Geophysical Research Abstracts
Vol. 20, EGU2018-10057-1, 2018
EGU General Assembly 2018

Laboratory investigations of Titan's enigmatic stratospheric ice clouds observed by CIRS during the Cassini mission

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Stratospheric ice clouds have been repeatedly observed in Titan's atmosphere by the Cassini Composite InfraRed Spectrometer (CIRS) since the Cassini spacecraft entered into orbit around Saturn in 2004. Most of these stratospheric ice clouds form as a result of vapor condensation processes, composed of a combination of pure and mixed nitriles and hydrocarbons. So far, the crystalline cyanoacetylene (HC_3N) ν_6 band at 506 cm^{-1} (Anderson *et al.*, 2010 and references therein) and a co-condensed nitrile ice feature at 160 cm^{-1}, dominated by a mixture of HCN and HC_3N ices (Anderson and Samuelson, 2011), have been identified in the CIRS limb spectra. However, the presence of other CIRS-observed stratospheric ice emission features, such as the ν_8 band of dicyanoacetylene (C_4N_2) at 478 cm^{-1} and the unidentified Haystack emission feature at 220 cm^{-1}, are puzzling since they have no associated observed vapor emission features. As well, recently, a massive stratospheric ice cloud system, called the High-Altitude South Polar (HASP) cloud, was discovered in Titan's early southern winter stratosphere at high southern latitudes, with an emission feature peaking near 210 cm^{-1} (Anderson et al., 2017). We are investigating these perplexing observed stratospheric ices to better understand their formation mechanisms, identify their chemical compositions, and determine their optical properties. For this purpose, we have performed transmission spectroscopy of thin films of pure and mixed nitrile ices (and some combined with benzene), from the near- to far-infrared spectral region (50 cm^{-1} to 11700 cm^{-1}), using the SPECtroscopy of Titan-Related ice AnaLogs (SPECTRAL) high-vacuum chamber. Their respective vapors were dosed at low temperatures from 30 K to 150 K and the resulting ices were analyzed at different times after deposition, from immediately after dosing to up to 24 hours post-dosing. This allows us to study their spectral evolution with time and temperature, to precisely identify the ice phase formation, and to compute their optical constants. The first surprising yet significant result reveals that the libration mode of HCN (166 – 169 cm^{-1}) is drastically altered by the surrounding molecules when mixing occurs in a co-condensed phase. For propionitrile ice, we observe peculiar temperature and time-driven ice phase transitions (as compared to other nitrile ices), revealed by significant spectral changes observed in the mid- and far-IR that cease once a stable crystalline phase is achieved. We have compared our laboratory spectra to the CIRS-observed stratospheric ice emission features. Here we present our findings, which may provide crucial inputs to deepen our understanding of Titan's stratospheric chemistry.