6.3 # RAMAN LIDAR MEASUREMENTS OF TROPOSPHERIC OZONE

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1. Introduction

Investigations of air quality are important in many applications, as well as in assessing how healthy the air is for humans to breathe. The most common methods used in atmospheric studies involve ground based instruments, tethersonde balloons, and aircraft. The problems in these data arise from the limitations in the instruments making the measurements. Although tethersondes do provide an opportunity to obtain vertical profiles of many important properties in the lower troposphere, their altitude is restricted, and their operations are limited due to high wind speeds. These balloons are also rather manpower intensive, and therefore costly. Aircraft are very useful in examining variations over regional scales, but they require good visibility for flight conditions, and they cannot be used continuously due to the expense of those operations. Ground based instruments simply do not have the capability of vertical profiling, and therefore an important dimension of understanding is lost. This paper shows how vertical profiling using remote sensing techniques, such as those obtained by the Lidar Atmospheric Profile Sensor (LAPS) instrument, provides a better understanding of atmospheric properties and improves our understanding of meteorological processes. It also shows that LAPS data corresponds well to data taken from other instruments. Specifically, ozone measurements obtained during the 1998, 1999, 2001, and 2002 Northeast Oxidant and Particle Study (NEOPS) campaigns are examined.

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2. Method

Measurements were gathered at the NEOPS field site in Philadelphia during the summers of 1998, 1999, 2001, and 2002 using the LAPS lidar instrument. This system uses a Nd:YAG laser pulsed at 30 Hz with an output power of 1.6 joules per pulse at the fundamental wavelength of 1064 nm. This laser fundamental is sent through two frequency-doubling harmonic crystals which provides laser beams at 532 nm (visible) and 266 nm (UV) [Mulik, 2000]. The UV allows day-time measurements as well as night-time. Raman scattering of these two wavelengths by molecules is received by the multi-wavelength detector incorporated into the LAPS instrument. These signals enable ozone, water vapor, optical extinction, and temperature to be measured.

Rotational Raman scatter is used to determine temperature, while vibrational Raman scatter is used to measure ozone, water vapor, and optical extinction. Ozone is measured using a DIAL analysis of the vibrational Raman shifts of N_2 (284 nm) and O_2 (277 nm), which lie on the steep side of the Hartley absorption band of ozone, Figure 1.

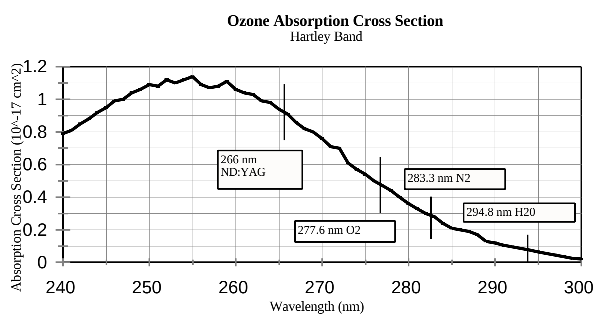


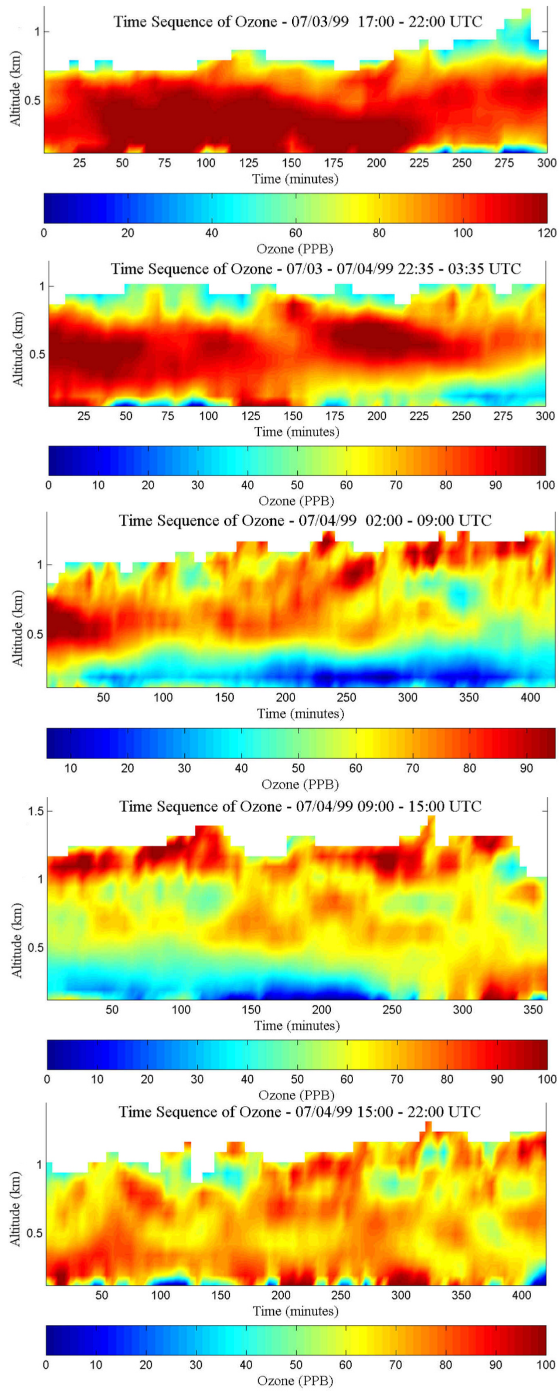
Figure 1. Hartley absorption band of ozone showing(from left to right) the 266 nm Nd:YAG laser, 277.6 nm O_2, 283.3 nm N_2, and 294.8 nm H_2O absorption.

