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Uptake, reactivity and light-absorbing products of carbonyl gases in acidic aerosols

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Two important aspects of the uptake and reactivity of carbonyl gases in acidic aerosols studied in our recent works will be presented:

1) the kinetics and mechanism of the reactions controlling the uptake and partitioning of the gases into the aerosols

2) The formation of light-absorbing products by these reactions.

Kinetic and mechanistic information has been obtained from the uptake of various carbonyl compounds (acetaldehyde, acetone, 2-butanone, and 2,4-pentanedione) by thin films of sulfuric acid solutions 0 – 96 % wt. at room temperature. Aldol condensation was identified as the main reaction of carbonyl compounds in acidic aerosols and a general expression for the rate constant as function of acid concentration is proposed. Based on this expression, the mass increase of Secondary Organic Aerosols in the atmosphere due to such reactions was found to be negligible. The reactivity of many other organic molecules and their contributions to SOA formation remains however to be explored.

The formation of products absorbing light in the near UV and visible range (200 - 1100 nm) by the same reactions has been evidenced. These processes could significantly impact the radiative properties of some types of aerosols believed until now to be non-absorbing. The formation of light-absorbing products was observed in concentrated (96 % wt. H_2SO_4) and moderately acidic solutions (50 and 25 wt. % H_2SO_4) with 6 different carbonyl compounds (acetaldehyde, acetone, propionaldehyde, butyraldehyde, 2-butanone, and trifluoroacetone) and with mixtures of compounds, indicating

that such processes should involve all the carbonyl compounds present in the atmosphere. In many cases exposure to UV-Vis light enhanced the formation of these products. Finally, a study between 273 and 314 K showed that the effect of temperature on the formation rate of the light-absorbing products is relatively small ($\sim$ 30 kJmol^{-1}), because of the opposite effects of temperature on the reaction rate constants and on the Henry's law coefficients. All these results suggest that these processes should be much more common in atmospheric aerosols than previously thought.