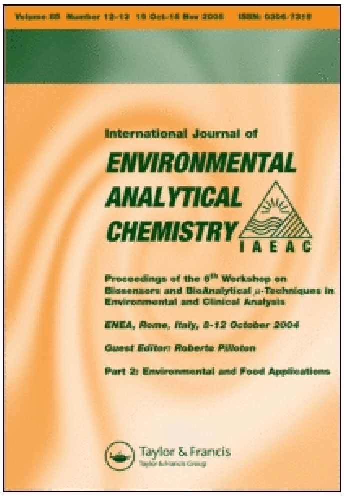

International Journal of Environmental Analytical Chemistry

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713640455

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To cite this Article Lazzarotto, Benot , Quaglia, Philippe , Simeonov, Valentin , Larchevěque, Gilles , Bergh, Hubert van den and Calpini, Bertrand(1999) 'A Raman Differential Absorption Lidar for Ozone and Water Vapor Measurement in the Lower Troposphere', International Journal of Environmental Analytical Chemistry, 74: 1, 255 — 261

To link to this Article: DOI: 10.1080/03067319908031430

URL: http://dx.doi.org/10.1080/03067319908031430

Intern. J. Environ. Anal. Chem., Vol. 74(1-4), pp. 255-261
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A RAMAN DIFFERENTIAL ABSORPTION LIDAR FOR OZONE AND WATER VAPOR MEASUREMENT IN THE LOWER TROPOSPHERE

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(Received 20 April 1998; In final form 24 July 1998)

A new way of measuring ozone and water vapor in the Planetary Boundary Layer (PBL) is proposed. The method is based on the simultaneous measurement of the Raman backscattering in the UV by O_2, N_2 and H_2O, using a single pump beam at 266 nm. The ozone concentration is retrieved from the differential absorption of the N_2 and O_2 Raman backscattered signals, while the water vapor is measured using the classical Raman scheme. We present some preliminary results showing daytime ozone measurements in good correlation with a point monitor.

Keywords: Raman; LIDAR; ozone; water vapor; UV

INTRODUCTION

The UV ***d****ifferential* ***a****bsorption* ***l****idar* (DIAL) technique is a well know technique successfully applied both to tropospheric and stratospheric studies [1,2,3]. However, strong uncertainties are caused by aerosols, mainly due to the unknown wavelength dependence of the aerosols backscattering coefficient [4]. In order to perform daytime boundary layer measurements of ozone and water vapor even in cases of high aerosol load, we are developing a new way to measure ozone concentration.

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By using a single excitation wavelength, namely the 4th harmonic of a Nd:YAG laser, vibrational Raman shifted wavelengths from atmospheric O_2 and N_2 are obtained. The two gases have well known Raman cross sections and constant mixing ratio. Such Raman shifted wavelengths are well suited for differential ozone measurements in the Hartley band. The two Raman backscattered signals are essentially independent of the uncertainties in the aerosol Mie scattering cross sections. Moreover, they are situated in the solar blind region, making daytime measurements possible. Furthermore the water vapor mixing ratio can be retrieved using the classical Raman method (i.e. using the relative intensity of the H_2O Raman backscattered line) [5].

This method, called Raman DIAL, has already been used successfully in the stratosphere under night time conditions for ozone measurements [6]. We have adapted this technique for daytime monitoring of the ozone in the PBL and the lower troposphere.

EXPERIMENTAL SET-UP

The optical layout is shown in Figure 1. The system is based on a quadrupled Nd:YAG laser which emits 266.1 nm radiation pulses of 120 mJ at 10 Hz, with a beam divergence of 0.5 mrad in a non coaxial configuration. In order to reduce the minimum altitude of the measurement (actually 250 m), some tests have been made with a coaxial configuration, but with no significant improvement. By using in a beam expander, we plan to reduce this value to around 100 m. The three backscattered Raman signals of interest are collected by a Newtonian telescope with a 20 cm diameter primary mirror, 60 cm focal length and 5 mrad field of view. The Raman signals are spectrally separated by a monochromator (500 mm optical path) with a background noise rejection of 10^5. The monochromator has a 1.1 nm / mm resolution using a 1.800 gr / mm grating. Additional rejection of the LIDAR backscattered signal at the pump wavelength is performed by two custom designed holographic bandpass filters, with an optical density 2.6 at 266 nm for each filter and up to 80% transmission for the three Raman wavelengths when tilted at 18 degrees. The bandpass filter transmission curves for a tilt of 0 and 20 degrees are shown in Figure 2.

