EGU21-727, updated on 05 Dec 2022
https://doi.org/10.5194/egusphere-egu21-727
EGU General Assembly 2021

Surface-Promoted Redox Reactions Occurs Spontaneously on Solvating Salt Surfaces

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Gas-particle interfaces play essential roles in the atmosphere and directly influence many atmospheric processes, including gas uptake, halogen chemistry, ozone depletion, and heterogeneous ice nucleation. However, because interfacial processes take place on molecular scales, classical bulk thermodynamic theories are often insufficient to describe interfaces. Also, interfacial processes are challenging to characterize and are often overlooked in current atmospheric chemistry.

For this study, ambient pressure X-ray photoelectron spectroscopy (APXPS) experiments were performed. A surface-promoted sulfate-reducing ammonium oxidation reaction is discovered to spontaneously take place on common inorganic aerosol surfaces undergoing solvation. Several key intermediate species including, S^0, HS^-, HONO, and $NH_{3(aq)}$ are identified as reaction components associated with the solvation process. Depth profiles of relative species abundance show the surface propensity of key species. The species assignments and depth profile features are supported by classical and first-principle molecular dynamics calculations. A detailed mechanism is proposed to describe the processes that lead to unexpected products during salt solvation. This discovery reveals novel chemistry that is uniquely linked to a solvating surface and has great potential to illuminate current puzzles within heterogeneous chemistry. Lastly, natural salts sampled from saline lakes and playas are examined for this behavior, and provide further evidence of the important roles this surface-promoted redox mechanism may play in nature.