Dual pure-rotational Raman channel design and implementation in a multiwavelength lidar system for the monitoring of aerosol optical properties.

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Overview

This work aims to quantify the improvement obtained with a purely rotational Raman (PRR) channel over a vibrorotational Raman (VRR) channel used in an aerosol lidar in terms of;

- Signal-to-noise ratio (SNR),
- Effective vertical resolution,
- Absolute and relative uncertainties associated to the retrieved aerosol optical (extinction and backscatter) coefficients.

Measurements were made with the European Aerosol Research Lidar Network/Universitat Politècnica de Catalunya (EARLINET/UPC) multi-wavelength lidar system enabling a PRR channel at 353.9 nm, together with an already existing VRR (386.7 nm).

Inversions were performed with the EARLINET Single Calculus Chain (SCC). It is the first time the SCC is successfully used to invert PRR signals.



The Raman lidar technique

Consists in measuring the backscattered radiation shifted by Raman effect from an abundant atmospheric species: Nitrogen (N_2) and diatomic Oxygen (O_2), with well-defined proportion in the atmospheric composition [1,2].

The principle lies in that, for a purely molecular atmosphere, the law followed by the molecule-specific Ramanshifted radiation collected by the lidar receiver is known, as it only depends (assuming that it does not fall in the absorbing spectrum of an atmospheric gas) on the species number concentration and the molecular scattering; hence, departures from this known law can be related to the extinction introduced by the aerosols.

The most common implementation is the combination of elastic signals and their associated N₂ Vibro-Rotational Raman (VRR) signal. However, VRR technique has several disadvantages:

- The significant wavelength shift of the VRR spectrum with respect to the excitation wavelength introduces an additional source of uncertainty.
- Low differential backscatter cross-section of the VRR spectrum [3].

To overcome these limitations pure rotational Raman (PRR) scattering can be used instead VRR scattering as this technique possess:

- Small spectral separation between the elastic and PRR lines (no extra source of uncertainty).
- Larger (about two to three orders of magnitude) differential backscatter cross-section of both N₂ and O₂.

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^{3.} Veselovskii, I.; Whiteman, D.N.; Korenskiy, M.; Suvorina, A.; Perez-Ramirez, D. Use of rotational Raman measurements in multiwavelength aerosol lidar for evaluation of particle backscattering and extinction. Atmos. Meas. Tech. 2015, 8, 4111–4122.

UV-PRR channel implementation

The European Aerosol Research Lidar Network / Universitat Politècnica de Catalunya (EARLINET/UPC) multi-wavelength lidar system [4,5] was modified enabling a new UV-PRR channel at 353.9 nm. Simultaneous PRR and VRR measurements were performed in order to evaluate the daytime/nighttime performances of both channels in different conditions of aerosol



Figure 1. Zemax optical layout. FB stands for fiber bundle, L for lens, D for dichroic mirror, BS for beam splitter, EP for eyepiece, IF for interferential filter and M for mirror. Blue/red/green rays have a divergence of 15°, 0° and -15°. 15° corresponds to the numerical aperture of the fiber bundle.

The detection of both PRR and VRR signals was made via interference filters employed to select their respective portions from the UV (PRR) and visible (VRR) spectra. Specifications of the filters are shown in Table 1.



Figure 2. (a) N2 and O2 pure rotational Raman spectra (left axis) and filter transmittance (right axis) (b) N2 vibro-rotational Raman spectrum (left axis) and filter transmittance (right axis). Excitation wavelength: 354.76 nm. The spectra shown correspond to a 300 K temperature and to a 1013 hPa pressure.

Table 1. Optical filters specifications

PRR Interference filters	PRR Filters	Alluxa Custom made	CWL: 353.9 nm, FWHM: 0.8 nm
VRR Interference filter	IF1	Barr Custom made	CWL: 386.7 nm, FWHM: 3 nm

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Results

Two nighttime and one daytime measurements are analyzed in this work. Specifications of the measurements as well as atmospheric parameters are shown in the Table 2.

