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High-sensitivity detection of HO_2 radical by cavity ring-down spectroscopy: prospects for future applications of narrow linewidth lasers

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The hydroperoxyl radical (HO_2) plays a key role in atmospheric chemistry. It reacts with NO to generate hydroxyl radical (OH) and nitrogen dioxide (NO_2), resulting in the HO_x (= OH + HO_2) cycle that governs the atmospheric oxidation capacity and the formation of air pollution, and a net production of ozone that determines the troposphere ozone budget. Quantitative measurement of its absolute concentration is very important. However, due to its short lifetime and low concentration (typically about 10^8 to 10^9 molecule/cm^3 under atmospheric condition), most of the currently used methods are indirect methods that require chemical conversion; direct measurement remains very challenging.

The cavity ring-down spectroscopy (CRDS) technique uses high reflectivity mirrors to increase the effective absorption pathlength to tens of kilometers, enabling very high detection sensitivity; the absolute concentration of the target absorbers can be quantitatively determined by the Beer-Lambert law, providing a powerful tool for direct measurement of free radicals. In this work, we report the development of a portable cavity ring-down spectrometer for direct and absolute measurement of HO_2 radical concentration using a distributed feedback (DFB) diode laser operating at 1506 nm. At a pressure of 100 mbar, a detection limit of ~ 7.3×10^7 molecule/cm^3 (1σ, 10s) was achieved with a ring-down time (τ_0) of 136 μs. The corresponding detection sensitivity was 1.5×10^{-11} cm^{-1}, which was close to the state-of-the-art performance.

In cavity ring down spectroscopy, the coupling efficiency of the laser beam into the cavity depends on the laser frequency tuning speed and the ratio of the laser linewidth to the cavity mode width. For the DFB laser system, the laser linewidth (~ 2 MHz) was about thousands of times larger than that of the cavity mode (~ 1.2 kHz), which results in the conversion of laser phase noise into amplitude fluctuation, making the cavity injection noisy and limiting the improvement of detection sensitivity. Here, by replacing the DFB diode laser with a narrow linewidth erbium-doped fiber (EDF) laser, the amplitude fluctuation caused by the laser phase noise was reduced and the cavity mode injection efficiency was improved. The sensitivity was improved to 3.9×10^{-12} cm^{-1} with a short data-acquisition time of 0.2 s. The one order of magnitude improvement makes further ambient applications look promising.