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Understanding the “Water-on” Surface Catalysis in the context of Atmospheric Chemistry

Content

The gas-particle surface is of essential importance as it directly determines many atmospheric processes, (George, Ammann et al. 2015) e.g., trace gases uptake, redox reactions, ozone depletion and heterogeneous ice nucleation. However, because surface processes take place on molecular scales, classical bulk thermodynamic theories are often insufficient to describe interfaces. For example, hygroscopic substrates are understood to solvate at a deliquescence relative humidity (DRH), but modern observational techniques have revealed that reversible water adsorption and reversible ion solvation already take place at a relative humidity (RH) lower than the DRH. (Kong, Castarède et al. 2020) Also, surfaces may spontaneously undergo abnormal chemical reactions that are thermodynamically unfavored and at first glance unpredictable.

Recently, we discovered a surface-catalyzed redox mechanism on typical inorganic aerosol surfaces when condensed water is solvating the surface. (Kong, Castarede et al. 2021) Specifically, on the surface of ammonium sulfate (a common proxy of atmospheric particles), ammonium (NH_4^+) ions and sulfate (SO_4^{2-}) ions react with each other spontaneously, forming various species containing differently oxidized nitrogen and sulfur elements (e.g., HONO , N_2 , S_0 , HS^-). (Kong, Castarede et al. 2021) The reaction theme is similar to a previously hypothesized sulfate-reducing ammonium oxidation (SRAO) mechanism. (Rikmann, Zekker et al. 2012) A major difference is that the SRAO reaction requires catalysts (e.g., bacterial) to overcome the energy barrier, but these water-involved reactions occur spontaneously on surfaces. In analogue to the previously discovered “on-water” catalysis (surface water in aqueous phase), (Ruiz-Lopez, Francisco et al. 2020) here the new mechanism is named “water-on” surface catalysis (surface water from gas phase). These surface processes are challenging to characterize and are often overlooked in current atmospheric chemistry but can be addressed utilizing novel analytical techniques and explorative curiosity-driven approaches.

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