Absolutely calibrated Raman lidar as a reference tool in atmospheric humidity profiling

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Abstract

Raman lidars exploit the proportionality between the intensity of scattered by a Raman process laser radiation and the number density of the scattering molecules to derive water vapor/air mixing-ratio profile. Water vapor/air mixing-ratio is directly proportional to the ratio of the measured intensities of Raman scattering from water vapor and nitrogen molecules. The coefficient of proportionality, commonly denoted as calibration constant, depends on the instrument parameters and the spectroscopic parameters of the scattering molecules and is the primary factor defining lidar measurement accuracy. Derivation of the calibration constant using the above mentioned parameters is possible but leads to high uncertainty. Therefore, Raman lidars are calibrated against reference instruments - a radio sonde or a microwave radiometer. The accuracy of such calibration is thus defined by the accuracy of the reference instrument. Since these reference instruments have inferior, compared to the potential Raman lidar accuracy, this type of calibration impairs the lidar accuracy.

A new first principle calibration method, based on the use of gravimetrically produced water vapor/air mixture will be presented. The method allows deriving the calibration constant with uncertainty lower than 0.1%, traceable to the primary standards of mass and length. Apart from the use for operational profiling, the calibrated in such a way lidar has the potential to become a reference in radio-sonde, microwave radiometer and GPS water vapor validation and/or calibration.

Introduction

Raman lidars have been used for high resolution vertical profiling of water vapor within the troposphere since the early 70 ies of the last century. Most of the measurements were performed for research purposes and at night time (1-5). The use of a narrow field-of-view (NFOV), narrow band (NB) receiver allows daytime operation at visible and near UV wavelengths with an operational range up to the mid troposphere (6).

The successful long-term operation of the first automated NFOV NB lidar motivated the Swiss (7, 8), the German (9), and the Dutch (10) meteorological services to develop operational water vapor lidars.

The Swiss <u>RA</u>man <u>L</u>idar for <u>M</u>eteorological <u>O</u>bservations – RALMO was developed and built by EPFL (Ecole Polytechnique Fédérale de Lausanne – Switzerland) as a co-funded project with Swiss Meteorological Service – MeteoSwiss (7, 8). Since August 2008 the lidar has been operated at the Aerological station of Payerne by MeteoSwiss with the support of EPFL. The lidar operates unattended and delivers water vapor profiles every half an hour except in case of precipitation or clouds below 800 m.

