Response to reviewers' comments on "Brown carbon aerosol in rural Germany: sources, chemistry, and diurnal variations" (egusphere-2024-1848)

The authors kindly thank the reviews for the careful review of the manuscript, and the helpful comments and suggestions, which improve the manuscript a lot. All the comments are addressed below point by point, with our responses in blue, and the corresponding revisions to the manuscript in red. All updates of the original manuscript are marked in the revised version.

Reviewer #1

This paper describes ambient air measurements conduced for one month in winter in Germany in 2021. In addition to standard instrumentation for O_3, NOx, particle sizes, etc, the key measurements were gas and particle phase composition using FIGAERO I-CIMS and an aethalometer. In addition to black carbon (BC) source apportionment, the goals of the work were to identify potential brown carbon (BrC) molecules in the gas and particle states, to study their diurnal variations, and to determine to what degree they contribute to the overall BrC measured by the aethalometer.

There is merit to this type of study because we need more molecular information about BrC molecules and their behavior, especially in regions which are not predominantly influenced by wildfire emissions, i.e., in this location the sources are presumably residential biomass burning and fossil fuel combustion. The findings are that there are both biomass burning (larger) and fossil fuel (smaller) contributions to BC, there were about 200 or so mass spectral features that may be BrC molecules, these features contribute about 10 percent to the total BrC absorption at 370 nm but a lower fraction of the total organic aerosol mass, gas phase BrC is largely photochemically generated during the day, and the ratio of gas phase BrC to particle phase BrC is much less than unity.

General comments

These are potentially interesting new measurements in Germany of quantities that have been measured at other locations. The methods and results are not particularly novel, and the paper needs to be much more quantitatively rigorous, especially with regard to uncertainties. I wonder whether this paper should be classified as an ACP "Measurement Report"? Overall, there is quite a bit of work to be done to get this paper ready for publication.

We modified the manuscript and added calibrations to make the results quantitative. In principle, we are open to convert our manuscript to a measurement report. However, we are not sure if this should be done at this stage and would like to know if the editor would advise us to do.

Specific comments:

1. My major criticism of this paper is the quantitative uncertainties in the measurements. In particular, unless I missed mention of them, there are no calibrations for the FIGAERO I-CIMS measurements. Rather, I believe that an "average sensitivity" was used for all mass spectral features, with the value taken from a literature study, i.e., with an entirely different instrument/operator. This is quite problematic, as CIMS instruments vary widely in sensitivity from one to another, even if operated in nominally the same manner. Calibration of at least a small

number of standard compounds is the minimum standard for field work, and increasingly many molecules are calibrated (or the voltage scanning method is applied) for I-CIMS work.

Indeed, the sensitivities of CIMS instruments can vary substantially and the sensitivity of the Iodide CIMS is known to vary over several orders of magnitude for different compounds. Application of an average, e.g. maximum, sensitivity can be useful to compare compounds for which standards are hardly available, like for highly oxygenated compounds. However, we agree that it is meaningful to calibrate our CIMS with some known BrC standard compounds. Therefore, we calibrated our instrument with four different nitro aromatic compounds (4-nitrophenol, 4-nitrocatechol, 4-methyl-5-nitrocatechol, and 2-methyl-4-nitrophenol).

