

## 2- $\mu\text{M}$ COHERENT DIAL FOR $\text{CO}_2$ , $\text{H}_2\text{O}$ AND WIND FIELD PROFILING IN THE LOWER ATMOSPHERE: INSTRUMENTATION AND RESULTS

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### ABSTRACT

We report on 2- $\mu\text{m}$  coherent differential absorption lidar (CDIAL) measurements of carbon dioxide ( $\text{CO}_2$ ), water vapour ( $\text{H}_2\text{O}$ ) absorption and wind field profiling in the atmospheric boundary layer. The CDIAL uses a Tm: fiber pumped, single longitudinal mode Q-switched seeded Ho:YLF laser and a fiber-coupled coherent detection. The laser operates at a pulse repetition frequency of 2 kHz and emits an output energy of 10 mJ with a pulse width of 40 ns (FWHM). Experimental horizontal and vertical range-resolved measurements were made in the atmospheric boundary layer and compared to co-located in-situ sensor data.

### 1. INTRODUCTION

Water vapor ( $\text{H}_2\text{O}$ ) and carbon dioxide ( $\text{CO}_2$ ) are the key atmospheric gases for water and carbon cycle studies in the earth system. They are the most important greenhouse gases that rule the global warming but they are also the key components of plants biological processes (photosynthesis, respiration). Knowledge of atmospheric  $\text{CO}_2$  and  $\text{H}_2\text{O}$  fields is of a major importance: (i) to improve our understanding of surface flux time and space heterogeneity (ii) to study the transport processes at different scales (iii) to monitor unambiguously anthropogenic emissions (for  $\text{CO}_2$ ) above a given site (city, industry, carbon storage and capture experiment) for the carbon has now a price in the framework of climate and energy policy. However, knowledge of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  fields is usually useless without some information of the dynamics at different scales in the atmosphere. The wind field is of major importance to understand the diffusivity processes of these gases and to link the concentrations to surface sources and sinks.

In the instrumental point of view,  $\text{CO}_2$  is an unprecedented challenge for optic based remote sensing techniques, as its variation is rather small in the atmosphere compared to  $\text{H}_2\text{O}$ . Typical natural variation due to its diurnal cycle (uptake by the vegetation, rectifier effect) are around 5% close to the surface but decrease to 0.1% for turbulent fluctuations in the convective boundary layer. Although differential absorption lidar experiments emerged in the last decade [1-3] the technology faced two main limitations to reach such precision: (i) a powerful and high PRF dual wavelength single mode emitter for coherent DIAL system and (ii) an efficient photodetector (i.e. avalanche photodiodes) in the near infrared for direct detection DIAL. In the framework of coherent DIAL system, a 2  $\mu\text{m}$  high PRF dual-wavelength single mode pulsed Ho:YLF oscillator has been achieved and fully characterized at LMD/CNRS [4]. In this paper we report for the first time on the use of such emitter in a CDIAL system for accurate range-resolved atmospheric  $\text{CO}_2$  absorption measurement. In addition we also demonstrate the ability of the lidar to make wind speed and  $\text{H}_2\text{O}$  absorption measurement.

### 2. EXPERIMENTAL SET-UP

The 2- $\mu\text{m}$  domain, in addition to be an eye safe domain, is characterized by  $\text{CO}_2$  and  $\text{H}_2\text{O}$  absorption lines that are well suited for DIAL measurements in the troposphere (Fig. 1): weak sensitivity to temperature ( $E'' \sim 300 \text{ cm}^{-1}$ ), non-interference with other gas absorption lines, suited optical depth for km-range of measurement. In addition this domain corresponds to Ho based crystal laser emission band. Using this technology, the LMD/CNRS developed during the last years a 2- $\mu\text{m}$  Ho:YLF emitter for DIAL application. The experimental set-up is presented in Figure 2.

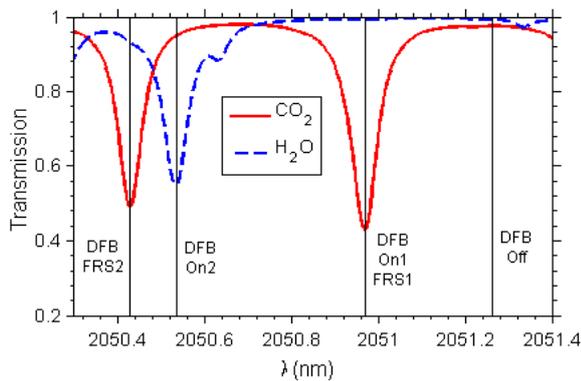


Fig. 1: 1-km horizontal single path atmospheric transmission in the 2.05- $\mu$ m band and coherent DIAL wavelengths position used for CO<sub>2</sub> (On1) and H<sub>2</sub>O (On2) differential absorption lidar measurements. H<sub>2</sub>O specific humidity is 10 g/kg and CO<sub>2</sub> dry air mixing ratio is 380 ppm.