With the combination of the monochromator and the two bandpass filters, we obtain over 10^{10} rejection at 266 nm. The f number of the monochromator and telescope are matched using two plano - convex lenses, a 40 mm focal length lens to collimate the light in the bandpass filters and a 60 mm focal length lens at the entrance of the monochromator. The two main Raman shifted wavelengths

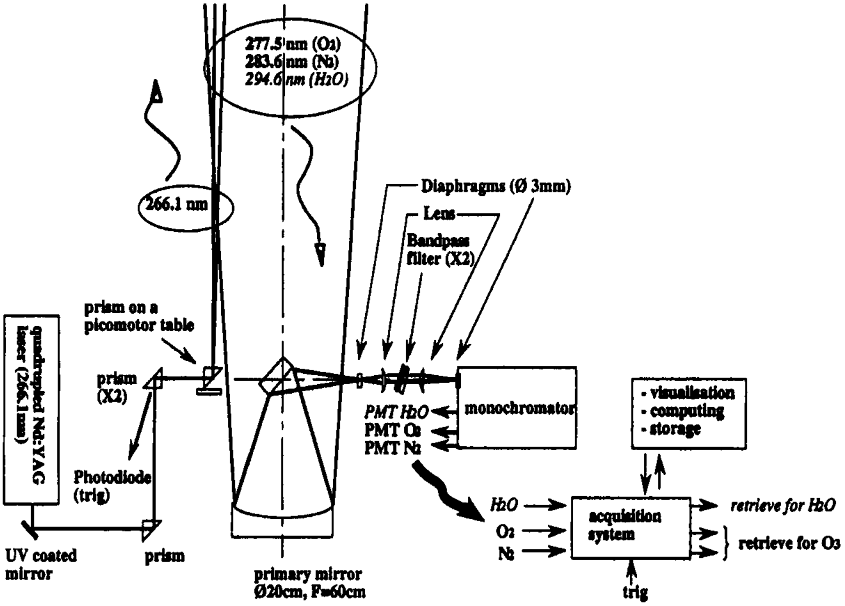

FIGURE 1 Experimental setup

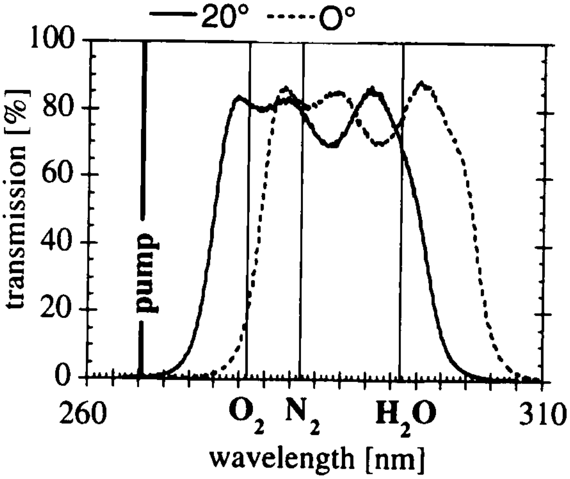

FIGURE 2 Bandpass filter transmission curves between 260 – 310 nm, at 2 different angles of the incident light

(by N_2 and O_2) at the output of the monochromator are simultaneously detected by two Hamamatsu H5783-06 photodetection modules, and the signals are recorded with a 12 bit - 20 MHz transient ADC recorder, with the control of the acquisition and treatment of the data performed by a PC. An additional photomultiplier tube (PMT) and acquisition channel is added for the water vapor retrieval.

DATA ANALYSIS

The Lidor equation for a vibrational Raman signal is the following [7] :

$$P_X(R) = P_0 K_X(\lambda)\frac{1}{R^2}\beta_X^R(R) n_X(R) O(R)$$

$$\exp\{-\int_0^R(\alpha_l(r)+\alpha_X(r)+n_{O_3}(R)\sigma_l(r)+n_{O_3}(R)\sigma_X(r))dr\}$$

Where X stands for O_2, N_2 or H_2O.

