Application of Raman Lidar Advancements in Meteorology and Air Quality Monitoring

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ABSTRACT

Raman lidar techniques provide vertical profiles of the key parameters needed for better understanding of meteorological and air pollution processes. The time sequence of atmospheric profiles is most valuable for understanding meteorological processes and describing the evolution of episodes and exposure associated with air pollution. The vibrational and rotational Raman lidar signals provide simultaneous profiles of ozone and airborne particulate matter, as well as several meteorological properties. The first operational prototype Raman lidar, Lidar Atmospheric Profile Sensor (LAPS), makes use of 2nd and 4th harmonic generated beams of a Nd:YAG laser to provide both daytime and nighttime measurements. The Raman scatter signals from vibrational states of water vapor and nitrogen provide robust profiles of the specific humidity in the lower atmosphere. The temperature profiles are measured using the ratio of rotational Raman signals at 530 and 528 nm from the 532 nm beam of the Nd:YAG laser. Optical extinction profiles can be determined from the measured gradients in each of several molecular species profiles by comparing with the molecular scale height. Wavelengths at 284 nm, 530 nm and 607 nm have been used routinely to determine profiles of optical extinction. The ozone profiles in the lower troposphere are measured using a DIAL analysis of the ratio of the vibrational Raman signals for nitrogen (284 nm) and oxygen (278 nm), which are on the steep side of the Hartley band of ozone. Examples from several measurement campaigns are used to demonstrate the utility of Raman lidar for describing the evolution of air pollution events. The examples presented have been selected to show the new level of understanding of air pollution events and meteorological processes that is gained from applications of lidar techniques.

Keywords: Lidar, Raman scatter, remote sensing, air pollution, optical extinction, tropospheric ozone

1. INTRODUCTION

Raman lidar has been used to provide vertical profiles of several of the key parameters needed during investigations of air quality and has contributed to our understanding of the evolution of air pollution episodes. Many of the physical processes, which are important in our atmosphere, are described in the literature of meteorology, however the Raman lidar has provided the first opportunity to observe those processes evolving in real time in the atmosphere. This paper shows some examples that demonstrate the value and contributions of Raman lidar to our understanding of the atmospheric physical processes with particular emphasis on understanding air pollution episodes.

Air pollution episodes have been linked to health risks and they influence our daily activities by causing changes in visibility that affect transportation and local aesthetics. The concern for our environment on a global scale is based upon the air pollution influences on the Earth's radiation balance from changes in concentrations of "greenhouse gases" and increasing aerosol particles that cause optical scattering. The two principal components that have been singled out as major air pollution concerns are ozone and airborne particulate matter (PM). Airborne particulate matter has been shown to be associated with increased hospital admissions for cardiovascular disease.[1,2] Ozone is a known toxic species that causes deleterious respiratory effects, particularly causing blisters in the respiratory tract, ageing of tissue, and complications for older individuals, those with asthma or other respiratory problems.[3,4] The increase in airborne particulate matter has changed the optical properties of the atmosphere by decreasing visibility which directly affects air traffic patterns and landing frequency of commercial aircraft, and by reducing the aesthetic appreciation of our national parks.[5] The increase of emissions into the atmosphere causes two competing mechanisms which affect the energy balance that controls our global climate. First, increased emission of chemical species leads to increases in the infrared

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absorption of the Earth's radiation, thereby causing global warming via the "greenhouse effect." Second, the increase in airborne particulate matter reduces the direct and indirect fluxes of solar radiation at the surface due to changes in the planetary albedo. Optical scattering by airborne particulate matter can result in relative decreases in incoming or outgoing fluxes of radiation that lead to a complicated non-linear response in Earth's surface temperature. The increase in airborne particulate matter is principally due to combustion products from transportation and power generation.[4] The goal of present research is to assist in understanding the physical and chemical processes that result in air pollution episodes and global environment changes, so that policies can be developed that will avert severe conditions and show the best directions for action.