Extinction is measured at 607 nm and 284 nm by using Raman signal profiles shifted by nitrogen from 532 nm and 266 nm respectively. The extinction of 530 nm is measured by using the rotational Raman signals of nitrogen and oxygen from the 532 nm transmitted wavelength [Philbrick, 2002]. Water vapor is determined by the ratio of the Raman signal for water vapor to the signal for molecular nitrogen. Finally, temperature is measured using the ratio of the rotational Raman scatter at two wavelengths.

3. Results

Lidar measurements taken during the 1999 and 2002 NEOPS campaigns are analyzed and compared with measurements from aircraft, tethersonde, and ground-based measurements, in order to show the usefulness and accuracy of the LAPS instrument.

Figure 2 shows time sequence plots obtained using the LAPS instrument during the summer 1999 NEOPS campaign in Philadelphia. Figure 2(a) shows ozone concentrations exceeding 120 ppb in the 0 to 600 meter range. At this time a warm front is moving in from the northwest. The convection within the Planetary Boundary Layer (PBL) mixes down the precursor materials, such as NO_X and VOC's, that were transported into the region [Nieuwstadt, 1996]. These precursors result in the photochemical production of ozone during the afternoon, and mixing causes this ozone to be distributed throughout the boundary later. The trend continues in the following graph until about 01:00 UTC (9 PM local). At this time, solar ultraviolet radiation necessary to create ozone at the surface is not present, and surface ozone decreases [Stull, 1997]. Ozone remains in the residual layer above an altitude of 300 meters as a stable surface layer develops below the nocturnal inversion [Stull, 1997]. In Figure 2(d), surface ozone increases once again due to three effects: solar energy is once again present to cause ozone production, downward mixing of ozone from the residual layer occurs, and ozone precursors are also mixed down. These effects are seen to begin near 13:00 UTC (9 AM local). Finally, in Figure 2(e), a large amount of convection, as well as surface ozone production, is seen to cause widespread distribution of ozone throughout the entire planetary boundary layer.



Figures 2a-e. Time sequence plots of ozone during an air pollution event from July 3, 1999 at 17:00UTC through July 4 at 22:00UTC

A University of Maryland aircraft was used to obtain profiles during spirals above the site of the summer 1999 NEOPS campaign. An example that compares measurements from the aircraft, a Millersville tethersonde, and the PSU LAPS Raman lidar instrument is shown in Figure 3. The tethersonde took measurements up to about 300 meters, while LAPS provides useful data to about 1000 meters, and the aircraft's spirals measure to 2500 meters. All three instruments correspond well between 100-250 meters. From 250 to 1000 meters, the LAPS

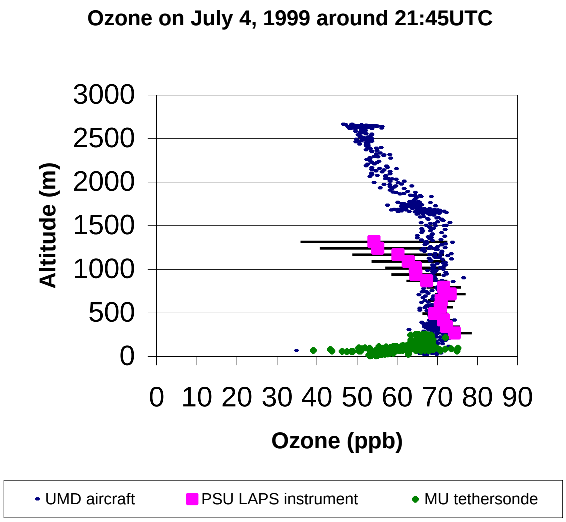


Figure 3. Ozone measurements verify within a small percent error the measurements taken with the LAPS instrument.