The six-day time series shown on Figure 1 demonstrate RALMO capability of revealing different meteorological events.
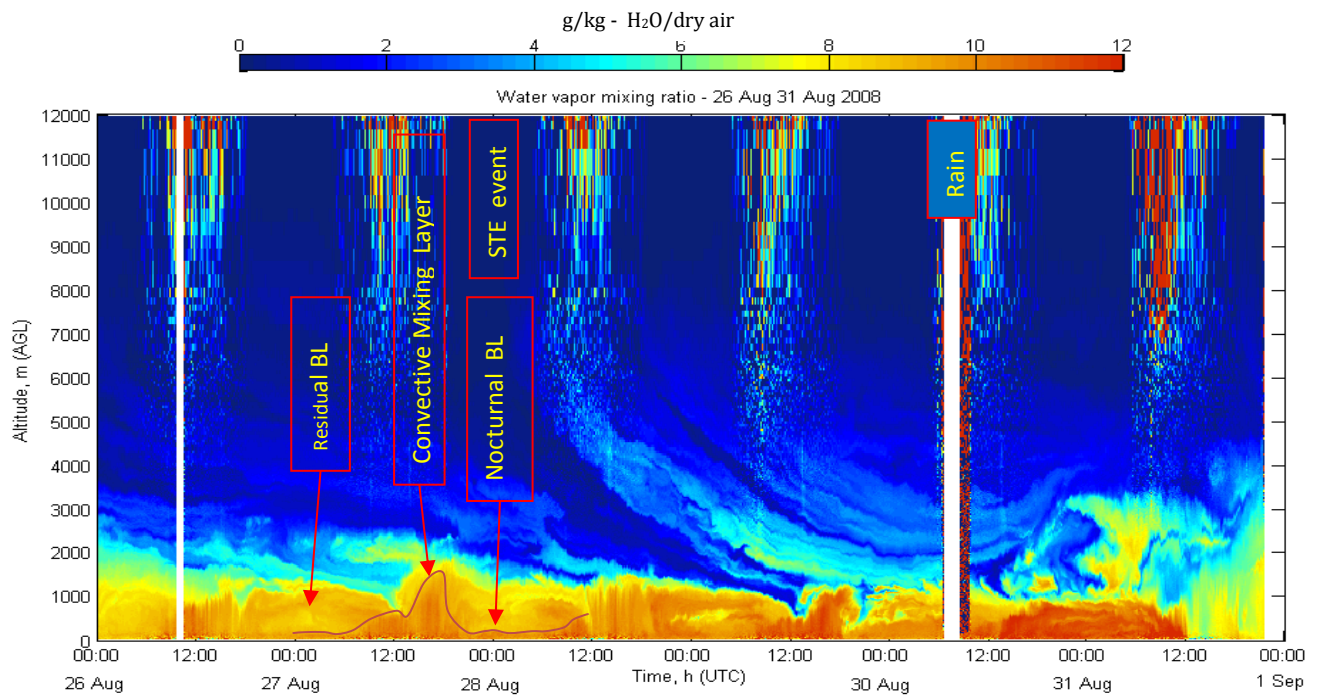

Fig.1 Six-day time series of water vapor with 10 min time resolution, spatial resolution 30 m to 4 km and up to 300 m at 12 km. All data are shown, including data with a statistical error > 10%. The noisy zones above 5000 m, around noon mark the increase in statistical error due to the solar background during the daytime. The white zones mark data gaps due to rain.

The analysis of RALMO long-term data series (8) revealed changes in the accuracy of the measurements attributed to changes in the lidar calibration constant requiring periodic recalibration by radiosonde data. The accuracy of such calibration is thus defined by the accuracy of the radiosonde data. Since radiosonde capacitive hygrometers have inferior, compared to the potential Raman lidar accuracy, this type of calibration impairs the lidar accuracy. The new calibration technique presented here was developed in order to improve the lidar accuracy. To avoid interruption of the regular operation of RALMO, the experiments reported here were done using the high resolution Raman lidar of EPFL (11). This lidar allows whole hemisphere measurements of water vapor mixing ratio with up to 1.2 m spatial and 1 s temporal resolution at distances to 500 m. The final goal of the experiments was to implement the new calibration technique on RALMO.

Theory

A LIDAR (LIght Detection And Ranging) is a laser-based, optical instrument, which allows remote profiling of atmospheric parameters such as, aerosol optical properties, humidity, temperature, trace gas concentration, wind speed and direction etc.. A lidar transmits short laser pulses into the atmosphere and detects and analyzes the backscattered light from the atmospheric molecules and particles. In the special case of inelastic light-matter interaction, known as Raman scattering, the backscattering efficiency is proportional to the number density of the scattering molecules. The scattered in the Raman process wavelength differs from the laser wavelength and is specific for each scattering compound thus giving high selectivity of the method.