The Raman lidar described here is applied as a tool to gain better understanding of the physical and chemical processes associated with air pollution episodes. The LAPS Raman Lidar developed at Penn State University is the first operational prototype Raman lidar. Raman lidar is a robust tool with a multi-function capability that can be employed to measure a wide range of meteorological and environmental properties.[6] The most important parameters for testing our understanding of air pollution models and calculations are the measured vertical profiles of ozone and particulate matter. The profiles of water vapor provide the best information on the dynamical processes in the atmosphere. An understanding and evaluation of models and calculations requires knowledge of the vertical profiles of the atmospheric properties and cannot be gained from either large networks of ground sites with local *insitu* measurements or from infrequent flights of instrumented aircraft. Only remote sensing of the vertical profiles of specific parameters can provide the needed data, and Raman lidar has the highest value in providing these results.

Raman lidar provides continuous time sequences of the vertical profiles of the key parameters in the lower atmosphere. The examples of lidar profiles shown here demonstrate some of the capabilities for measuring the atmospheric properties. The ozone profiles in the lower atmosphere are measured directly from the absorption by ozone in the Hartley band. The particulate matter profiles are determined from the measurements of optical extinction and backscatter at both visible and ultraviolet wavelengths. The important meteorological parameters, temperature and water vapor, are measured with high resolution in time and altitude. Temperature profiles are obtained from the rotational Raman scattered intensity. The water vapor profiles of the specific humidity are obtained directly from the ratio of the vibrational Raman radiation scattered by water vapor and molecular nitrogen. The water vapor is a particularly important tracer of the tropospheric dynamics and is the best marker of the thickness of the planetary boundary layer that describes the dilution volume for the chemical species emissions. The Raman lidar techniques used to investigate air pollution events are described in the following section. Examples from the results of several investigations are shown to provide examples of the capability of Raman lidar as a tool for investigations of the physical and chemical processes during air pollution events.

2. RAMAN LIDAR MEASUREMENT TECHNIQUES

Raman scattering is one of the processes that occurs when optical radiation is scattered from the molecules of the atmosphere. It is most useful because the vibrational Raman scattering provides distinct wavelength shifts for species according to specific vibrational energy states of the molecules, and rotational Raman signals provide measurements directly related to the atmospheric temperature.[6] Figure 1(a) shows a diagram of the vibrational and rotational energy levels that are associated with Raman scatter. When a photon scatters from a molecule, the redistribution of the charge cloud results in a virtual energy state. Most of the atmospheric molecules reside in the ground vibrational level because the vibrational excitation corresponds to relatively large energy transitions (tenths of eV for simple molecules like nitrogen and oxygen) compared to the thermal energy available. After scattering occurs, most of the events result in the return of the molecule to the ground state and the emitted photon has the energy of the initial photon plus/minus the random thermal velocity of the molecule that is the Doppler broadening. A small fraction of the transitions (order of 0.1%) result in giving part of the photon energy to the molecule, and ending in the first vibrational level (a Stokes transition). The emitted photon energy is decreased by exactly the energy of the vibrational quanta for that molecule. For the small fraction of molecules existing in the vibrational excited level, the unlikely anti-Stokes transition is possible. The relative intensities of the scattering signals are indicated by the scattering cross-section values at 532 nm in Figure 1(b). The wavelengths of vibrational Raman back scatter signals from the molecules of the water vapor and molecular nitrogen are widely separated from the exciting laser radiation and can be easily isolated for measurement using modern filter technology and sensitive photon counting detectors.[7] The ratio of rotational Raman signals at 528 nm and 530 nm provides a measurement of atmospheric temperature.[8,9] All of the molecules of the lower atmosphere

