Abstract— Water vapor is an important greenhouse gas due to its high concentration in the atmosphere (parts per thousand) and its interaction with tropospheric aerosols particles. The upward convection of water vapor and aerosols due to intense heating of the ground leads to aggregation of water particles or ice on aerosols in the air forming different types of clouds at various altitudes. The condensation of water vapor on aerosols is affecting their size, shape, refractive index and chemical composition. The warming or cooling effect of the clouds hence formed are both possible depending on the cloud location, cover, composition and structure. The effect of these clouds on radiative global forcing and therefore on the short and long term global climate is of high interest in the scientific world. A major interest is manifested in obtaining accurate vertical water vapor profiles simultaneously with aerosol extinction and backscatter in the meteorological and remote sensing fields all around the globe in an effort to understand the hygroscopic properties of aerosols.

In previous work, simultaneous measurements of RH with backscatter measurements from a surface nephelometer were used to probe the hygroscopic properties of aerosols. However, most of these measurements were not able to probe the high RH domain since such high RH is rare for surface altitudes. For this reason, experiments using a 355nm raman water vapor and aerosol lidar at the ARM site were used. Capable of providing simultaneous backscatter and RH profiles, and performing the experiments under low altitude cloud decks insured stable well mixed layers as well as probing RH profiles to above 95% which is required for the differentiation of different aerosol hygroscopic models.

Keywords—elastic-Raman lidar, aerosol hygroscopicity, backscatter

I. INTRODUCTION

In this paper, we explore the nature and variability of urban aerosol hygroscopic properties using multi-wavelength Raman lidar measurements at 355nm, as well as backscatter measurements at 532nm and 1064nm. The addition of the longer wavelength channels allows us to more accurately validate the homogeneity of the aerosol layer as well as provide additional multiwavelength information that can be used to validate and modify the aerosol models underlying the hygroscopic trends observed in the Raman Channel. In particular, we show that aerosols backtracked to different sources (bio-mass, maritime due to sea breeze) have different hygroscopic behavior which are compared to theoretical models.

In addition, in support of our hygroscopic measurements, we also discuss our calibration procedures for both the aerosol and water vapor profiles. The calibration algorithm we ultimately use for the water vapor measurements are twilight measurements where water vapor radiosonde data from the OKX station (Brookhaven laboratory) in NYS, are combined with total water vapor obtained from a GPS MET station. These sounds are then time correlated with independent near surface RH measurements to address any bias issues that may occur due to imperfect calibration due to lidar overlap issues and SNR limitations in seeing the water vapor at high altitudes. The results of our calibrations are then presented.

II. METHODOLOGY

A. Elastic-Raman Lidar

The chemical property of water vapor of condensation different types of aerosols found in the atmosphere is changing the chemical composition, shape and size of this particles, and at the same time it affects their optical properties and the direct radiative forcing [1]. High Cirrus clouds and contrails created by airplanes are formed from amalgams of water or ice crystals suspended in air cooled below the saturation temperature, allowing the moisture to form into small drops on microscopic particles called condensation nuclei that can be water molecules (homogeneous nucleation) or other metallic nitrogen oxides, carbon dioxide etc (heterogeneous formation of aerosols)[2]. While the impact of high altitude clouds on global warming is larger than their cooling effect (long waves albedo is 0.4-0.7), the lower altitude clouds in change have an overall cooling effect [1],[3]. Therefore good measurements of water vapor in the atmosphere (troposphere) are necessary. One of the methods of monitoring the water vapor mixing ratio in the atmosphere is by using lidar systems. The possible lidar interactions are split in two big mechanisms the elastic and inelastic ones. The elastic mechanism can be described by the Rayleigh and Mie scattering theories.

