# Atmospheric Aerosol Distribution and Cloud Properties using Raman Lidar

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***Abstract*-Atmospheric aerosol measurements are essential to better understand their role as a feedback process in modifying the Earth's radiation budget as well as their influence on human health. The PSU Raman lidars have been used for a number of years, in different locations, to measure the profiles of molecular nitrogen, molecular oxygen, water vapor and the rotational Raman scatter (the mixture of all molecular species) at both visible and ultraviolet wavelengths, and also ozone in the ultraviolet. An aerosol extinction profile, which provides an excellent method to describe aerosol distribution, is then obtained from the incremental extinction based on the difference in the slope of the measured profiles from the major molecular components known vertical gradient. The datasets collected by the PSU Raman lidars, during different campaigns, allow us to infer atmospheric aerosol size variations over different altitude regions by using extinction variations as a function of wavelength to describe the changes in the size distribution of aerosols. This variation in the extinction profiles at different wavelengths can also be used together with the water vapor profiles to study the formation, growth and dissipation of cloud structures. The change in the size of the cloud particles during the different stages can also be observed in the multi-wavelength aerosol extinction. The growth of a cloud, as particles absorb water, is usually associated with a decrease in the water vapor concentration around the cloud. Similarly, a dissipating cloud is characterized by an increase in the water vapor concentration surrounding the region.**

## I. Introduction

Aerosols play vital roles in balancing the Earth's radiation budget, in applications of electro-optic sensors, and they are of increasing importance due to their impact on human health from air pollution. Studies have shown tropospheric aerosols to be directly related to a myriad of health problems [1]. Particles having a diameter less that 2.5 microns, referred to as $PM_{2.5}$, are considered to be of greater risk to human health because a large number of these particles are associated with the emissions from combustion products and carry carcinogenic materials. Particles having a diameter less than 10 microns ($PM_{10}$) are also considered to be a risk to human health but this older standard appears to be less descriptive of their pollution hazard. The propagation of light through the atmosphere is also strongly affected by the presence of aerosols, and hence causes a significant deterioration in the performance of electro-optic sensors. In order to mitigate the effects of aerosols, research efforts are focused on identifying and predicting production and transport mechanisms as well as ascertaining number densities and species concentrations. Raman lidar provides one of the best techniques for describing the aerosol distributions and studying cloud properties. Because the gradient of the vertical profile of the primary constituents is well known, the difference in the slope of the measured profile from the molecular scale height can be used to determine the incremental extinction and thereby generate an aerosol extinction profile. By taking the ratio of extinction at three different wavelengths we can describe changes in particle size and density as a function of time and altitude.

## II. Laps Raman Lidar Instrument

The Lidar Atmospheric Profile Sensor (LAPS) instrument was built by the staff and graduate students of the Applied Research Laboratory and the Electrical Engineering Department of Penn State University for the U.S Navy. It was the fifth Raman lidar developed by the group since 1978 and was the first prototype prepared as an operational Raman lidar. The LAPS instrument uses Raman lidar techniques to simultaneously provide profiles of water vapor, temperature, ozone and optical extinction [2]. Measurements of the atmospheric properties are obtained from the return signal intensity at the transmitted wavelength as well as at Raman shifted wavelengths. LAPS was first tested onboard a U.S. Navy ship, the USNS SUMNER during September and October 1996 in the Gulf of Mexico and Atlantic Ocean. Since then it has been used in a number of research investigations, which have provided a large dataset that will now be used for investigations of aerosol and cloud properties. PSU's LAPS Raman lidar is a rugged instrument and was designed for automatic operation to enable measuring in virtually any environment at any given time.

The Raman scattering technique is advantageous because of its rugged and rigorous quantitative measurement capabilities using a single transmitted wavelength. Raman scatter signals can be used to identify a trace constituent and quantify it relative to the major constituents of a mixture [3]. The LAPS instrument uses the vibrational Raman scattered signals to measure water vapor, ozone and optical extinction, and uses the rotational Raman scatter signals to measure temperature. It collects the rotational Raman backscatter signals at 528 nm and 530 nm and the vibrational Raman backscatter signals at 607 nm, 660 nm, 277 nm, 284 nm and

295 nm. The 607 nm and 660 nm signals are the 1st Stokes vibrational Raman shifts from the $N_2$ and $H_2O$ molecules in the atmosphere excited by the second harmonic (532 nm) of the Nd:YAG laser. The 277 nm, 284 nm and 295 nm signals correspond to the 1st Stokes vibrational Raman shifts from the $O_2$, $N_2$, and $H_2O$ molecules in the atmosphere excited by the fourth harmonic (266 nm) of the Nd:YAG laser. The LAPS instrument measures the water vapor mixing ratio by taking the ratio of the signals from the 1st Stokes vibrational Raman shifts for water vapor and nitrogen. Profiles of water vapor can be obtained during the day (295/284) and the night (660/607) using the ultraviolet and visible laser wavelengths [4].

