 28th-30th July, 2021 (b1-b5). Three particulate organic nitrate molecules ($C_5H_7O_7N$, $C_{10}H_{15}O_8N$, $C_{15}H_{23}O_8N$) detected by the FIGAERO-CIMS are plotted in (b5).
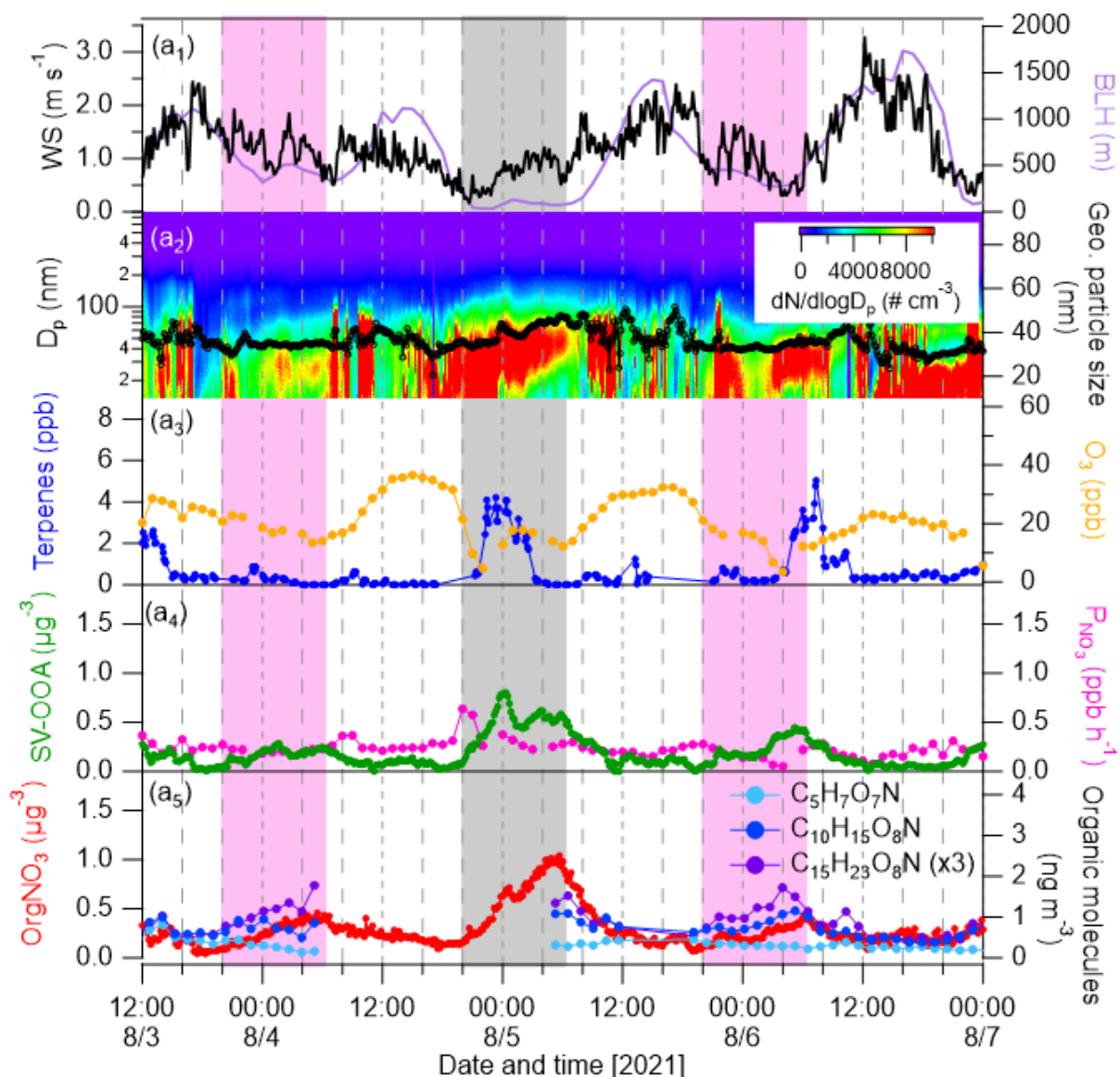
Figure S17. Cases showing the nighttime non-particle growth events as marked in pink shaded areas. Time series of wind speed, boundary layer height, particle number size distributions and geometric mean particle size, and mixing ratios of terpenes factor and O_3, production rate of nitrate radicals (PNO_3), and mass concentrations of SV-OOA1 and organic nitrate calculated from the AMS during 3rd-7th August, 2021. Three particulate organic nitrate molecules ($C_5H_7O_7N$, $C_{10}H_{15}O_8N$, $C_{15}H_{23}O_8N$) detected by the FIGAERO-CIMS are plotted in (a5).

Minor comments:

L. 115: "The site is mainly composed of pine trees". Please precise which pine are dominating the forest as BVOC emissions can differ from one pine species to another. It is important as the authors claim that pine trees are dominating BVOC emissions.

Response: The site is mainly composed of pine trees including Scots pine and European beech. We add this information in the revised manuscript.

Lines 116-117: Geographically, the campus is mostly surrounded by the Hardwald forest composed of mainly pine trees (e.g., Scots pine and European beech), which are important contributors for BVOC emissions.

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