#### RAMAN LIDAR MEASUREMENTS OF AIRBORNE PARTICULAR MATTER

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# ABSTRACT

Raman Lidar techniques have been used to provide remote sensing measurements of the aerosol optical extinction profiles from the particles in the atmosphere, as well as water vapor, temperature and ozone profiles. The variation of optical extinction associated with aerosols with respect to atmospheric conditions is useful in understanding the evolution of pollution events. Several cases of extinction, water vapor and ozone lidar results from the NARSTO-NE-OPS program in Philadelphia during summers of 1998, 1999 and 2001 have been investigated. Examples of lidar results are compared with other measurements such as PM sampling and radiosonde measurements. Knowledge of aerosol optical properties assumes significant importance in the wake of studies strongly correlating airborne particulate matter with adverse health effects.

# 1. Introduction

Characterization of airborne particulate matter has been a major challenge to researchers. Knowledge of the aerosol optical properties assumes significant importance in the wake of studies strongly correlating airborne particulate matter with adverse health effects. The small aerosol component, PM2.5, is of most concern to human health because it can be easily inhaled deep into the lungs. Along with health issues, significant aerosol particle distributions have implications for natural environment aesthetics and climatic change conditions. Increasing aerosol loading of the atmosphere can lead either to an increase or decrease in the mean global temperature of Earth, depending on the optical properties in the visible and infrared portion of the spectrum due to the size distributions of aerosol particles. Additionally, airborne particle distributions have a significant influence on visibility, which effects routine aircraft traffic, and has been implicated in climatic change issues. The typical visual range, compared to clean molecular atmosphere, is around 50-67% in the western United States and 20% in eastern United States [Albritton, 1998].

#### 2. Method

Lidar techniques have been used to make remote sensing measurements of the aerosol optical extinction from the particles in the atmosphere, as well as water vapor, temperature and ozone profiles. The Lidar Atmospheric Profile Sensor (LAPS) instrument, which was developed and demonstrated as an operational prototype for the U.S. Navy, is the primary instrument used to obtain the results presented in this paper. LAPS has the capability of measuring meteorological conditions and atmospheric properties such as vertical profiles of water vapor, ozone and extinction during both the day and night conditions [Philbrick, 1998a]. We have used Raman lidar techniques to measure the optical extinction and scattering properties as part of the NARSTO-NE-OPS (NorthEast - Oxidant and Particle Study) [Philbrick, 1998b] during the summers of 1998, 1999 and 2001.

The LAPS system uses rotational Raman scatter to measure temperature, and vibration Raman scattering to measure profiles of water vapor, ozone, and optical extinction. Ozone is obtained from a DIAL (Differential Absorption Lidar) analysis of the Raman shift of N<sub>2</sub> (285 nm) and O2 (276 nm), which occur on the steep side of the Hartley absorption band of ozone. The signal profiles from Raman scatter signals at wavelengths of 607, 530 and 284 nm are directly analyzed to determine the optical extinction profiles. Airborne particulate matter can be analyzed using the gradients in optical Raman scattering profiles to determine optical extinction and to describe changes in particle size and density as the function of time and altitude [Philbrick and Lysak. 1998 and O'Brien et al. 1996]. Extinction is the total attenuation due to scattering and absorption of molecular and aerosol particles in the atmosphere. For wavelengths selected, the extinction is the predominantly from optical scattering due to airborne particulate matter. A telescope form factor is used to aid in correcting and analyzing the surface level signals (up to an altitude 800 m) [Jenness, et. al. 1997]. The telescope form factor is introduced to correct the received signal vertical profile at low altitudes where the out-of-focus ray bundle overfills the detector, which is caused by the near field effect below 800 meters. The form factor used in this analysis is calculated from the return signal curves obtained under clear weather conditions.

7.7

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Extinction is calculated by first subtracting the background signal from the Raman backscattering raw photon count, it is subsequently corrected for range and telescope form factor, and then extinction is determined from the signal gradient [Measures,1992; Ansmann et al.,1992]. The extinction is determined using the basic lidar equation, assuming the absorption and multiple scattering effects are negligible over the interval. The extinction profile can be simply calculated using Beer-Lambert law:

$$a = -\frac{\ln(I_1 / I_2)}{2 x_{12}}$$

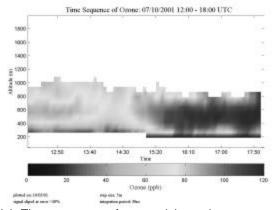
where  $\dot{a}$  is the extinction coefficient and x equals the range bin size in meters. I<sub>1</sub> and I<sub>2</sub> refer to the received signal power from the altitudes 1 and 2 respectively.

During NE-OPS summer 2001 campaign, the rawinsode sounding technique was used to establish the background meteorology condition to provide data to verify the lidar calibration constants. Twelve sondes were released to measure the temperature, relative humidity, pressure and wind speed and the results were compared with lidar measurements.

