Tropospheric CO_2 DIAL measurements

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Abstract

The Global Carbon Cycle has been significantly perturbed by human activities in the last two centuries. The current network seems limited to fully understand the fundamental processes and predict the future rate of increase of atmospheric CO_2 and its impact on future climate. In this context, a 2-µm Heterodyne Differential Absorption Lidar (HDIAL) has been developed and operated at IPSL/LMD to monitor CO_2 mixing ratio in the atmospheric boundary layer. One objective is to demonstrate our capability to monitor CO_2 from air- and later space platform.
First, the HDIAL performances are tested at 10 m above the ground in horizontal line of sight and the absolute mixing ratio measurements compared to routine *in situ* measurements. The Doppler capability is fully used to understand the air masses advection and to analyse possible discrepancies between HDIAL and in situ measurements in the broader context of the Paris area.
Secondly, the HDIAL was used to make vertical measurements in the boundary layer. We also demonstrated its capability to make CO_2 measurements in the free troposphere using clouds as hard targets.

1. Introduction

The atmospheric carbon dioxide (CO_2) is one of the main contributors to the greenhouse effect. A global monitoring ultimately from space is foreseen as a key issue to quantify the sources and sinks at a regional scale and to better understand the links between the various components of the carbon cycle. However, the measurement is very demanding for an accuracy of 0.5-1 % (i.e. 1-3 ppm) in the lower troposphere is needed to significantly improve model predictions. In situ sensors can achieve such a high performance but in a few dedicated locations in the framework of a global network or even less frequently using aircraft. Remote sensors like Lidar can complement the existing ground based network and could ultimately be operated in space. In the meantime a Differential Absorption Lidar (HDIAL) operating from the ground or an airborne platform could provide with CO2 column content information for both science application and technology demonstration. Preliminary results obtained with a 2 µm Heterodyne DIAL looking horizontally in the atmospheric boundary layer (ABL) have been reported showing that a 1 % relative accuracy can be achieved. [1]
In this paper, the HDIAL abilities are developed to make atmospheric CO_2 vertical measurements in the boundary layer and in the free troposphere. Vertical measurements in the boundary layer using aerosols as backscatters are discussed and compared with ground-based in-situ measurements. However, as the return signal from aerosols is sometimes too weak and noisy that we cannot detect it, we analyse possibilities to make measurements using clouds as hard targets. This possibility is also used to make measurements in the free troposphere and will be useful for measurements from a space based instrument.

2. The 2 µm Heterodyne DIAL system

The HDIAL is based on a Ho:Tm:YLF laser transmitter. The specifications are listed in Table 1. A Ho:Tm/YLF laser is well suited for DIAL measurement due to its tunability over several CO_2 lines in the 2.05-2.65 µm range.

Pulse energy	10mJ
Pulse repetition rate for a wavelength pair ON-OFF	5 Hz
Pulse width/ Line width	230 ns/ 2.5 MHz
OFF-line	2064.41 nm
ON-line	2064.00 nm
Telescope aperture	100 mm
Detection	Dual balanced InGaAs photodiodes (η=70%)
Spectral control	Photoacoustic cells with CO_2 and H_2O at 1000 hPa
Signal digitization	8 bits/ 125 MHz
Signal processing Estimators	- Squarer - Levin-like

Tab.1.Experimental set up

As shown in Figure 1, a Ho:Tm/YLF pulsed laser is pumped by a 5W-10Hz-pulsed Alexandrite laser in a ring configuration with an acousto-optic unit

(OAM 2) for Q switching and a Lyot filter for tuning. Line narrowing and fine frequency control are done by injection seeding using cw lasers.

An opto-acoustic modulator (OAM 1) is used to generate a 25 MHz intermediate frequency between the cw local oscillator and the pulsed laser. There are actually two injection-seed lasers involved, one Ho:Tm:YLF for the absorbing wavelength (ON-line seed) and a Ho:Tm:LuLiF$_4$ for the non-absorbing wavelength (OFF-line seed). The 2nd wavelength is also used for wind profiling using the Doppler technique. Each seeder laser includes a 780-nm-cw-diode laser, an etalon and a PZT-adjustable output coupler for fine tuning.

