Sulfate Aerosol Formation Rates in the Presence of Different Bases: Ammonia, Methylamine and Mixed Ammonia-Methylamine

Nana Appiah-Padi*, Berhane Temelso, George C. Shields
*Lewisburg Area High School, Lewisburg PA 17837
Department of Chemistry, College of Arts & Sciences, Bucknell University, Lewisburg PA 17837

Aerosol particles in the atmosphere reflect back sunlight and regulate the lifecycle of clouds. Our work focused on ternary sulfate aerosols containing methylamine (MA, CH_3NH_2), ammonia (A, NH_3), the combination of the two (MA-A) and sulfuric acid dimers (S_2) and compared their effectiveness in new particle formation in the atmosphere. We chose MA_2, A_2, and MA-A because recent experiments have shown that the presence of heterogeneous bases (MA-A) enhances aerosol formation significantly more than homogeneous bases (A_2 and MA_2). This work attempts to explain this phenomena using computational tools.

To sample the possible configurations, we used genetic algorithm (GA) with the PM7 and SCC-DFTB semiempirical methods. The unique low energy structures from the final pool of 1500 structures for PM7 and DFTB were minimized using the PW91/ 6-31G* density functional method. These structures were finally run through PW91, M06-2X and wB97X-D methods with 6-311++G** basis sets. Although experimental measurements showed that there was a greater new particle formation rate for the MA-A system, followed by MA_2 and A_2, the computed binding energy of the clusters follows a different order: MA_2 > MA-A > A_2. The reasons for these differences between computation and experiment will be explored.

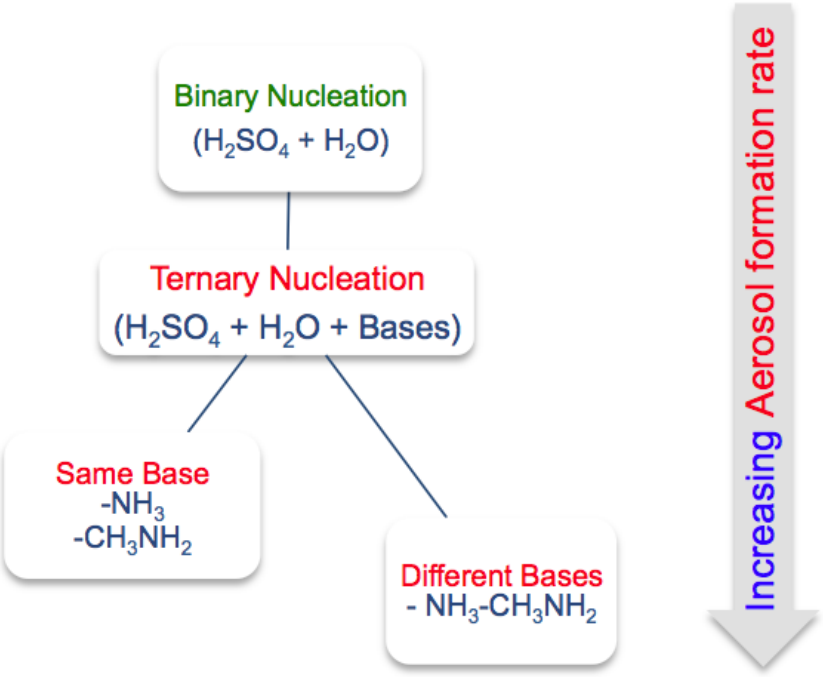

Figure 1: Aerosol formation rates are higher for ternary systems with two different bases than those with same bases.