Self-catalytic reaction of SO_3 and NH_3 to produce sulfamic acid and its implication to atmospheric particle formation

With the support of the National Natural Science Foundation of China, a research team including a group led by Professor Zhang XiuHui (张秀辉) from Beijing Institute of Technology, a group led by Professors Xiao Cheng Zeng and Joseph S. Francisco from the University of Nebraska-Lincoln, a group led by Professor Ge MaoFa from the Institute of Chemistry, Chinese Academy of Sciences, and collaborators from the University of Helsinki, made a major advancement in the study of new particle formation in highly polluted regions. This team work was published in *JACS* (2018, 140(35): 11020—11028).

Aerosol new particles are a main source of particulate matter in atmosphere and cloud condensation nuclei. High concentration of particular matter in troposphere can have significant impacts on environmental quality, regional climate, and human health. Especially, new particle formation (NPF) events tend to occur more frequently in megacities in China because of the higher content of SO_2 and NH_3 molecules being released in atmosphere through industry, transportation, and anthropogenic processes. However, the molecular physicochemical mechanism underlying the NPF, especially the one related to the realistic pollution events in highly polluted regions, are still open questions, due largely to the limiation of today's experimental techniques in measuring time-dependent dynamics of NPF.

In the reported work, the self-catalytic reaction between SO_3 and NH_3 was shown by using quantum chemical calculation and the atmospheric cluster dynamics code. The team of researchers identified a new and competitive loss pathway of SO_3 when abundant ammonia (NH_3) species are present in the atmosphere. The team found that the effective rate constant for the bimolecular SO_3-NH_3 reaction can be high enough such that the new loss pathway for SO_3 becomes as competitive as the conventional loss pathway of SO_3 with water. The final product of the self-catalyzed reaction, sulfamic acid, then participates in NPF and, in particular, can enhance the rate of NPF originating from sulfuric acid and dimethylamine by about a factor of two. The new chemical mechanism has implication to atmospheric modeling of particulate matter, especially for events in highly polluted regions in China.

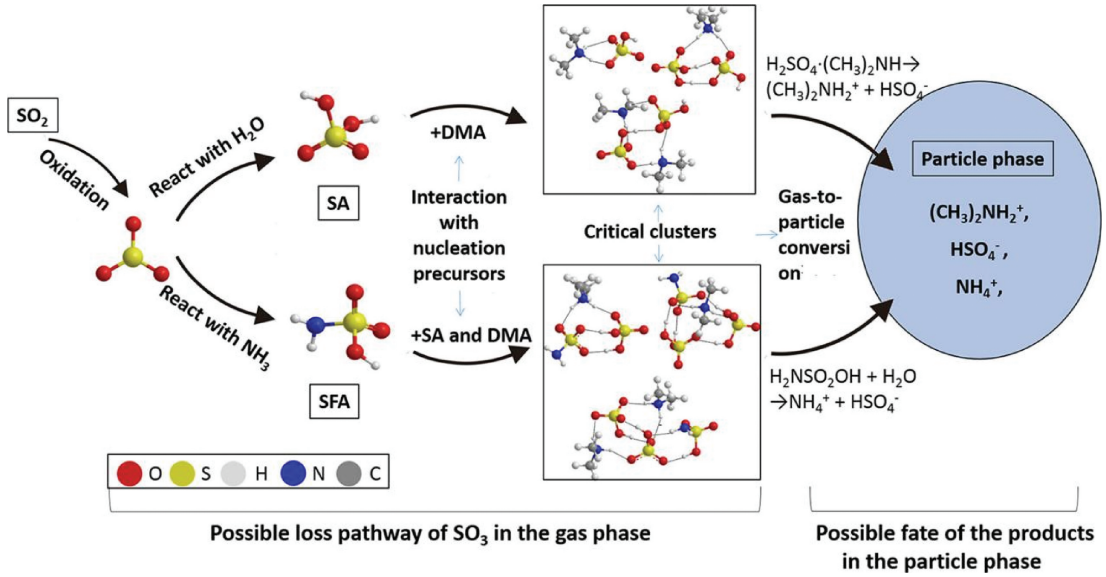

Figure The possible fate of pollutant SO_3 in the atmosphere.