The Raman method (1) for lidar water vapor profiling uses Raman-shifted backscatter from atmospheric water vapor $P_{H_2O}(R)$ and nitrogen $P_{N_2}(R)$ to retrieve water vapor mixing ratio $q(R)$ at a distance R as:

$$q(R) = k_{H_2O} \frac{P_{H_2O}(R)}{P_{N_2}(R)} \tau(R) \qquad (1)$$

where $\tau(R)$ is a term correcting the differences in the atmospheric transmission at the water vapor and nitrogen Raman wavelengths. The coefficient of proportionality k_{H_2O}, commonly denoted as the lidar calibration constant, can be presented as:

$$k_{H_2O} = \frac{M_{H_2O} n_{N_2}}{M_{air} n_{air}} \frac{\int T_{N_2}(R,\lambda) \eta_{N_2}(R,\lambda) \sigma_{N_2}(\lambda) d\lambda}{\int T_{H_2O}(R,\lambda) \eta_{H_2O}(R,\lambda) \sigma_{H_2O}(\lambda) d\lambda} \qquad (2)$$

where: M_X and n_X are the molecular mass and the number density of species X (air denotes dry air), $\sigma_X(\lambda)$ is the Raman cross-section of species X, and $T_X(\lambda,R)$ and $\eta_X(\lambda,R)$ are the instrument transmission function and the PMT efficiency of the respective Raman channel X.

The calibration constant is commonly obtained by comparison of a lidar profile to data from a reference instrument: collocated radiosonde, microwave radiometer, or GPS (2, 3, 6, 7). The thus derived calibration constant has low accuracy mostly because of the limited accuracy of the reference instruments (12), and the unaccounted for atmospheric influence $\tau(R)$. Another essential drawback of this calibration method is that the lidar and the reference instrument nearly always sample different air masses with different time and space resolutions leading to additional systematic errors.

In another approach, known as "absolute" calibration, k_{H2O} is calculated using Equation 2 and measured or modeled spectral and instrumental parameters (4, 13). Currently this method has accuracy that is even lower than the accuracy of the method mentioned above, mostly because of the low accuracy of the measured (modeled) Raman cross-section functions, and the measured instrumental functions $T_x(R, \lambda)$ and $\eta(R, \lambda)$.

In the method for calibration presented here, k_{H_2O} is derived using Equation 1 and backscatter signals measured with the lidar receiver in a calibration cell filled with reference water vapor/air mixture. The reference mixture is produced by mixing known masses of water vapor and dry air, which yields an absolute mixing ratio. Therefore the obtained in this way lidar calibration constant is absolute. The new calibration method eliminates the unavoidable in the currently used Raman lidar calibration method errors and uncertainties induced by the reference instrument. Since the measurement is taken from short distance the term $\tau(R) = 1$. The use of stable calibration concentrations allows for longer acquisition series thus reducing the statistical errors. An essential condition for the successful application of the method, however, is to achieve range independence of the instrumental functions in Equation 2, which at present is a problem for the existing water vapor Raman lidars. We managed to satisfy this condition in the lidars built at EPFL by using a novel design of the lidar receiver (7, 11).

Experimental setup

In the experiment we employ backscattering (180°) but not the classical 90° Raman scattering setup to avoid errors related to differences between 180° and 90° scattering cross sections. This configuration has the advantage of detecting the backscattered light as in real lidar observations and allows using bigger scattering volumes. The main challenge in such configuration is to eliminate the parasite light from elastic scattering and auto-fluorescence.

