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Optical properties, chemical characterization and loss pathways of brown carbon molecules in rural Germany

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Brown carbon (BrC) affects the global radiation balance by absorbing light in the ultraviolet (UV) and visible regions (Laskin et al., 2015). However, the molecular characterization and loss pathways of BrC component are still not fully understood. Here, we studied the characteristics of BrC at a rural site in Southwest Germany in winter 2021.

In this study, we measured light absorption of aerosol particles at multiple wavelengths with two Aethalometers (AE33 and MA200). Volatile organic compounds (VOCs) and less oxygenated organic aerosol were measured with PTR-MS coupled with a CHARON particle inlet. Furthermore, the molecular compositions of oxygenated organic aerosol compounds were measured by a filter inlet for gases and aerosols coupled to a high-resolution time-of-flight chemical ionization mass spectrometer (FIGAERO-HR-TOF-CIMS; hereafter CIMS) employing iodide ions.

We identified 178 BrC molecules showing a good correlation (R=0.8) with the absorption at 370 nm (abs_{370}) of BrC. It indicates that the 178 BrC molecules extracted from around two thousand molecules measured by FIGAERO-CIMS were reliable based on the double bond equivalent/carbon number ratio (DBE/C) of each molecule being higher than 0.5 and less than 0.9. The mass concentrations of 178 BrC molecules in the particle and gas phase were 63±33 ng m^{-3} and 6.4±3.0 ng m^{-3}, respectively. The 178 BrC molecules only account for 0.9±0.6% of $PM_{2.5}$ mass concentration, but they can explain 3.7±3.5% of total absorption of BrC at 370 nm, assuming an average mass absorption coefficient at 370 nm (MAC_{370}) as 9.5 m^2 g^{-1}.

Figure 1 shows that the particulate BrC molecules had significant diurnal variations with higher concentrations at nighttime. In contrast, the gaseous BrC molecules showed higher concentrations at daytime. In addition, the volatility of BrC in the gas phase was also higher at daytime. This indicates that the BrC molecules in the particle phase partitioned into the gas phase at daytime. Finally, the BrC molecules show a negative correlation (r=0.5) with O_3, and the O/C ratios of BrC were higher during daytime. These indicated that BrC molecules in the particle phase were photo-oxidized during the day.

[1] Laskin, A., (2015) Chemical Reviews 115, 4335–4382

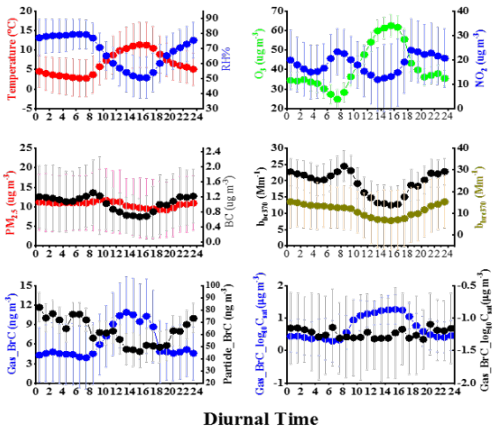

Figure 1. Diurnal variation of temperature, relative humidity (RH), O_3, NO_2, $PM_{2.5}$, BC, b_{bc370}, b_{brc370}, gas BrC, particle BrC, gas-BrC volatility, and particle-BrC volatility during the winter campaign.