Geophysical Research Abstracts
Vol. 20, EGU2018-15130, 2018
EGU General Assembly 2018

Elucidating contributors of aerosol acidity and the explanation of elevated pH during severe winter haze in Beijing

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Aerosol acidity is a key parameter in determining atmospheric heterogeneous reactions, influencing both the thermodynamic equilibrium processes (namely gas-particle partitioning) and the chemical kinetics (namely the reaction rate constants). Heterogeneous reactions in aerosol water are suggested as the dominate contributor to the severe haze formation in China, while the major pathway is largely determined by aerosol acidity. Under more acidic conditions, sulfate production is dominated by transition metal ions catalyzed oxygen oxidation, while under near-neutral conditions the NO_2 oxidation pathway could excel. Higher aerosol pH of 4∼7 during the extreme Beijing hazes is estimated, despite the NH3-buffering effect which was shown to keep southeastern US aerosol pH within 0∼2. The NH3-buffering effect, or the effect that gas-phase NH3 have to increase by 10 times in order to increase aerosol pH by 1 unit, can well explain the long-term trend of aerosol acidity dependence on SO_2 emissions in southeastern US. However, some recent studies suggested that this effect would sometimes disagree with observed gas-particle partitioning, potentially due to influence of organic compounds. Discrepancies exist among southeastern US cases, let alone between southeastern US and winter Beijing cases. Here we proposed the governing equation of aerosol acidity, and show that under Beijing winter conditions, the NH3-buffering effect could still exist but would buffer in much higher pH ranges of 4∼7. That is due to the combination of lower temperature, higher gas-phase NH3, higher non-volatile cations and higher nitrate in Beijing winter hazes. Moreover, size heterogeneity of aerosol compositions can further elevate overall sulfate production rate through NO_2 oxidation. Other influencing factors, including the transition metal ions, relative model uncertainty and organic compounds, are also investigated.