instrument and the aircraft show very similar values. Near the upper limit of the LAPS measurements, error bars become larger. For measurements under 100 meters, ground-based ozone-meters are relied upon. However, this graph shows well the accuracy of the LAPS instrument between 100 and 1000 meters. A higher power laser would be required to reduce errors at higher altitudes.

Figure 4 shows another example of the usefulness of the LAPS vertical profiling abilities. On July 1, 2002, around 17:00 UTC (700 min), ozone reached levels near 120 ppb from ground level to 500 meters. This high level of ozone continued until about 20:00 UTC (900 min). However, fairly high levels of ozone remained throughout the night in the residual layer between 300 and 500 meters. As will be verified in Figure 5, the small peak of ozone at 8:00 EDT (12:00 UTC or about 1800 min in Figure 4) is due to this residual layer mixing down to the surface. Finally, it is seen that a large peak of ozone occurs on July 2, 2002 at 16:00UTC. This is near noon local time, and the increase in ozone is due to photochemistry. Precursors transported into the area react to form O_3.

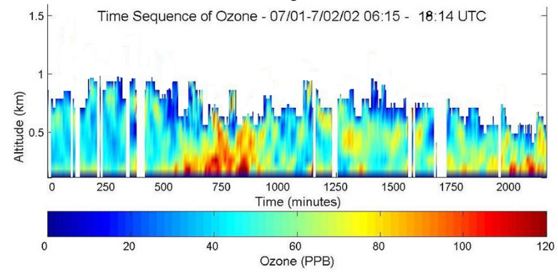
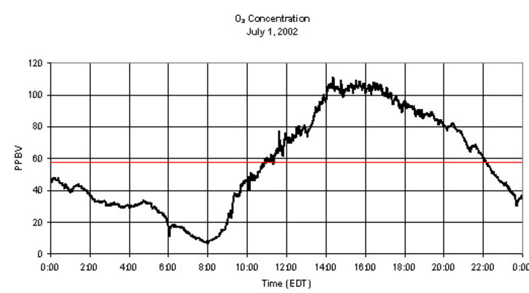
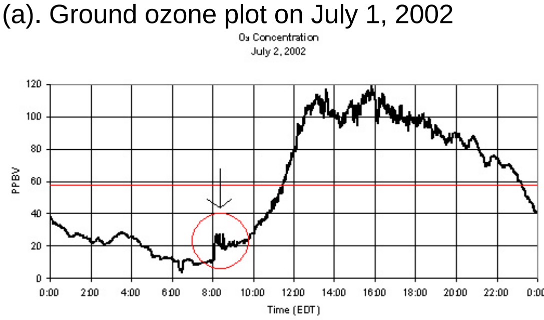


Figure 4. 36 hour ozone time sequence taken with the LAPS instrument during the summer 2002 NEOPS-DEP campaign.

Figure 5(a) shows the increased ozone at the ground level at 13:00 EDT (17:00 UTC) on July 1, and Figure 5(b) shows the increased ozone on July 2 at 12:00 EDT (16:00 UTC). Although lidar results are inaccurate near the ground, the lidar provides the distribution of ozone at upper levels and corresponds well with ozone meter measurements taken at the ground. The lower altitude limitations on the lidar results are overcome by using ground-based measurements compared with lidar measurements above 50 meters and then using a smooth transition over the first three points.



(a). Ground ozone plot on July 1, 2002



(b). Ground ozone plot on July 2, 2002

Figure 5. Ground based ozone measurements taken from a Millersville University ozone meter.

4. Conclusion

The LAPS instrument has been proven to provide accurate, robust measurements of atmospheric processes by comparison with more traditional instruments. Through its vertical profiling techniques, it provides unique insight that other meteorological instruments simply cannot match. Time sequences, which show vertical movement of ozone and other airborne particles, are most useful in testing and developing atmospheric models. Because continuous measurements can be obtained with relatively low manpower and upkeep costs, instruments such as LAPS should be considered as the next step for advancing meteorological technology.

5. Acknowledgements

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