"After the filed campaign, the calibration of 4-nitrophenol, 4-nitrocatechol, 2-methyl-4-nitropehnol, and 4-methyl-5-nitrocatechol was utilized to characterize the sensitivity factor of nitro aromatic compounds (NACs). Each NACs was dissolved into methanol to about 10 ng μL^{-1} as a standard NACs solution. Different volume (1, 2, 5, and 10 µL) of the standard NACs solution was deposited on a PTFE filter using an accurate syringe. The deposited filter was heated by FIGAERO-iodide-CIMS with ultra-high purity nitrogen following a thermal desorption. The filters were then desorbed in the same way as for the field samples. Every volume of the standard solution was repeated three times. The sensitivity factors of our iodide CIMS for 4-nitrophenol, 4-nitrocatechol, 2-methyl-4-nitropehnol, and 4-methyl-5-nitrocatechol were 0.80 ± 0.44, 0.50 ± 0.32, 0.96 ± 0.52, 0.97 ± 0.63, respectively. The sensitivity factors are the ratios of measured concentrations by FIGAERO-CIMS from the calibration vs "standard concentrations", where measured concentrations were calculated using a nominal maximum sensitivity of 22 cps/ppt (Salvador et al., 2021). The average sensitivity factor of all four NACs was 0.81 ± 0.53. We used this average sensitivity factor to estimate the concentrations of other potential brown carbon molecules in this study."
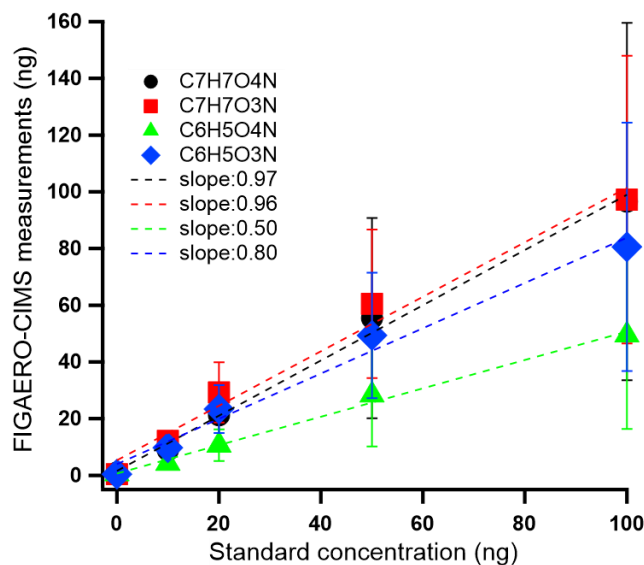

Figure S9. Calibration of FIGAERO-CIMS with four different nitro aromatic compounds. Blue: 4-nitropehnol; Green: 4-nitrocatechol; Red: 2-methyl-4-nitropehnol; Black: 4-methyl-5-nitrocatechol.

2. Moreover, calibrations for particle bound species (such as levoglucosan) can be performed with the FIGAERO by depositing known amounts of these molecules on the collecting filter. Thus, the authors have to better justify their reports of absolute amounts of BrC molecules. If they have not calibrated themselves, I do not believe they can report an absolute amount.

We agree and calibrated our FIGAERO-CIMS with the important biomass burning tracer levoglucosan. The calibration method was same as NACs calibration. The sensitivity factor was determined to 0.40 ± 0.14 corresponding to sensitivity of 9 ± 3 cps/ppt.

We added a few sentences as follow:

"The sensitivity factor of levoglucosan was 0.40 ± 0.14 in this study (Figure S10). We used the sensitivity factor of 0.40 ± 0.14 to estimate the concentrations of molecules, which are not identified as potential BrC molecules."
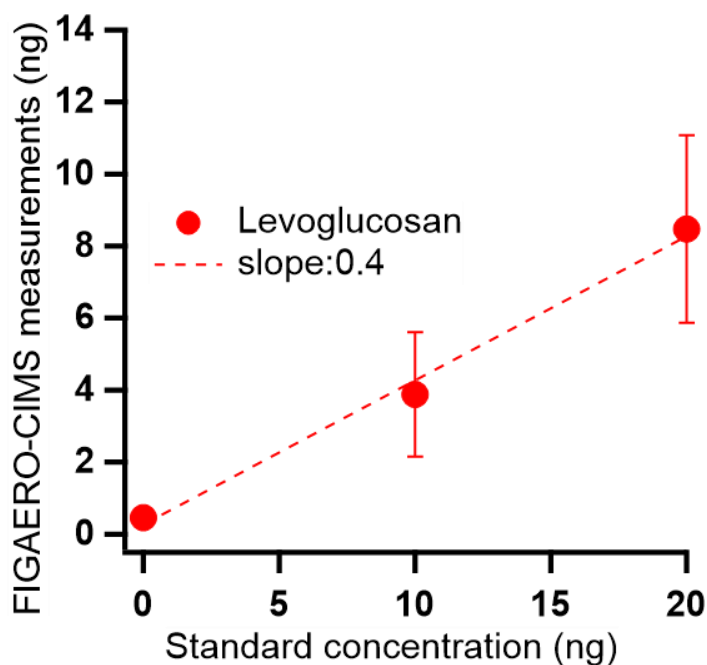

Figure S10. A calibration of FIGAERO-CIMS with levoglucosan. The measurement concentrations are calculated using a maximum sensitivity of 22 cps/ppt (Salvador et al., 2021).