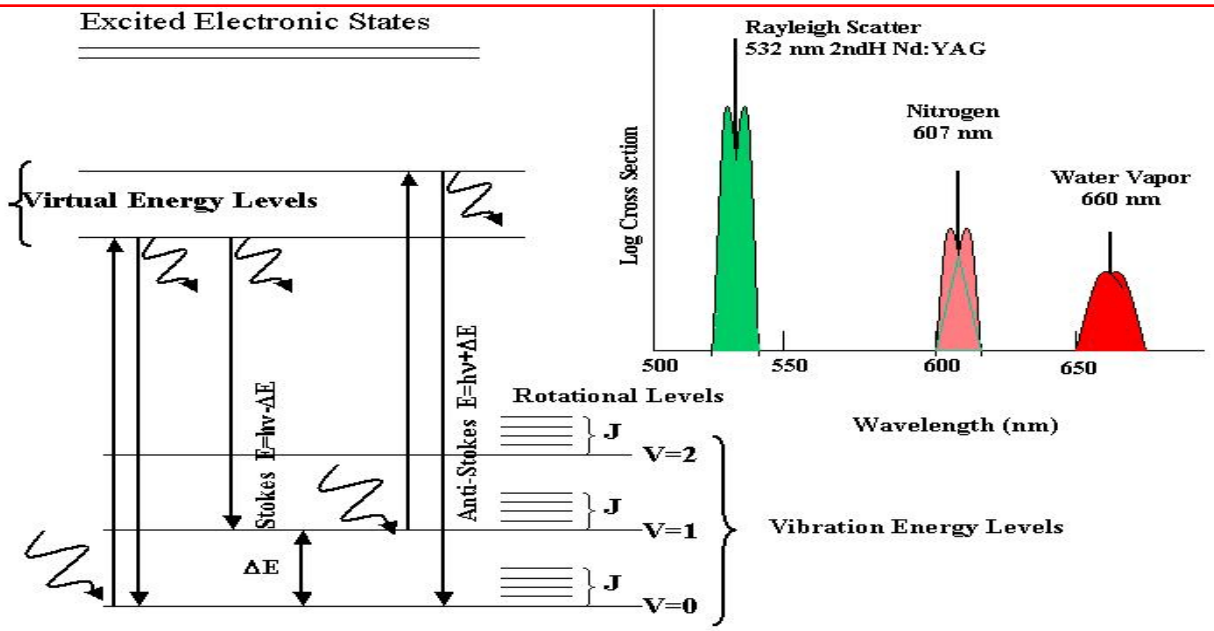

Figure 1. (a) The energy diagram of a molecule illustrating photon scattering raising the molecule to a virtual level from the ground (V=0) state, the emitted photon has the same energy, only broadened by thermal Doppler velocity. In a small number of cases, the return is Stokes shifted to the first vibrational level (V=1), and much less likely is the anti-Stokes transition when the process starts in an upper level. The rotational states (J-levels) are populated by thermal excitation. (b) The relative intensities of the Stokes vibrational Raman shifts of oxygen, nitrogen and water vapor are indicated for illumination of atmospheric molecules with the 532 nm laser. The distributions of relative rotational states are indicated by envelopes of the individual lines.[6]

are distributed in the rotational states according the local temperature. By measuring the ratio of scattered signals at two wavelengths in this distribution, the temperature can be directly measured. In order to push the lidar measurement capability into the daylight conditions, we use the "solar blind" region of the spectrum between 260 and 300 nm. The "solar blind" region is darkened by the stratospheric ozone absorption of ultraviolet radiation. Night time measurements are made using the 660nm/607nm (H_2O/N_2) signal ratio from the doubled Nd:YAG laser radiation at 532 nm. Daylight measurements are obtained using the 295nm/284nm (H_2O/N_2) ratio from the quadruple Nd:YAG laser radiation at 266 nm. A small correction for the tropospheric ozone must be applied. That correction can be obtained from the ratio of the O_2/N_2 signals 278nm/284nm, and this analysis also provides a direct measure of the ozone profiles in the lower troposphere.[10] The Raman techniques, which use ratios of the signals for measurements of water vapor and temperature, have the major advantage of removing essentially all of measurement uncertainties, such as any requirement for knowledge of the absolute sensitivity and non-linear factors.[11] Optical extinction profiles are measured using the gradient of the measured molecular profile compared with that expected for the density gradient.[12-16]

2.1. Water Vapor Measurements

The specific humidity, or water vapor mixing ratios, are determined by taking the ratio of the signals from the 1st Stokes vibrational Raman shifts for water vapor and nitrogen. The measurements are made with laser lines at visible (532 nm) and ultraviolet (266 nm) wavelengths. The visible measurements (660/607) are available at night and the ultraviolet measurements (294/284) are available day and night. The ultraviolet profiles are limited to the first 3 km because of signal loss due to the large scattering cross-section. The ultraviolet water vapor instrument calibration value has remained relatively constant for the LAPS during the past five years, however the visible sensitivity has shown significant changes on a few occasions, possibly due to an inadvertent overload the photomultiplier tube during daylight. Investigation of the stability of the instrument has shown that the variations between the meteorological balloon sonde water vapor and the lidar are about $\pm$4%, and this is approximately the value expected due to the spatial and temporal differences.[16]

