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Aerosol pH and regime transition of sulfate formation during a severe winter haze event in northern China

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Understanding the formation mechanism of severe haze is crucial for the development of efficient pollution control strategy. Recently, multiphase reactions in aerosol water has been for the first time suggested as an important source of sulfate aerosol during severe haze (Zheng et al. 2015; Cheng et al. 2016; Wang et al. 2016). Though several oxidation mechanisms have been recognized, the dominant oxidation pathway is still under debate reflecting a missing consensus. Based on a model survey with Weather Research and Forecasting model coupled with Chemistry (WRF/Chem), we investigated the variability of aerosol pH and regimes of sulfate formation through multiphase oxidations during the haze episodes in January of 2013. Our results show a large spatial and temporal variability in the aerosol pH and sulfate formation regimes. Surface aerosol pH shows a clear diurnal variation with low pH during daytime and high pH during nighttime for most cases. Aerosol pH tends to decrease with increasing altitude in the boundary layer. For the scenario best reproduces the observations in Beijing, NO_2 , O_3 and TMI reactions all can dominate the oxidation pathways of sulfate in northern and middle Beijing-Tianjin-Hebei (BTH) areas. H_2O_2 pathway has the greatest contribution over the most southern BTH areas. With the increasing height, O_3 pathway and gas phase oxidation by OH radicals become more important. Moreover, our study also suggests that, emissions of crustal particles, NH3 and soluble iron/manganese have great impacts on aqueous phase chemistry, and should be better constrained in future studies.