R = ct/ 2: distance LIDAR – probed volume (t is the time for the backscattered signal to come from this volume and c is the velocity of the light).

$P_X(R)$ [W]: received energy for the component X.

P_0 [W]: emitted energy of the laser.

$K_x(\lambda)$ [no dimension]: constant which takes into account the optical efficiency of the transmission and detection part for the lidar (telescope surface, quantum efficiency of the detector, optical transmission coefficients, ...).

β_X^R (R) [cm^2/mol]: Raman cross section for the vibrational transition of the corresponding element X.

$n_X(R)$ [#mol/cm^3]: density of the molecules X.

O(R): optical overlap recovery function.

$\alpha_X(r)$(resp. $\alpha_l(r)$) [cm^{-1}]: extinction coefficient for the Raman shifted wavelength of the element X (resp. for the laser emitted wavelength).

$\sigma_x(r)$(resp. $\sigma_l(r)$) [cm^2/mol]: ozone absorption cross section for the Raman shifted wavelength of the element X (resp. for the laser emitted wavelength).

Raman shifts are 1555 cm^{-1} for O_2, 2331 cm^{-1} for N_2 and 3651 cm^{-1} for H_2O. With the 266.1 nm pump beam, the vibrational Raman wavelengths are correspondingly 277.5 nm for O_2, 283.6 nm for N_2 and 294.6 nm for H_2O [8]. The corresponding ozone cross section for the Raman shifted wavelengths are $490.6*10^{-20}cm^2/mol$ for O_2 and $296.3*10^{-20}$ cm^2/mol for N_2 [9].

Taking $S_X=\ln(R^2P_X(R))$, with $S_{N_2}(R)$ and $S_{O_2}(R)$ we can retrieve the ozone concentration:

$$n_{O_3}(R) = \frac{\frac{d}{dr}(S_{N_2}(R)) - \frac{d}{dr}(S_{O_2}(R)) - [\alpha_{O_2}(R) - \alpha_{N_2}(R)]}{\sigma_{O_2}(R) - \sigma_{N_2}(R)}$$

By detecting the H_2O Raman signal at 294.6 nm, the water vapor profile can be calculated by applying the method proposed in [4]: with a normalization of P_{H_2O} and in taking into account P_{N_2}, the water vapor density is calculated as:

$$n_{H_2O}(R) = n_{N_2}(R)\frac{P_{H_2O}(R)}{P_{N_2}(R)}\frac{\beta^R_{N_2}}{\beta^R_{H_2O}}\frac{K_{N_2}}{K_{H_2O}}$$
$$\exp\{\int_0^R[\alpha_{H_2O}(r) - \alpha_{N_2}(r) + n_{O_3}(R)(\sigma_{H_2O}(r) - \sigma_{N_2}(r))]dr$$

RESULTS AND DISCUSSION

With the presented configuration, Raman signals have been acquired in the analog detection mode for altitudes up to 1000 m. The high rejection ratio of 266nm was confirmed by the absence of the elastic lidar echo in O_2 and N_2 Raman channels caused by low level clouds. Two typical range corrected signals obtained with the system are shown in Figure 3, for the O_2 and N_2 Raman shifted wavelengths, based on a 4000 shots average. The graph shows the difference in the slopes of the curves due to different ozone absorption cross-sections. The O_2 shifted signal corresponds to the *on* wavelength (i.e. the more absorbed beam) and the N_2 shifted signal to the *off* wavelength. No degradation of the signal due to background daylight has been observed.

In figure 4 we show a comparison between the ozone concentration measured by a point monitor 12 m above the ground and by the Raman DIAL values started at 262 m above the ground. The calibrated ozone point monitor was started at 08:01pm on the 20th of March and gave values every 1'26", with a 2ppb resolution, until 02:38pm the 21st of March. The LIDAR was operated from 11:12pm on the 20th of March till 08:16pm on the 21st of March, with a 4000 shots average implying that each ozone versus height profile took 06'40". The DIAL data points are obtained with a binning of 150 meters (20x7.5 m) and we show here the first layer of the profile at 262 m above the ground level. Error bars indicate the 67% confidence intervals of the measurements. The comparison shows the same behavior for both measurements (reduction of ozone content during the night and increase during the morning) and no reduction of the ozone measurement quality during daytime. The different altitudes of the two measurements do not allow to conclude quantitatively on the accuracy of the Raman DIAL measurement.

