Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-894-RC2, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "In-cloud formation of secondary species in iron-containing particles" *by* Qinhao Lin et al.

Anonymous Referee #1

Received and published: 13 November 2018

General comments

The authors presented single-particle measurements of iron-containing aerosols to analyze the formation of secondary species. The work conducted in this paper may contribute to improve our understanding of effect of iron on the formation of secondary species, although more careful discussions are required for the iron chemistry in aerosol and cloud water. I have some comments and questions to improve this paper.

Specific comments

p.9, l.187, Figure S2: Please indicate the upper size of the particles.

p.12, l.249: Please present the references and specify the in-cloud chloride formation

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Discussion paper

and irradiation effect.

p.12, l.256: Did you measure HCl concentration? What are the sources of HCl in the region?

p.15, l.319: The Fenton reaction is a sink of ferrous iron. Please consider the redox cycling for the role of the Fenton reaction in the formation of aqueous-phase OH.

p.18., l.378: The photolysis of Fe-oxalate complexes could reduce the net amount of oxalate formation in cloud droplet, even though the Fenton reaction increases the formation rate of aqueous-phase OH (Sorooshian et al., 2013; Lin et al., 2014). Thus, other mechanisms (e.g., strong complexation of iron with organic compounds) are needed to enhance the oxalate concentration in Fe-rich particles. Please consider the effect of Fe chemistry on the net formation of oxalate and discuss the mechanisms to enhance the oxalate concentration in Fe-rich particles.

p.18., l.384: The formation of stable complexes (e.g., calcium oxalate) is assumed for the SOA yield to remain entirely in the particulate phase when the water is evaporated. This is different from the suppression of oxalate-promoted Fe dissolution in the model (Ito, 2015), which is based on the laboratory measurements (Chen and Grassian, 2013). At low concentration of oxalate, complexation of oxalate with iron on the surface of the particle is suppressed through formation of Fe-oxalate complexes in solution. Less content of Ca-containing particles suggests low pH in aerosol water. It was proposed that the competition is more significant at low pH. Thus, excess oxalate is needed to enhance Fe dissolution in aerosols. Please consider the effect of the ligand concentration on Fe dissolution in aerosols and discuss the implications.

p.19, l.398: Please reconsider this. The Fenton reaction increases the formation rate of aqueous-phase OH, but the net amount of oxalate formation in cloud droplet is reduced by the photolysis of Fe-oxalate complexes. Please also consider the pH upon droplet evaporation when the ammonium species may enhance the SOA yield.

References

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Ito, A.: Atmospheric Processing of Combustion Aerosols as a Source of Bioavailable Iron, Environ. Sci. Technol. Lett., 2(3), 70–75, doi:10.1021/acs.estlett.5b00007, 2015.

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Sorooshian, A., Wang, Z., Coggon, M. M., Jonsson, H. H., and Ervens, B.: Observations of sharp oxalate reductions in stratocumulus clouds at variable altitudes: organic acid and metal measurements during the 2011 E-PEACE Campaign, Environ. Sci. Technol., 47, 7747–7756, https://doi.org/10.1021/es4012383, 2013.

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