The Ho:YLF crystal is positioned in a ring cavity and is pumped by a commercial 100 W linearly polarized Thulium fiber laser from IPG Photonics. The advantage of such configuration compared to Nd:YAG pumped OPO emitter is (i) the compactness of the system for a given power and (ii) that no stringent characteristic is required for the pump laser, i.e. seeding, pulse repetition frequency (PRF) and beam quality [5,6]. The

Ho:YLF oscillator is sequentially seeded by two fiber-coupled Distributed FeedBack laser diodes (DFB) at 2050.97 nm (On-line1) for CO<sub>2</sub> or 2050.53 nm (On-line2) for H<sub>2</sub>O and 2050.26 nm (Off-line) (Fig. 1). On-line DFB is indeed frequency locked (offset locking) to a frequency reference system (FRS) using a low-pressure pure CO<sub>2</sub> absorption cell and Pound-Drever-Hall (PDH) technique (external frequency modulation). A similar PDH technique is used to lock the cavity length (i.e. a longitudinal mode) to the On-line seeder wavelength. Off wavelength is located in a free absorption window close to the R30 CO<sub>2</sub> line so that Off-line CDIAL signal serves as a reference. The laser was operated continuously and provided linearly polarized 10 mJ On-Off pulse-pair of approximately 40 ns duration at a repetition rate of 1 kHz. The beam was collimated with a built-in expanded 100-mm aperture telescope. The overall system is implemented in a container and a scanning device enables to make 3D measurements. The atmospheric molecules and particles backscattered signal is collected by the same telescope and optically mixed with the seeders on a fiber-coupled balanced InGaAs photodiode detection.

