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Raman scattering investigation of VOCs in interaction with ice particles

Sébastien FACQ, Adriana OANCEA, Cristian FOCSA, and Bertrand CHAZALLON
Université de Lille1, 59655 Villeneuve d'Ascq, France (facq@phlam.univ-lille1.fr)

Cirrus clouds that form in the Earth's upper troposphere (UT) are known to play a significant role in the radiation budget and climate [1]. These clouds that cover about 35% of the Earth's surface [2] are mainly composed of small ice particles that can provide surfaces for trace gas interactions [3]. Volatile Organic Compounds (VOCs) are present in relative high abundance in the UT [4][5]. They promote substantial sources of free OH radicals that are responsible for driving photochemical cycles in the atmosphere. Their presence can both influence the oxidizing capacity and the ozone budget of the atmosphere. VOCs can interact with ice particles via different trapping processes (adsorption, diffusion, freezing, and co-deposition, i.e., incorporation of trace gases during growing ice conditions) which can result in the perturbation of the chemistry and photochemistry of the UT. Knowledge of the incorporation processes of VOCs in ice particles is important in order to understand and predict their impact on the ice particles structure and reactivity and more generally on the cirrus cloud formation. This proceeds via the in-situ characterization of the ice condensed phase in a pressure and temperature range of the UT.

An important mechanism of UT cirrus cloud formation is the heterogeneous ice freezing process. In this study, we examine and characterize the interaction of a VOC, i.e., ethanol (EtOH) with ice particles during freezing. Vibrational spectra of water O-H and EtOH C-H spectral regions are analysed using confocal micro-Raman spectroscopy. Information at the molecular level on the surface structure can be derived from accompanying changes observed in band shapes and vibrational mode frequencies. Depending of the EtOH content, different crystalline phases have been identified and compared to hydrates previously reported for the EtOH-water system. Particular attention is paid on the effect of EtOH aqueous solutions cooling rate and droplet sizes on the phases formed. These results are finally compared with those obtained by co-deposition trapping process.

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