2.2. Ozone Measurements

The Raman vibrational 1st Stokes shifts from molecular nitrogen and oxygen are used to obtain the ozone profiles. Since the concentration ratio of these two principal molecular constituents is constant within 10 ppm in the lower atmosphere, any variation in the vertical profile of this ratio can be associated with the integrated absorption due to ozone.[17] Figure 2 shows the location of the Raman shifted wavelengths on the sloped side of the Hartley Band. Using the laboratory measured cross-sections in a DIAL lidar inversion analysis, the concentrations of ozone can be calculated.[6,12] This technique eliminates the task of tuning and stabilizing the frequency and relative power of transmitted wavelengths as required for typical DIAL measurements. The fact that the nitrogen and oxygen molecules scatter a known fraction of the two Raman wavelengths in each volume element makes the technique a very robust measurement that depends only on the absorption cross sections and the statistical errors in the signals.

2.3. Optical Extinction Measurements

The extinction coefficient is made up of components due to absorption by chemical species and particles, and scattering by molecules and particles.[18] The Raman scatter signals from the major molecular species provide direct measurements of the optical extinction. The backscatter signal at the transmitted wavelength exhibits a profile that combines molecular and particle scattering, and it is difficult to analyze for significant properties, except cloud height. However, analysis of the Raman profiles from molecular scattering signals can provide unique vertical profiles of optical extinction. The LAPS instrument measures the optical extinction profiles from the gradients in each of the measured molecular profiles, at 607, 530 and 284 nm. The wavelength dependent optical extinction can be used to describe changes in the particle size distribution as a function of altitude for the important small particle sizes. These measurements can also be interpreted to describe the air mass parameter and atmospheric optical density. Measurements of optical extinction are based upon gradients in the molecular profiles, using the N_2 vibrational Raman scattering or the rotational Raman lines in a region with weak temperature dependence. The calculation is easily applied to the rotational Raman signal at 530 nm because it is so close to the 532 nm transmitted wavelength that no significant wavelength dependent difference exists. By first calculating the extinction at 532 nm from the 530 nm path, it is possible to calculate the optical extinction at 607 nm without assuming a wavelength dependence for aerosol scattering.

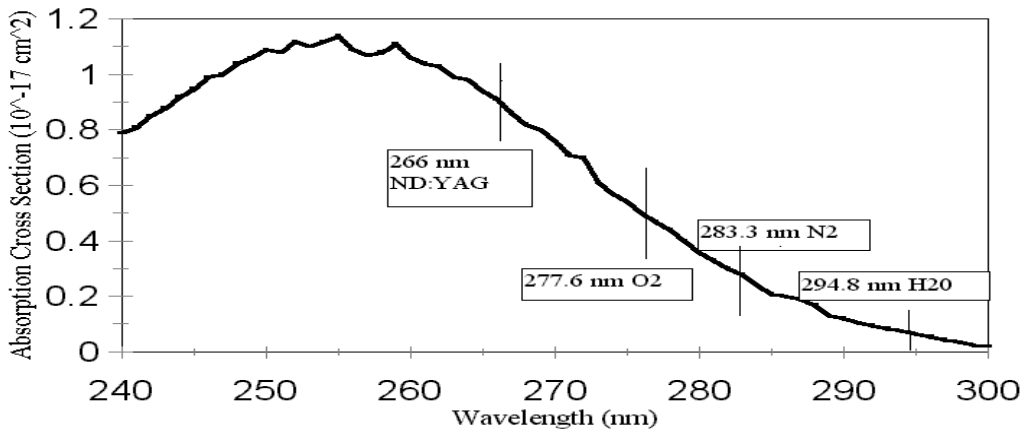

Figure 2. The absorption cross-section of the Hartley band of ozone is shown with the incident and scattered wavelengths indicated.[19]