AERONET measurements are level 1.5.						
Case	Units	N1	N2	D1		
Conditions		Nighttime	Nighttime	Daytime		
Date		11/3/2020	11/3/2020	21/3/2020		
Start time	UT	18:57	18:57	12:46		
Temporal resolution	Hours	3	1	3		
Nearest AERONET						
Time	UT	17:05	17:05	12:59		
AOD_{440}		0.15	0.15	0.26		
$AE_{440-675}$		1.27	1.27	0.71		
Probable airmass origin		Local	Local	Local, dust in the FT		

Table 2. Main characteristics of the measurements used in this work. TheAERONET measurements are level 1.5.

The first nighttime measurement (N1) lasted for 3 h. The second nighttime study case (N2) corresponds to the first hour from N1. This 1-h-averaged measurement was chosen to examine the effect of temporal resolution on the quality of the retrievals. The daytime measurement (D1) lasted for 3 h as well.

In N1 and N2 aerosols were of local origin and accumulated mostly in the planetary boundary layer: AOD_{440} is equal to 0.15 and $AE_{440-675}$ to 1.27. In D1 most of the locally originated aerosols are in the PBL; mineral dust particles are present in the free troposphere up to 4.5–5 km. This situation is associated with a higher AOD_{440} (0.26) and a low $AE_{440-675}$ (0.71).



Signal-to-noise ratio

To get a quantitative comparison between PRR and VRR preprocessed (glued) signals Signal-to-noise ratio (SNR) was calculated computing estimators of the mean value μ and the standard deviation σ over a 9-samples continuous sliding interval of the signal. SNR is estimated as μ/σ . To smooth the SNR calculation, a model was fitted to the σ estimator [6,7].



Continuous lines plotted in the Figure 3 show PRR-SNR, dotted lines correspond to VRR-SNR.

When using PRR instead of VRR, the measurements show a gain in SNR of a factor 2.8 and about 7.6 for 3-h nighttime and daytime measurements, respectively.

PRR enhancement is quite noticeable in D1 study case, in which background noise contribution is of consideration, SNR for the PRR channel shows quite stable with no high fluctuations as for the VRR channel.



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Retrieved products: aerosol extinction and backscatter coefficients

The retrieval of the aerosol extinction and backscatter coefficients combining PRR and VRR signals with elastic signals has been performed by using the Single Calculus Chain (SCC) from the European Aerosol Research Lidar Network (EARLINET) [8–10]. It is the first time that the SCC is used to retrieve aerosol optical products from a combination of PRR and elastic signals. The optical processor module of the SCC is called EARLINET Lidar Data Analyzer (ELDA). ELDA was configured to retrieve backscatter and extinction coefficients at the same effective vertical resolution (product lidar ratio and extinction) using the Raman method [11].

Figure 4. Aerosol backscatter and extinction coefficients, lidar ratio and effective vertical resolution from PRR and VRR signals for case (a) N1, (b) N2 and (c) D1. On the plot of the vertical resolution, the ratio of the resolutions PRR to VRR is reported (black line, top axis). Time-height plots of the range-square corrected signal (in arbitrary units) are reported in the far-right plot. In (a) and (b) the black horizontal dashed line represents the lowest height for which the extinction coefficient is below the detection limit Δ_{DL} . In (c) it represents the highest height for which extinction coefficient from VRR is retrieved. The legend in the first plot of Figure 4a applies to all plots.



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Discussion

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To comprehend PRR improvement over VRR associated to the retrieved aerosol optical coefficients three key parameters were analyzed:

- Effective vertical resolution (EVR, associated to the ٠ averaging needed for the estimation of the final products),
- Absolute uncertainties ($\sigma_{\alpha aer}$), ٠
- Relative uncertainties ($\sigma_{\alpha a er} / \alpha_{a er}$).

For comparison purposes, the extinction coefficient is the chosen product for the analysis, as it is the only Raman product that depends only on the Raman signal (Backscatter coefficient calculation channel requires extra calibration based on the extinction retrieval).

Figure 5 shows histograms with layer-mean values of EVR, $\sigma_{\alpha a e r}$ and $\sigma_{\alpha a e r} / \alpha_{a e r}$ of both PRR and VRR retrievals in the main surface aerosol layer, below the detection limit ($\alpha_{aer} > \Delta_{DL}$).