3. On a related note, the authors appear to dismiss this uncertainty after acknowledging it: Line 131 "These values have high uncertainty with several orders of magnitude. However, this is still a reasonable method to measure the organic aerosol in atmosphere." They need to justify why this approach is "reasonable".

We agree that this text is unclear and modified it as follows:

"In this study, BrC molecules were identified and partially quantified in atmospheric aerosol by FIGAERO-CIMS. Please note that the iodide CIMS has sensitivities varying over several orders magnitude for different compounds e.g. of different oxidation states (Lopez-Hilfiker et al., 2016).

Therefore, the quantitative interpretation is limited to the small amount of compounds for which we could do calibration with authentic standards. Keeping this in mind, it can still be meaning to a relative comparison of the large number of high oxidized compounds assuming the same sensitivity."

4. In an analogous manner, aethalometer measurements require care to interpret, with corrections for on-filter scattering and loadings. Although the authors mention these uncertainties, they do not provide quantitative estimates for them.

To address this point, we added the following sentences on the uncertainty and calibration of aethalometer measurements in section 2.4.

"Since our aethalometer has been used two loading spots, the loading effect was corrected by a Dual-spot loading compensation algorithm (Drinovec et al., 2015). To further address the scattering effect (Yus-Díez et al., 2021), we did comparison experiments in the Aerosol Preparation and Characterization (APC) chamber (Huang et al., 2018). Black carbon was injected into the APC chamber by using the PALAS soot generator (GfG 1000, Palas) (Saathoff et al., 2003). The APC chamber was connected to a photoacoustic spectrometer (PAS) operating at three wavelengths (405, 520, and 658 nm) (Linke et al., 2016) and an aethalometer AE33. As shown in Figure S11, for three wavelengths (370, 520, and 660 nm), the correlation slopes were 1.88, 1.94, and 1.98, respectively. The average multiple-scattering correction factor was 1.90 ± 0.06 in this study."
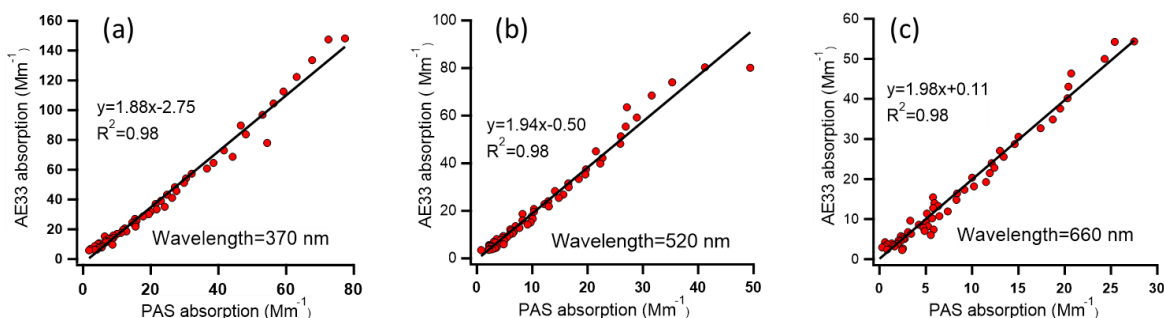

Fig. S11. Comparison of aerosol absorption coefficient measured by the photoacoustic spectrometer (PAS) and the aethalometer AE33 at 370 nm (a), (b), 520 nm, and 660 nm (c).

5. Likewise, the paper performs BC source apportionment, and it decouples BC absorption from total absorption to arrive at BrC absorption. There are many ways to do these analyses. The paper should justify the methods chosen.

