

Lidar detection of carbon dioxide in volcanic plumes

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ABSTRACT

Volcanic gases give information on magmatic processes. In particular, anomalous releases of carbon dioxide precede volcanic eruptions. Up to now, this gas has been measured in volcanic plumes with conventional measurements that imply the severe risks of local sampling and can last many hours. For these reasons and for the great advantages of laser sensing, the thorough development of volcanic lidar has been undertaken at the Diagnostics and Metrology Laboratory (UTAPRAD-DIM) of the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA). In fact, lidar profiling allows one to scan remotely volcanic plumes in a fast and continuous way, and with high spatial and temporal resolution. Two differential absorption lidar instruments will be presented in this paper: BILLI (BrIdge voLcanic LIdar), based on injection seeded Nd:YAG laser, double grating dye laser, difference frequency mixing (DFM) and optical parametric amplifier (OPA), and VULLI (VULcamed LIdar), based on injection seeded Nd:YAG laser and optical parametric oscillator (OPO). The first one is funded by the ERC (European Research Council) project BRIDGE and the second one by the ERDF (European Regional Development Fund) project VULCAMED. While VULLI has not yet been tested in a volcanic site, BILLI scanned the gas emitted by Pozzuoli Solfatara (Campi Flegrei volcanic area, Naples, Italy) during a field campaign carried out from 13 to 17 October 2014. Carbon dioxide concentration maps were retrieved remotely in few minutes in the crater area. Lidar measurements were in good agreement with well-established techniques, based on different operating principles. To our knowledge, it is the first time that carbon dioxide in a volcanic plume is retrieved by lidar, representing the first direct measurement of this kind ever performed on an active volcano and showing the high potential of laser remote sensing in geophysical research.

Keywords: laser remote sensing, differential absorption lidar, gas detection, volcanic phenomena

1. INTRODUCTION

Ten active volcanos¹ and many dormant volcanos are located in central and Southern Italy. Their craters are often very close to densely populated urban areas. In order to assess the potential risks, Italian Civil Protection National Service (DPC) supports continuous monitoring and restricts the access to dangerous zones. In this framework, the Diagnostics and Metrology Laboratory (UTAPRAD-DIM) of the Frascati Research Center of the Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA) carries out research on differential absorption lidar (DIAL)² of carbon dioxide in volcanic plumes. Carbon dioxide has been chosen because its anomalous release has been shown to anticipate volcanic eruptions³.

Two systems are under development: BrIdge voLcanic LIdar (BILLI)⁴, funded by the ERC (European Research Council) project BRIDGE, and VULcamed LIdar (VULLI) funded by the ERDF (European Regional Development Fund) project VULCAMED. The first one is based on injection seeded Nd:YAG laser, double grating dye laser, difference frequency mixing (DFM) and optical parametric amplifier (OPA), and the second one relies on injection seeded Nd:YAG laser and optical parametric oscillator (OPO).

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BILLI has been mounted in a small truck and carried out the 3D profiling of the volcanic plume of Pozzuoli Solfatara in October 2014⁵. It is the first time that carbon dioxide in a volcanic plume is retrieved by lidar, although laser remote sensing has already been applied to sound aerosol load⁶, sulfur dioxide⁷ and water vapor⁸ in volcanic plumes: up to now, to our knowledge, lidar soundings of CO₂ have been performed, but not in volcanic plumes⁹, and differential absorption measurements of volcanic CO₂ have been carried out, but they were not range resolved¹⁰. VULLI underwent a first test at the end of 2014, is under upgrade at UTAPRAD-DIM and it is expected to operate in a volcanic area within 2015. BILLI and VULLI can also be used for the measurement of wind speed⁸, thanks to their ability of aiming the laser beam in different directions. The main specifications of the two DIAL systems are listed on Table 1.

Table 1. Main specifications of the two DIAL systems.