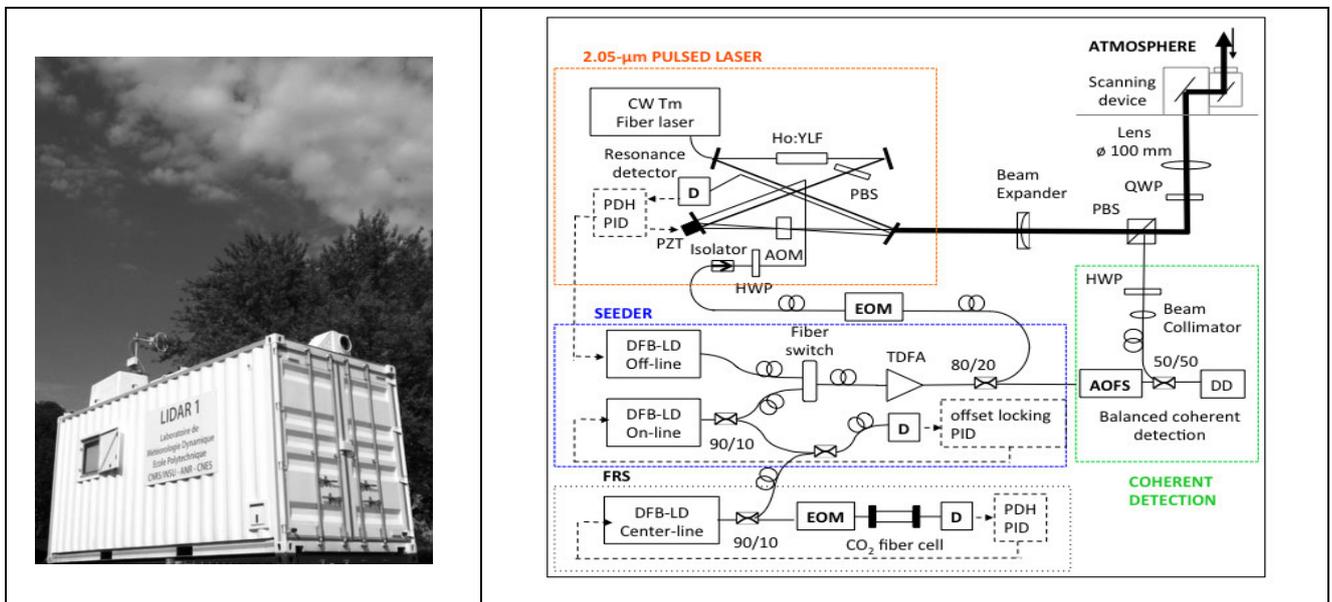


Fig. 2: Left: Coherent DIAL and in-situ mobile station for turbulence, CO<sub>2</sub> and H<sub>2</sub>O cycle studies in the atmosphere. Right: 2- $\mu$ m coherent DIAL experimental set-up. EOM: electro-optical modulator; AOM: acousto optical modulator; PID: proportional integral derivative; PDH: Pound Drever Hall; AOFS: acousto optic frequency shifter; TDFA: Thulium doped fiber amplifier; PBS: polarizer beam splitter; HWP: half wave plate; QWP: quarter wave plate; PZT: piezoelectric transducer.

The signal is digitized on a 400 MHz – 14-bits FPGA based high-speed data acquisition card. Real time signal processing consists in a 50-m range gate Discrete Fourier Transform accumulation over 4 s (2000 shot averaging for each wavelength). Post-processing uses Squarer and Levin-like estimators to deliver atmospheric On and Off backscattered signal power and frequency at the wanted time and space resolution. Further MATLAB based software provides a real time view of lidar reflectivity, radial velocity and CO<sub>2</sub> or H<sub>2</sub>O differential absorption coefficients.

### 3. RESULTS

#### 3.1 Accurate CDIAL range-resolved atmospheric CO<sub>2</sub> absorption measurement and mixing ratio estimates

Using the CDIAL measurements of the CO<sub>2</sub> absorption coefficient, one may estimates the CO<sub>2</sub> dry air mixing ratio with additional spectroscopic and meteorological data:

$$X_{CO_2} = \frac{\alpha}{n_a \Delta \tilde{\sigma}_{CO_2}} = \frac{kT(1+X_{H_2O})}{p} \frac{\alpha}{(\tilde{\sigma}_{CO_2,On} - \tilde{\sigma}_{CO_2,Off})} \quad (1)$$

where  $\alpha$  is the differential absorption coefficient due to CO<sub>2</sub> absorption only,  $n_a$  is the dry air density,  $\Delta \tilde{\sigma}_{CO_2}$  is the CO<sub>2</sub> differential absorption cross section,  $k$  the Boltzman constant,  $T$ , the temperature,  $p$  the pressure and  $X_{H_2O}$  the dry air water vapor mixing ratio.

In a preliminary experiment the CDIAL was located at the second floor of the LMD building and we made horizontal measurement at ~15 m height above Ecole Polytechnique campus. In parallel, a PICARRO in-situ gas analyzer made some routine measurements. Figure 3 shows some examples of horizontal profiles of CO<sub>2</sub> dry air mixing ratio in late afternoon (19:00) and early morning (07:00). The time and space resolutions were 15 min and 100 m, respectively. In-situ measurements are indicated with the star markers for comparison. The experimental standard deviation for each profile is indicated in Fig.3(b). This accounts for both instrumental and natural standard deviation. The instrumental error accounting for CDIAL absorption measurements may be estimated using the high frequency data power spectrum (4 s - 50 m) and looking for the white noise level. The results indicate an

instrumental standard deviation of 0.5% (2 ppm) up to a range of 500 m and 2% (8 ppm) at 1 km (solid line in Fig. 3). This error is confirmed by the CNR-based SNR calculation method [7] at least up to 700 m (dashed line in Fig.3(b)). The experimental standard deviation is nevertheless much higher (between 4 to 8 ppm up to 1 km) as a result of badly mixed anthropogenic CO<sub>2</sub> emissions in the nocturnal boundary layer. This is confirmed by the in-situ sensor standard deviation which shows 2 ppm at 19:00 and 6 ppm at 07:00 (typical PICARRO instrumental error is 0.2 ppm).

