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Wavelength choice for lidar detection of carbon dioxide in volcanic emissions

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ABSTRACT

The true magnitude of CO_2 emissions from volcanic activity is poorly constrained, limiting our understanding of the natural carbon cycle. CO_2-sensitive lidars could be used to measure the distribution of CO_2 in a volcanic plume, thereby allowing volcanic CO_2 fluxes to be measured directly. The recently begun ERC research project CO2VOLC aims to produce such an instrument based on the differential absorption lidar (DIAL) technique. In this paper we investigate the ON and OFF wavelengths which offer optimal CO_2 detection and identify the spectral requirements of the lidar transmitter, in the context of commercially available solid-state laser sources.

KEYWORDS: remote sensing, lidar, DIAL, atmospheric CO_2, volcanic emission

1. INTRODUCTION

Lidar [1] has been used to profile the volcanic plume of Mount Etna during the 2008 eruption [2]. Vertical profiles of extinction coefficient were retrieved up to an altitude above ground level of 5000 m. The system was able to accurately track the spatiotemporal evolution of the volcanic plume thanks to a spatial resolution of 15 m and a temporal resolution of 1 minute. Differential absorption lidar (DIAL) [1] has been deployed to study the Stromboli volcano plume in 2009 [3]. It measured water vapour concentration in cross sections of the plume and wind speed at the crater. Wind speed was retrieved by correlation technique [1]. Lidar returns were obtained up to a range of 3 km. The spatial resolution was 15 m and the temporal resolution was 20 s. By combining these measurements, the water vapour flux in the Stromboli volcano plume was determined.

DIAL is based on the detection of the backscattered photons from laser pulses transmitted to the atmosphere at two different wavelengths. At one wavelength (λ_{OFF}), the light is scattered primarily by air molecules and aerosols, whereas at the other one (λ_{ON}), it is also absorbed by the gas under study. The difference between the two recorded signals is thus related to the gas concentration. More precisely, the DIAL equation can be written [1]:

$$C(R) = \frac{1}{2[\sigma(\lambda_{ON}) - \sigma(\lambda_{OFF})]} \frac{d}{dR} \ln\left[\frac{n(R, \lambda_{OFF})}{n(R, \lambda_{ON})}\right] \quad (1)$$

where C and σ are the concentration and the absorption cross-section of the gas, respectively, R is the range from the system and n is the number of detected photons.

CO_2 fluxes from active volcanoes have been measured only episodically in past, the few available data being incomplete, sparse, and inaccurate. This paucity of information derives from the inherent technical complexity of measuring

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volcanic CO_2: CO_2 is challenging to be measured spectroscopically using remote sensing techniques because of its high concentrations in the background atmosphere.

The volcanic CO_2 concentration is typically only a few ppm within a km of the source, and in this situation the plume width is typically a few hundred meters, making satellite measurements challenging due to the large observing footprint. DIAL has already been used to measure atmospheric CO_2 [4] and is therefore the most promising option for measuring volcanic CO_2 emissions with ground-based and airborne systems.

Since none of the currently available remote sensing techniques is sensitive enough to distinguish the volcanic CO_2 signal over the overwhelming atmospheric signal, nearly all previous estimates of CO_2 fluxes have been indirect. A major objective of the ERC research project CO2VOLC is to design and test in the field a new airborne DIAL for the direct CO_2 flux measurement.

The lidar will be positioned near the target (volcanic plume or gaseous atmospheric dispersion of volcanic gases) that will be probed with the laser beam. With such a configuration, the scanning of the plume will be carried out in a plane or car roughly perpendicular to its axis.

2. Wavelength choice

Carbon dioxide absorbs strongly in the 15, 4.2, 2.1 and 1.6 µm bands (in order of decreasing strength) [5]. Unfortunately, in the first two bands viable lasers are not available and atmospheric backscattering is rather low. Menzies and Tratt [6] suggest to use the 2.1 and 1.6 µm bands and calculate the cross sensitivity of CO_2 measurement to temperature variation. According to them, this cross sensitivity is minimum for the absorption peaks at 1571.406 and 2069.532 nm. As a consequence, these wavelengths are good candidates for λ_{ON}. In order to choose λ_{OFF}, we looked at the nearest wavelength:

- with minimum in the CO_2 absorption,
- in a region where H_2O absorption is as close as possible to that at λ_{ON} (in order to minimize cross sensitivity to water vapour) and
- in a region where H_2O absorption is weakly dependent on the wavelength (in order to minimize the effect of inaccuracies in λ_{OFF}).

