Geophysical Research Abstracts, Vol. 7, 09888, 2005
SRef-ID: 1607-7962/gra/EGU05-A-09888

Temperature dependency of ice mixtures formed by co-condensation of gaseous formaldehyde and H_2O: a micro-Raman spectroscopic study

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The formaldehyde-ice system was recently shown to play an important role in the upper troposphere and snowpack chemistry (Winkler et al., 2002, Dominé & Shepson, 2002). Formaldehyde (H_2CO) is an important intermediate in the photochemical oxidation of atmospheric hydrocarbons and is closely linked to the production of HO_x radicals that control photochemical cycles involving ozone budget in the atmosphere. The interaction of trace gases (such as for e.g. formaldehyde) with ice (from ice clouds or ice in snowpack) can perturb this chemistry. The formation of solid ice mixtures by gas condensation (H_2O and trace gases) corresponds to processes occurring in the atmosphere which can affect the ice structure and modify the nature and composition of the incorporated species, in ice or in the gas phase. In this context, we performed a preliminary study by micro-Raman spectroscopy and ab-initio calculations on the effects of freezing on water-formaldehyde aqueous solutions of different concentrations (Chazallon et al., 2005). Crystallization of different phases (stable between 163 K-213 K and 213 K – 234 K) was induced by increasing the temperature. The frozen ice mixtures were found to be composed of ice Ih and formaldehyde polymers of different chain lengths, as suggested by our infrared laser desorption work (Mihesan et al., 2004). The purpose of this work is focused on the characterization by micro-Raman spectroscopy of co-condensed H_2CO:H_2O gas mixtures of different concentrations (Chazallon et al., 2005). An amorphous ice-mixture is formed during the condensation process at $\sim$ 90 K. As temperature increases, a structural re-arrangement occurs

with the formation of a distinct amorphous phase at $\sim$110 K and a crystalline phase at $\sim$ 140 K, close to the temperature of the amorphous-cubic transition in pure ice. At $\sim$150 K, a different crystalline phase can be identified. Only crystalline ice Ih remains after a further heating above $\sim$ 180 K. A comparison of these results with those obtained on the frozen aqueous formaldehyde solutions will be discussed in the context of physics and chemistry of the atmosphere.

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