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Summertime $PM_{2.5}$ ionic species in four major cities of China: nitrate formation in an ammonia-deficient atmosphere

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Abstract. Strong atmospheric photochemistry in summer can produce a significant amount of secondary aerosols, which may have a large impact on regional air quality and visibility. In the study reported herein, we analyzed sulfate, nitrate, and ammonium in $PM_{2.5}$ samples collected using a 24-h filter system at suburban and rural sites near four major cities in China (Beijing, Shanghai, Guangzhou, and Lanzhou). Overall, the $PM_{2.5}$ mass concentrations were high (with a mean value of 55–68 gμgm^{-3}), which reflects the long-known particulate pollution in China's large urban centers. We observed very high concentrations of sulfate and nitrate at the Beijing and Shanghai sites, and, in particular, abnormally high levels of nitrate (24-h average concentration up to 42 gμgm^{-3} and contributing up to 25% of the $PM_{2.5}$ mass) in the ammonium-poor samples. The Beijing and Shanghai aerosols were characterized by high levels of aerosol acidity (~220–390 nmol m^{-3}) and low levels of in-situ pH (−0.77 to −0.52). In these samples, the formation of the observed high concentrations of particulate nitrate cannot be explained by homogeneous gas-phase reaction between ammonia and nitric acid. Examination of the relation of nitrate to relative humidity and aerosol loading suggests that the nitrate was most probably formed via the heterogeneous hydrolysis of N_2O_5 on the surface of the moist and acidic aerosols in Beijing and Shanghai. In comparison, the samples collected in Lanzhou and Guangzhou were ammonium-rich with low levels of aerosol acidity (~65–70 nmol m^{-3}), and the formation of ammonium nitrate via the homogeneous gas-phase reaction was favored, which is similar to many previous studies. An empirical fit has been derived to relate fine nitrate to aerosol acidity, aerosol water content, aerosol surface area, and the precursor of nitrate for the data from Beijing and Shanghai.

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