DIAL system		BILLI	VULLI
Transmitter	Pulse energy	25 mJ	17 (2014), 25 (2015) mJ
	Pulse duration	8 ns	< 6 ns
	Repetition rate	10 Hz	10 Hz
	Wavelengths	1.6, 2.1 μm	1.57–2.08 (2014), 1.5–3.4 (2015) μm
	Laser linewidth	0.04 cm^{-1}	0.15 (2014), 0.02 (2015) cm^{-1}
	Beam divergence	0.5 mrad	2 (2014), 0.5 (2015) mrad
Receiver	Mirror coating	Al	Al
	Clear aperture	300 mm	300 mm
	Focal length	900 mm	1200 mm
Detector module	Photodiode	InGaAs PIN	InGaAs PIN
	Diameter	1 mm	1 mm
	Responsivity	1.2 A W ⁻¹	1.2 A W ⁻¹
	Gain	5.1×10 ⁴ V A ⁻¹	5.1×10 ⁴ V A ⁻¹
	Noise equivalent power	10 pW Hz ^{-1/2}	10 pW Hz ^{-1/2}
	Bandwidth	0 ÷ 10 MHz	0 ÷ 10 MHz
Analog-to-digital converter (ADC)	Dynamic range	14 bit	14 bit
	Sampling rate	100 MS s ⁻¹	100 MS s ⁻¹

CO₂ absorbs in the 15, 4.2, 2.1 and 1.6 μm bands (in order of decreasing strength)¹¹. Unfortunately, in the first two bands viable lasers are not available and atmospheric backscattering is rather low, so the 2.1 and 1.6 μm bands have been suggested for its detection¹². Nevertheless, the DIAL measurement of CO₂ remains a difficult task because the absorption lines are narrow and weak¹³.

As powerful, tunable and narrow-linewidth laser source we chose:

- a dye laser with difference frequency mixing (DFM) and optical parametric amplifier (OPA) for BILLI,
- an optical parametric oscillator (OPO) for VULLI.

Tm,Ho:YLF and fiber lasers have been discarded⁴ mainly for their limited tunability (few tenths of nm) that can prevent to choose the best absorption line¹⁴.

2. BILLI (BRIDGE VOLCANIC LIDAR)

From 13 to 17 October 2014, BILLI measured carbon dioxide inside the volcanic plume of Pozzuoli Solfatara in Contrada Pisciarelli (Campi Flegrei crater area, Naples, Italy)⁵. Lidar retrievals were in good agreement with conventional techniques, yet based on completely independent and significantly different approaches. Each CO₂ profile was obtained averaging 200 lidar returns at 10 Hz, corresponding to a temporal resolution of 20 s. The spatial resolution was 1.5 m. A total plume scan was retrieved combining 20-30 profiles, i.e. in less than 10 minutes.



Figure 1. Left) Location of lidar and plume, and most northern/southern directions of the laser beam during the measurements. Center) Volcanic plume emitted by the Pozzuoli Solfatarata in Contrada Pisciarelli. The lidar is mounted in a small truck (inside the white circle). Right) Lidar inside the truck. The laser beam is aimed by two large elliptical mirrors (major axis: 450 mm) mounted in two black boxes (inside the white circle).

BILLI was located at latitude $40^{\circ}49'46.28''\text{N}$ and longitude $14^{\circ}08'50.51''\text{E}$ (Figure 1), and the laser beam scanned typically the heading angles from 196° to 234° at an elevation from 0° to 18° . The laser beam was backscattered by the plume and by the rockface (Figure 2). Each lidar return can be divided into two portions, one inside the plume and another outside it, where the CO_2 concentrations equal to the natural background ($C_0=400$ ppm). This latter includes two segments, respectively before (length: L_1) and after (length: L_2) the plume interception. Assuming that inside the plume the CO_2 concentration corresponding to the i -th ADC channel (C_i), is proportional to the lidar signal in the i -th ADC channel (S_i), the optical depth of the laser path can be written as:

$$OD = \Delta\sigma \left[C_0 (L_1 + L_2) + \Delta R k \sum_i S_i \right], \quad (1)$$

where $\Delta\sigma$ is the CO_2 differential absorption cross section and k is the proportionality constant between C_i and S_i . The proportionality assumption is very reasonable because the light backscattering is due to aerosol that is dominated by water droplets, and H_2O and CO_2 are usually proportional in volcanic plumes.

