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Uptake of carbonyl compounds by acidic aerosols: reaction kinetics, mechanisms and light absorbing products.

W. Esteve and B. Noziere

westeve@rsmas.miami.edu
University of Miami / RSMAS

Reactions of volatile organic compounds in acidic aerosols have been shown recently to have 2 important aspects : organic aerosol growth and optical properties of acidic aerosols. Aldol condensation, the acid-catalyzed polymerization of carbonyl compounds, is a likely candidate to enhance the flux of organic matter from the gas phase to the condensed phase in the atmosphere. Until now these reactions have only been characterized for conditions relevant to synthesis, and remote from atmospheric ones.

In this work, we present a complete laboratory study of the uptake and kinetics of gas-phase carbonyl compounds known to be present in the atmosphere (acetone, acetaldehyde, 2-butanone,...) in sulfuric acid aiming at obtaining general information on the reactivity of carbonyl compounds in acidic aerosols. Cross-reactions between aldehydes and ketones, and aldehydes and alcohols have also been investigated. The studies have been performed using a rotating wetted-wall reactor coupled to a mass spectrometer. The uptakes and rate constants have been determined at room temperature as a function of acidity (0-96 % wt. H_2SO_4) for self-reactions and cross-reactions of acetone, and 2,4-pentanedione, 2-butanone, propanal, and at temperature between 248 and 300 K for acetone in 96 % wt. H_2SO_4. Mechanisms are discussed in term of aldol condensation, and compared to the organic chemistry literature. Spectroscopic studies have also been undertaken by measuring the absorption index of sulfuric acid solutions exposed to gas-phase carbonyl compounds. Kinetic information of the formation of light absorbing products have thus been determined, and are discussed in term of effect for atmospheric aerosols.