The Rayleigh scattering is relevant for molecular gases including N₂O₅ where the diameter is much smaller than the transmitted lidar radiation and can be defined by Measures (1992) as “Laser radiation elastically scattered from atoms or molecules is observed with no change of frequency”. The Mie scattering theory is for particulates (spherical) where the
diameter is proportional to the laser wavelength and is defined in
the same book as “Laser radiation elastically scattered from
small particulates or aerosols (of size comparable to
wavelength of radiation) is observed with no change in
frequency. There is no wavelength change in either mechanism
described above. The elastic lidar equation is written as:

\[ P_L(z) = \frac{K_s(\lambda_g, z)}{z^2} n_e(z) \frac{\pi \sigma(\lambda_g, z)}{d\Omega} \]

\[ \star \exp \left[ -2 \int_0^z [\alpha_{\lambda_g}(z) + \alpha_{\lambda_e}(z)] dz \right] + b_{\lambda_e}(z) \]

where \( P_L \) is the detected Raman lidar backscatter signal

The second interaction mechanism where the “Laser
radiation inelastically scattered from molecules is observed
with a frequency shift characteristic of the molecule (hv - hν =
E)” is called Raman Scattering\[1\]. The considered Raman
transition is the radiation generated at longer wavelength from
excitation (Stokes or red shift)\[2\]. In other words when a
substance is subjected to an incident exciting wavelength, it
exhibits the Raman effect. It reemits secondary light at
wavelengths that are shifted from the incident radiation.
The magnitude of the shift is unique to the scattering molecule,
while the intensity of the Raman band is proportional to the
molecular number density.

The method of monitoring the water vapor mixing ratio
in the atmosphere by use of lidar is the detection of shifted
radiation defined above as the Raman effect. The water vapor
Raman lidar technique uses the ratio of rotational-vibrational
Raman scattering intensities from water vapor and nitrogen
molecules, which is a direct measurement of the atmospheric
water vapor mixing ratio. The backscatter radiation detected by
the lidar is expressed as a function of range.

The water vapor and nitrogen Raman signals (inelastic lidar
equation) are given by:

\[ P_R(z) = \frac{K_s(\lambda_g, z)}{z^2} n_e(z) \frac{\pi \sigma(\lambda_g, \lambda_e, z)}{d\Omega} \]

\[ \star \exp \left[ -\int_0^z [\alpha_{\lambda_g}(z) + \alpha_{\lambda_e}(z)] dz \right] + b_{\lambda_e}(z) \]

where \( P_R \) is the detected Raman lidar backscatter signal
corresponding to nitrogen and water vapor, \( K_s \) is a system
function that is dependent on the optical transmission and
detector efficiency, the overlap function and the area of the
collector mirror of the telescope, \( n_e \) is the number density of
Raman active molecule, \( \frac{\pi \sigma(\lambda_g, \lambda_e, z)}{d\Omega} \) is the differential
Raman backscatter cross-section, \( \alpha_{\lambda_g}(z) \) is the aerosol and
molecular extinction coefficient at the laser wavelength and
and \( \alpha_{\lambda_e}(z) \) is the aerosol and molecular extinction coefficient
at the Raman wavelength and \( b_{\lambda_e}(z) \) is the background
signal.

B. Calibration and water vapor measurements

By taking the ratio of the two signals one can obtain the
water vapor mixing ratio profile, equation 3.

\[ R_{H,O}(z) = \frac{K_{H,O}}{K_{N,O}} \left[ \frac{\frac{d\sigma}{d\Omega}}{T_{H,O}} \right] \frac{P_{H,O}}{P_{N,O}} \]

where \( T \) is the transmission for the two wavelengths through
the atmosphere and \( M \) represents the mass of water vapor in
atmosphere and dry air. The water vapor profile may be
retrieved as expressed in equation 3.

\[ q_{H,O}(z) = \frac{K_{H,O}}{K_{N,O}} \left[ \frac{\frac{d\sigma}{d\Omega}}{T_{H,O}} \right] \frac{P_{H,O}}{P_{N,O}} \]

where \( q \) is the water vapor mixing ratio in \([g/Kg\text{ dry air}]\) or in
[ppmv]. Hence \( q_{H,O}(z) = C \Gamma \star \left[ \frac{S_{H,O}}{S_{N,O}} \right] \)

where \( \Gamma = \frac{T_{N,O}}{T_{H,O}} \approx 1 \) and \( C \) is the calibration function. Due to
little aerosol influence on the 407nm and 387nm Raman
channels this technique is relatively free of systematic errors
(aerosol effects), can be operated from the ground (the
radiation is not absorbed by the water vapor itself) and its use is
relatively simple. Its main limitation is the relatively small
backscatter Raman cross-sections, that result in weak Raman
signals compared with the elastic backscatters or the electronic
noise induced in daytime by solar radiation.