We used the Aethalometer model to obtain BC associated with fossil fuel (BC$_{ff}$) and wood burning (BC$_{wb}$). The calculation function is given as below:

$$BC_{wb} = \left[\frac{b_{abs}(470nm) - b_{abs}(950nm) * \left(\frac{470}{950}\right)^{-aff}}{\left(\frac{470}{950}\right)^{-awb} - \left(\frac{470}{950}\right)^{-aff}}\right]/b_{abs}(950nm) * BC$$

The absorption Ångström exponents (α) for fossil fuel and wood burning were αff and αwb, respectively. One of the largest sources of uncertainty in the Aethalometer model is related to the section of αff and αwb values (Healy et al., 2017; Zotter et al., 2017). In addition, the αff was typically in the range of ∼ 0.8 – 1.2 in ambient air whereas αwb can vary from 1.6 to 2.2 (Saarikoski et al., 2021). However, we used the αff and αwb values as 0.95 and 1.68 to calculate the BC source (Helin et al., 2018), since our measurement site is in a rural area and nearby a suburban area.

We included this as follow:

"One of the largest sources of uncertainty in the Aethalometer model is related to the section of αff and αwb values (Healy et al., 2017; Zotter et al., 2017). In addition, the αff was typically in the range of ∼ 0.8 – 1.2 in ambient air whereas αwb can vary from 1.6 to 2.2 (Saarikoski et al., 2021). However, we used the αff and αwb values as 0.95 and 1.68 to calculate the BC source (Helin et al., 2018), since our measurement site is in a rural area and nearby a suburban area."

We added some sentences to describe how to determine BrC absorption:
"We assumed that the absorption from dust and other aerosol was negligible. Hence, the absorption was only contributed from BC and BrC. Therefore, Abs(λ) can be divided in BC and BrC absorption:

$$Abs = Abs_{BrC}(\lambda) + Abs_{BC}(\lambda) \tag{1}$$

where $Abs_{BrC}(\lambda)$ is the absorption caused by BrC at the following aethalometer wavelengths, λ = 370, 470, 520, 590, or 660 nm while $Abs_{BC}(\lambda)$ is the absorption contributed by BC at the same wavelength (Wang et al., 2019). To determine $Abs_{BC}(\lambda)$ at each wavelength, we assumed that BC was the only absorber at λ = 880 nm, and thus the $Abs_{BC}(\lambda)$ (λ = 370, 470, 520, 590, and 660) can be extrapolated from the following equation:

$$Abs_{BC}(\lambda) = Abs_{880} \times \left(\frac{\lambda}{880}\right)^{-AAE_{BC}} \tag{2}$$

where AAE_{BC} represents the spectral dependence of $Abs_{BC}(\lambda)$, and a value of 1.0 was chosen for AAE_{BC} based on previous studies in Germany (Teich et al., 2017). Finally, one can obtain the $Abs_{BrC}(\lambda)$ as follows:

$$Abs_{BrC}(\lambda) = Abs(\lambda) - Abs(880) \times \left(\frac{\lambda}{880}\right)^{-AAE_{BC}} \tag{3}$$

6. Moreover, it uses a literature value for the MAC value of BrC. How variable are these values from one site to another? The MAC value could be strongly dependent on the type of BrC being analyzed.
The mass absorption coefficients of potential BrC molecules vary substantially from molecule to molecules (Moschos et al., 2024). Indeed, some weakly absorbing compounds may sometimes contribute with a higher mass fraction and some highly absorbing compounds may dominate the absorption despite of small mass fractions at other locations. In order to estimate the potential light absorption of the 171 potential BrC molecules, we assumed an average MAC value for them in a similar way as we have done previously (Jiang et al., 2022). This allows to estimate the order of magnitude of the potential BrC absorption.

To make this clear we have modified the text in section 3.3 as follows:

"In order to calculate the light absorption from the other 171 potential brown carbon molecules identified, we assumed an average MAC value of 9.5 m^2g^{-1} at 370 nm for all BrC molecules to estimate their absorption (Jiang et al., 2022). So far, the MAC_{370} of most potential brown carbon molecules are still unknown. In addition, since the potential BrC molecules detected by FIGAERO-CIMS could have isomers, we did not calibrate mass absorption coefficients of 171 potential BrC. Despite these uncertainties, we think it is reasonable to estimate the order magnitude of the total BrC absorption based on this assumption."

