P1.1 QUANTIFYING THE EFFECT OF HUMIDITY ON AEROSOL SCATTERING WITH A RAMAN LIDAR

Raymond Rogers*, Kevin McCann, and Raymond Hoff
University of Maryland Baltimore County, Baltimore, Maryland.

1. INTRODUCTION

Aerosols play a large role in contributing to climate change, however the aerosol indirect effect is poorly understood at best. Understanding how the optical properties of aerosols vary with changing relative humidity is an important factor in gaining knowledge of this effect (Charlson, 1992, Tang, 1996). As aerosols particles take up water vapor in an environment of changing relative humidity, depending on the aerosol composition, their radii will increase and their index of refraction will decrease towards that of water.

At the present there have been several theoretical and experimental investigations into hygroscopic effects on aerosol optical properties. Typically humidified nephelometers or humidified tandem differential mobility analyzers are used to make these measurements. These instruments are unable, however, to give accurate results above 85% relative humidity (Wulfmeyer, 2000). To this end, several elastic, differential, and Raman lidar studies have been used to observe hygroscopic growth of aerosols (Ferrare, 1997, Feingold, 2000). The lidar method has several advantages over other measurements of the optical properties as a function of relative humidity. A lidar is able to remotely probe the atmosphere at ambient relative humidity, not requiring instrumentation to control air samples at different relative humidities. In this way, a lidar is able to obtain aerosol growth in the actual atmosphere rather than in the lab. Another advantage of this method is that a Raman lidar is able to detect relative humidities nearing 100% with errors less than 20% in the daytime (Ansmann, 1992), giving growth curves up to much higher relative humidity than other experimental methods.

This study illustrates the use of a Raman lidar, the Atmospheric Lidar EXperiment (ALEX), to measure the hygroscopic properties of an aerosol mass found over Baltimore, MD during the summer of 2004. In particular, daytime measurements will be examined. The measurements are compared to Mie code simulation of aerosol growth with relative humidity. Additionally, the data are used to determine the hygroscopic growth factor, defined by Charlson (1992) as ratio of the light scattering coefficient at 80% relative humidity to the light scattering coefficient at 30% relative humidity. The hygroscopic growth factor has been identified as a key factor in the aerosol indirect effect.

2. MEASUREMENTS

2.1 *Lidar measurements*

The ALEX system is a three channel Raman lidar transmitting at 355 nm, capable of making measurements of backscatter, extinction, and water vapor mixing ratio as functions of height. The vertical resolution of the measurements is 30 m. When combined with a temperature profile from a radiosonde, profiles of relative humidity are obtained from the water vapor mixing ratio data.

The extinction and relative humidity profiles may be directly compared to obtain a growth curve on days where the water vapor mixing ratio was constant with height and the lapse rate is unstable. These conditions indicate that the boundary layer is well-mixed, thus any change in extinction must be due to

* *Corresponding author address:* Raymond Rogers, 1000 Hilltop Circle, University of Maryland, Baltimore County, Room 220, Baltimore, MD 21250; email: rrogers@umbc.edu

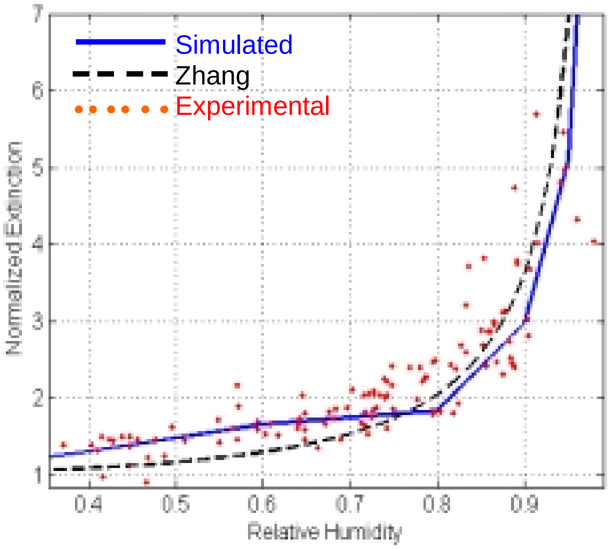

Figure 1. Calculated (blue), Zhang (black), and Experimental (red) curves of extinction, normalized to extinction at 30 % relative humidity, as functions of relative humidity. These curves are all normalized to a dry value of extinction.

a decreasing temperature causing an increase in relative humidity.

The data shown in Figure 1 are a combination of the extinction profiles and the relative humidity profiles from the ALEX system on June 24, 2004. On this day, the boundary layer was determined to be well-mixed based on the criteria discussed above. A total of nine five-minute averaged profiles, ranging in height between 1 km and 2 km, were used in this example. The extinction data are normalized to a "dry" extinction value of 0.15 km^{-1} in order to compare to the Mie simulations.

2.2 *Mie simulation*

The Mie theory routines, described in Bohren and Huffman (1983) and coded in Matlab by Matzler (2002) were used to compute the light scattering coefficients. In conjunction with the Mie simulation, a growth model described by Hanel (1976) was used to obtain the light scattering coefficients as functions of relative humidity.