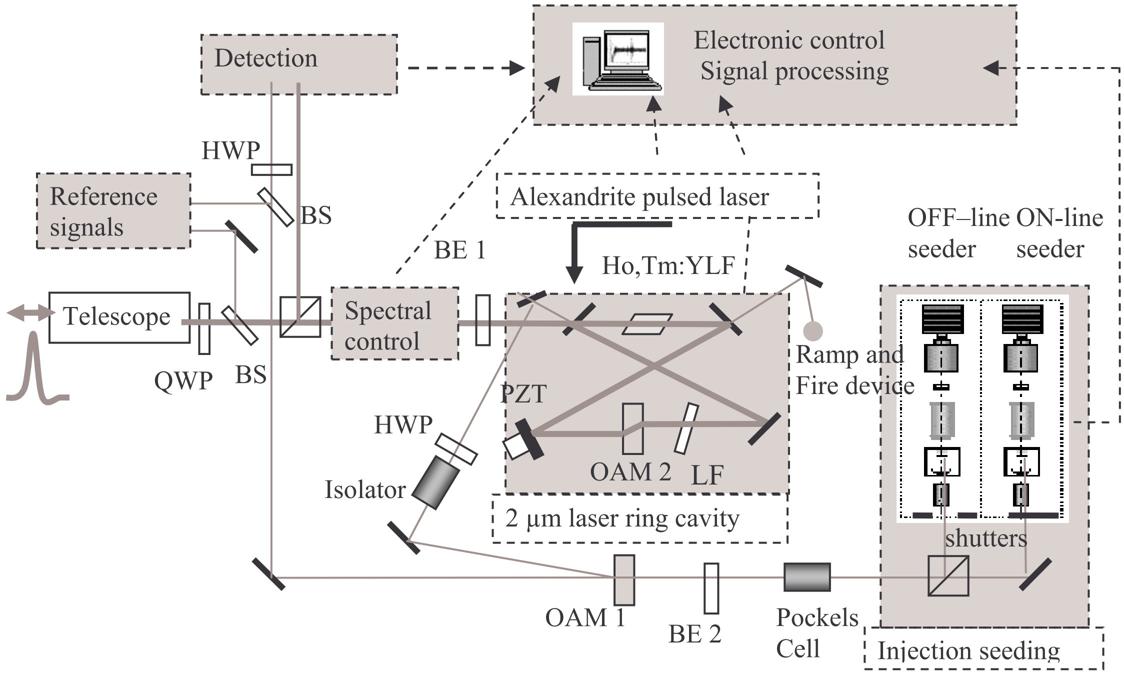

Fig. 1. Experimental set up of the 2.06 μm HDIAL. BS, beam splitter, HWP, half-wave plate, QWP, quarter-wave plate, LF, Lyot filter, OAM, opto-acoustic modulator, BE, beam expander.

OFF-line and ON-line lasers are locked thanks to a photoacoustic absorption cell containing CO_2 (or H_2O) at low pressure for fine tuning on the absorption line of interest. A switching of the ON and OFF probing wavelengths is made possible by 2 shutters and a Pockels cell (Fig. 1). A small fraction of the transmitted beam is mixed with the local oscillators to record the ON- OFF beat frequency on a shot-to-shot basis on a reference InGaAs photodiode. The atmospheric signal is mixed with the two local oscillators on two 75 µm-InGaAs photodiodes in a balanced detection arrangement. After digitization, later processing is achieved with software developed in MATLAB programming language. The ON-line and OFF-line signals are recorded with the corresponding CO_2-photoacoustic cells signals to be analysed for cross section absorption line correction.[2] A Levin-like filter is used for signal processing for power and frequency estimates. The power spectra are accumulated on several consecutive lidar shots and the resulting spectrum is correlated with a 4 MHz FWHM Hanning filter, slightly larger than the ~ 2.5 MHz FWHM for the pulse power spectrum. A squarer estimator is also used for power estimates as a double check.

3. CO_2 molecule spectroscopy for Lidar application

An important issue in DIAL for atmospheric CO_2 density measurements is the selection of the proper absorption line to match an optimal double pass optical depth of ~ 1.3.[2] Other conditions are set on non-interference with H_2O absorption lines and a low temperature dependence of the cross section (i.e the lower state level energy (E'') needs to be around ~ 300 cm^{-1} for tropospheric measurements). Unfortunately, because of limited tuning emitter, these conditions were not satisfied in practice for the ON-line (Fig.2). The ON- and OFF-line were tuned to 2064.4 nm and 2064.0 nm, respectively. The CO_2 line cross-sections were corrected for temperature and pressure dependence using the meteorological information provided by in situ sensors and MM5 meso-scale model for horizontal and vertical HDIAL measurements.

More recently, experimental investigations in the 1.6 and 2 µm domains by GSMA team enables us to use more accurate spectroscopic data.[3] Moreover, given that the cross section corrections were made using meteorological and photoacoustic cell information, we estimate that the total error on the cross section amounts to ~ 1 %.

