Preliminary results on the determination of ozone concentration in the atmosphere using the DIAL system

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Differential absorption lidar (Light Detection and Ranging) technique (DIAL) offers ways to perform measurements for the retrieval of ozone vertical profiles (concentration ppbv) up to 12 km during the nighttime and up to 8 km during the daytime. The system used for this type of retrievals is a DIAL lidar based on a Nd:YAG laser source and Raman shifting techniques to generate several wavelengths in the ultraviolet for the differential absorption technique. This paper presents the methods used in the ozone retrievals and the first profiles over the city of Bucharest.

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1. Introduction

The study of tropospheric ozone is of great importance due to the fact that it plays a key role in determining the oxidizing capacity of the lower troposphere. Here, it is the primary source of the hydroxyl radicals (OH), witch in turn are responsible for the removal of most atmospheric pollutants (by chemical interactions) [1, 2]. Extended studies of the tropospheric ozone are therefore important not only from the atmospheric chemistry perspective but also from the atmospheric models and global change perspective.

The in situ sampling of ozone from balloons and aircrafts are inadequate due to its large spatial and temporal variability in the troposphere, therefore remote sensing techniques are more appropriate. The use of differential absorption techniques (DIAL techniques) for ground based remote measurements of O3 in the troposphere permits periodic measurements above that a specific location.

Differential absorption lidars (DIAL) in UV can provide a high spatial and temporal resolution as well as continuous measurements, suitable for ozone tropospheric studies. Due to its high complexity, only some ozone DIAL systems can perform routine ozone measurements, through which the physico-chemical equilibrium of the troposphere and the stratosphere can be determined and understood [3, 4, 5].

The aim of this paper is to present the techniques used for the ozone retrievals and the preliminary results of DIAL measurements over Bucharest. Our lidar proposes the use of several on-off pairs for the extension of the dynamic range of the system. Section 2 will describe the methodology and Section 3 presents results and discussions. Finally, Section 4 summarizes our conclusions.

2. Methodology

2.1 Theoretical background

The DIAL system is based on the differential absorption of light. Basically the DIAL transmits two wavelengths: an "on-line" wavelength that is absorbed by the gas of interest (in our case ozone) and an off-line wavelength that is not absorbed. The differential absorption between the two wavelengths is a measure of the concentration of the gas as a function of range. DIALs are essentially dual-wavelength elastic backscatter lidars that transmits a pair, or several pairs of wavelengths (on and off) for the retrieval of concentration for a certain gas of interest.

If we consider the "on" and "off" received lidar signals as P_{on} and P_{off}, the concentration of gas in a range interval between R_1 and R_2 can be determined from the ratio of the lidar signals at the 'on' and 'off' wavelengths. The basic formula for concentration retrieval is:

$$N_a = \frac{1}{2\Delta\lambda}\left\{\frac{\partial}{\partial R}\ln\frac{P_{on}(R,\lambda_{on})}{P_{off}(R,\lambda_{off})} - \frac{\partial}{\partial R}\ln\frac{\beta(R,\lambda_{on})}{\beta(R,\lambda_{off})} + 2[\alpha(R,\lambda_{on}) - \alpha(R,\lambda_{off})]\right\} \quad (1)$$

From this, we can derive:

$$N_a = \frac{1}{2(R_2-R_1)(\lambda_{on}-\lambda_{off})}\ln\frac{P_{on}(R_1)\cdot P_{off}(R_2)}{P_{off}(R_1)\cdot P_{on}(R_2)} - \frac{1}{2(R_2-R_1)(\lambda_{on}-\lambda_{off})}\ln\frac{\beta_{on}(R_1)\cdot\beta_{off}(R_2)}{\beta_{off}(R_1)\cdot\beta_{on}(R_2)} - \frac{1}{\lambda_{on}-\lambda_{off}}(\lambda_{on}-\lambda_{off}) \quad (2)$$

Where σ_{on}-σ_{off} is the difference between the absorption cross section at the on and off wavelengths, σ_{on}, σ_{off} are the volume backscattering coefficients for the on and off wavelengths, P_{on}, P_{off} are the signal powers received from ranges R_i and R_j and α is the extinction coefficient [6, 7]. The average concentration can be determined by dividing N_a to the molecular number density of air. The second and third term in equation (1) is the backscatter and extinction correction. The second term can be neglected under conditions of spatially homogeneous backscatter. The third term is usually smaller than the second but must be taken in consideration under conditions of low atmospheric visibility (high extinction coefficients).