Optical extinction, which is a measure of the total attenuation of a laser beam due to scattering and absorption in the atmosphere, is obtained directly from the slope of the molecular profiles compared to their expected hydrostatic gradient. The LAPS instrument measures the optical extinction profiles from the gradients in each of the measured molecular profiles, at 607 nm, 530 nm and 284 nm. For these wavelengths the extinction is mainly from optical scattering due to airborne particulate matter. The extinction coefficient can be derived directly from the Raman lidar equation [5]. The Raman lidar equation can be written as,

$$P(\lambda_R, z) = E_T(\lambda_T)\xi_T(\lambda_T)\xi_R(\lambda_R)\frac{c\tau}{2}\frac{A}{z^2}\beta(\lambda_T, \lambda_R)\exp\left[-\int_0^z [\alpha(\lambda_T, z') + \alpha(\lambda_R, z')]dz'\right] \quad (1)$$

where $z$ is the altitude of the scattering volume element, $\lambda_T$ is the wavelength transmitted, $\lambda_R$ is the wavelength received, $E_T(\lambda_T)$ is the light energy per laser pulse transmitted at wavelength $\lambda_T$, $\xi_T(\lambda_T)$ is the net optical efficiency of all transmitting devices at wavelength $\lambda_T$, $\xi_R(\lambda_R)$ is the net optical efficiency of each receiving device at wavelength $\lambda_R$, $c$ is the speed of light, $\tau$ is the time duration of the laser pulse, $A$ is the area of the receiving telescope, $\beta(\lambda_T,\lambda_R)$ is the back scattering cross section of the volume element for the laser wavelength $\lambda_T$ at Raman shifted wavelength $\lambda_R$, and $\alpha(\lambda,z')$ is the extinction coefficient at wavelength $\lambda$ at range $z'$. The extinction coefficients in (1) can be written as,

$$\alpha(\lambda_T, z) + \alpha(\lambda_R, z) = \alpha_{\lambda_T}{}^{mol-sca}(z) + \alpha_{\lambda_T}{}^{aer-sca}(z) + \alpha_{\lambda_R}{}^{mol-sca}(z) + \alpha_{\lambda_R}{}^{aer-sca}(z) + \alpha_{\lambda_T}{}^{abs}(z) + \alpha_{\lambda_R}{}^{abs}(z) \quad (2)$$

where $\alpha_{\lambda}{}^{mol-sca}(z)$ and $\alpha_{\lambda}{}^{aer-sca}(z)$ are the extinction coefficients due to molecular and aerosol scattering at the transmit and receive wavelengths, and $\alpha_{\lambda}^{abs}$ are the molecular and aerosol extinction coefficients due to optical absorption. The molecular scattering contribution to the extinction can be easily taken into account. The selected visible wavelengths do not correspond to molecular absorption features in the atmosphere, and the only molecular absorption of significance in the ultraviolet is ozone. The aerosol profile measurements of extinction include aerosol scatter and aerosol absorption contribution. The aerosol extinction coefficient at the visible wavelengths can be expressed by rewriting (1) as,

$$\alpha_{\lambda_R}{}^{aer} = \frac{\frac{d}{dz}\left[\ln\frac{N(z)}{P(z)z^2}\right] - \alpha_{\lambda_T}{}^{mol}(z) - \alpha_{\lambda_R}{}^{mol}(z)}{1 + \frac{\lambda_T}{\lambda_R}} \quad (3)$$

