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Recent progress towards measuring the clumped isotopic composition of carbon dioxide with laser spectroscopy

David Nelson (1), Zhennan Wang (2), David Dettman (2), Barry McManus (1), Jay Quade (2), Kate Huntington (3), and Andrew Schauer (3)

(1) Aerodyne Research, Inc., Billerica, United States, (2) Dept. of Geosciences, University of Arizona, Tucson, United States, (3) Dept. of Earth and Space Sciences, University of Washington, Seattle, United States

We describe our latest results using Tunable Infrared Laser Differential Absorption Spectroscopy (TILDAS) to measure the clumped isotope ratio of gaseous carbon dioxide samples and describe our plans to extend this technique to the measurement of carbon dioxide samples derived from carbonate minerals. Our method uses two lasers to measure the four isotopologues involved in the 16O13C18O equilibrium. The gas samples consist of carbon dioxide diluted in nitrogen. These samples are trapped at low pressure (30 to 60 Torr) in a low volume ($\sim$200 ml) optical multi-pass cell with a path length of 36 meters. Each sub-sample is probed for <2 minutes. Each sub-sample measurement is bracketed with measurements of a working reference gas. A complete sub-cycle requires a measurement time of $\sim$4 minutes and consumes 3.8 micro-moles of CO_2. Our best results to date show repeatability of 0.015 per mil for D638 which is the infrared spectroscopic equivalent of D47. Furthermore, when we execute multiple sub-cycles drawing from the same original sample mixture, we often observe noise reduction consistent with random noise statistics. Hence, after averaging 3 sub-cycles we observe repeatability of 0.009 per mil. This requires 12 minutes of time and 11.5 micro-moles of CO_2. We also present preliminary calibration results obtained from the measurement of pure CO_2 samples equilibrated at a wide range of temperatures.