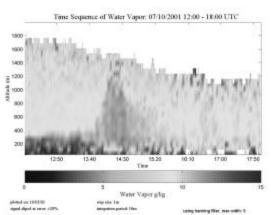
The surface level particulate matter was measured by Harvard School of Public Health during the NE-OPS summer campaign of 1998, 1999 and 2001. During summer 1998, PM2.5 was sampled using CAMMS and TEOM instruments, and PM 10 was sampled using TEOM data continuously from August 3 to August 22 with 10 minute averaging. During summer 1999, PM2.5 was sampled using CAMMS continuously from June 28 to August 15 with 10 minute averaging.

# 3. Results

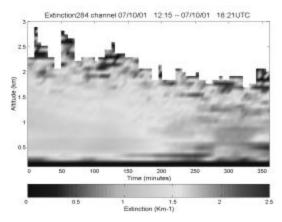
Several sets of lidar measurements from the NARSTO-NE-OPS investigations in Philadelphia during summer 1998, 1999 and 2001 have been analyzed and compared with other measurements.



(a). Time sequence of water mixing ratio.



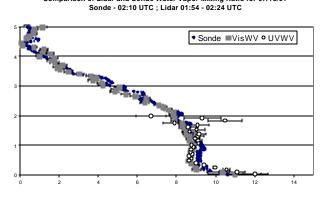
(b). Time sequence of ozone concentration.



(c). Time sequence of Extinction at wavelength 284 nm.

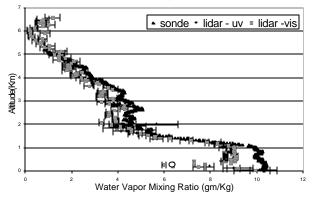
Figure 1. Sequence of plots of an air pollution event that occurred from 12:00UTC to 18:00 UTC of July 10 of the NE-OPS study of 2001.

Figure 1 shows sequence of plots obtained using LAPS instrument during an air pollution event that occurred during the NE-OPS study of 2001. This figure depicts a time sequence of Raman lidar data, which is integrated for thirty minutes and displayed with a sliding window at five-minute steps for profiles of water vapor mixing ratio, ozone and optical extinction. The event on 10 July 2001 is unusual because of the rapid onset observed. During the early morning hours, the boundary layer does not show its normal growth rate due to an upper haze layer that reduced the solar flux and could have been responsible for transport of ozone and PM precursors into the area. At 10AM local time, there is a sudden rise in the PBL from 300 m to 1500 m in about 30 minutes and the ozone concentration increases almost immediately afterward, while the optical extinction measurements indicate that the particulate matter increased substantially about one hour later. This response would be expected from precursor material advected into the region being suddenly mixed to the lower troposphere to produce the rapid rise in ozone and PM observed. Further investigation of the event should clarify the episode.



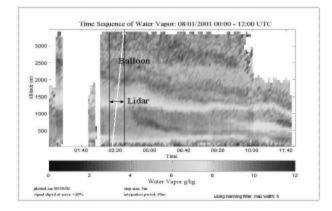
Comparison of Lidar and Sonde Water Vapor Mixing Ratio for 07/10/01

# (a). Lidar data taken from 1:54 UTC to 2:24 UTC of July 10, 2001; Rawinsonde was released 02:10 UTC.



Comparison of Lidar and Sonde Water Vapor Mixing Ratio for 08/01/01 Sonde - 03:19 UTC ; Lidar - 3:05 - 3:34 UTC

(b). Lidar data taken from 03:05 UTC to 03:34 UTC of August 1, 2001; RawinSonde was released 03:19 UTC.

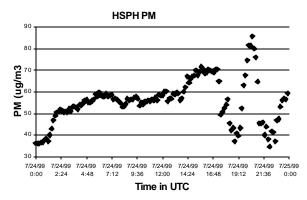


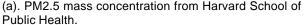
(c). Time sequence plot of water vapor mixing ratio from 00:00 UTC to 12:00 UTC of August 1, 2001, which is during the sonde release.

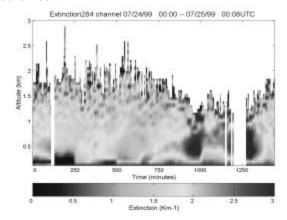
Figure 2. Comparison of lidar and rawinsonde water vapor mixing ratio.

Figure 2 (a) shows the vertical profile of water vapor mixing ratio measured by rawinsonde and LAPS lidar using visible and ultraviolet wavelengths between 1:54 UTC to 2:24 UTC of July 10, 2001. The visible channel water vapor mixing ratio extends to a higher altitude than the ultraviolet channel because its signal is less attenuated by scattering.