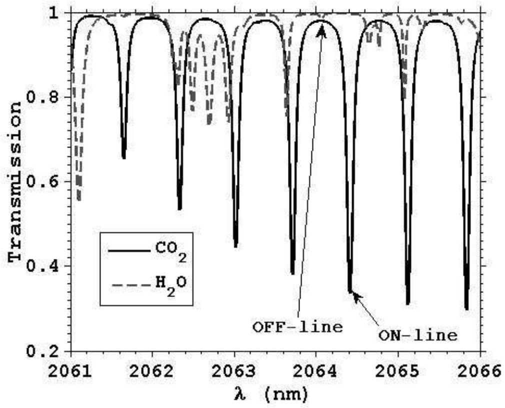

Fig.2. Transmission spectrum using the GEISA or HITRAN database with Voigt line shapes for CO_2 (375 ppm) and H_2O (10 g.kg^{-1}) in the 2.060-2.065 µm range for a 1 km vertical path starting from the ground level.

4. Vertical CO_2 HDIAL measurements

Looking horizontally at 10-m above the ground, CO_2 HDIAL density measurements have already shown results in good agreement with in-situ measurements.[1] These measurements enable us to validate the DIAL system. Vertical measurements were achieved on November 5, 2004 (N05 case).

4.1 Data presentation and analysis

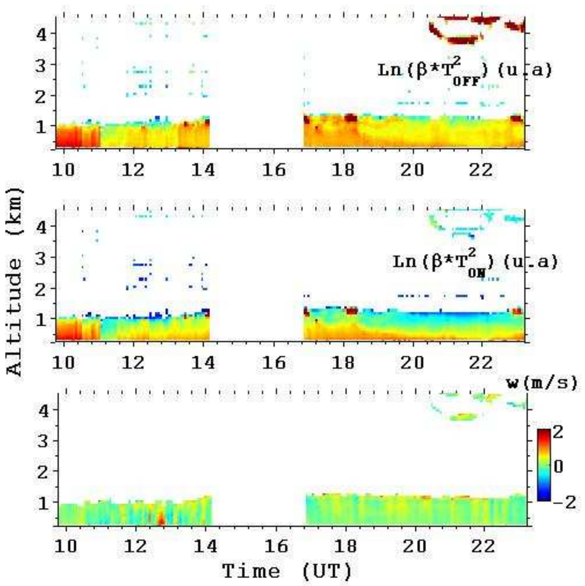

Fig. 3: Time-height contour plots of OFF- and ON-line backscatter signals (upper) and vertical velocity (lower) taken with the HDIAL on November 05, 2004. Colour plot are for $Ln\left(\beta.T^2_{OFF/ON}\right)$ in arbitrary unit (red is for higher return signal). Range and time resolution are respectively 75 m and 3 min.

Figure 3 shows HDIAL measurements in terms of OFF- and ON-line backscatter signals and vertical velocity. N05 case is characterized by a weak and late development of the PBL. Stratocumulus clouds appear at 14 UT and remain presents in the evening. Weak vertical velocities ($|w| < 0.5\ m.s^{-1}$) ensure a vertical mixing during the night.

4.2 Using the "slope method" for aerosols backscatters

To retrieve the mean CO_2 mixing ratio in the boundary layer we use the slope method. [1] The optical depth between range 0 and R is:

$$\tau(0,R) = \frac{1}{2}\ln\left(\frac{\langle P(\lambda_{OFF},R)\rangle}{\langle P(\lambda_{ON},R)\rangle}\right) \qquad (1)$$

where $\langle P(\lambda_{OFF},R)\rangle$ and $\langle P(\lambda_{ON},R)\rangle$ are the mean OFF- and ON-line spectral powers in a range gate. The relative error on optical depth measurement is :

$$\frac{\sigma(\tau)}{\tau} = \frac{1}{2\tau}\sqrt{\frac{1}{SNR^2_{OFF}} + \frac{1}{SNR^2_{ON}} - 2\frac{\rho(\langle P_{ON}\rangle,\langle P_{OFF}\rangle)}{SNR_{OFF}SNR_{ON}}} \qquad (2)$$

where $\rho(\langle P_{ON}\rangle,\langle P_{OFF}\rangle)$ is the cross correlation coefficient between return signals $\langle P_{ON}\rangle$ and $\langle P_{OFF}\rangle$ and SNR_{ON}, SNR_{OFF} the ON- and OFF-line signal to noise ratio. The optical depth numerical bias is:

$$\delta(\tau) = \frac{1}{4}\left(SNR^{-2}_{ON} - SNR^{-2}_{OFF}\right) \qquad (3)$$