The experiments were carried out with a configuration resembling biaxial lidar with probed volume inside a calibration cell. The scheme of the experimental setup is presented in Figure 2. The cell is 1.8 m long with a cross section of 0.3 x 0.284 m. The long cell walls are made of glass and the sidewalls of Al alloy. The pumping beam enters the cell trough a 1 m focal length lens and exits trough a low auto-fluorescence window. The content of the cell can be changed trough valve-controlled gas inlet and gas exit ports installed on the opposite sidewalls.
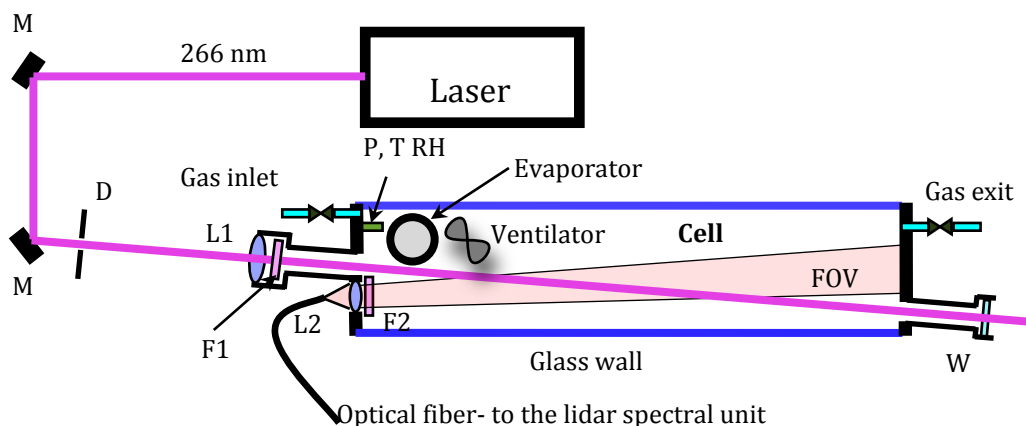

Fig. 2 Experimental setup: L1-Transmitter lens, L2-Receiver lens, F1-Laser-line filter, F2-Long-pass filter, W-Output window, M-Steering mirrors, D-Diaphragm, FOV-receiver's field of view, P, T, RH- Pressure, temperature and relative humidity sensors.

A combined T/RH sensor installed inside the cell is used for temperature/ relative humidity monitoring and the gas pressure is measured by an electronic pressure gauge.

In the experiments we used the laser, the spectral unit and the photodetectors of the EPFL's high spatial/temporal resolution lidar. The laser beam is delivered to the calibration cell by steering mirrors. The radiation, backscattered from the probed volume, is collected by a fused silica lens and focused on a 400 μm optical fiber. The fiber delivers the light to the lidar spectral unit (a prism polychromator). The signal visualization and acquisition is carried out by a 1 GHz, 5GS/s oscilloscope - LeCroy Waverunner 6100.

Reference humidity generation

The accuracy and the precision of the calibration constant depend directly on the accuracy and the precision of the water vapor mixing ratio of the reference mixture. To produce the reference mixture precisely weighed quantity of liquid water is evaporated directly into the calibration cell. The cell is filled beforehand with known amount of dry air. The water vapor mixing ratio is calculated directly as a ratio of the mass of evaporated water to the mass of dry air in the cell. Since mass is a fundamental quantity, the method yields an absolute measurement of water vapor mixing ratio. The mass of the dry air is calculated from the air density and the cell volume. The dry air density is derived from the measured cell pressure p and temperature T as:

$$\rho = \frac{pM_a}{ZRT} \qquad (3)$$

where M_a is the molar mass of dry air, R is the molar gas constant and Z is the compressibility factor

The dry gas is produced by passing air with RH below 2% through a liquid nitrogen trap and a filter. The produced by this method dry air has sufficiently low water content for our purposes (estimated at hundreds of ppb to several ppm).

Calibration experiment

The calibration constant was derived from measurements taken at five humidity levels with water vapor mixing ratios ranging from 2.4 to 13.4 g/kg. The lower limit was chosen above 10% RH to avoid uncertainties related to low water vapor content and possible diffusion of wet air inside the cell. The higher limit was chosen below 70% RH to prevent uncertainty related to water condensation and increased wall adsorption. The water vapor-air mixing ratio of the cell content was verified by measuring the relative humidity and temperature with precise capacitive hygrometer and Pt 1000 thermometer.

