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CO_2 profiling in the lower troposphere using the Raman lidar technique: preliminary results

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Abstract: Measurements of CO_2 vertical profiles in the lower troposphere are of great importance for climate research and monitoring, in particular for carbon sources and sinks characterization. In the present work we illustrate CO_2 mixing ratio measurements carried out by the Raman lidar system CONCERNING (COmpact RamaN lidar for Atmospheric CO_2 and ThERmodyNamic ProfilING). The calibration method adopted takes advantage of the lidar background signal, proportional to the zenithal sky radiance. Measurements for a specific case study are illustrated and discussed. The obtained profile is compared with satellite data from an OCO-2 overpass showing a good level of agreement (within one standard deviation).

1. Introduction

Knowledge of the vertical distribution of CO_2 in the lower troposphere is crucial for climate research and monitoring. It serves as a critical component in reconstructing our planet's carbon balance, providing valuable insights into quantifying both carbon sources and sinks.

The roto-vibrational Raman lidar technique, employed to measure water vapor mixing ratio profiles, has been investigated over the past decades for its application in CO_2 profile measurements [1–3]. Despite its high potential, the technique faced challenges due to the low signal-to-noise ratio (SNR) associated with CO_2 roto-vibrational Raman signals. However, recent advancements in laser technology, as well as improvements in spectral selection and signal detection devices, have renewed interest in the technique, leading to the development of a new generation of lidar systems specifically designed for its exploitation.

In the frame of the project CONCERNING (COmpact RamaN lidar for Atmospheric CO_2 and ThERmodyNamic ProfilING), a Raman lidar has been developed. This instrument is capable to provide accurate and high-resolution profiles of CO_2 and water vapor mixing ratio, temperature, particle backscattering, extinction and depolarization ratio. In this work we present preliminary results of the CO_2 profile measurements carried out by this system.

2. CO_2 Raman technique

The CO_2 mixing ratio is obtained from roto-vibrational Raman backscattered signals through the following equation:

$$x_{CO2}(R) = F_{cal} \Delta T_{CO2-N2}(R) \frac{S_{CO2}(R)}{S_{N2}(R)} \quad (1)$$

where $S_{CO2}(R)$ is the CO_2 roto-vibrational Raman lidar signal at wavelength λ_{CO2}, while $S_{N2}(R)$ is a temperature-independent reference signal (for the purpose of this research effort we are considering the N_2 roto-vibrational Raman signal), F_{cal} is a calibration constant, $\Delta T_{CO2\text{-}N2}(R)$ is a differential transmission term, which accounts for the different atmospheric transmission by molecules and aerosols at λ_{CO2} and λ_{N2}. The wavelength λ_{CO2} can be selected to be coincident with two strong vibrational bands. Few roto-vibrational O_2 lines lie within these spectral bands and represent a potential source of contamination for the CO_2 Raman lidar. λ_{CO2} was chosen to be coincident with the CO_2 vibrational band centred at 371.7 nm due to its lower contamination by O_2 lines [4].

The calibration constant F_{cal}, usually retrieved via comparison with in-situ measurements, was here calculated independently using the day-time background signal, proportional to the zenithal sky radiance. This technique, first described by Sherlock et al. [5,6] for water vapour Raman lidar measurements, is here adapted to CO_2 measurements:

$$F_{cal} = 0.7808 \frac{\frac{\partial \sigma_{N2_{eff}}}{\partial \Omega}}{\frac{\partial \sigma_{CO2_{eff}}}{\partial \Omega}} \frac{K_{N2}}{K_{CO2}} \quad (2)$$

$$\frac{\partial \sigma_{X_{eff}}}{\partial \Omega} = \sum_i \frac{\partial \sigma_X(\lambda_i)}{\partial \Omega} t_X(\lambda_i) \quad (3)$$

$$\frac{K_{N2}}{K_{CO2}} = \frac{L_{\lambda CO2}}{L_{\lambda N2}} \frac{S_{BN2}}{S_{BCO2}} \quad (4)$$

$$L_{\lambda X} = \int L(\lambda) t_X(\lambda) d\lambda \quad (5)$$

where $\frac{\partial \sigma_{X_{eff}}}{\partial \Omega}$ is the effective cross section (i.e., the sum over λ_i of the Raman cross section weighted by the filter transmittance t_X), $L_{\lambda X}$ is the effective radiance (i.e., the spectral radiance L weighted by the filter transmittance), and S_{BX} is the measured background signal. The Raman cross sections were computed as in Di Paolantonio et al. [4], and the spectral radiance was estimated with the SCIATRAN radiative transfer model [7].