For the UV wavelength the absorption is significant due to ozone absorption and hence the extinction equation includes the compensation for the ozone absorption as given below,

$$\alpha_{\lambda_R}{}^{aer} = \frac{\frac{d}{dz}\left[\ln\frac{N(z)}{P(z)z^2}\right] - \alpha_{\lambda_T}{}^{mol}(z) - \alpha_{\lambda_R}{}^{mol}(z) - \alpha_{\lambda_T}{}^{Abs}(z) - \alpha_{\lambda_R}{}^{Abs}(z)}{1 + \frac{\lambda_T}{\lambda_R}} \quad (4)$$

## III. Results

The LAPS lidar has a distinct advantage in being able to measure optical extinction at different wavelengths and we use this to infer particle size variation by taking ratios of the extinction coefficients at the different wavelengths. Fig. 1 shows a model calculation of the ratios of the extinction coefficients of 530 nm/284 nm and 607 nm/530 nm. The calculation was done assuming only spherical particles using Mie theory. We see that the ratios of the extinction coefficients provide us with information about variations in the size of particles. When the particle size is small compared to wavelength, the scattered intensity follows Rayleigh's theory and is inversely proportional to the fourth power of the

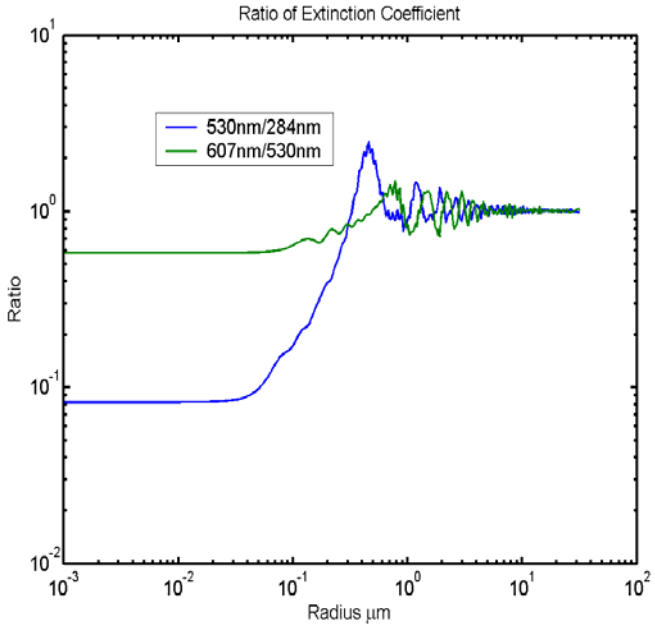



Figure 1. Ratios of extinction coefficients as a function of particle size calculated using Mie theory.

wavelength while the scatter cross-section increases as the sixth power of radius. For accumulation mode particles, where the size range is from 0.05 μm to 1 μm, the ratios of the extinction coefficients are size dependent. For coarse mode particles the ratios lose their size dependence and approach unity.

Fig. 2 shows an example of the optical extinction measurements made at the three wavelengths during the SCOS97 measurement program. The vertical profiles show the variation of extinction with altitude. By comparing with the model calculations we observe that large particles seem to dominate in the lower atmosphere, from the surface at 1.2 km up to about 1.7 km, as the UV and visible extinction coefficients imply the presence of accumulation mode particles with sizes near the wavelengths of visible light. At higher altitudes, between 1.7 km and 4 km, the ultraviolet extinction is much greater than the visible extinction and this suggests a distribution of smaller particles in this region (refer to Fig. 1). We also observe two layers with no significant wavelength dependence above 4.5 km where the scattering is due to large particles in a cloud [6].

Fig. 3(a) and 3(b) show the time sequence plots of extinction at the ultraviolet and visible wavelengths on the night of August 16, 1999 at Philadelphia during the NARSTO-NEOPS campaign. During this time period we see several aerosol cloud layers advect through the laser beam and the analysis of the ratio of the extinction coefficient of 530nm/284nm shows the results relative to particle size variations in regions inside and surrounding the cloud layers. The ratio of the extinction coefficients for different times, as the clouds pass though the laser beam, is shown in Fig. 4. The different periods of integration in Fig. 4 are shown with similar colored lines in Fig. 3. We see in Fig. 3(a) and 3(b) the presence of a cloud at 0045 UTC and we see the expected increase in the extinction coefficient ratios in Fig. 4. The ratio is very close to 1 inside the cloud and this suggests that the cloud is formed by relatively large size particles (>1μm). The ratio of 530 nm to 284 nm is also higher near the ground and this indicates a higher concentration of larger aerosol particles at the surface layer. The ratio of the extinction coefficients at the different times, as shown in Fig. 4, also depicts the evolution of the cloud in terms of particle size variations. Comparing the time sequences with the ratio plot we see that as the cloud begins to dissipate the ratio begins to fall to lower values, which follows the expectation due to the decrease in cloud particle sizes. We can also infer a slight increase in particle sizes from 1 km to 1.4 km in Fig.4, and this growth of particles is evident in the time sequences plots as a corresponding increase in extinction in that region. Comparing with the model calculation of the ratios, the size difference between the top and bottom of the 1 km and 1.4 km region is about 150 nm. Also, the particle size change at 1.4 km between 0045 UTC and 0125 UTC is seen to be about 70 nm. Fig. 3 is also important because it shows the capability of the LAPS Raman lidar to look through dense clouds. This is advantageous because it enables us to study the top and bottom layers of various cloud and aerosol layers.

