Linking Neutral and Charged Sulfuric Acid - Ammonia and Sulfuric Acid - Dimethylamine Clusters

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New-particle formation is an important source of aerosols and cloud condensation nuclei in the atmosphere (Merikanto et al. 2009). But, despite its importance, the process is still poorly understood. The main problem about understanding new-particle formation is the size of the nucleating clusters. They are formed by only few molecules, and they have diameters close to 1 nm. Therefore they are well below present detection limits if they are electrically neutral. Very recent developments in cutting-edge high-resolution mass spectroscopy (APi-TOF instrument) are starting to permit the unambiguous chemical characterization of charged atmospheric clusters from molecular scale upward (Junninen et al. 2010). However, these experiments alone yield no information on the neutral clusters. A charger can be included before the mass spectrometer (e.g. CI-APi-TOF), but depending on the charging mechanism used in the instruments, some cluster types may not be charged, others may be broken up and others may be chemically completely altered by the process.

We have used quantum chemical calculations (Ortega et al. 2012) to calculate the evaporation rates of charged sulfuric acid-ammonia and sulfuric acid–DMA clusters. Figures 1 and 2 compare the evaporation rates of neutral clusters (Ortega et al. 2012) with the evaporation rates of the corresponding charged clusters and the evaporation rates of the most stable charged clusters.

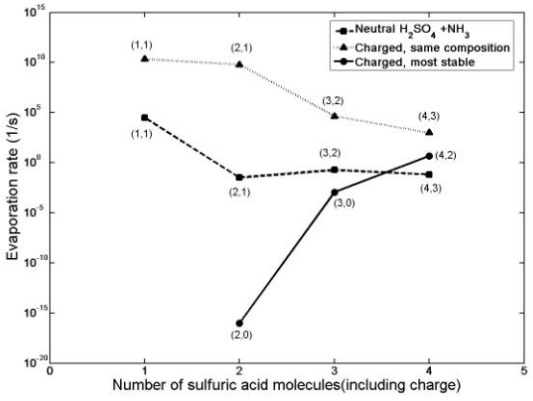

Figure 1. Evaporation rates of ammonia containing clusters.

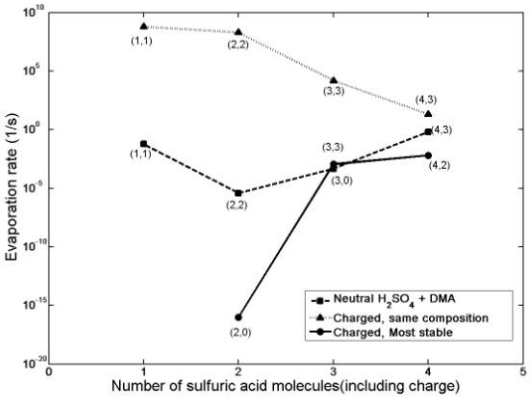

Figure 2. Evaporation rates of DMA containing clusters.

We found that ammonia and DMA molecules will evaporate from small negatively charged sulfuric acid clusters. Ammonia will evaporate from clusters with three or less sulfuric acid molecules and DMAwill evaporate from clusters with two or less sulfuric acid molecules. This shows how, although small neutral clusters containing base molecules are stable, they will lose the base molecules when charged and be detected as charged pure sulfuric acid clusters. These results can explain why small charged ammonia clusters were not observed by APi-TOF during the CLOUD experiment (Kirkby et al. 2011).

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