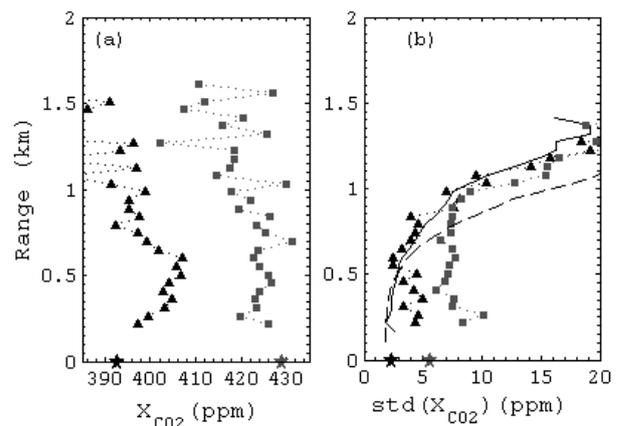


Fig. 3. (a) Examples of horizontal CDIAL CO<sub>2</sub> dry air mixing ratio profile. Time and space averaging are processed over 15 min and 100 m, respectively. (b) Experimental standard deviation calculated over a time gate of 3 h around each CO<sub>2</sub> profile. The instrumental error accounting for CDIAL absorption measurements is estimated using the high frequency data power spectrum (solid line) and using the CNR (dashed line) [7]. The different symbols are for the PICARRO in-situ gas analyzer (stars) and the CDIAL (squares) at 19:00 (black) and 07:00 (grey) in local time.

#### 3.2 Preliminary test of CDIAL H<sub>2</sub>O absorption measurement and specific humidity estimates

Using the nearby H<sub>2</sub>O absorption line close to the R30 CO<sub>2</sub> absorption line (Fig.1), we tested the CDIAL ability for water vapor dry air mixing ratio profiling in the boundary layer. Figure 1 shows that On2 wavelength is not entirely free from CO<sub>2</sub> absorption. However, the low variation of atmospheric CO<sub>2</sub> along the CDIAL line of sight has a negligible effect of H<sub>2</sub>O profile CDIAL retrieval. Practically, in-situ CO<sub>2</sub> mixing ratio measurements are usually sufficient to correct the

H<sub>2</sub>O CDIAL differential absorption from this interference. One may estimate the dry air H<sub>2</sub>O mixing ratio in [g/kg] by:

$$X_{H_2O} = \left( \frac{\alpha - n_a X_{CO_2} \Delta \tilde{\sigma}_{CO_2}}{n_a \Delta \tilde{\sigma}_{H_2O} - \alpha} \right) \frac{10^3 \cdot M_{H_2O}}{M_{air}} \quad (2)$$

where  $\alpha$  is the total differential absorption coefficient due to CO<sub>2</sub> and H<sub>2</sub>O absorption,  $\Delta \tilde{\sigma}_{H_2O}$  is the H<sub>2</sub>O differential absorption cross section,  $M_{H_2O}$  and  $M_{air}$  are the water and the air molar masses and  $X_{CO_2}$  the dry air CO<sub>2</sub> mixing ratio provided by the in-situ sensor.

For the experimental measurements, the CDIAL was installed in a container and used the scanning device to look vertically. Figure 4 shows an example of a vertical profile and a comparison with a radiosonde profile. The time and space resolution were 10 s and 100 m. Although the radiosonde was not at the right time and space of the CDIAL measurement, there is a good agreement of the two profiles. The CDIAL profiles fluctuations are interpreted as both instrumental error and natural fluctuations due to turbulence. The experimental standard deviation is 10% up to 1 km.

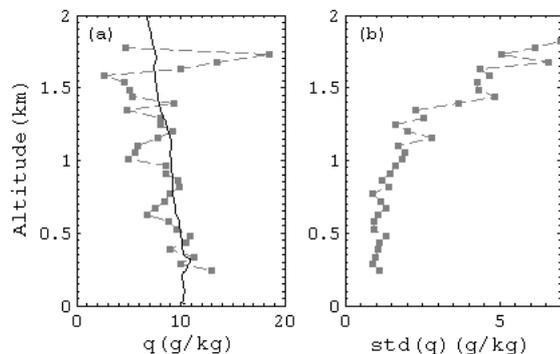


Fig. 4. (a) Example of CDIAL vertical H<sub>2</sub>O dry air mixing ratio profile at 04:16 pm in local time. Time and space averaging are processed over 10 s and 100 m, respectively. Solid line is for the 10-km apart radiosonde profile at 01:00 pm. (b) Experimental standard deviation calculated over a time gate of 10 min.

#### 4. CONCLUSION

To conclude, we demonstrate the 2- $\mu$ m Ho emitter based coherent DIAL ability to measure CO<sub>2</sub> and H<sub>2</sub>O absorption coefficient in the atmospheric boundary layer. Precision and time and space resolutions seem to be sufficient for 3-D monitoring application in the boundary layer for

CO<sub>2</sub> and H<sub>2</sub>O. Concerning turbulence-based fluctuations, the CDIAL may match the required performances for H<sub>2</sub>O but not for CO<sub>2</sub> where both precision and time resolutions are not sufficient.

#### ACKNOWLEDGEMENT

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