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On the Stability and Dynamics of Atmospheric Pre-Nucleation Clusters VILLE LOUKONEN, University of Helsinki, Department of Physics, Helsinki, FI-00014 University of Helsinki, Finland, I-FENG W. KUO, Lawrence Livermore National Laboratory, Chemical Sciences Division, Livermore, CA 94550 USA, MATTHEW J. MCGRATH, Department of Biophysics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan, HANNA VEHKAMAKI, University of Helsinki, Department of Physics, Helsinki, FI-00014 University of Helsinki, Finland — Atmospheric new-particle formation is a complex physical phenomenon with far-reaching consequences: currently, the role of aerosols is one of the main uncertainties in predicting the climate change. However, the molecular-level particle formation mechanisms are poorly understood. It is believed that sulfuric acid is the key player with possible contributions from various base molecules, ions or organics. Here we present results from first-principles molecular dynamics simulations of molecular clusters of sulfuric acid and two atmospherically relevant bases, ammonia and dimethylamine. The dynamics and stability of the studied clusters (sulfuric acid)$_n$•(ammonia)$_{n-1}$ and (sulfuric acid)$_n$• (dimethylamine)$_n$ where $n = 2, 3, 4$ were probed for 45 ps at T=300K at BPE/TZV2P level of theory. The stability of the clusters is largely dependent on the H-bonding patterns and in most cases the equilibrium patterns emerged within the first 10 ps. Curiously, even after the equilibrium was reached the clusters showed pronounced bond rearrangement: the number of bonds remained the same, but the individual atoms forming the bonds changed. Regardless of this behavior, the clusters remained bound together.

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