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Characterization of Particle Hygroscopicity by Raman Lidar: Selected Case Studies from the Convective and Orographically-Induced Precipitation Study

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Abstract. The characterization of particle hygroscopicity has primary importance for climate monitoring and prediction. Model studies have demonstrated that relative humidity (RH) has a critical influence on aerosol climate forcing. Hygroscopic properties of aerosols influence particle size distribution and refractive index and hence their radiative effects. Aerosol particles tend to grow at large relative humidity values as a result of their hygroscopicity. Raman lidars with aerosol, water vapor and temperature measurement capability are potentially attractive tools for studying aerosol hygroscopicity as in fact they can provide continuous altitude-resolved measurements of particle optical, size and microphysical properties, as well as relative humidity, without perturbing the aerosols or their environment. Specifically, the University of Basilicata Raman lidar system (BASIL) considered for the present study, has the capability to perform all-lidar measurements of relative humidity based on the application of both the rotational and the vibrational Raman lidar techniques in the UV. BASIL was operational in Achern (Black Forest, Lat: 48.64° N, Long: 8.06° E, Elev.: 140 m) between 25 May and 30 August 2007 in the framework of the Convective and Orographically-induced Precipitation Study (COPS). The present analysis is focused on selected case studies characterized by the presence of different aerosol types with different hygroscopic behavior. The observed behavior, dependent upon aerosol composition, may range from hygrophobic to strongly hygroscopic.

Keywords: Hygroscopicity, Particle, Raman, Lidar.

PACS: 92.60.H; 92.60.Jq

INTRODUCTION

The characterization of particle hygroscopicity has a primary role in climate monitoring and weather forecast. Hygroscopic properties of aerosols, their size distribution and composition may alter the Earth's radiative budget [1]. Model studies have demonstrated that relative humidity has a critical influence on aerosol climate forcing [2]. At the moment the dependence of aerosol particles' hygroscopic growth on relative humidity and particle chemical composition is poorly understood. The swelling of hygroscopic aerosol particles at large relative humidity values has important implications in terms of aerosol direct effect on climate [3].

Lidar systems are efficient tools for the characterization of particle hygroscopicity because of their capability to characterize aerosol optical properties and atmospheric humidity (relative humidity) within the same atmospheric column. Specifically, the University of Basilicata Raman lidar system (BASIL) considered in the present study has the capacity to perform all-lidar measurements of relative humidity based on the combined application of the rotational and the vibrational Raman lidar techniques in the UV.

Particle backscatter may increase as a result of particle hygroscopic swelling at great relative humidity values. Few attempts have been performed to measure particle backscatter or extinction over a large range of relative humidities. Reference [4] and [5] measured particle backscatter as a function of relative humidity for values not exceeding 80%. Reference 3 reported measurements of particle backscatter at relative humidity levels close to 100%, revealing a strong hygroscopic growth of aerosol particles when relative humidity values exceed 80-85 %. In [3] a water vapor differential absorption lidar (DIAL) system was used to study water vapor uptake by aerosols, considering a methodology applicable only in cloudy conditions and in presence of an adiabatic convective boundary layer. The present document considers the application of a Raman lidar system which includes water vapor and temperature measurement capability, thus allowing simultaneous measurements of relative humidity and aerosol optical properties in any atmospheric circumstance.

LIDAR SYSTEM AND RESULTS

The University of Basilicata Raman Lidar system (BASIL), which is used in the present research effort, has the ability to provide simultaneous multi-wavelength particle backscatter, extinction and depolarization measurements, together with measurements of the water vapor mixing ratio and temperature profiles [6],[7],[8]. The measurement of these two latter parameters translates into the capability to perform all-lidar measurements of relative humidity. A description of the procedure used to determine RH from Raman lidar measurements is provided in [9], together with an estimate of the uncertainties affecting this measurement. The application of Raman lidars allows to measure atmospheric humidity close to saturation conditions. BASIL can also provide continuous altitude-resolved measurements of particle optical, size and microphysical properties without perturbing the aerosols or their environment. BASIL was operational in Achern in the frame of the Convective and Orographically-induced Precipitation Study [10–13]. During COPS, BASIL collected more than 500 hours of measurements, distributed over 58 measurement days and 34 intensive observation periods (IOPs). In the present work we focus our interest on particle backscattering measurements at 355 and 532 nm to reveal aerosol hygroscopic behavior and quantify it at variable relative humidity values.

