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Modeling of evolution of aerosol properties during winter haze episodes over a megacity cluster in North China: roles of regional transport and heterogeneous reactions

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Regional transport and heterogeneous reactions played key roles in haze formation in a megacity cluster centered on Beijing in North China. In this study, the updated Nested Air Quality Prediction Model System (NAQPMS) and a Lagrange trajectory model named as HYSPLIT were employed to investigate the evolution of aerosols on number concentration, size distribution and aging degree in Beijing during six haze episodes from November 15 to December15, 2016, as a part of Air Pollution and Human Health-Beijing (APHH-Beijing) winter campaign in 2016. The model showed reasonable performance not only in mass concentrations of $PM_{2.5}$and its components in Beijing, but also number concentrations, size distribution and aging degrees. We found that regional transport played an innegligible role in haze episodes, with contributions of 14%-31% to surface $PM_{2.5}$ mass concentrations. Regional transport of secondary inorganic aerosols were more significant than primary aerosols (30-63% .vs. 3-12%). The chemical transformation of SO_2 in the transport pathway from source regions to Beijing was the major form of SO_4^{2-} regional transport. We also found that sulfate formed outside Beijing, by SO_2 emitted in Beijing, can be flowed back to Beijing and significantly affected the haze formation. In the transport pathway, aerosols experienced a significant aging process, which altered mass ratio of coating-to-BC (R_{BC})and size distribution of number concentrations. During episodes, geometric mean diameter (GMD) increased to $\sim$120 nm at the ending site (Beijing) from less than 100nm at the staring site. And the R_{BC} increased from 2$\sim$4 to 4$\sim$8. These changes would affect regional radiation and climate change. In haze episodes with high humidity, averaged contributions of gas and aqueous chemistry, heterogeneous chemistry and primary sulfate were comparable. Impact of primary emissions mostly concentrated in light-medium pollution levels, while heterogeneous chemistry was more significant in high polluted levels.