As mentioned above one might observe that the method
requires the use of a calibration constant to be used in the
retrieval

\[ C = \frac{K_{H,O}}{K_{N,O}} \left[ \frac{\frac{d\sigma}{d\Omega}}{T_{H,O}} \right] \frac{P_{H,O}}{P_{N,O}} \]

The calibration methods used in this paper are presented in
detail in [5].
III. ANALYSES AND RESULTS

The analyses of the hygroscopic properties of the aerosols have been done for the day of September 25, 2006 due to the low cloud presence in the planetary boundary layer and increasing relative humidity with height. The lidar image of the day has been plotted in figure one.

To have a better understanding of the vertical profiles of the lidar returns fig. 2 is plotting the five channels returns versus altitude at the different wavelengths. The profiles have been averaged between 11:35 and 12:35 of the above mentioned day.

The relative humidity retrieved from the lidar water vapor channel and radiosonde temperature profile [6] and calibrated by methods described in section II B. The local surface measurements obtained from a GPS instrument confirm the 39%-40% relative humidity at the ground.

By performing ratios of the normalized backscatter ratio the number concentration of particles in the atmosphere is cancelled as one can observe from the formula (7). Therefore, we may perform vertical profile analysis on the scattering ratios without concern for the actual number density of the scattering objects in the backscattering ratio dependence of relative humidity. Using algorithms to determine the backscatter at 355nm and 1064nm [4-6] one can obtain the ratio of the two obtained backscattered profiles at the two wavelengths as described by (7). Several attempts have been done to determine different models of extinction or backscatter dependence on relative humidity [9-11] but none of them had
the advantage of using the two simultaneous wavelength measurements as a quantification of the microphysical properties of aerosol particles.

After simplifications the above expression can lead to the simple equation (8)

\[
\frac{\sigma_s(RH)}{\sigma_s(0)} \frac{355nm}{1064nm} = \left[ \int \frac{Q}{\lambda} \frac{2\pi r}{m} dr \right]_{355}^{1064}
\]

(8)

The normalization was performed with respect to the dry particles at ~60% relative humidity due to the high uncertainties in the low altitude lidar measurements caused by the overlap function.

As it can be observed for given wavelengths and known backscattering ratio respectively extinction ratio one may retrieve the particle radius and refractive indexes of particles by simply solving the system of two equations and two unknowns. As well, one could assume known complex refractive indexes of different dry particles that could be present in the urban atmosphere like sulfates (\((NH_4)_2SO_4\) or NaCl and use only the overlap function.

As it can be observed for given wavelengths and known backscattering ratio respectively extinction ratio one may retrieve the particle radius and refractive indexes of particles by simply solving the system of two equations and two unknowns. As well, one could assume known complex refractive indexes of different dry particles that could be present in the urban atmosphere like sulfates \((NH_4)_2SO_4\) or NaCl and use only the backscatter ratio to determine the aerosol particle size. The normalized scattering of the hygroscopically grown particles with respect to the scattering of the dry particles is then just a function of relative humidity (measurable), linear mass increase coefficient real and imaginary components of the refractive indexes, and molecular masses of the pure water and dry particles known from existing models [11].

In figure below the normalized backscatter ratios of the 355nm backscattering to the 1064nm backscattering have been plotted for models 1,3,5,6 [11] and CCNY model retrieved from the vertical profiles measured by the multiwavelength Raman lidar. Models 1,3,5,6 are represented by aerosol collected during summer at Mainz in 1966, maritime aerosol collected during 13-16th April 1969, urban aerosol at Mainz, in January 1970 and respectively aerosol collected at the foothills of Hohenpeissenberg (1000m altitude).

IV. Conclusions

The results of the model analysis taken at wavelengths 300nm and 1064nm (those nearest our experimental arrangement) indicate a clear dependence of the backscattering and total geometric cross section on relative humidity which we observe to be in qualitative agree. More mathematical investigations of the new proposed analyses should be performed on the lidar data to determine the degree of dependency of the retrievals on different microphysical properties of aerosol. Therefore the new proposed

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Fig 5. Normalized aerosol backscatter ratio of the 355nm and 1064nm lidar returns