2.2 Experimental set-up

Our DIAL system uses state of the art hardware to obtain accurate ozone profiles up to 12 km during the night and up to 8 km during the day. The general specifications of the DIAL system are:

- altitude range: 1.5 - 12 km
- vertical resolution of ozone profiles: from 100 up to 750 meters
- raw data resolution: 7.5 meters
- integration time: 30 - 60 minutes
- concentration accuracy: ~in parts per billion (ppb) range

2.2.1 Emission box - laser transmitter

The emission box is equipped with a Quanta Ray Nd:YAG laser, emitting a total of 110 mJ on the forth harmonic (266 nm), at 10 Hz repetition frequency. The emitted laser beam at 266 nm pumps two low pressure Raman cells, containing H2 and D2 (Fig. 1a). The four output beams obtained through non-resonant stimulated Raman scattering (SRS) effect, have an energy varying from 10 to 40 mJ at 266 nm, 289 nm, 299 and 316 nm, respectively. These four laser beams are transmitted to the atmosphere, off-axis from the receiving telescope [8].

2.2.2 Receiving box - optical receiver

The optical receiver system is based on a Newtonian telescope, used to collect the backscattered lidar signals. This telescope has a 400 mm focal length and a 7 mm aperture. Combined with the distance between the emitting axes and the telescope axes, the optical setup of the telescope allows the complete overlap only at about 1 km (Fig. 1b).

A grating spectrometer is used to spectrally separate the four wavelengths and redirect them to the PMT's (photomultipliers). A licel TR 20 is used to process and digitize the lidar signal [8].

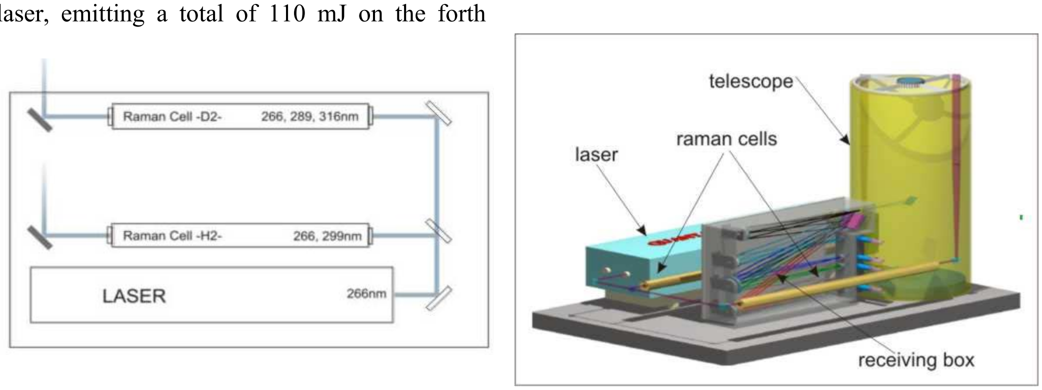

Fig. 1a, b. Emission and receiving box (a: emission: laser, optics, Raman cells, alignment optics; b: emission: laser, optics, Raman cells, alignment optics; receiving: telescope and detection box).

Due to different absorption rates (different cross sections of each wavelength), on-off pairs were used for different ranges. For the lower troposphere (1 to 3 km), the 266-289 nm pair was used, for the mid troposphere (3 to 6 km) the 289-299 nm pair was used and for higher altitudes (up to 12 km) the 299-316 nm pair was used.

Wavelengths 289 and 299 were used both as "on" and "off" channels. This was possible due to different cross section rates between the channels. First, the 289 channel was used in respect with 266 that has higher absorption rate than the first one. In the second step, the 289 channel was used in respect with 299 that has a lower absorption rate than 289. Fig. 2 shows different absorption rates between the channels used in the ozone retrieval.

