Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.14163

Optical Characterization, Seasonality, and Sources of Brown Carbon in Fine Aerosols from Tianjin, North China: Year-round Observations

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Brown carbon (BrC) is an important atmospheric absorber of ultraviolet radiation and plays a key role in climate radiative forcing and photochemistry. To investigate the physicochemical characteristics and radiative forcing of BrC in North China, fine aerosols ($PM_{2.5}$) were collected at an urban site in Tianjin over a 1-year period. They have been studied for UV absorption and excitation emission matrix (EEM) of the BrC extracted into water (WSBrC) and of the water-insoluble (WI)BrC extracted into methanol (MS(WI)BrC), using a three-dimensional fluorescence spectrometer. Average light absorption of both WSBrC ($Abs_{365,WSBrC}$) and MS(WI)BrC ($Abs_{365,MSBrC}$) in each season were distinct from that in other seasons, with higher mass absorption efficiency (MAE) at 365 nm in winter, than in autumn and summer. More chromophores dissolved in organic solvents were demonstrated by $Abs_{365,MSBrC}$ stronger than $Abs_{365,WSBrC}$ at short wavelength. The higher humification index (HIX) together with lower biological index (BIX) and fluorescence index (FI) in WSBrC in summer, suggesting the strong photooxidation and secondary chemical processing of aerosols in summer, whereas in winter, those parameters in MS(WI)BrC were opposite to the former case, implying that the aerosols were significantly influenced by primary emissions and less aging. Based on EEM, the types of chromophores in BrC were divided into humic-like substance (HULIS), including low-oxygenated and high-oxygenated species, and protein like compounds (PLOM). The contribution of humic fluorophore to the fluorescence intensity of WSBrC was more than 60%, indicating that humic fluorophore played a leading role in the fluorescence properties. In MS(WI)BrC, the average contribution of PLOM to fluorescence intensity was higher in summer (25.6%) than in other seasons, indicating that biological activity increased in summer. The direct radiation absorption caused by WSBrC and MS(WI)BrC combinedly in the range of 300-400 nm is accounted for about 40% to the total radiation (range, 300-700 nm), which emphasized that the radiation balance of the Earth's climate system is substantially affected by the BrC and should be considered in the radiative forcing models. These results illustrated the light absorption properties of BrC in metropolis aerosols and emphasized its significant contribution to radiative forcing.