Fig. 1 illustrates the time evolution of the particle backscattering coefficient at 532 nm, $\beta_{532}(z)$, from 07:00 UTC on 30 July 2007 to 20:00 UTC on 30 July 2007. Data points in this figure and in the forthcoming figures are characterized by a time and vertical resolution of 5min and 15 m, respectively.

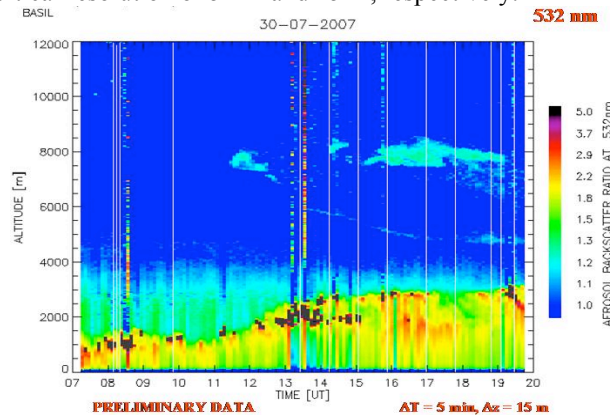


FIGURE 1. Time evolution of the particle backscattering ratio at 532 nm has measured by BASIL from 07:00 to 20:00 UT on 30 July 2007. The figure clearly indicates the presence of stratiform clouds which are visible above 1800 m (black features) during the time interval 13:00-15:00 UTC.

Fig. 2 illustrates the scatter-plot of $\beta_{532}(z)$ versus RH for the time interval 13:00-15:00 UTC. We focused our attention on the vertical region 500-1800 m below the clouds. Data points below 500 m were not included in the plot because being affected by a bias associated with overlap issues (partial superimposition between the laser beam and the telescope field-of-view). For the purpose of this preliminary study we are considering RH values obtained from the radiosonde launched from Supersite R at 14:01 UTC. The figure reveals a monotonic (smoothly varying) variability of $\beta_{532}(z)$ versus RH. $\beta_{532}(z)$ is found to undergo an increase of approximately 86 % from a values of $8.6 \times 10^{-7} \text{ m}^{-1} \text{ sr}^{-1}$ at 50 % to a value of $1,6 \times 10^{-6} \text{ m}^{-1} \text{ sr}^{-1}$ at 90 %.

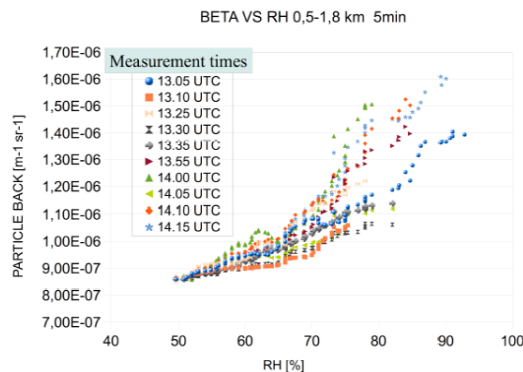


FIGURE 2. Scatter-plot of $\beta_{532}(z)$ versus RH for 30 July 2007.

A monotonic (smoothly varying) hygroscopic behavior was also reported by Pan et al. [14] in a study focusing on a rural area near Beijing. Pan et al. [14] reveal a slowly monotonic increase in the scattering coefficient with increasing RH and no evidence of deliquescence. This kind of behavior was conjectured by these authors to be ascribable to the presence of urban/anthropogenic polluted conditions. Pan et al. [14] reported an aerosol hygroscopic growth factor $f(RH=80\%)$ of 1.57, which was conjectured in inverse proportion to hydrophobic organic carbon matter.

