Heterogeneous oxidation reactions relevant to tropospheric aerosol chemistry studied by sum frequency generation GRACE STOKES, AVRAM BUCHBINDER, JULIANNE GIBBS-DAVIS, KARL SCHEIDT, FRANZ GEIGER, Northwestern University — Unsaturated organic molecules (terpenes) that commonly form molecular films on tropospheric aerosols can be oxidized by ozone, influencing the microphysics of cloud formation and thus the earth’s climate. Using a laboratory approach that combines organic synthesis with surface spectroscopy, we track the ozone oxidation reactions of tropospherically relevant terpenes bound to glass surfaces that serve as mimics for mineral dust. Specifically, vibrational broadband sum frequency generation (SFG) is used to study a number of tailor-made terpene-modified glass surfaces and to track their interactions with ozone in real time. Exposure of these surfaces to ppm levels of ozone at 1 atm and 300 K yield initial reaction probabilities that are significantly higher than corresponding gas phase reactions. SFG spectra help elucidate the molecular orientations of the surface-bound terpenes and the accessibility of reactive C=C bonds. Our work shows the successful use of SFG spectroscopy to determine heterogeneous atmospheric reaction probabilities and bridges the gap between atmospheric aerosol science and surface spectroscopy.

Grace Stokes
Northwestern University

Date submitted: 01 Nov 2007

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