3. LAPS INSTRUMENT

The LAPS lidar instrument is the fifth Raman lidar that we have prepared and includes the lessons learned during those developments. The LAPS instrument is the first operational prototype Raman lidar and was demonstrated for the US Navy in 1996.[16, 20, 21] The long term goal for this instrument development is to complement the current balloon sonde profiling and enable continuous measurements to meet the requirements for future meteorological data. The shipboard testing of the Lidar Atmospheric Profile Sensor (LAPS) instrument demonstrated its ability to measure the principal atmospheric properties and capability for automated operation under a wide range of meteorological conditions. Profiles are currently obtained at each minute, with a vertical resolution of 75 meters from the surface to 7 km. The LAPS

instrument includes several sub-systems to automate the operation and provide the real-time results. Also, the instrument includes an X-band radar which detects aircraft as they approach the beam and automatically protects a 6 degree cone angle around the beam. Table 2 lists the primary characteristics of the LAPS lidar and Table 3 lists the measurements obtained and the typical altitude range of the data products expected.

Table 2. LAPS Lidar characteristics

Transmitter	Continuum 9030 -- 30 Hz 5X Beam Expander	600 mj @ 532 nm 130 mj @ 266 nm
Receiver	61 cm Diameter Telescope	Fiber optic transfer
Detector	Seven PMT channels Photon Counting	528 and 530 nm -- Temperature 660 and 607 nm -- Water Vapor 294 and 285 nm -- Daytime Water Vapor 276 and 285 nm -- Raman/DIAL Ozone
Data System	DSP 100 MHZ	75 meter range bins
Safety Radar	Marine R-70 X-Band	protects 6^{o} cone angle around beam

Table 3. Measurements made by the LAPS lidar instrument

Property	**Measurement**	**Altitude**	**Time Resolution**
Water Vapor	660/607 Raman 294/285 Raman	Surface to 5 km Surface to 3 km	Night - 1 min. Day & Night - 1 min.
Temperature	528/530 Rot. Raman	Surface to 5 km	Night (30 min.)
Ozone	276/285 Raman/DIAL	Surface to 2 - 3 km	Day and Night (30 min.)
Optical Extinction at 530 nm	530 nm Rot. Raman	Surface to 5 km	Night (10 to 30 min.)
Optical Extinction at 607 nm	607 N_2 - 1st Stokes	Surface to 5 km	Night (10 to 30 min.)
Optical Extinction at 285 nm	285 N_2 - 1st Stokes	Surface to 3 km	Day and Night (30 min.)

4. RESULTS AND EXAMPLES OF APPLICATIONS TO AIR QUALITY

The LAPS instrument uses Raman lidar techniques to simultaneously provide the profiles of water vapor, temperature, ozone and optical extinction. The goal of this paper is to introduce some of the measurements using examples from the recent application of the techniques to investigations of air quality. The results presented here were obtained during the measurements from a research program referred to as the North American Research Strategy for Tropospheric Ozone - Northeast - Oxidant and Particle Study (NARSTO-NE-OPS).[22] The later program has been conducted by a consortium of universities and government laboratories and is focused on understanding the air quality in the urban corridor extending through the northeastern states.

The results described in this paper are examples taken from the NARSTO-NE-OPS field campaign during summer of 1999 at the Philadelphia field site. The theme developed with these examples shows the importance of vertical profiling of the atmospheric properties to understand the processes controlling the environment and demonstrates the utility of Raman lidar. In Figure 3, a time sequence of surface measurements of ozone (measurements obtained by Richard Clark, Millersville University), during part of the month of July 1999, show the diurnal variation in the production and loss of ozone near the surface. The results presented here include measurements from the 17 and 25 July, which were days of high air pollution levels.