The ratio of water vapor to nitrogen Raman signals was calculated after subtracting the respective backgrounds and compared to the reference water vapor mixing ratio. The result from the comparison is presented in Figure 3.
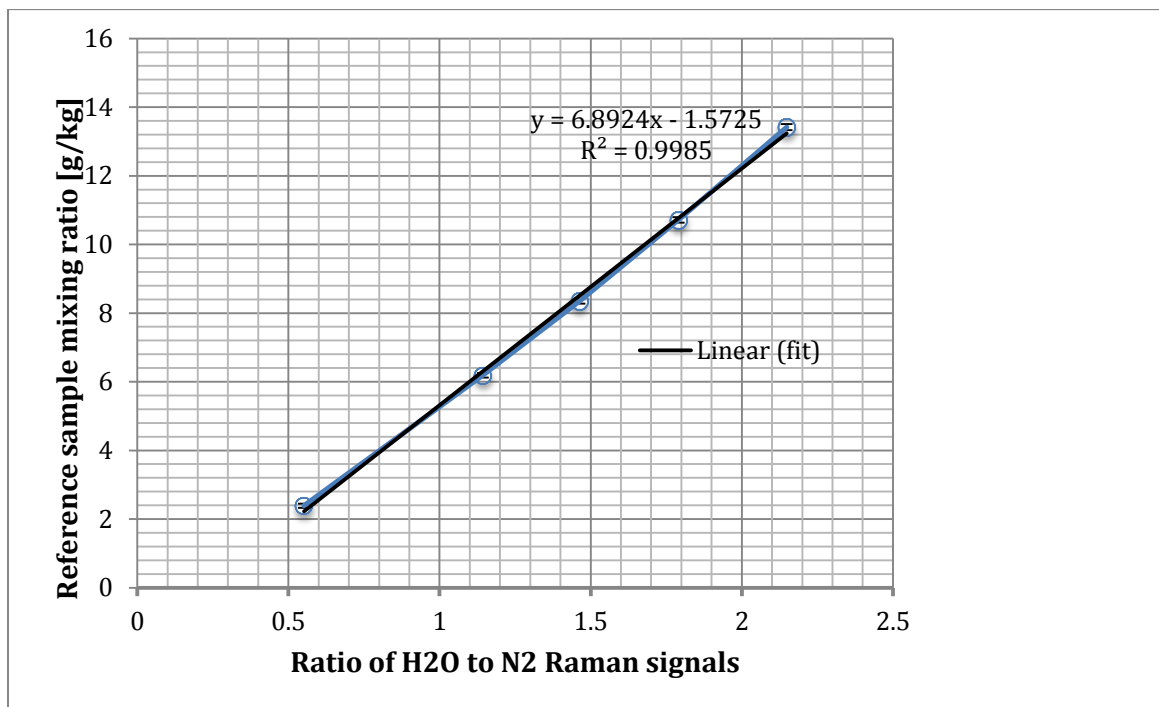

Fig.3 Ratio of water vapor to nitrogen Raman signals versus water vapor mixing ratio of the reference sample. The points on the blue line are measured values and the black line is the linear fit. Note the mixing ratio error bars.

The calibration constant is derived as the slope of the linear fit to the five measurement points. The observed offset corresponds to a leak of nitrogen signal into the water channel. The leak is a result of the close positioning in space of the water vapor and nitrogen channels with the nitrogen signal about one order of magnitude stronger than the water vapor one. The amount of this leak was measured during the polychromator alignment and correlates well with the offset of the calibration curve. The calibration results correlate within 5% with the lidar calibration performed in open air against relative humidity sensors positioned along the laser beam.

Conclusions

A new method for absolute calibration of water vapor Raman lidars was developed. A lidar calibrated with the described method has the potential to become reference instrument for atmospheric profiling of water vapor and can be used for validation and calibration of other instruments for water vapor measurements, such as microwave radiometer, balloon-borne radiosondes, and GPS.

The method was used to calibrate the EPFL high-resolution Raman lidar. Development of a calibration facility for calibrating the Raman Lidar for

Meteorological observations (RALMO) of MeteoSwiss is foreseen. The absolute calibration of RALMO will contribute to establishing the MeteoSwiss aerological station in Payerne as a WMO reference site for upper air humidity measurements.

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