We, in addition, focused our attention on the case study of 1-2 August 2007 data. Fig. 3 shows the values of the particle backscattering coefficient at 355 nm, $\beta_{355}(z)$ versus the RH for the time interval 22:00-23:00 UTC and between the vertical interval 3-4.5 km a.g.l. Data are characterized by a time and vertical resolution respectively of 2 min and 120 m and the number of points plotted is approximately 600. Fig. 3 reveals a marked increase in particle backscattering when RH exceeds 75%, which testifies the swelling tendency of the observed aerosol particles for RH values exceeding the deliquescent point. This trend is compatible with partially soluble aerosol particles characterized by a deliquescent growth. Data points for $RH < 75\%$ and for $RH > 75\%$ are fitted with two distinct straight lines having different slopes, $c < 75\%$ and $c > 75\%$, respectively, with $c > 75\%$ being $1.3 \times 10^{-7} \text{ m}^{-1} \text{ sr}^{-1} \text{ \%}^{-1}$, i.e. approximately twelve times larger than $c < 75\%$ ($1.1 \times 10^{-8} \text{ m}^{-1} \text{ sr}^{-1} \text{ \%}^{-1}$).

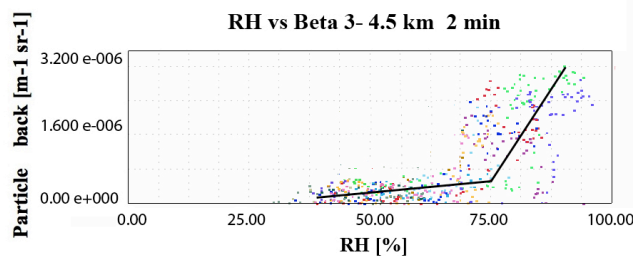


FIGURE 3. Scatter-plot of $\beta_{355}(z)$ versus RH for 1-2 August 2007.

Data points in Fig. 3 allow to also compute the aerosol growth factor [1]:

$$f(RH)_\beta = \frac{\beta(RH)}{\beta(RH_{ref})} \quad (1)$$

which expresses the backscatter enhancement in terms of lidar backscatter relative to that at a lower reference value of RH, RH_{ref} , (for example the deliquescent point). Values of $f(RH)$ are as large as 2 for $RH=85\%$, in agreement with values reported by Feingold et al.[15] and Pahlow et al. [1].

Our attention was lastly focused on the case study of 4 June 2007. In Fig.4 we illustrate the variability of the particle backscattering coefficient at 355 nm, $\beta_{355}(z)$, as a function of relative humidity in the height area 1.7-3.7 km and for the time period 17:38-18:38 UTC. The figure, likewise the one for 30 July, shows a monotonous trend in the hygroscopic behavior. A slow increase of backscatter with increasing relative humidity is present, with no evidence of deliquescence.

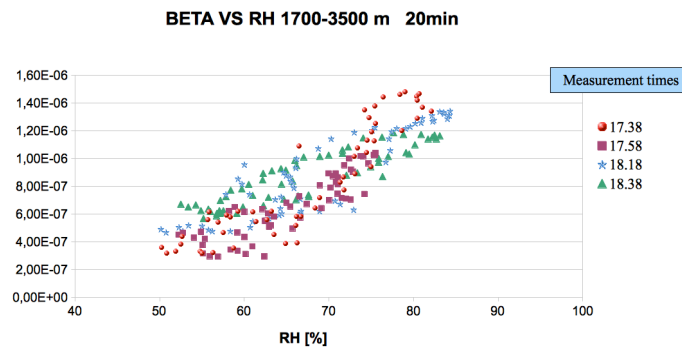


FIGURE 4. Scatter-plot of $\beta_{355}(z)$ versus RH for 4 June 2007.

For the above considered case studies we intend to extend the analysis in the future to other aerosol optical properties (as the backscattering at 1064 nm and the particle extinction coefficient at 355 and 532 nm).

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