7. What uncertainties are there in the total organic aerosol mass loading given that there was no measurement of it during the campaign?

Unfortunately, our aerosol mass spectrometer (HR-ToF-AMS, Aerodyne) was not available due to a technical problem during the time of this measurement campaign. Therefore, we have no independent quantification of the total organic aerosol mass loadings. However, we estimated the total organic mass as a fraction of 50 ± 20 % of $PM_{2.5}$ which is a typical fraction for this season and region at the location in Germany (Song et al., 2022; Song et al., 2024). According to this assumption, the organic mass detected by FIGAERO-CIMS based on calibrated sensitivity factors was 37 ± 20 % of the estimated total organic mass. This is in a similar range as observed in previous studies (Ye et al., 2021). We modified the manuscript text as follow:

"We have no independent quantification of the total organic aerosol mass loadings. However, we estimated the total organic mass as a fraction of 50 ± 20 % of $PM_{2.5}$ which is a typical fraction for at the location (Song et al., 2022; Song et al., 2024). According to this assumption, the average organic mass concentration was 4.5 ± 3.1 µg m^{-3}. The organic mass detected by FIGAERO-CIMS based on calibrated sensitivity factors was 37 ± 20% of the estimated organic mass. This is in a similar range as observed in previous studies (Gao et al., 2022; Ye et al., 2021)."

8. The paper does not provide a justification for how BrC molecules are identified from 1000's of mass spectral features, aside from providing a reference. How accurate are the mass fittings and the calculation of DBE and elemental composition for each feature? In other words, are these fittings unique for only one elemental formula? The paper should identify the BrC mass spectral features identified, with some indications of their intensities. Were any mass spectral features observed in both the gas and particle phase spectra? It would be interesting to know this, and a partition coefficient could be calculated.

Lin et al., (2018) assigned potential brown carbon compounds in the plot of DBE vs the number of carbon and nitrogen atoms per molecule (Figure S12). They employed high-resolution mass spectrometry to analyze biomass burning organic aerosol. We used this method to assign 178 potential BrC molecules (including 7 NACs) in the particle phase and 31 potential BrC molecules (including 4 NACs) in the gas phase. Figure 1 shows the mass spectra of organic aerosol detected by FIGAERO-CIMS in the gas and particle phase. The partitioning coefficients of potential BrC molecules are given in Table S6.

We added the following sentences to the text:

"Lin et al., (2016, 2018) employed high-resolution mass spectrometry to analyze biomass burning organic aerosol. They assigned potential brown carbon compounds according to the correlation of double bond equivalents (DBE) with the number of carbon atoms per molecule (Figure S12). We used this method to assign 178 potential BrC molecules (including 7 NACs) in the particle phase and 31 potential BrC molecules (including 4 NACs) in the gas phase, as shown in Figure 1 in the corresponding mass spectra. The gas to particle phase partitioning coefficients of those semi volatile potential brown carbon molecules which could be measured in both phases with sufficient sensitivity are listed in table S6."
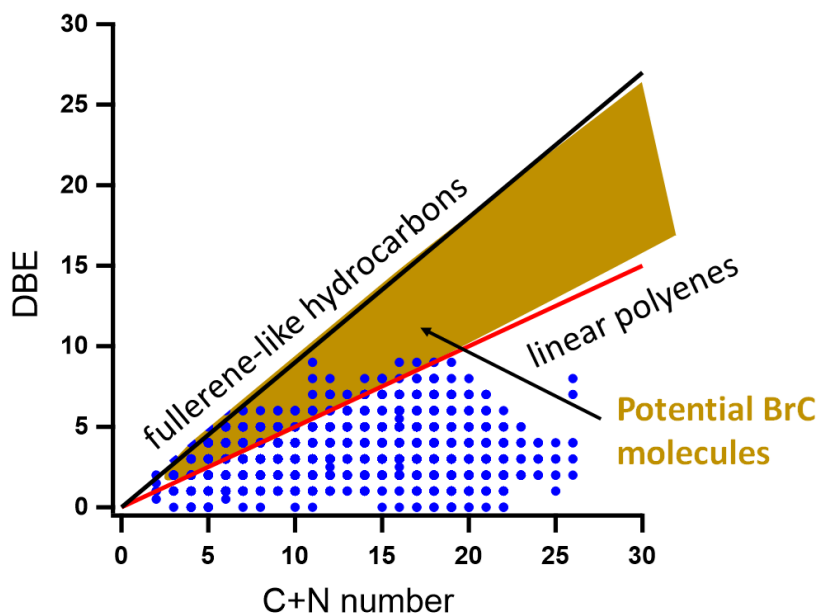