In the model, the aerosol radius distribution, obtained from AERONET (Holben, 1998), was assumed to grow cubically with relative humidity. An onsite IMPROVE sampler (Sisler, 2000) indicated that the majority of the aerosol mass on the 24^{th} was sulphate. Based on this observation, a dry index of refraction of 1.54 was assumed, with some absorption included in the model by setting the imaginary index of refraction to 0.05 (Hanel, 1976). Similar studies varied the index of refraction cubically (Hanel, 1976, Hoff 1996) with relative humidity to take into account that the aerosols hydrate. At 99% relative humidity, the scattering medium was assumed to be primarily water, with an index of refraction of 1.34. The Mie theory results are shown in Figure 1.

3. RESULTS AND DISCUSSION

The dependence of the light scattering coefficient on relative humidity as found by simulation and experimental data is shown in Figure 1. There is good agreement with the simulated growth curve (blue) and the measured data (red). Also shown in Figure 1 is an empirical relationship by Zhang (1994), in good agreement at higher relative humidities. This curve lies slightly below the other curves because Zhang's relationship is normalized to unity at zero relative humidity, while the Mie simulated curve is normalized to 30% relative humidity.

The observed data can also be used to estimate a value of the hygroscopic growth factor. These data imply a hygroscopic factor of 2.1, slightly higher than Charlson's value of 1.7 ± 0.3. This may be because the observed data do not extend down to 30 % relative humidity; they were normalized to an extinction value at a higher relative humidity.

4. CONCLUSIONS

A Raman lidar has been used to measure the modification of the light scattering by relative humidity in the daytime. These measurements have been compared to a Mie theory simulation with good results. A value of the hygroscopic growth factor, a key element in the aerosol indirect effect, has been estimated for this case. More data are required to estimate this parameter for the typical Baltimore aerosol.

It is promising that a simple model gives results generally consistent with the observed data. Future work will include testing the sensitivity of the model to various parameters such as the initial radius distribution and the index of refraction. Additionally, the 2004 data will be examined for further cases of hygroscopic growth to compare with the simulation, specifically cases that capture different aerosol composition and size distributions.

5. REFERENCES

Ansmann, A., M. Riebesell, U. Wandinger, C. Weitkamp, E. Voss, W. Lahmann, W. Michaelis, 1992: Combined Raman Elastic-Backscatter Lidar for Vertical Profiling of Moisture, Aerosol Extinction, Backscatter, and Lidar Ratio, *Appl. Physics B*, **54**, 2469-2479.

Bohren, C. R., and D. Huffman, 1983, *Absorption and Scattering by Small Particles*, John Wiley and Sons, New York,

Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hansen, J.E., and Hoffman, D.J., 1992: Climate forcing by anthropogenic aerosols, *Science*, **255**, 423-430.

Feingold, G, and B Morley, 2003: Aerosol hygroscopic properties as measured by lidar and comparison with in-situ measurements," *J. Geophys. Res*., **108**, D11, 4327.

Ferrare, R, 1997: The applicability of a scanning Raman lidar for measurements aerosols and water vapor, PhD Thesis, University of Maryland, College Park.

Hanel, G., 1976: The properties of atmospheric aerosol particles as functions of relative humidity at thermodynamic equilibrium with surrounding moist air, *Geophy*., **19**, 73-188.

Holben B. N., T. F. Eck, I. Slutsker, D.Tanre, J. P. Buis, A. Setzer, E.Vermote, J. A. Reagan, Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, 1998: AERONET - A federated instrument network and data archive for aerosol characterization, Rem. Sens. Environ., **66**, 1-16.

Hoff, R. M., L. Guise-Bagley, R. M. Staebler, H. A. Wiebe, J. Brook, B. Georgi, T. Dusterdiek, 1996: Lidar, Nephelometer and In-situ Aerosol Experiments in Southern Ontario, *J. Geophys. Res. D.* **101**, 19,199-19,209,

Matzler, C, 2002: MATLAB functions for Mie scattering and absorption,"Institute of Applied Physics, University of Bern Research Report, vol. 2002-08,.

Tang, I. N. 1996: Chemical and size effecs on hygrocopic aerosols on light scattering coefficients, J*. Geophysh Res D* **101**, 19245-19250.

Sisler, J. and W. Malm, 2000: Interpretation of trends of PM2.5 and reconstructed visibility from the IMPROVE Network, *J. Air Waste Manage*, **50**, 775-89.

Wulfmeyer, V., Feingold, G. 2000: On the relationship between relative humidity and particle backscattering coefficient in the marine boundary layer determined with differential absorption lidar." *J. Geophys. Res.,* **105**, 4,729-4,741.

Zhang, X., B. Turpin, P. McMurry, S. Hering, and M Stolzenburg, 1994 : Mie theory evaluation of species contributions to 1900 wintertime visibility reduction in the Grand Canyon." *J. Air Waste Manage. Assoc*., **44**, 15.3-162.