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Wintertime aerosol chemistry and haze evolution in an extremely polluted city of North China Plain: significant contribution from coal and biomass combustions

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The North China Plain (NCP) frequently encountered heavy haze pollution in recent years, particularly during wintertime. In 2015-2016 winter, the NCP region suffered several extremely severe haze episodes with air pollution red alerts issued in many cities. In this work, we investigated the sources and aerosol evolution processes of the severe pollution episodes in Handan, a typical industrialized city in the NCP region, using real-time measurements from an intensive field campaign during the winter of 2015-2016. The average ($\pm$ 1σ) concentration of submicron aerosol (PM_1) during December 3, 2015 – February 5, 2016 was 187.6 ($\pm$ 137.5) μg m^{-3}, with the hourly maximum reaching 700.8 μg m^{-3}. Organic was the most abundant component, on average accounting for 45% of total PM_1 mass, followed by sulfate (15%), nitrate (14%), ammonium (12%), chloride (9%) and BC (5%). Positive matrix factorization (PMF) with multi-linear engine (ME-2) identified four major organic aerosol (OA) sources, including traffic emissions represented by a hydrocarbon-like OA (HOA, 7% of total OA), industrial and residential burning of coal represented by a coal combustion OA (CCOA, 29% of total OA), open and domestic combustion of wood and crop residuals represented by a biomass burning OA (BBOA, 25% of total OA), and formation of secondary OA (SOA) in the atmosphere represented by an oxygenated OA (OOA, 39% of total OA). Emissions of primary OA (POA), which together accounted for 61% of total OA and 27% of PM_1, are a major cause of air pollution in this region during the winter. Our analysis further uncovered that, primary emissions from coal combustion and biomass burning together with secondary formation of sulfate (mainly from SO_2 emitted by coal combustion) are important driving factors for haze evolution. However, the bulk composition of PM_1 showed comparatively small variations between less polluted periods (daily $PM_{2.5}$ $\leq$ 75 μg m^{-3}) and severely polluted periods (daily $PM_{2.5}$ $>$ 75 μg m^{-3}), indicating relatively synchronous increases of all aerosol species during haze formation. The case study of a severe haze episode, which lasted 8 days starting with a steady build-up of aerosol pollution followed by a persistently high level of PM_1 (326.7 – 700.8 μg m^{-3}), revealed the significant influences of stagnant meteorological conditions on acerbating air pollution problems in the Handan region. The haze episode ended with a shift of wind which brought in cleaner air masses from the northwest of Handan and gradually reduced PM_1 concentration to $<$ 50 μg m^{-3} after 12 hours. Furthermore, aqueous-phase reactions under higher relative humidity (RH) were found to significantly promote the production of secondary inorganic species (especially sulfate), but showed little influence on SOA.