Figure S12. Plot of the double bond equivalent (DBE) vs numbers of carbon and nitrogen atoms according to our measurements following the procedure described by Lin et al., (2018). The lines indicate DBE reference values of linear conjugated polyenes (red solid line) and fullerene-like hydrocarbons with DBE=0.9*C (black solid line). Data points inside the yellow shaded area are potential BrC molecules. (cf. Lin et al., 2018).
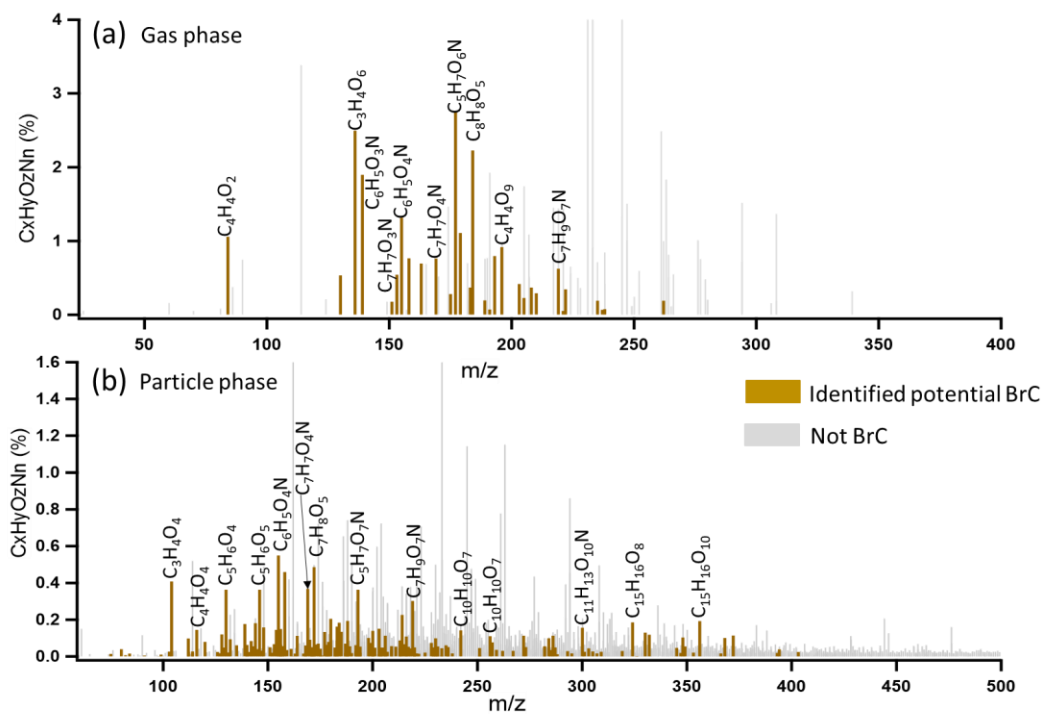
Figure 1. CIMS mass spectra of organic aerosol measured by FIGAERO-CIMS for a biomass burning event on March 1st, 2021, a: gas phase, b: particle phase. The CI source employs reactions of I⁻ ions, which convert analyte molecules into [M+I]⁻ ions. Legends above MS features correspond to neutral molecules. The brown peaks in mass spectra were assigned as potential BrC molecules, while the gray peaks refer to the other organic molecules.

