SI TRACEABLE SATURATED ABSORPTION SPECTROSCOPY OF OZONE AT THE 50 KHZ LEVEL USING A QUANTUM CASCADE LASER AT 9.5 MICROMETRE

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Significant progress on the quantitative spectroscopy of ozone in the UV[1,2,3] and the IR[4,5,6,7,8] has been made recently, addressing the long-standing issue of the missing consistency between UV and IR spectroscopic data for the remote sensing of ozone.[9] However, some uncertainty remains whether these improvements suffice to account for the striking observation of opposite trends in the global tropospheric ozone burden derived from satellite instruments (e.g. OMI or TROPOMI operating in

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the UV versus IASI in the IR)[10] or for the inconsistencies in the ozone retrieval from different IR bands using the same instrument.[11] Unresolved issues in the IR spectral region at 9.5 μm are uncertainties and inconsistencies in current ozone broadening[12] and pressure shift[13] parameters, as well as the question of appropriately representing molecular line shapes.[14] These issues are best investigated using SI traceable instrumentation which provides spectral resolution surpassing that of typical high resolution Fourier transform spectrometers.

Based on previous work at LPL,[15,16] we have developed a new SI traceable high resolution laser spectrometer, which will be able to address these questions. A quantum cascade laser at 9.5 μm is frequency stabilized on an optical frequency comb itself stabilized to a 1.55 μm ultra-stable laser transmitted by fiber link through the REFIMEVE[17] network from LNE-SYRTE. There it is measured against primary standards, thus assuring SI traceability of the frequency measurements. As a first demonstration of the spectrometer perfomances, we report on line positions in the R-branch of the ozone ν_3 band from saturated absorption spectroscopy with line center determinations at the 50 kHz level. This is almost three orders of magnitude more accurate than the current uncertainty index ($< 10^{-3}$ cm^{-1}) given in the HITRAN 2020 database.

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