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Demonstration of range-resolved detection of stable water isotopologues by differential absorption lidar

Jonas Hamperl¹, Jean-Baptiste Dherbecourt¹, Myriam Raybaut¹, Julien Totems², Rosa Santagata¹, Jean-Michel Melkonian¹, Antoine Godard¹, Patrick Chazette², Cyrille Flamant³

¹DPHY, ONERA, Université Paris Saclay, Palaiseau, France

²Laboratoire des Sciences du Climat et de l'Environnement (LSCE), UMR 1572, CEA-CNRS-UVSQ, Gif-sur-Yvette, France

³Laboratoire Atmosphères Milieux et Observations Spatiales (LATMOS), UMR 8190, CNRS-SU-UVSQ, Paris, France

Author e-mail address: jean-baptiste.dherbecourt@onera.fr

Abstract: We present range-resolved measurements of the stable water isotopologues H_2^{16}O and HD^{16}O in the lower troposphere enabled by a ground-based differential absorption lidar based on a high-energy parametric laser source. © 2022 The Author(s)

1. Introduction

Observations of stable water isotopologues in the lower atmosphere can provide valuable insights into the variety of processes governing the global hydrological cycle. The provision of such data with sufficient vertical resolution in the lower troposphere (0–3 km) has the potential to significantly increase the accuracy in the predictions made by atmospheric general circulation models [1]. For this purpose, differential absorption lidar (DIAL) is a promising technique which has been successfully demonstrated for water vapor and other greenhouse gases [2]. In order to quantify the relative abundance in HD^{16}O (δD), the method requires collocated measurements of both the main isotopologue H_2^{16}O and HD^{16}O with an equivalent relative precision, which has not been demonstrated so far despite recent progress on multiple wavelengths DIALs [3], and the proven capability of some approaches to probe isotopic water vapor lines [4]. In this paper we present a DIAL instrument operating at 1.98 μm , for which calculations indicate that range-resolved measurements of δD should be achievable [5]. We show first results for the water isotopologues H_2^{16}O and HD^{16}O along a horizontal line of sight.

2. Lidar setup

The DIAL transmitter (Fig. 1) is based on a doubly-resonant nested-cavity optical parametric oscillator (NesCOPO) [6] combined with an optical parametric amplification (OPA) stage pumped by a Nd:YAG laser delivering 12 ns pulses at a repetition rate of 150 Hz. The NesCOPO provides single-frequency emission tunable in a 1 nm window around 1982.5 nm suitable for addressing H_2^{16}O and HD^{16}O absorption lines (Fig. 2).

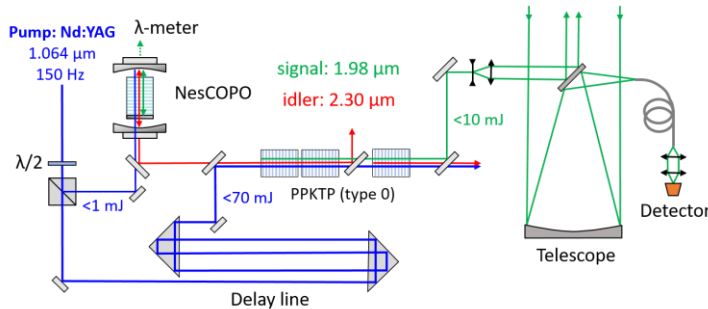


Fig. 1. Lidar setup

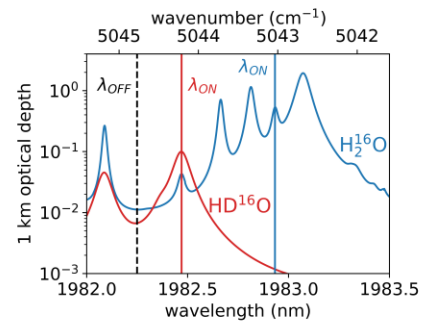


Fig. 2. Water vapor absorption lines and DIAL on/off wavelengths

The OPA stage consists of three high-aperture (5x5 mm²) PPKTP crystals [7]. For enhanced conversion efficiency, the idler beam is filtered prior to the last crystal. With pump energy pulses of 70 mJ, output energies at 1.98 μm are in the range of 6–9 mJ. Pulse-to-pulse wavelength monitoring is realized with a commercial wavemeter (High Finesse WS6 IR). Wavelength control and on/off switching is accomplished by adjusting the NesCOPO cavity length using piezoelectric transducers. The amplified signal beam passes a beam expander resulting in a divergence full-angle of 300 μrad . The lidar receiver consists of a 10-inch Newtonian telescope which focuses the backscattered