Table S6. Gas to particle phase partitioning coefficients of those semi volatile potential brown carbon molecules which could be measured in both phases with sufficient sensitivity (average and standard deviation).

Number	Formula	Average g/p ratio	Std g/p ratio
1	C3H5O6N1	0.81	0.67
2	C4H4O2	9.7	10.1
3	C4H5O6N1	2.1	1.8
4	C4H5O7N1	1.8	1.8
5	C5H5O8N1	0.8	0.5
6	C5H6O4	0.2	0.4
7	C5H7O6N1	3.2	4.4
8	C5H7O7N1	0.3	0.3
9	C6H6O5	0.1	0.1
10	C6H6O10	1.3	0.9
11	C6H7O6N1	0.4	0.3
12	C6H7O7N1	0.6	0.6
13	C6H7O8N1	0.8	0.4
14	C6H5O3N1	0.5	2.1
15	C6H5O4N1	0.1	0.1
16	C7H7O3N1	0.7	0.4
17	C7H7O4N1	0.3	0.2
18	C7H9O6N1	0.4	0.3
19	C7H9O7N1	0.5	0.6
20	C7H9O8N1	0.4	0.4
21	C8H8O5	0.5	0.3
22	C8H9O4N1	0.3	0.2
23	C9H10O9	0.6	0.4

9. Can a non-parametric wind direction analysis be provided to aid source apportionment?

Such an analysis can of course be an additional indicator for certain sources. However, for our wind measurements, we see a strong impact of channeling by the buildings nearby which inhibits a useful analysis.
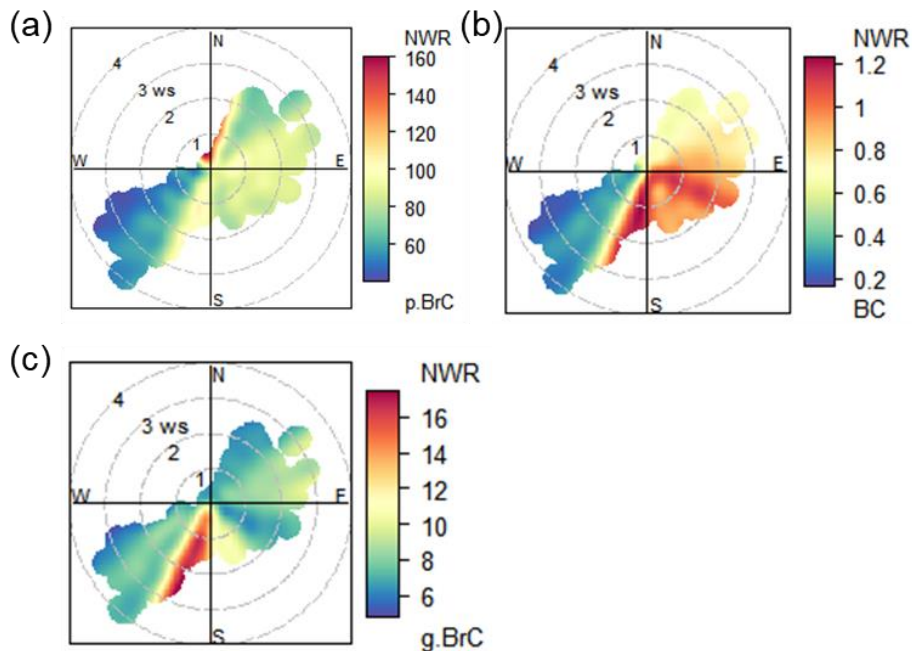

Figure. Non-parametric wind regression (NWR) plots of particle-phase BrC (a), BC (b), and gas-phase BrC (c), respectively. Note: different scales of concentration are used, BrC: ng m^{-3}, BC: μg m^{-3}.

10. Line 267.   Photochemical activity forms ozone and is not the only cause of aging.

We agree and reformulated the text as follows:

"Therefore, the higher fraction of IVOC in the gas phase at daytime is most likely caused by secondary formation e.g. photochemical conversion/aging because of higher oxidant levels as indicated e.g. by higher concentration of ozone at same time (Figure 4c) (Saarikoski et al., 2021)."

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