The variation in the extinction profiles at different wavelengths is also used along with the water vapor profiles to observe the formation, growth and dissipation of cloud structures. The water vapor concentrations have been seen to decrease in regions surrounding a growing cloud as the particles grow in size by absorbing the water. Also, the water vapor concentrations are found to increase as

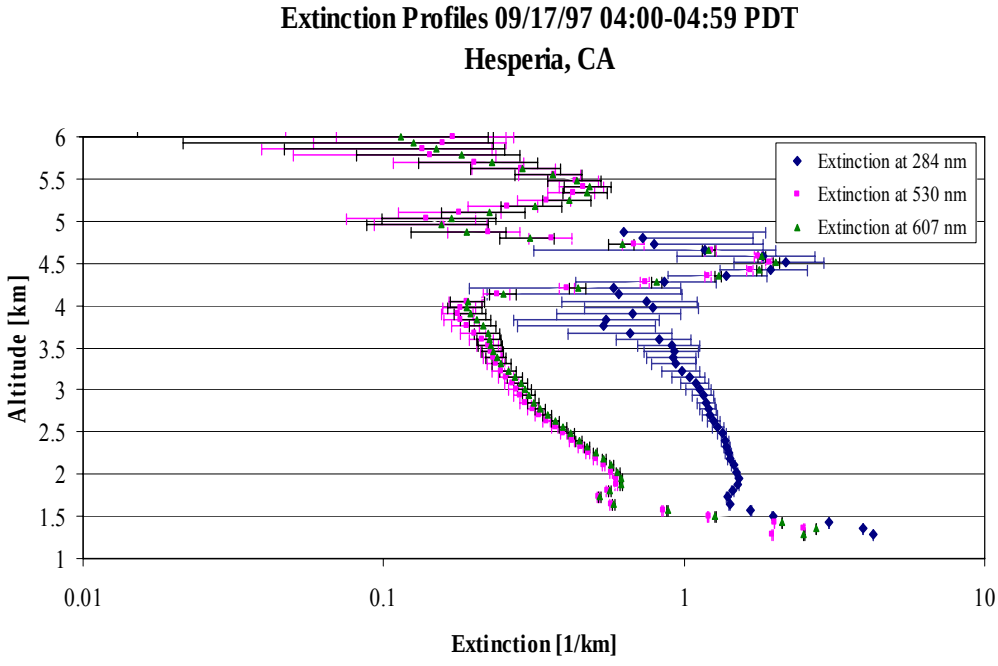
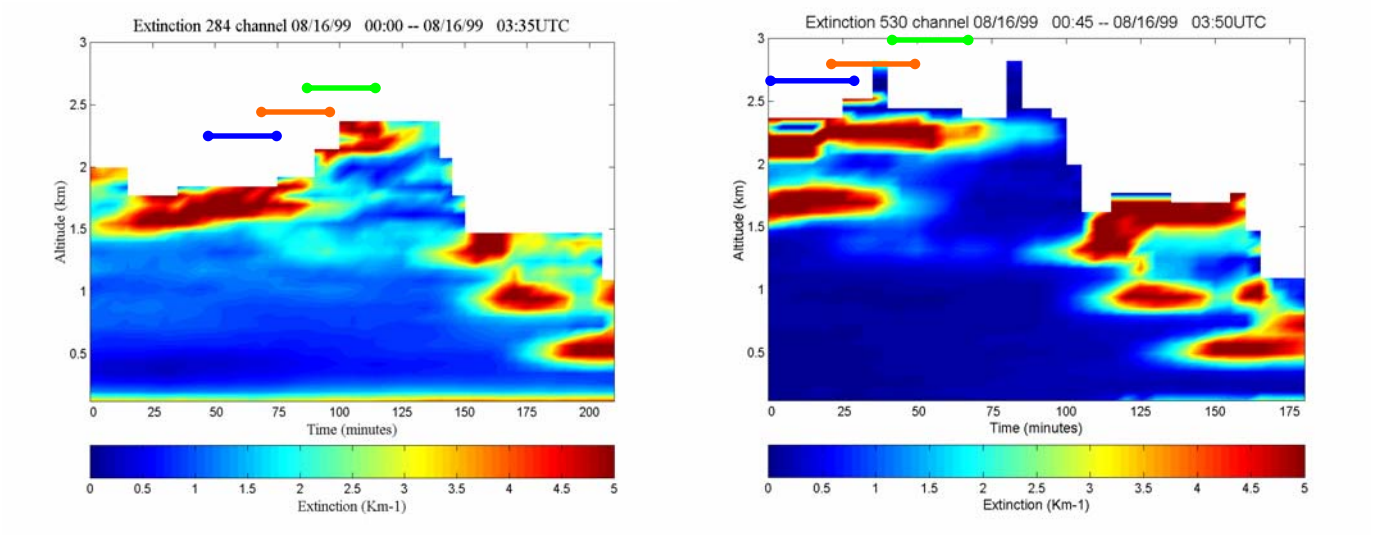



