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Secondary new particle formation initiated by sulfuric acid-amine nucleation in Beijing

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Secondary new particle formation is an important source of the number concentration of atmospheric aerosols. Despite relatively high coagulation sinks contributed by pre-existing aerosols, intensive new particle formation occurs frequently in polluted atmospheric environments such as in urban Beijing. Considering the measured concentrations of sulfuric acid and organic compounds, the contrast between the high coagulation sink and the frequent intensive NPF events in urban Beijing indicates an efficient nucleation mechanism. Based on long-term atmospheric measurements conducted at the campus of Beijing University of Chemical Technology, we show that sulfuric acid-amine nucleation is a governing mechanism to initiate new particle formation in urban Beijing. The molecular-level mechanism of sulfuric acid-amine nucleation, especially with low amine concentrations and high aerosol concentrations, are discussed. We present evidence for the existence of the missing amine molecules in the measured H_2SO_4-amine clusters. A neutral cluster needs to be ionized before it is detected by a mass spectrometer. Deprotonation or clustering with an additional reagent ion changes the stability of the original neutral cluster. Therefore, the amine molecules in neutral H_2SO_4-amine clusters may dissociate before detection. Combining measurements and cluster kinetic simulations, we show that although not directly detected, a considerable proportion of H_2SO_4 monomers exist in the form of $(H_2SO_4)_1(amine)_1$, where the amine is most likely to be dimethylamine or trimethylamine. The evaporation rate of $(H_2SO_4)_1(amine)_1$ is moderate and forming $(H_2SO_4)_1(amine)_1$ is a critical step for H_2SO_4-amine nucleation. According to nucleation theory, $(H_2SO_4)_1(amine)_1$ is the critical cluster at a low amine concentration, whereas H_2SO_4-amine nucleation may occur without a free energy barrier at a high amine concentration. The clustering between $(H_2SO_4)_1(amine)_1$ and $(H_2SO_4)_n(amine)_n$ is a major reaction pathway for the initial growth of H_2SO_4-amine clusters. These findings are supported by the measured H_2SO_4 dimer concentration and its dependencies on amine concentrations and temperature in urban Beijing. Besides, the enhancement of cluster growth rate due to synergy between amines and ammonia are discussed.