Optical Detection of Radiocarbon (^{14}C) Below Modern Levels by Cavity Ring-down Spectroscopy

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We report the optical detection of radiocarbon (^{14}C) in biogenic carbon dioxide (CO_2) samples with a fraction modern carbon of $F < 1$ using a mid-infrared laser spectrometer. The table-top instrument operates on the principles of cavity ring-down spectroscopy in the linear absorption regime by measuring the mole fraction of absorbers $\chi=(\int\alpha(\nu)d\nu)/(cnS)$, where c is the speed of light, n is the total number density, S is the absorption line strength, α is the measured absorption coefficient, and ν is the laser frequency. The absorption coefficient coefficient α is related to the time constant of the optical resonator in the presence of molecular absorbers τ by the simple equation $\alpha=1/(c\tau)-1/(c\tau_0)$, where τ_0 is the empty-cavity time constant. The optical detection of radiocarbon by linear absorption spectroscopy is therefore fundamentally different from the seminal laser experiments based on non-linear saturated absorption cavity ring-down (SCAR). Measurements of χ_{14} for biogenic and petrogenic CO_2 samples, respectively, were used to determine the limits of our first-generation radiocarbon spectrometer capable of definitively distinguishing samples of CO_2 with $F < 1$.

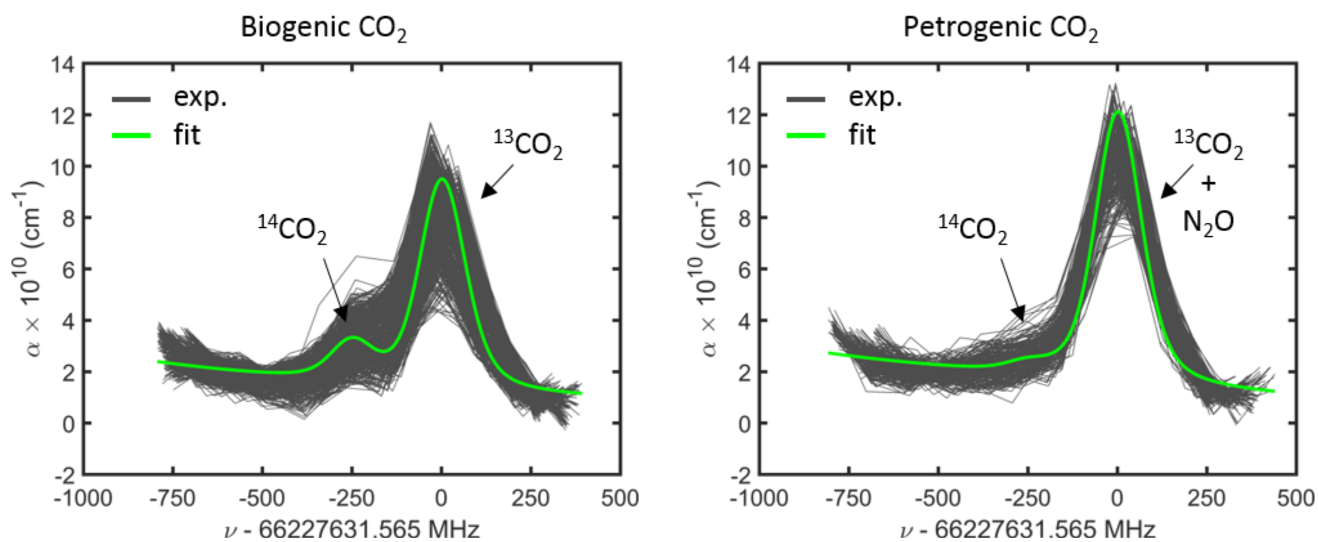

Figure 1. Left. Spectrum of radiocarbon dioxide ($^{14}CO_2$) for a biogenic sample of CO_2 with $F = 0.86$. Right. Spectrum of a petrogenic sample of CO_2 with $F = 0$. Fraction modern (F) was independently measured for each sample by a commercial accelerator mass spectrometry laboratory. Nearby $^{13}CO_2$ and N_2O interferences were mitigated by reducing the sample temperature to $T = 220$ K. The spectrometer comprises a quantum cascade laser, fast optical switch, high-finesse optical resonator and cold cell, low-noise photoreceiver, analog-to-digital converter, and computer software for signal processing.