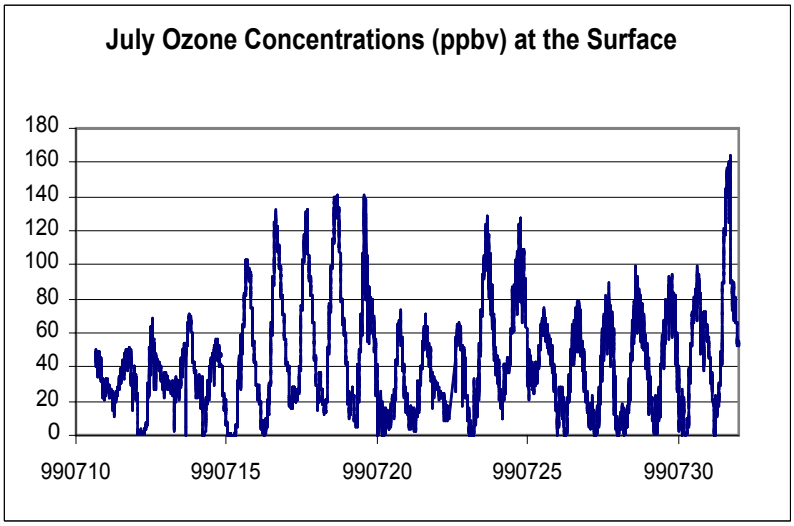

Figure 3. The surface measurements of ozone at the NARSTO-NE-OPS site in Philadelphia during part of July 1999 show the time periods of major ozone episodes, 15-19, 23-25 and 31 July (R.D. Clark - Millersville University).

Two periods of time have been selected to show the insight gained with the time sequences of atmospheric properties that can be measured with a Raman lidar. Figure 4 shows the development of a high concentration of ozone at altitudes below 300 meters during the late afternoon between 4 and 7 PM local time on 25 July 1999. The corresponding panel shows the results from the simultaneous measurements of water vapor. The distribution of the ozone in the lower troposphere can be variable and does not always correlate well with the concentrations measured at the ground. During the afternoon hours, strong convection periods usually do tend to produce uniform vertical profiles of ozone and water vapor. One interesting feature of this example is that the water vapor concentration is very low in the same region where the ozone is highest. In several cases of high ozone concentrations, there appears an anti-correlation of the water vapor concentration. The correlations of high ozone concentrations with low water vapor concentrations are observed sufficiently often to suggest a significant physical or chemical causal connection. The fact that the maximum concentrations are late in the afternoon compared with the peak period of photochemical activity suggests a transport mechanism that could bring dry air with pollutants and precursors into the region.

The importance of transport mechanisms also comes from examining the nighttime results shown in Figure 5. Figure 5 shows a nighttime sequence of profiles of water vapor and attention is drawn to the dry layer observed between 400 and 800 meters altitude during the night. The lower panel shows the results from the PSU Radar/RASS instrument at the NE-OPS site. The wind velocity arrows show the direction of the wind at each point in the time sequence versus altitude plot (North is upward pointing). Additional information on the wind speed is given by the length of the arrow and by the color assignment in the diagram. The measurements show that a significant wind velocity develops between 00 and 11 hours UTC (8 PM to 7 AM local time) in the altitude range between 400 and 800 meters. The general direction of this wind is from the west-southwest and the speed reaches 15 m/s. The dynamical feature of the nocturnal low level jet (LLJ) provides an interesting transport phenomenon when there exists the right combination of topography (Appalachian slope) and circulation pattern (high pressure cell in the southern states and Atlantic region). Most of the major air pollution episodes that we have observed were accompanied by the development of a nocturnal low level jet. Nocturnal low-level jets (LLJ) are suspected to transport significant quantities of pollutants and precursors over large distances. The LLJ formation could just be a natural consequence of the circulation pattern, which also brings the quasi-stagnation of the air mass and the clear conditions that permit ultraviolet photochemical production that produces air pollution. However, the LLJ does also transport air parcels above the nocturnal boundary layer over great distances at night, and these parcels of air can contribute pollutants and precursors to production during the following day. It will require some careful modeling to decide the actual importance of the LLJ in development of an air pollution episode.