Looking at the absorption coefficients from HITRAN [7] (Fig. 1 and Fig. 2) we suggest λ_{OFF} at 1569.906 in the 1.6 µm band and 2068.359 nm in the 2.1 µm band. Cross sensitivity of water vapour is low: a 50% relative humidity at 296 K will imply a bias in the carbon dioxide measurements of 0.29 and 0.031 ppm in the 1.6 and 2.1 µm bands, respectively.

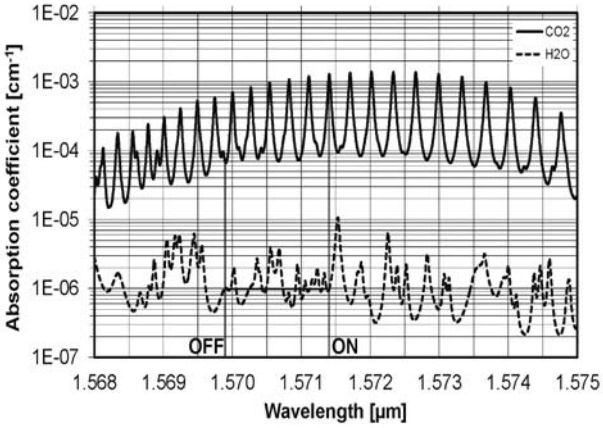

Fig. 1. Absorption coefficient of carbon dioxide and water vapour around 1.57 µm at 296 K and 1 atm. Graphic based on the public database HITRAN [7]. Data retrieved from the free access website http://hitran.iao.ru

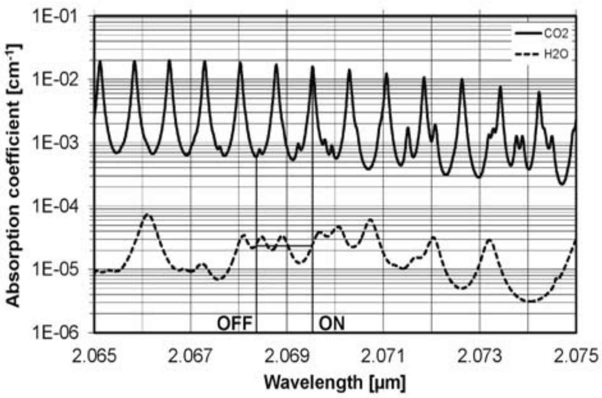

Fig. 2. Absorption coefficient of carbon dioxide and water vapour around 2.07 µm at 296 K and 1 atm. Graphic based on the public database HITRAN [7]. Data retrieved from the free access website http://hitran.iao.ru

Table 1. Wavelengths and linewidths of the laser source for DIAL detection of carbon dioxide.

	1.6 μm band	2.1 μm band
λ_{ON}	1571.406 nm 6363.728 cm^{-1}	2069.532 nm 4832.010 cm^{-1}
λ_{OFF}	1569.906 nm 6369.808 cm^{-1}	2068.359 nm 4834.751 cm^{-1}
Linewidth	0.04 nm 0.15 cm^{-1} 5 GHz	0.06 nm 0.15 cm^{-1} 5 GHz

The full width at half maximum (FWHM) linewidth of the CO_2 absorption has been evaluated with HITRAN [7] considering the gas mixture of the US Standard Atmosphere 1976 [8] (mean latitude summer model) at 296 K and 1 atm. It resulted in both cases 0.15 cm^{-1}. As a consequence, the laser source of the DIAL system should have a linewidth narrower than 0.15 cm^{-1}, at least for λ_{ON}: λ_{OFF} lies in between two peaks whose spacing is about 1 cm^{-1}.

The wavelengths and linewidths of the laser source for DIAL detection of carbon dioxide have been summarized in Table 1.

3. Signal modelling

The lidar signals have been simulated for a vertically pointing system, as described below.