OD can be measured as follows:

$$OD = \ln \left[\frac{S_R(\lambda_{OFF})/S_L(\lambda_{OFF})}{S_R(\lambda_{ON})/S_L(\lambda_{ON})} \right], \quad (2)$$

where S_R is the lidar signal from the rockface, S_L is the signal due to the scattering, inside the laboratory, of some photons of the transmitted laser pulse (this peak gives the exact time of pulse transmission and is proportional to the transmitted energy, thus providing the signal normalization) and $\lambda_{ON}/\lambda_{OFF}$ is the more/less absorbed wavelength used in the DIAL experiment.

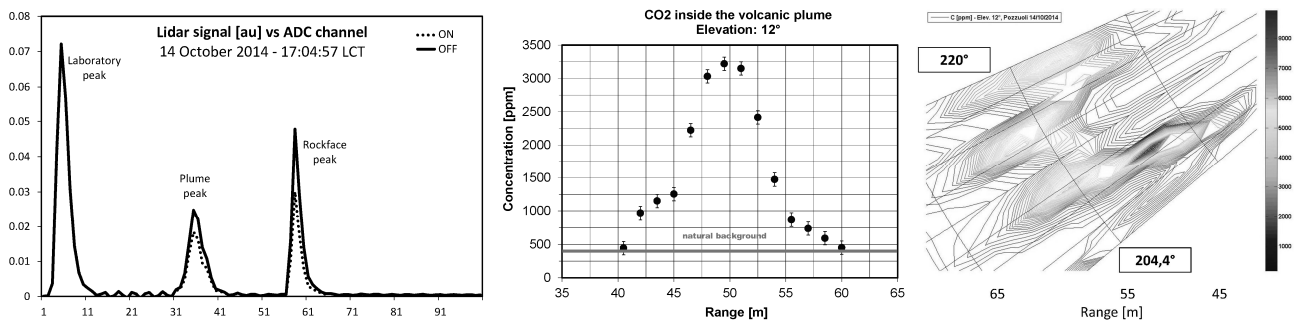


Figure 2. Example of: Left) lidar signal. Center) CO_2 profile. Right) Plume scan (the vent correspond to the grey spot).

Once determined OD, equation (1) can be solved for k , the only remaining unknown, and C_i can be simply calculated as the product of k and S_i (Figure 2). Sometimes, the plume was so optically thick that the rockface was not detected. In this case, in equation (2) the signal peak due to the plume was used instead of that due to the rockface, and carbon dioxide was retrieved only before the peak due to the plume, applying equation (1) with $L_2=0$.

The measurement error was dominated by inaccuracy in wavelength setting. This is why, back to the Frascati Research Center, we implemented a photo-acoustic cell filled with pure CO_2 at atmospheric pressure and temperature, close to the laser exit, as shown in figure 3. Using new software developed for this application, we are able to change the transmitted wavelength (by moving the stepper motor inside the resonator cavity of the dye laser) and to record the photo-acoustic signal (averaging 10 laser shots, corresponding to 1 s). Once corrected the wavelength for a small shift (0.12 cm^{-1}), the agreement between experiment and theory¹³ is very good (figure 3).

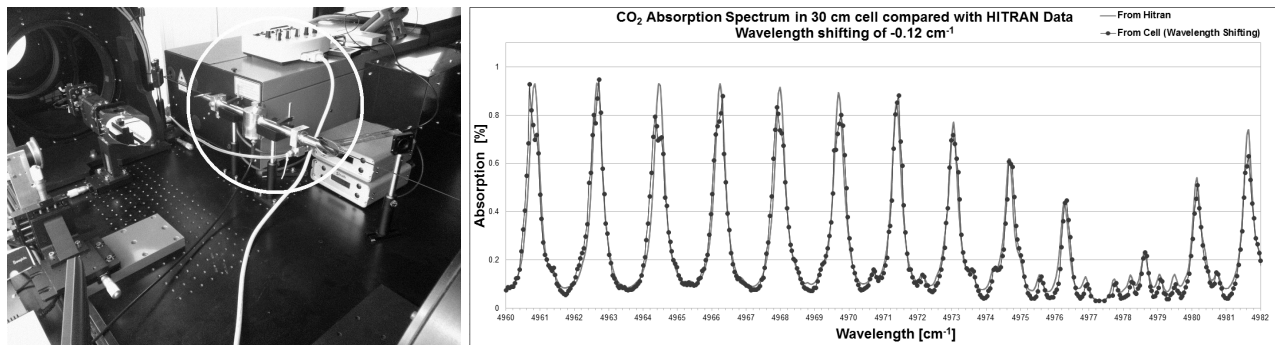


Figure 3. Left) Photo-acoustic cell (inside the white circle). Right) Theoretical (line) and experimental (circles and line) CO_2 absorption spectrum.