3. System setup

The Raman lidar system CONCERNING was developed by the Lidar Group of the School of Engineering of the University of Basilicata, in close collaboration with the Institute of Marine Sciences of the Italian National Research Council and Sapienza – University of Rome [8]. The relevant instrumental characteristics of the lidar system are summarized in Table 1. The characteristics of the interferential filters used in this study with their central wavelengths (CWL), full-width at half-maximum (FWHM), and measured peak transmissions (T) are listed in Table 2.

Table 1. System setup

Laser	Seeded Nd:YAG (diode-pumped)
Energy / Power	100 mJ / 10 W @ 354.7 nm
Frequency	100 Hz
Reception channels	• Elastic total (354.7 nm) • Elastic ∥ (354.7 nm) • Elastic ⊥ (354.7 nm) • N_2 (386.7 nm) • H_2O (407.5 nm) • CO_2 (371.7 nm) • Low J (354.3 nm) • High J (352.9 nm)
Telescope	D 500 mm, F 1800 mm, f/3.6
Field of view	0.55 mrad
Full overlap	~ 800 m
Detectors	PMTs, 43% nominal peak quantum efficiency
Filter bandwidths	0.15–0.5 nm
Vertical sampling	3.75 m (analog & photon counting)
Acquisition system	16 bit, 40 MHz analog, 800 MHz photon counting

Table 2. Filter characteristics

Channel	CWL	FWHM	T
N_2	386.7 nm	0.3 nm	83 %
CO_2	371.7 nm	0.15 nm	60 %

4. Results and discussion

The measurements in the here presented case study were performed on 22-01-2024 in a semi urban site. The system was located at the School of Engineering of the University of Basilicata (40.647477 N, 15.807204 E, 710 m a.s.l.), Potenza, Italy. Being the approach robust with respect to the diurnal cycle of the CO2 mixing ratio, the day-time portion of the measurement session was used for the calibration, night-time (17:00-19:00 UTC) measurements were instead used for the CO_2 mixing ratio profile retrieval.

Figure 1 shows the measured CO_2 mixing ratio profile. The vertical resolution is 150 m up to 2400 m, 300 m up to 3600 m, and 450 m in the higher portion of the profile. The measurements were integrated over a 2-hour time window.

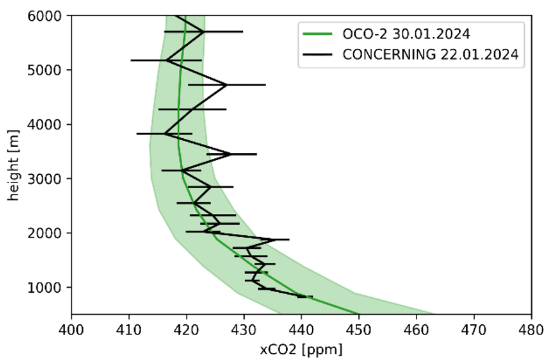

Figure 1. Comparison between lidar (black) and OCO-2 (green) measured CO_2 mixing ratio profiles. The uncertainty of the measurements is represented by horizontal bars and shaded green, respectively.

The obtained profile has an uncertainty < 5 ppm up to 4 km, enough to highlight a clear gradient slightly below 2 km. Measurements under 800 m were discarded, being potentially compromised by overlap issues. In absence of in-situ measurements, a comparison was made with the closest OCO-2 [8] satellite overpass (40.560715 N, 15.84124 E, 30-01-2024 11:51 UTC), with an agreement being within one standard deviation.

This case study demonstrates the potential of the instrumental setup and of the adopted technique. Further work will consist in the reduction of the system-derived uncertainties (e.g., improvement of the system thermomechanical stability, removal of undesired electromagnetic noise still affecting photon counting channels), comparison with in-situ CO_2 measurements, and measurements coincident with OCO-2 overpasses. Lastly, multiple measurement sessions will be carried out in order to evaluate the stability of the calibration factor and study the time evolution of the mixing ratio profile over extended time periods.

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6. References

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