



Reactive Uptake of Ammonia to Secondary Organic Aerosols: Kinetics of Organonitrogen Formation

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Organonitrogen compounds originating from the heterogeneous uptake of NH_3 or amines by secondary organic aerosol (SOA) has received significant attention recently. This is primarily due to its potential contribution to brown carbon (BrC), which can absorb solar radiation and affect climate. In addition, particle phase Organonitrogen species may represent a means of altering regional nitrogen cycles and/or nitrogen deposition patterns through the sequestering of ambient ammonia which is ultimately deposited downwind. Several reduced nitrogen forming heterogeneous reactions have previously been proposed, including Schiff base and/or Mannich reactions between NH_3 , ammonium salts or amines and organic carbonyl functional groups in particles.

In order to assess and model the possible impact of Schiff base, Mannich or other N-forming reactions (via NH_3) on the radiative forcing ability of ambient SOA and/or its impact on N-deposition, the kinetics of such heterogeneous reactions are required, and yet remain largely unknown. In the current study, the uptake kinetics of NH_3 to form organonitrogen compounds in SOA derived from the ozonolysis of α -pinene and the OH oxidation of m-xylene is reported for the first time from experiments performed in a 9 m³ smog chamber equipped with a High Resolution Time-of-Flight Aerosol Mass Spectrometer. The results demonstrate that particle bound organonitrogen compounds are mainly formed by NH_3 uptake onto newly formed SOA (~ 1 hr), but relatively little onto more aged SOA. The uptake coefficients of NH_3 to form organonitrogen compounds (between 0-150 min) are on the order of 10^{-4} - 10^{-3} and are prominently dependent upon particle acidity. Following 6 hours of reaction, the total organonitrogen mass contributed up to 10.0 ± 1.5 wt% and 31.5 ± 4.4 wt% to the total SOA mass from the ozonolysis of α -pinene and OH oxidation of m-xylene. The influence of VOC precursors, seed particle acidity and gaseous NH_3 concentration on the obtained uptake coefficients is described. The implications of the kinetics on atmospheric BrC and N-deposition are also discussed.