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Traceable air humidity measurements by means of frequency-stabilized cavity ring-down spectroscopy.

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Water vapor is the most important constituent of the Earth's atmosphere, being responsible for about 70% of the known atmospheric absorption of sunlight and the majority of the greenhouse effect. Therefore, air humidity in terms of volume concentration of water is a key parameter to be measured for understanding the global climate processes. In reason of that, accurate water vapor measurements are of the utmost importance, especially for radiative transfer modeling. However, water vapor content of the upper troposphere and lower stratosphere is very difficult to be measured accurately and the need of water vapor amount fraction measurements traceable to the SI units is becoming a growing request among the scientific community. In the last decades, diode laser absorption spectroscopy has represented a very powerful method for trace gas detection and measurements. In particular, if a high-finesse optical cavity is employed, extremely high levels of detection sensitivity can be reached. Furthermore, the use of a frequency-stabilized ring-down measurement strategy can enable for absolute determinations of the molecular density, while preserving the highest levels of sensitivity.

In this work, we report on our recent efforts toward the development of a new concept of frequency-stabilized cavity ring-down spectroscopy (FS-CRDS). The spectrometer, based on a pair of phase-locked extended cavity diode lasers, operates at 1.39 μm wavelength. One of the two lasers acts as reference oscillator, while the second laser (namely, the probe laser) is actively controlled so that its emission frequency maintains a given offset with respect to the frequency of the reference laser. The probe laser is injected into an ultra-stable high finesse cavity having a finesse of about 10^5 and made with a 43-cm long ZERODUR spacer. A tracking servo-loop electronics was implemented for cavity ring-down measurements with high temporal resolution, while scanning the offset-frequency of the probe laser around a given center frequency. The complete characterization of the cavity was performed, also measuring an empty cavity ring-down time (τ_0) of about 60 μs. Repeated determinations revealed a precision of about 0.1% on a time scale of few seconds, while an Allan-Werle analysis, performed on 25000 ring-down events over a period of about 30 minutes, reveals that a minimum Allan deviation (σ_A) of 2×10^{-8} μs could be reached in correspondence of an integration time of 200 s. This value allows for an estimation of 2×10^{-10} cm^{-1} as limit of detection (LOD), to which corresponds a noise-equivalent absorption (NEA) coefficient of 5×10^{-11} cm^{-1}Hz$^{-1/2}$. Moreover, the dual laser approach allowed us to build an absolute and highly-reproducible frequency scale underneath the absorption spectra, thus satisfying one of the main requirements for absolute and traceable determinations. Our approach appears to be very promising, showing all the requisites to act as a primary method for determinations of amount of substance for water vapor, at trace levels.