The atmosphere has been modelled with the US Standard Atmosphere 1976 [8] (mean latitude summer model). Aerosol backscattering and extinction have been calculated according to the classical paper by Shettle and Fenn [9] (urban model with 50% relative humidity). The values tabulated in that paper have been used at the ground level and a decrease proportional to air density has been applied. For the water vapour, the mixing ratio corresponding to a 50% relative humidity at ground level has been retained, while for the carbon dioxide the value of 391.57 ppm has been chosen, according to the 2011 average recorded at the Mauna Loa Observatory [10].

A Gaussian plume has been added to the above mentioned atmospheric profiles, with the following characteristics:

- range from the lidar system: 250 m,
- width: 100 m,
- aerosol peak height: 20% of the ground level,
- water vapour peak height: 20% of the ground level,
- carbon dioxide peak height: 10 ppm.

As far as the lidar system is concerned, the pulse energy is 10 mJ, the optical efficiency 50%, the receiver diameter 15 cm and the time resolution 10 ns.

ON and OFF lidar signals in the 1.6 and 2.1 μm bands have been shown in Fig. 3 and Fig. 4, respectively. A small deflection on the signals can be noticed around 250 m, due to the aerosol peak. Although the signals are smaller in the 2.1 μm band, due to lower backscattering, the difference between the ON and OFF signals seems too small for the 1.6 μm band, at least for the short distance of 250 m that could be likely be used in an airborne measurement.

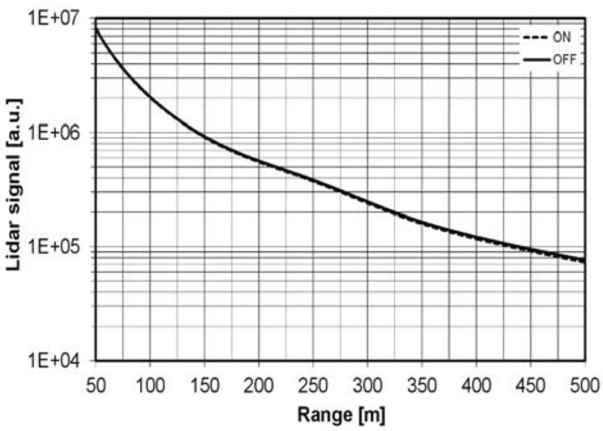

Fig. 3. ON and OFF lidar signals at around 1.57 μm.

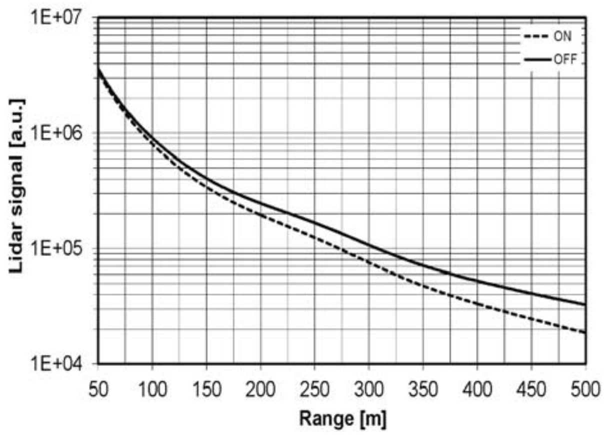

Fig. 4. ON and OFF lidar signals at around 2.07 μm.

4. CONCLUSIONS

DIAL measurements of carbon dioxide in volcanic emissions should be performed in the 1.6 or 2.1 μm bands. In order to reduce cross sensitivity to temperature variation and water vapour the DIAL wavelengths should be chosen as follows: in the 1.6 μm band λ_{ON} = 1571.406 nm and λ_{OFF} = 1569.906 nm; in the 2.1 μm band λ_{ON} = 2069.532 nm and λ_{OFF} = 2068.359 nm.

It is advisable that the laser source of the DIAL system has a linewidth narrower than 0.15 cm^{-1}.

A preliminary modelling of the lidar signals indicates that the 2.1 μm band is more appropriate for an airborne system (closer to the plume than a ground-based system). Another advantage of this band is the lower cross sensitivity to water vapour.

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