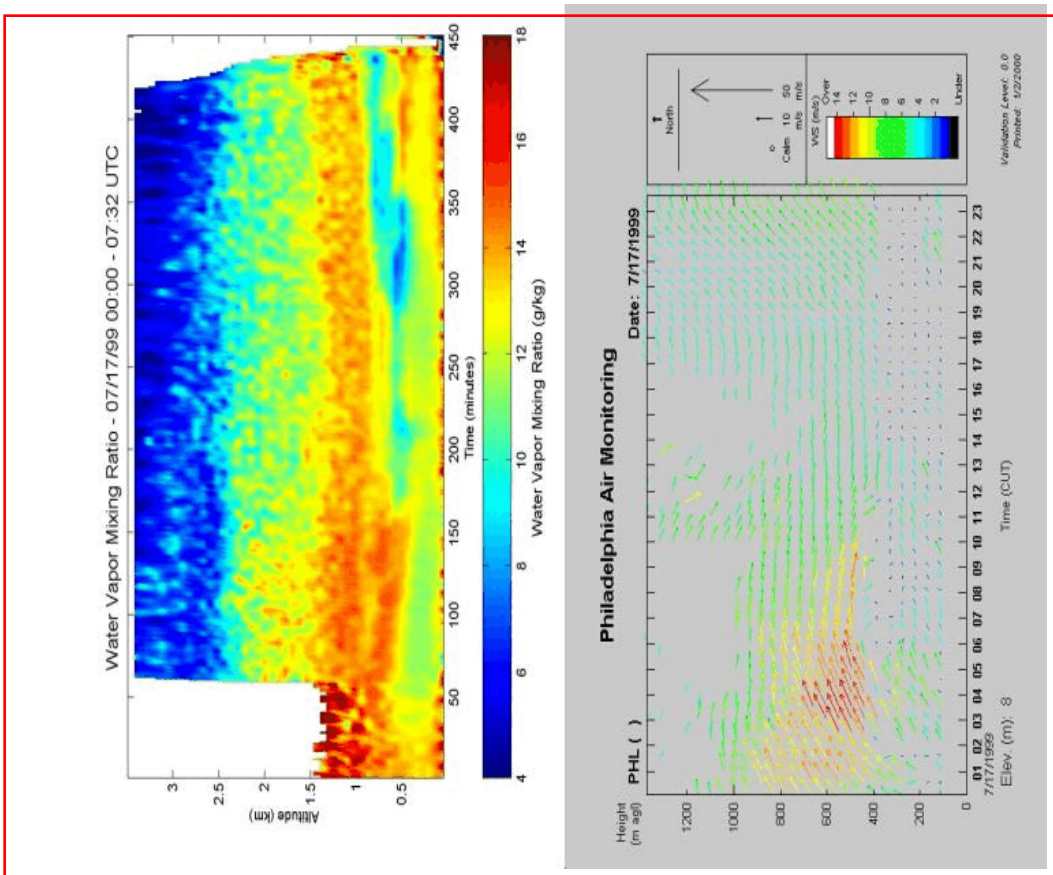

Figure 5. The time sequence of the water vapor profiles on 17 July 1999 shows a dry layer between 400 and 800 meters that is associated with the low level jet shown in the wind velocity profiles from a Radar/RASS sounder.