Figure 5. Comparative histograms of layer-mean values of the (a) vertical resolution, (b) absolute uncertainty and (c) relative uncertainty in the main surface aerosol layer ($\alpha_{aer} > \Delta_{DL}$) retrieved by the SCC from PRR and VRR signals for cases N1, N2 and D1. Black plus signs (right axis) are the ratio PRR to VRR. For layer-mean values exceeding the selected vertical scale the numerical value is reported in black on the top of the bar.

For nighttime measurements the effective vertical resolution is reduced abot 17%, for daytime is reduced about 20%. The absolute uncertainty is more than double for cases N1 and N2, and much larger for D1, divided by a factor about 10. The corresponding relative uncertainties are 24.2, 19.3 and 94.0% for N1, N2 and D1 respectively. The highest reduction occurs for the daytime case.



Conclusions

A pure rotational Raman channel at 353.9 nm has been implemented in the EARLINET/UPC multi-wavelength lidar system. This new channel detects backscattered signals produced by the PRR effect of atmospheric N2 and O2 excited by the emission wavelength of 354.7 nm. Spectral filtering was obtained by cascading two extremely narrow and steep interference filters with an equivalent (i.e., for the two filters in cascade) CWL at 353.9 nm, FWHM < 0.8 nm, transmission at peak > 70% and OD8 at 354.7 nm.

PRR and elastic signals have been inverted for the first time with the EARLINET Single Calculus Chain and profiles of aerosol extinction and backscatter coefficients have been retrieved successfully in daytime and nighttime conditions.

To fully quantify the improvement of pure rotational over vibro-rotational Raman signals, simultaneous measurements at the VRR wavelength of 386.7 nm were performed. Two cases were taken with different aerosol loads and vertical structures. The signal-to-noise ratio was found in agreement with the theory: an increase of a factor 2.8 and ~7.6 was observed for 3-h nighttime and daytime measurements, respectively, when using PRR vs. VRR. Improvements in terms of retrieved optical properties are measured in terms of the reduction of the effective vertical resolution, EVR, and of the uncertainties (absolute, $\sigma \alpha aer$, and relative, $\sigma \alpha aer/\alpha aer$) associated to the extinction coefficient in the main aerosol surface layer when using PRR retrievals vs. VRR ones. For long (3 h), nighttime measurements EVR is reduced by 17%, $\sigma \alpha aer$ is divided by 2 and and $\sigma \alpha aer/\alpha aer$ is divided by 3 when using PRR instead of VRR. During daytime and also 3-h measurements EVR is reduced by 20%, $\sigma \alpha aer$ is divided by 10 and $\sigma \alpha aer/\alpha aer by 7$ when using PRR instead of VRR. In the daytime case, the VRR extinction coefficient is retrieved in a limited height range (<2.2 km). This prevents the SCC from finding a suitable calibration range in the search height range of 4–8 km for the retrieval of the backscatter coefficient, so the advantage of using PRR instead of VRR is particularly evident in daytime conditions. For short (1 h), nighttime measurements EVR is reduced by a little more than 2 and $\sigma \alpha aer/\alpha aer$ is divided by a little less than 2 when using PRR instead of VRR.

To get a full description of this work you may check:

Zenteno-Hernández, J.A.; Comerón, A.; Rodríguez-Gómez, A.; Muñoz-Porcar, C.; D'Amico, G.; Sicard, M. A Comparative Analysis of Aerosol Optical Coefficients and Their Associated Errors Retrieved from Pure-Rotational and Vibro-Rotational Raman Lidar Signals. Sensors 2021, 21, 1277. https://doi.org/10.3390/s21041277



Future work

The next stage for this work is to enable a PRR channel at 530.2 nm in the visible sub-section of the EARLINET/UPC lidar system. Following the same methodology as with the previous UV-PRR channel.

A first theoretical analysis has shown that the effective PRR Raman spectra is about 2 orders of magnitude larger than VRR. With this in mind, and as the work this far suggests, PRR channel implementation is expected to provide higher SNR for the received signals as well as low absolute and relative uncertainties for the retrieved products ($\alpha \ll \beta$) in comparison with the current VRR channel at 607 nm.



Figure 6. Pure rotational and vibro-rotational Raman spectra for N₂ and O₂. Excitation wavelength: 532.14 nm, T=300 K, P=1013 hPa.



