V-R Ion-Molecule Reactions in the Troposphere

Ion chemistry in the troposphere is the most complicated among all level of earth's atmosphere because of the presence of a variety of trace compounds. We have studied ion-molecule reactions in the troposphere by investigating ion mobility distribution and its dependence on reaction time, pressure and temperature using an ion mobility spectrometer.[1,2] In order to confirm the ion-molecule reactions occurring in the troposphere, we have developed a high-resolution ion mobility/mass spectrometer which is capable of chemical identification of ion species forming ion peaks in mobility spectra. Using this spectrometer, we have been investigating the ion processes in conditioned laboratory air as well as in natural air. Experiments have been extended to the study on the role of ion-induced nucleation in the troposphere.

References
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V-R-1 Measurements of Mobility and Mass Spectra of Tropospheric Ions

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An ion mobility/mass spectrometer (Figure 1) has been developed for the purpose of investigating the ion evolution in the lower troposphere. Total ion mobility spectra, mass spectra, and mass-resolved mobility spectra were obtained for positive and negative ions produced and aged for 0.02–0.5 s in the ambient air. In this range of aging time, positive ions were observed to evolve from $NH_4^+(H_2O)_n$ to the ions of pyridines and amines that have higher proton affinities. It is also demonstrated that mass-resolved mobility spectra could be useful to analyze the detailed pathway of the evolution. Four series of hydrated cluster ions were observed in the negative ion mass spectra, which are believed to be ions of formic acid ($HCOO^-(H_2O)_n$) and oxalic acid ($COOHCOO^-(H_2O)_n$) along with $NO_2^-(H_2O)_n$ and $NO_3^-(H_2O)_n$. This suggests that organic acids play important roles in the negative ion evolution in the lower troposphere.

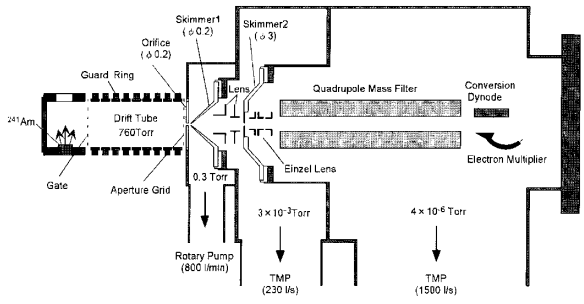

Figure 1. Schematic view of the ion mobility/mass spectrometer.

V-R-2 Experimental Study of Ion-Induced Nucleation in the Troposphere

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There is a growing interest in the ion-induced nucleation in the troposphere.[1] In order to investigate the role of tropospheric ions in the aerosol formation, measurements of mass spectra for positive and negative ions generated by α-ray radiolysis in the $NH_3/SO_2/H_2O$/Air mixture were made by using an ion mobility/mass spectrometer. In the negative ion mass spectra at low SO_2 and H_2O concentrations (Figure 1a), SO_2^-, SO_3^-, SO_4^-, and SO_5^- were observed as major SO_2-originated ions, which indicates that SO_2 were directly ionized by ion-molecule reactions. With increasing the concentrations of SO_2 and H_2O (Figure 1c), HSO_4^- and $HSO_4^-{\cdot}H_2SO_4$ ions became dominant, suggesting that SO_2 were primarily converted into H_2SO_4. This result agrees with our previous experiments that charged particle fraction decreased with increasing the concentrations of SO_2 and H_2O.[2,3] It was also observed that the addition of NH_3 led the decrease in H_2SO_4.

References
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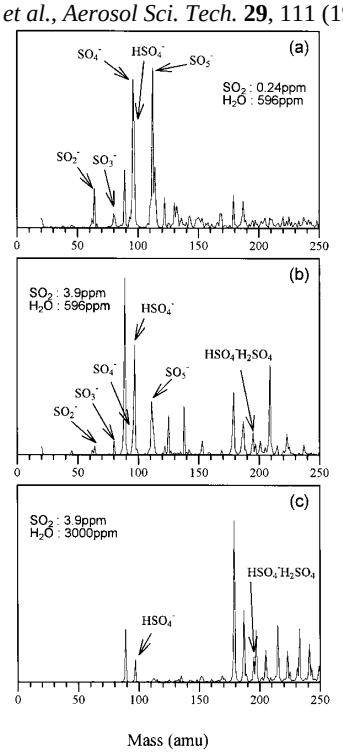

Figure 1. Mass spectra of negative ions generated by α-ray radiolysis in the SO_2/H_2O/Air mixture.