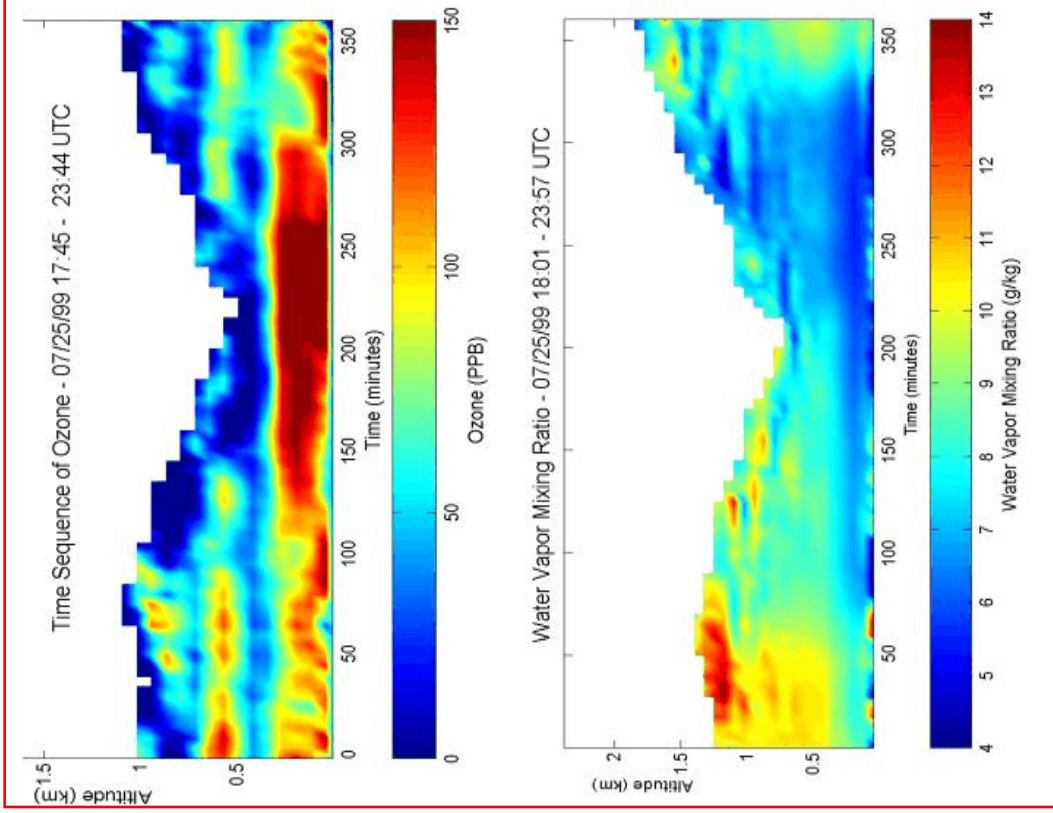

Figure 4. The time sequence of the Raman lidar profiles of ozone and water vapor during the afternoon and evening of 25 July 1999 show the anti correlation of water vapor and ozone concentrations.

5. CONCLUSIONS

Raman lidar measurements provide the key results for understanding the physical/chemical processes associated with air pollution episodes. The primary goals of the research undertaken in these environmental programs are to investigate and model the physical and chemical processes important in understanding the evolution of air pollution events. By measuring the variations in species concentrations and the meteorological factors controlling transport, it is possible to identify the local and distant sources that contribute to increased concentrations of ozone and PM2.5. The results are used to develop and test models, which fully predict the distribution of air pollutants to test regulatory measures. The measurement programs also provide an opportunity to improve and validate new measuring techniques needed for process monitoring.

Measurements of the variations in the profiles of optical extinction, water vapor, and ozone provide valuable insight into the evolution of pollution events. The water vapor profiles show the growth of the boundary to observe transport from the nighttime reservoir and describe the thickness of the mixing volume, which is available for dilution of the chemical species. These measurements show the dynamical processes occurring in the lower atmosphere that control the chemical species distributions by vertical mixing, horizontal transport, and storage of precursor materials in elevated layers. The details revealed in the time sequences from the lidar data add a new dimension to understand the evolution of air pollution episodes. Combining the Raman lidar with other remote sensing measurements, such as Doppler radar, provides the complete set of parameters for testing model predictions, evaluating dynamical processes (vertical and horizontal), investigating atmospheric turbidity, and describing the meteorology of the lower atmosphere with improved temporal and spatial resolution.

The atmospheric dynamics and general meteorological processes are the major factors that control the development and evolution of air pollution episodes investigated in the northeastern region. The rise in air pollutant concentrations to hazardous levels is usually associated with quasi-stationary air masses where pollutant species can accumulate and be processed by ultraviolet driven photochemistry. The major air pollution event periods are associated with air masses that are transported into the region from the west and southwest. Signatures of the nocturnal low level jets, which transport air parcels into the region, are frequently associated with major ozone enhancements on the following afternoon. The normal formation of the nocturnal inversion changes the atmospheric stability and cuts off the vertical transport, thereby resulting in a nighttime storage reservoir and transport layer aloft for ozone and other species. The morning growth of the daytime convective boundary layer mixes materials stored in the nighttime reservoir rapidly to the ground. This fast increase in the temperature of some species can result in thermal decomposition of active molecules that to contribute to photochemical production of ozone.

Additional dynamical control is observed during frontal passages, which can initially concentrate pollutants ahead of their passage and then rapidly removal of air pollutants during the passage. In summary, the dynamical processes of the atmosphere, that transport horizontally over regional scales and vertically mix aloft layers to the ground, exert the first level control over air pollution events.

6. ACKNOWLEDGMENTS

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