Figure 2 (b) shows another example of the vertical profile of water vapor mixing ratio taken from rawinsonde and LAPS lidar measured at visible and ultraviolet wavelengths from 3:05 UTC to 3:34 UTC on August 1, 2001. The top of night time boundary layer is observed at about 1.1 km. The difference in water vapor mixing ratio of lidar data with rawinsonde data between 0 and 1 km is due to the changes in the atmosphere that occur during the 30-miniute integration of lidar data. Figure 2 (c) shows the time sequence plot of water vapor during the sonde release. Notice the higher moisture in the PBL at the time of the balloon release. The time sequence of lidar profiles provides more useful and accurate picture of atmospheric structure than single instrumented balloon flights.



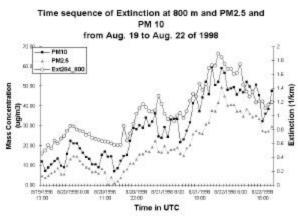




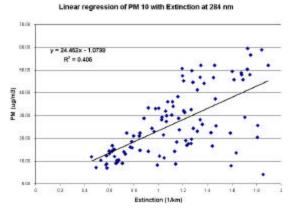
(b). Time sequence of Extinction at 284 nm from Lidar.

Figure 3. Comparison of extinction at 284 nm with PM2.5 mass concentration during July 24, 1999.

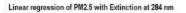
Figure 3 shows a pollution event from NE-OPS study on July 24, 1999. Figure 3 (a) shows PM2.5 mass concentration. Figure 3 (b) gives the corresponding extinction at 284 nm for the same time period. The PM2.5 was sampled using a 10-minute average. The comparison shows that the surface measurements of PM2.5 and the lower altitudes of the lidar profile exhibit a strong correlation. The striking point to realize is that the PM variations aloft are quite significant and that we can observe the vertical extent of the airborne particulate matter using lidar. The extinction shows a strong correlation with particulate matter. It has been conclusively established from a number of data sets that the changes in extinction are strongly correlated with the change of mass concentration of particular matter. A sequence of plots of extinction and particulate matter measurement results are shown in Figure 4, 5 and 6. Both the extinction and particulate matter were averaged for 60 minute periods in these presentations. Due to the telescope form factor, the extinction has a large error below 800 meters and so we have used the extinction there to represent the surface conditions on the assumption that the atmosphere is uniform in boundary layer. This is, of course, a relatively poor approximation but it does serve to show the relationship between the PM and the optical extinction.

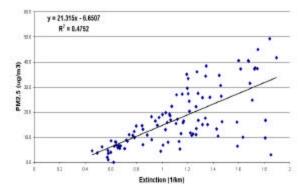


(a). Comparison of PM10, Pm2.5 with optical extinction.



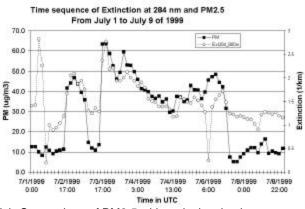
(b). Linear Regression of PM10 with optical extinction.

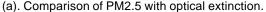


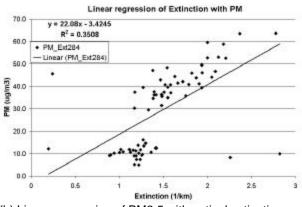


(c). Linear regression of PM2.5 with optical extinction.

Figure 4. Comparison of PM concentration with Extinction at 284 nm, August 15 to August 22, 1998.

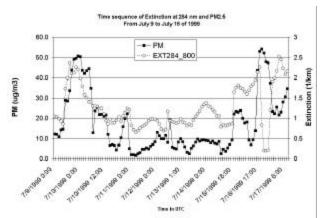




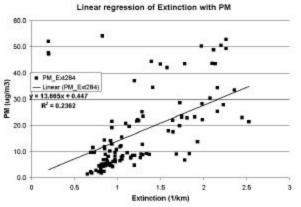


(b) Linear regression of PM2.5 with optical extinction

Figure 5. Comparison of PM2.5 mass concentration with optical extinction at 284 nm, July 1 to July 9, 1999.



(a). Comparison of PM2.5 with optical extinction.



(b). Linear regression of PM2.5 with optical extinction.

Figure 6. Comparison of PM2.5 mass concentration with optical extinction at 284 nm, July 9 to July 22, 1999.

# 4. Conclusion

Lidar has the capability of measuring vertical profiles of meteorological conditions and atmospheric properties in a time sequence which provides a much more useful measure of atmospheric structure than conventional techniques. A strong relationship has been observed between extinction measured from lidar and surface PM mass concentration. These results show that we can describe the vertical distribution of the airborne particulate matter using Raman lidar and thereby describe the evolution of air pollution episodes more accurately. Future analysis will examine the advection of precursor materials and meteorological control factors that govern the particulate components of the lower atmosphere. The vertical distribution of airborne particulate matter, which can be measured by Raman lidar, is expected to provide a critical test for the evaluation and development of air quality models.

#### ACKNOWLEDGEMENTS

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