After time averaging, the SNRs are high (> 100). Moreover, with an optical thickness on ground near 1 and an emitted energy ratio E_{ON}/E_{OFF} = 3 the SNRs at both wavelengths are of the same order.[2] Therefore, the bias on each optical thickness is usually negligible (< 10^{-4}).
The optical depth can also be written as:

$$\tau(0,R) = \int_0^R \rho_{CO_2}(r)n_a(r)\left(\tilde{\sigma}_{ON}(r) - \tilde{\sigma}_{OFF}(r)\right)dr \qquad (4)$$

where $\rho_{CO_2}(R)$ is the CO_2 mixing ratio, $\tilde{\sigma}_{ON}$ and $\tilde{\sigma}_{OFF}$ the ON- and OFF-line effective absorption cross-section (accounting for spectral shift), n_a is the dry-air density.
From Eq. 8 it comes:

$$\rho_{CO_2}(r) = \frac{1}{n_a(r)\Delta\tilde{\sigma}(r)}\frac{d\tau(0,r)}{dr} \qquad (5)$$

where $\Delta\tilde{\sigma}(r) = \tilde{\sigma}_{ON}(r) - \tilde{\sigma}_{OFF}(r)$

Error reduction on CO_2 mixing-ratio requires an averaging over several range gates. A fruitful approach consists in calculating the cumulated optical depth as a function of range according to Eq. 1. Then, the CO_2 mixing ratio is computed as the slope (gradient) of the cumulated optical depth as a function of range. In practice, the mean CO_2 mixing ratio is obtained by a mean square least fit of the optical depth (accounting for standard deviation) as a function of range. This technique corresponds to the maximum likelihood estimate of the slope for normally distributed noise.

5. Preliminary outdoor results and comparison against in situ measurements

Figure 4 displays both HDIAL and in-situ CO_2 density measurements on November 5, 2004. HDIAL measurements are calculated using GSMA and HITRAN (or GEISA) spectroscopic data. The temporal resolution is 3 min. Statistical error amounts to 15 up to 20 ppm depending on independent samples and PBL height. A 10-point sliding averaging (30 min averaging) reduces the statistical error within 5 ppm.

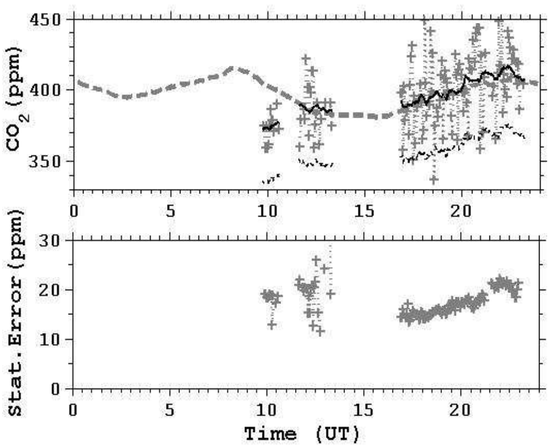

Fig. 4: (a) CO_2 mixing ratio measurements by HDIAL for a 1000 shot pair averaging (cross), 10 points sliding averaging using GSMA spectroscopic database (solid line) and HITRAN database (dotted line) and LSCE in-situ routine measurements (dashed line). (b) Statistical relative error on HDIAL CO_2 measurements due to the slope retrieval technique only.

During winter, energy and fuel combustions and reduced mixing layer height are the main contributors to CO_2 increases and diurnal variations in the boundary layer. As the following measurements took place in mid November, we must consider that CO_2 density is strongly influenced by anthropogenic sources. Such anthropogenic increases of CO_2 may be seen at 0 and 9 UT (Fig. 4).

Between 0930 and 1130 UT, HDIAL measurements are made in the residual layer (RL). CO_2 density amounts to 375 ± 3 ppm, namely, in agreement to airborne in-situ tropospheric measurements in November, 2004. A strong difference appears during this period between RL-HDIAL measurements and in-situ ground-based measurements, which were made in the nocturnal boundary-layer (NBL).
After 1130 UT, the ABL is rising (Fig. 3). HDIAL and in-situ measurements are made in the mixed layer and are in good agreement within 5 ppm as expected. It is worth noticing that HITRAN (or GEISA) database entails underestimation of CO_2 density. During the evening, Figure 3 shows that vertical mixing still occurred due to stratocumulus clouds. CO_2 density increases and HDIAL measurements are still in good agreement with in-situ measurements.
Using mid-high clouds in the free troposphere on N05 case (Fig. 3) we investigate the possibility to make DIAL measurements using these kinds of diffuse targets.

6. Conclusion

A 2 µm Heterodyne DIAL has been developed and operated by IPSL/LMD to assess the possibility to monitor the CO_2 mixing ratio in the troposphere using both boundary layer and cloud particles as diffused targets. HDIAL CO_2 mixed layer measurements are in good agreement with in-situ ground-based measurements. The statistical error is about 15-20 ppm (3 min of averaging) and it can be reduced within 5 ppm (~ 1.5 %) using a 5 point smoothing (15 min of averaging). Spectroscopic data accuracy from GSMA measurements is within ~ 2 %. Furthermore, the DIAL system proved its ability to distinguish RL and NBL CO_2 densities and to make measurements in the free troposphere.

References

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