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Dual-carbon isotopic characterization of carbonaceous aerosol reveals different primary and secondary sources in Beijing and Xi'an during severe haze events

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To mitigate haze pollution in China, a better understanding of the sources of carbonaceous aerosols is required due to the complexity in multiple emissions and atmospheric processes. Here we combined the analysis of radiocarbon and the stable isotope ^{13}C to investigate the sources and formation of carbonaceous aerosols collected in two Chinese megacities (Beijing and Xi'an) during severe haze events of "red alarm" level from December 2016 to January 2017. The haze periods with daily $PM_{2.5}$ concentrations as high as ~400 μg m^{-3} were compared to subsequent clean periods (i.e., $PM_{2.5}$ < median concentrations during the winter 2016/2017), with $PM_{2.5}$ concentrationsbelow 100 μg m^{-3} in Xi'an and below 20 μg m^{-3} in Beijing. In Xi'an, liquid fossil fuel combustion was the dominant source of elemental carbon (EC; 44%–57%), followed by biomass burning (25%–29%) and coal combustion (17%–29%). In Beijing, coal combustion contributed 45%–61% of EC and biomass burning (17%–24%) and liquid fossil fuel combustion (22%–33%) contributed less. Non-fossil sources contributed 51%–56% of organic carbon (OC) in Xi'an and fossil sources contributed 63%–69% of OC in Beijing. Secondary OC (SOC) was largely contributed by non-fossil sources in Xi'an (56 ± 6%) and by fossil sources in Beijing (75 ± 10%), especially during haze periods. The fossil vs. non-fossil contributions to OC and EC did not change drastically during haze events in both Xi'an and Beijing. However, compared to clean periods, the contribution of coal combustion to EC during haze periods increased in Xi'an and decreased in Beijing. During clean periods, primary OC from biomass burning and fossil sources constituted ~70% of OC in Xi'an and ~53% of OC in Beijing. From clean to haze periods, the contribution of SOC to total OC increased in Xi'an, but decreased in Beijing, suggesting that contribution of secondary organic aerosol formation to increased OC during haze periods was more efficient in Xi'an than in Beijing. In Beijing, the high SOC fraction in total OC during clean periods was mainly due to elevated contribution from non-fossil SOC. In Xi'an, a slight day-night difference was observed during the clean period, with enhanced fossil contributions to OC and EC during the day. This day-night difference was negligible during severe haze periods, likely due to enhanced accumulation of pollutants under stagnant weather conditions.