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Novel quantum-cascade laser based spectrometer for high precision isotopic ratio measurements of atmospheric CO_2

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Continuous and in-situ monitoring of the isotope content of atmospheric carbon dioxide is of prime interest in the biosphere–atmosphere exchange research. We present the design and laboratory results of a compact mid-IR spectrometer developed for such purposes using an pulsed quantum cascade laser (QCL) from AlpesLasers operating at 4.3 μm. Employing thermoelectrically cooled (TEC) components for both the IR laser light source and detector, the instrument can operate in a complete cryogenic-free mode that greatly facilitates field applications. Furthermore, the carefully selected spectral range in the ν_3 ro-vibrational band of CO_2 allows for simultaneous concentration measurements of the three main stable carbon dioxide isotopologues ($^{16}O^{12}C^{16}O$, $^{16}O^{13}C^{16}O$ and $^{18}O^{12}C^{16}O$) in ambient air. The isotopic ratio measurement is performed using differential absorption spectroscopy. A dual multipass-cell arrangement is used to analyze the difference between the sample spectra and the simultaneously acquired reference spectra. This approach significantly improves the spectrometer performance due to the cancellation of correlated noise components specific to this method. The instrument precision has been evaluated using the Allan variance technique. The $^{13}CO_2/^{12}CO_2$ ratio has a 1 s standard deviation of 0.6 $^{o}/_{oo}$, while the variance minimum at 430 s corresponds to a minimum σ_{Allan} of 0.04 $^{o}/_{oo}$. This clearly demonstrates the feasibility of high precision in-situ isotope ratio measurements by the above mentioned optical technique. Additional efforts for the development of an adequate calibration method and the integration of optimized TEC detectors, which are expected to give even better performance, are currently undertaken.