radiation onto a 900 μm -multimode fiber. The resulting full field-of-view angle is 1.2 mrad. The fiber output is imaged onto a commercial InGaAs amplified photodiode (300 μm diameter) with a 3.5 MHz electronic bandwidth.

3. First demonstration of water isotopologues detection by DIAL

DIAL tests along a horizontal path were conducted on 16 April 2021 under relatively dry conditions (ca. 2.5 g/kg water vapor mixing ratio) in the suburban Paris region. Water vapor H_2^{16}O and the isotopologue HD^{16}O were addressed at 1982.93 nm and 1982.47 nm, respectively (Fig. 2). The two-way differential optical depth $\Delta\tau$ is calculated as the logarithm of the ratio of off-line to on-line return signals. The mixing ratio of each isotopologue at a range R is then proportional to the slope of $\Delta\tau$ within a range gate ΔR evaluated by a least-squared regression. The HD^{16}O retrieval requires a correction due to the H_2^{16}O absorption contribution (Fig. 2). The upper panels of figure 3 show the recorded on/off lidar signals expressed as signal-to-detection-noise ratio (SNR), with a dominant noise contribution from the transimpedance amplifier (1.3 pW $\text{Hz}^{-1/2}$ noise-equivalent power). The lower panels of figure 3 show the retrieved volume mixing ratios (VMR). The signals were recorded and averaged over 30 minutes successively for each species. The range-gate resolution ΔR for both data sets is 150 m. For H_2^{16}O only, a reference measurement was provided by an in-situ Picarro CRDS (Cavity Ring Down Spectrometer) analyzer located in the vicinity of the lidar instrument showing fair agreement (5% relative deviation) with the first DIAL data point.

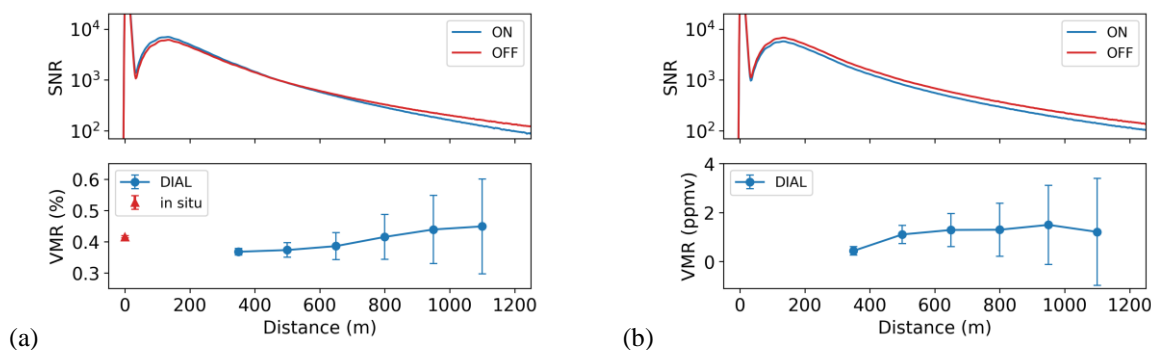


Fig. 3. Raw (no energy normalization) averaged DIAL signals over 30 min and retrieved VMR for H_2^{16}O (a) and HD^{16}O (b).

This DIAL experimental demonstration on water isotopologues is a first important step toward range-resolved δD quantification in the vertical direction. For this purpose, future work will focus on the characterization of errors and biases as well as on increasing the power performance of the transceiver for higher sensitivity.

Acknowledgments

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