Geophysical Research Abstracts,
Vol. 11, EGU2009-10143, 2009
EGU General Assembly 2009

NO2 trace measurements by optical-feedback cavity-enhanced absorption spectroscopy

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In order to reach the sub-ppb NO_2 detection level required for environmental applications in remote areas, we are developing a spectrometer that exploits a technique that we introduced several years ago, named Optical-Feedback Cavity-Enhanced Absorption Spectroscopy (OF-CEAS) [1]. It allows very sensitive and selective measurements, together with the realization of compact and robust set-ups as was subsequently demonstrated during measurements campaigns in harsh environments [2,3]. OF-CEAS benefits from the optical feedback (OF) to efficiently inject a cw-laser in a high finesse cavity (typically F >10 000). Absorption spectra are acquired on a small spectral region ($\sim$1 cm^{-1}) that enables selective and quantitative measurements at a fast acquisition rate ($\sim$10 Hz) with a detection limit of several 10^{-10} cm^{-1} as reported in this paper. Spectra are obtained with high spectral resolution ($\sim$150 MHz) and are self calibrated by cavity rind-down measurements regularly performed (typically every second).

Therefore, OF-CEAS appears very attractive for NO_2 trace detection. This work is performed in the blue spectral region where NO_2 has intense electronic transitions. Our setup involves a commercial extended cavity diode laser (ECDL) working at room temperature around 411nm. A first setup was developed [4] to demonstrate that OF sensitivity of ECDL is fully consistent with this technique, initially introduced with distributed feedback diode lasers in the near infrared region.

In this paper we will report on a new set-up developed for in-situ measurements with proper mechanical, acoustic and thermal insulation. Additionally, new data processing was implemented allowing real time concentration measurements. It is based on a reference spectra recorded under controlled conditions by OF-CEAS and used later to fit the observed spectra. We will present measurements performed with calibrated NO_2 reference samples demonstrating a good linearity of the apparatus. The minimum detectable absorption loss is estimated by considering the standard deviation of the spectra. We achieved better than 2x10^{-10} cm^{-1} for a single spectrum recorded in less than 100ms at 100mbar. This limit constitutes an improved of more than one order of magnitude as compare to the previous measurements reported in [4]. It leads to a detection limit of 3x10^8 molecules/cm^3, corresponding to about 150pptv at 100mbar. At atmospheric pressure the same measurement would yield a detection limit of 15pptv assuming we can maintain the same level of sensitivity. But currently, works are under development to transfer the low minimum detectable absorption limit, already obtained for one spectrum, to a set of real time measurements. These are now limited by amplitude fluctuations of a few ppb from one spectrum to another one.

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