Effect of Ammonia on Hydration of Sulfuric Acid Dimer

Dechen Yangkyi, Berhane Temelso, George C. Shields
Department of Chemistry, College of Arts and Sciences, Bucknell University, Lewisburg, PA 17837

Aerosols play a major role in global climate by countering the warming effect of greenhouse gases, but knowledge of their formation and subsequent growth are still very limited. Sulphuric acid and water are key ingredients for aerosol formation, but this binary system alone is not enough to explain observed aerosol formation as was previously thought, without contribution from other compounds. One of the major contributing compounds is ammonia. Ammonia is very abundant in the atmosphere, and its strong basicity stabilizes sulfuric acid - water clusters very well and therefore, enhances growth.

In this study, we looked at both neutral and ionic $(H_2SO_4)_2NH_3(H_2O)_n$ system where n=0-6. We adopted a genetic algorithm (GA) employing the PM7 and SCC-DFBT semi-empirical methods to perform configurational sampling. The two semi-empirical methods performed equivalently in our initial benchmarks. We performed high level quantum mechanical calculations using MP2 due to its ability to model hydrogen bonding well and reasonable computational cost. A comparison of our results with those from previous literature for systems like $(H_2SO_4)(NH_3)(H_2O)_n$, $(H_2SO_4)_2(CH_2NH_3)(H_2O)_n$ and $(H_2SO_4)_2(H_2O)_n$ will be discussed. Also, the implication of those findings for aerosol formation will be explored.

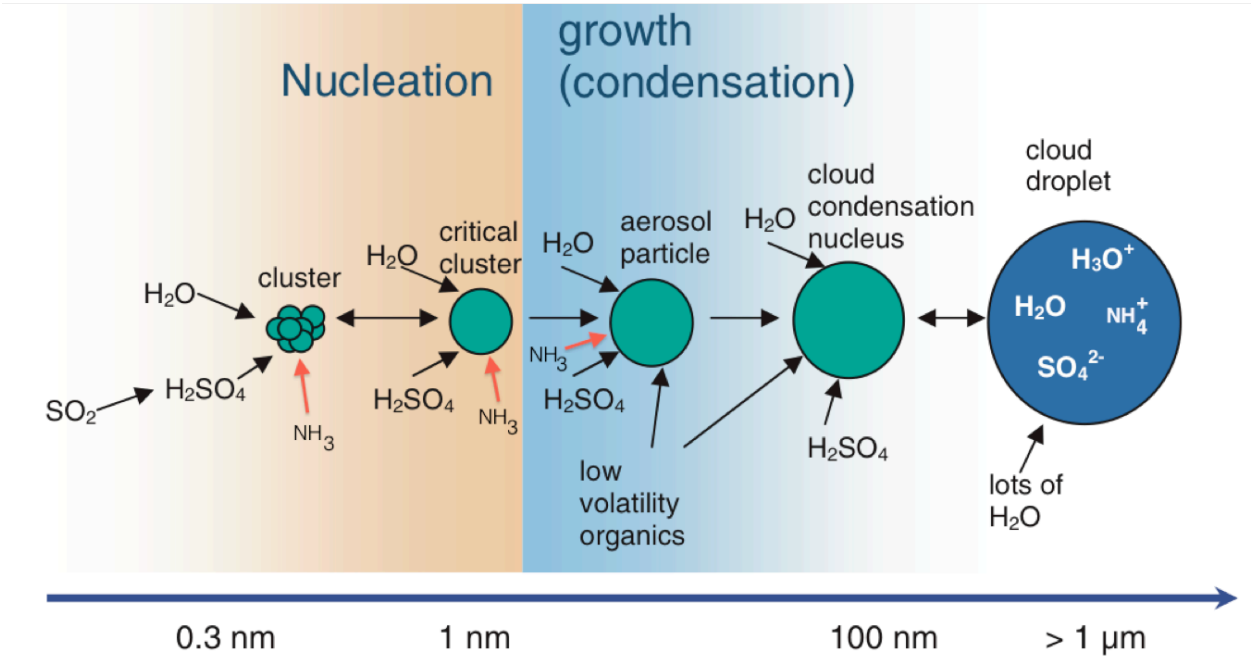

Figure 1. Aerosol nucleation and growth mechanisms. Figure adapted from Curtius, J. EPJ Web of Conferences 2009, 1, 199-209.