3. VULLI (VULCAMED LIDAR)

A first version of VULLI has been completed in 2014 (figure 4). It reached the following specifications:

- spectral range 1.57–2.08 μm ,
- linewidth 0.15 cm^{-1} ,
- power 170 mW,
- pulse duration less than 6 ns.

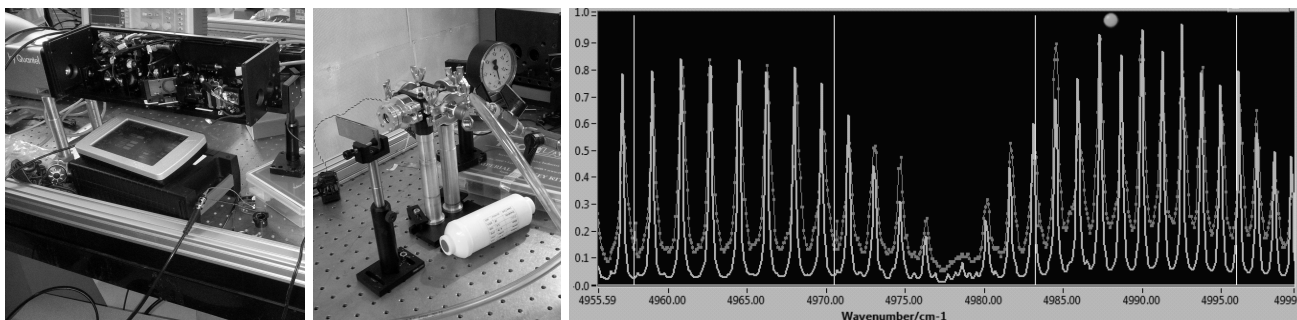


Figure 4. Left) The OPO opened during the test of the laser source of VULLI. Center) Photo-acoustic cell. Right) Theoretical (line) and experimental (circles and line) CO_2 absorption spectrum.

The spectral purity of the laser source has been checked with a cell filled with CO₂ (figure 4), equipped with a microphone to record the photo-acoustic absorption spectrum. Theoretical¹³ and experimental spectrum are in excellent agreement (figure 4), demonstrating that VULLI is ready to be tested on the field, with the purpose of measuring carbon dioxide in the atmosphere.

Notwithstanding these encouraging results, the system will be upgraded in 2015, in order to reduce even more its linewidth, with the final goal of reaching a linewidth of 0.02 cm⁻¹ (single longitudinal mode). Other specifications of VULLI will evolve as indicated in Table 1.

4. CONCLUSIONS

BILLI and VULLI, two differential absorption lidar for 3D profiling of carbon dioxide in volcanic plumes, are under development at the UTAPRAD-DIM laboratory of the Frascati Research Center of ENEA.

Both the laser sources developed for BILLI and VULLI are “single pieces”, realized according to our specifications by world leading manufacturers: the great challenge was to realize laser sources transmitting very narrow linewidths and operating in the harsh environment characteristic of volcanic zones. Moreover, they must reach high power and emit nearly simultaneously two wavelengths. The source chosen for BILLI is more conservative: it is more reliable and has a better quality of the laser beam (circular profile, smaller size and lower divergence). On the contrary, the source chosen for VULLI requires still development, but is less complex, large, heavy and expensive. Our strategic choice, effective up to now, was to differentiate the technical approaches, in order to increase the probability of success of our volcanic activity: in fact BILLI carried out the first lidar measurement of carbon dioxide in a volcanic plume and VULLI promises to follow the example of his older brother.

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