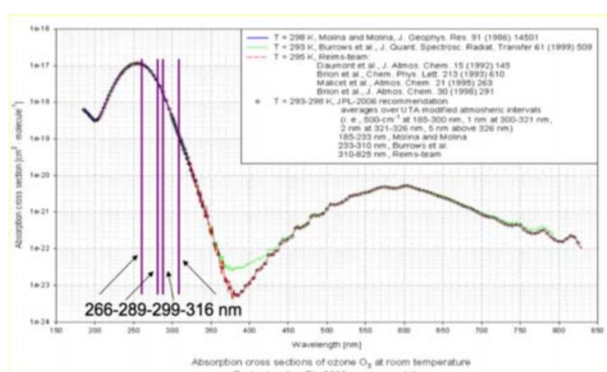

Fig. 2. Different cross sections between the channels used in DIAL retrieval.

3. Results and discussions

For ozone studies it is important to choose only low turbulence days, free of clouds or dust layers intrusions. As a case study, the 9th of June 2010 was ideal for this type of measurements. The Range Corrected Signal (RCS) for this day shows a clear day, with a well-mixed Planetary Boundary Layer (PBL), ideal for ozone investigations (see Fig. 3 a and b).

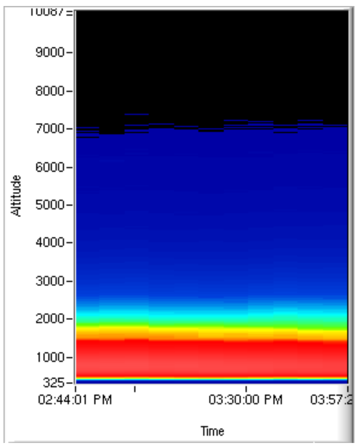

Fig. 3a. RCS at 289nm on 09.06.2010.

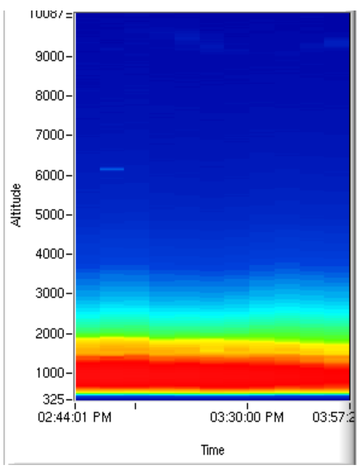

Fig. 3b. RCS at 316nm on 09.06.2010.

The difference between Fig. 3a and b above 5 km is determined by different absorption cross-sections of ozone at these specific wavelengths. The cross sections are determining the absorption rates on each specific channel. For the 289 nm channel, the cross section is two orders of magnitude larger than for the 316 nm channel (σ_{289}=1.488 10^{-18} cm^2 and σ_{316}=4.946 10^{-20} cm^2) [9].

The ozone profile for this case study was obtained by averaging one hour measurements using Eq. 2 for data processing. During the first stage, only the first term of equation 1 was used. This first term represents the pure ozone retrieval without any backscatter or absorption correction. Usually these corrections are the result of the different behavior of light (scattering and absorption properties) at different wavelengths. These differences are not encountered in the first term of Eq. 2. The backscatter and extinction correction were neglected due to the consideration that all the wavelengths used in the retrieval were relatively close to each other and the differences in behavior is negligible. Fig. 4 shows the ozone concentration obtained using all three “on”-”off” pairs. In this figure we can see a good correlation between 289-299 and 299-316 pairs between 5 and 7 km. In 266-289 we can observe a decrease in concentration after 1.5 km. This could be the effect of strong absorption rate at 266 nm (σ_{266} = 8.74 × 10^{-18} cm^2) [9]. This effect can also be observed on the corresponding RCS for 266 nm (see Fig. 5).

In the lower part of the profile, between 1 and 1.5 km (Fig. 5), we can see a significant increase of ozone concentration for the same pair (266-289 nm) which is an artifact, due to the contribution of different overlap functions at these heights. This difference in overlap functions is normal for a system that uses multiple emission axes. One can improve the assessment of ozone concentration in the lower region (up to 3 km) by determining the overlap function for the channels involved, either experimental, or theoretical, every time measurements are performed.