Figure 2. One-hour integrated vertical profiles of optical extinction at 284 nm, 530 nm and 607 nm on September 17, 1997.

the cloud begins to dissipate. Fig. 5 and Fig. 6 show time sequence plots of extinction and water vapor mixing ratio measured using the LAPS Raman lidar during the SCOS97 campaign. We see the presence of a growing cloud in the extinction measurements at around 2100 UTC. The water vapor measurements show high water vapor mixing ratio values in the region of the cloud and lower values in regions surrounding the cloud. The reduced water vapor mixing ratio values surrounding the cloud is attributed to the absorption of water by the aerosols to form the cloud. As the particles begin to grow they start to absorb excess water from the surrounding regions. Conversely, we have observed in a number of data sets that a dissipating cloud is characterized by a region surrounded by excess water vapor.




(a) Time sequence plot of extinction at 284 nm

(b) Time sequence plot of extinction at 530 nm.

Figure 3. Time sequence plot of extinction on August 16, 1999.

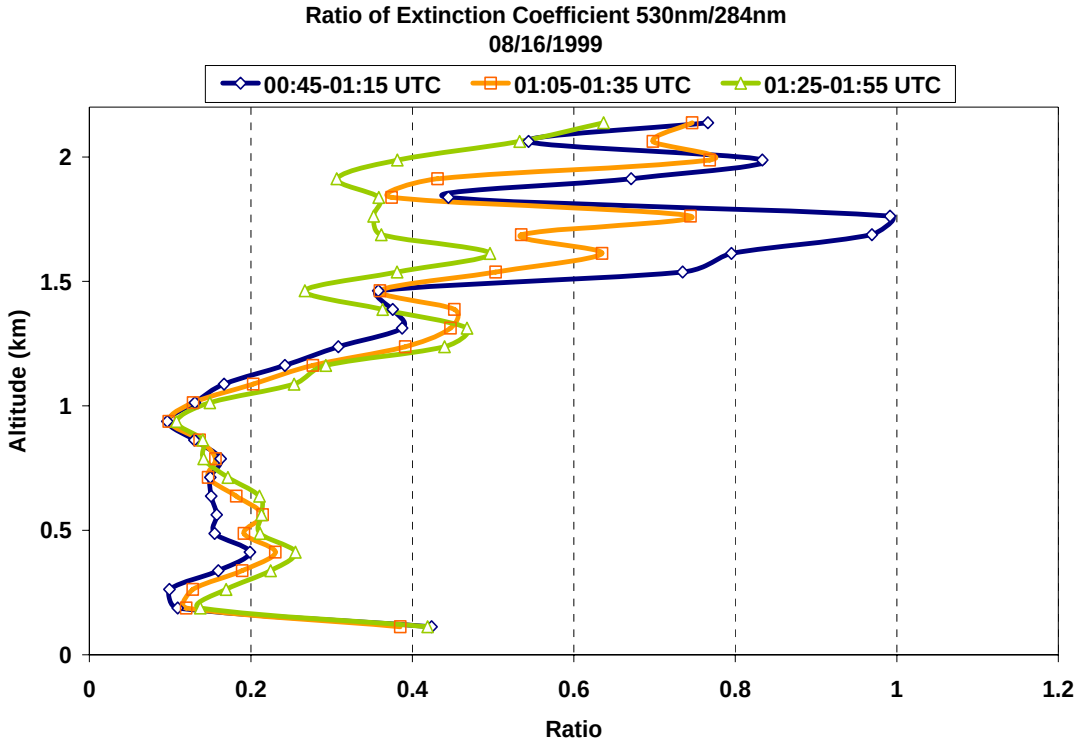



Figure 4. Ratio of extinction coefficient of 530 nm to 284 nm on August 16, 1999.

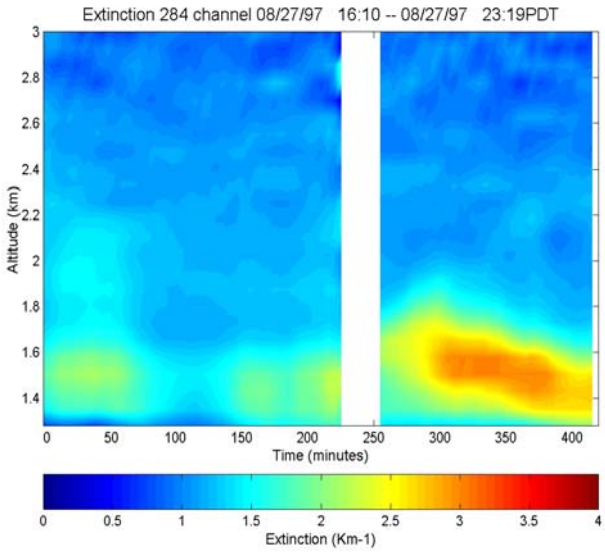



Figure 5. Time sequence plot of extinction on August 27, 1997.