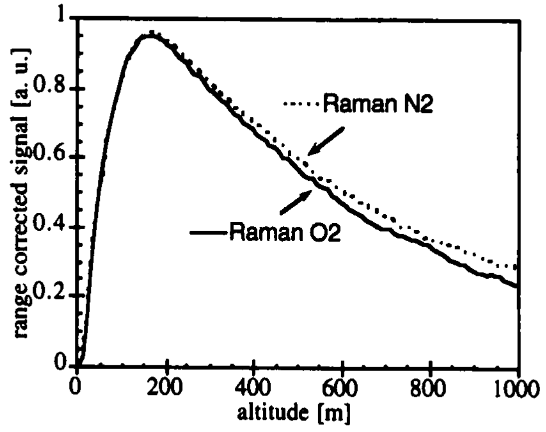

FIGURE 3 Typical range corrected Raman signal for N_2 and O_2 signals

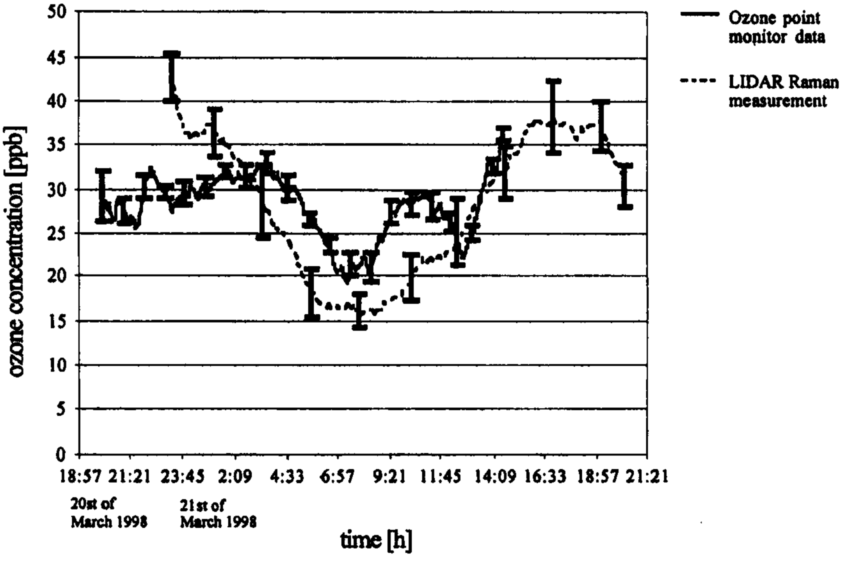

FIGURE 4 Comparison between a calibrated ozone point monitor at 12 meters above the ground level (AGL) and a LIDAR profile measured around 262m AGL on the 20 - 21 of March 1998

In order to asses the effect of some possible differences in the sensitivity (and the spatial uniformity [10] of the two PMTs, we have done some measurement using a single PMT configuration [11] in which we acquire one channel after the other, thus yielding an ozone profile every 13 minutes. The retrieved ozone values were not realistic, mainly due to fluctuation of the atmosphere on such time scale. This check will be done in the near future using a fast switch (chopper) between the channels.

CONCLUSIONS

The principle and design of a new single wavelength excitation Raman-DIAL for daytime ozone measurement in the boundary layer has been proposed. The relatively simple and cheap combination of a commercial spectrometer and holographic band pass filters in the receiving part has proved to efficiently reject background daylight and elastic scattering from the emitted laser wavelength. Comparison with an ozone point monitor shows a good correlation for the ozone retrieval, even during daytime with a short averaging time (6'40"). By using a third photomultiplier tube, we will be able to retrieve the water vapor mixing ratio simultaneously. Using the photon counting technique instead of the analog mode, we hope to extend the measurement range up to the free troposphere.

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