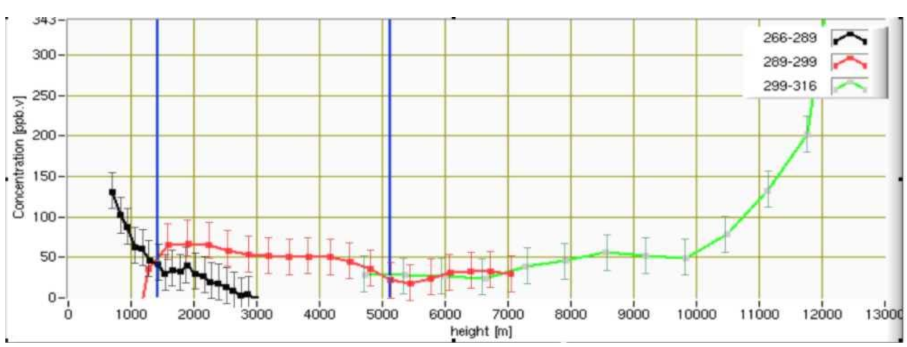

Fig. 4. Ozone profile from 09.06.2010. Overlap between different “on” - “off” pairs.

Above 10 km, we can observe a significant increase of ozone concentration. This increase is not unusual; it indicates a regular behavior of ozone in the lower stratosphere.

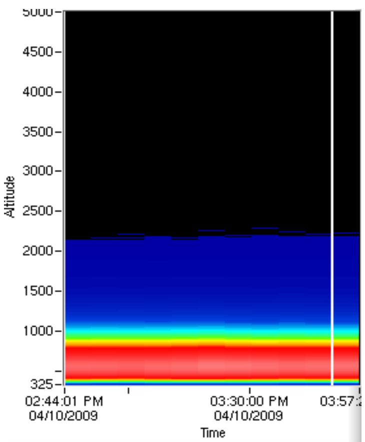

Fig. 5. RCS at 266 nm.

3.1 Error analysis

A complete error analysis can be obtained using error propagation techniques already debated in several publications [10], [11]. In short terms, one must take in consideration the statistical errors involved in dial ozone concentration retrieval as follows:

$$e_s = \frac{1}{2n\Delta\lambda_{onoff} \cdot \Delta Z N^{1/2}} \sqrt{\frac{2}{SNR_1^2} + \frac{2}{SNR_2^2}} \quad (3)$$

Where n = ozone concentration at range z, ΔZ= range resolution, Δσ = cross section difference between the on and off channels, N = number of laser shots and SNR is the signal to noise ratio for the "on" and "off" channels:

$$SNR_i = P_i / (P_i + P_{bi})^{1/2} \quad (4)$$

The P_i represents the number of photons at channel i and P_{bi} represents the background noise at channel i.

The total error is feasible to within 5% accuracy up to altitudes of 15 km for both daytime and nighttime conditions. This value can be achieved for an integration time reaching 15 min (9000 laser shots for a 10Hz repetition rate). In the PBL the accuracy falls down to 30% due to the aerosol contribution [8].

4. Conclusions

In this study we have used a DIAL system for ozone concentration retrieval for a case study June 9, 2010, measuring near Bucharest, in Magurele.

Overlapped ozone concentrations using different pairs are an indicator of good performance of the DIAL system. During this preliminary study, we have obtained a good agreement between 5 and 7km using both the 289-299 and the 299-316 pairs. The discrepancy seen between the 266-289 and 289-299 pairs, in the 1-2 km range indicates a bad performance of the system at this altitude range, probably due to optical misalignments.

For better accuracy of ozone concentration retrieval, the contribution of aerosols must be taken into consideration especially in the lower part of the troposphere (second and third term in Eq. 1).

The high values for ozone concentration near ground levels are artifacts due to both the contribution of aerosol in the PBL as well as the different overlap functions for the channels used in determination of concentration in the lower part of the troposphere.

For a better retrieval of ozone concentration at low altitudes, ways of decreasing the distance of full overlap must be found. A solution could be the determination of the overlap function either theoretical or experimental every time measurements are performed.

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