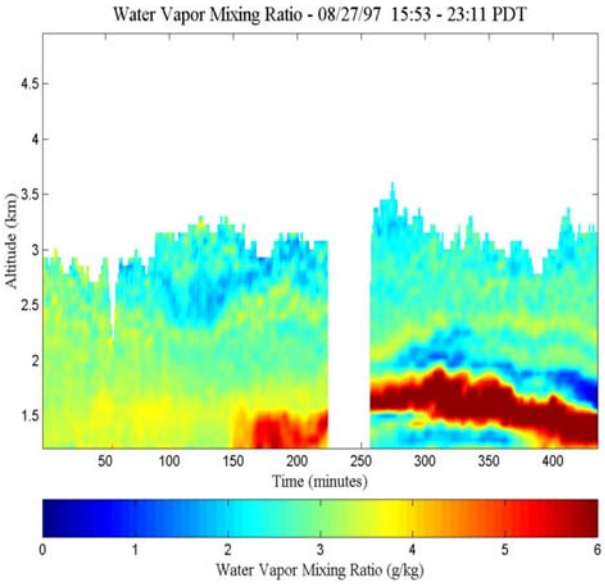



Figure 6. Time sequence plot of water vapor mixing ratio on August 27, 1997.

## IV. Conclusions

Raman lidar techniques provide a direct measure of aerosol and cloud properties by simultaneously measuring optical extinction profiles at several different wavelengths. The variations in aerosol size distribution can be described based upon the ratio of the extinction coefficients at the different wavelengths. The ratio of the extinction coefficients is seen to be size dependent for accumulation mode particles, with size range from 50 nm to 1 $\mu$m, and size independent for larger particles. We see that the analysis of the ratios provide a means to determine changes in the size distribution of aerosols in the atmosphere and also to describe formation processes in cloud layers. The multi-wavelength extinction profiles are also used along with the water vapor profiles to observe the evolution of cloud structures. Water vapor concentrations have been seen to decrease in regions surrounding a growing cloud and increase as the cloud begins to dissipate. A combination of the above techniques will be used in the future to investigate microphysical properties of clouds using data that has been collected in the several measurement campaigns.

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