6.1 ADVANCES IN UNDERSTANDING URBAN AIR POLLUTION FROM THE NARSTO-NEOPS PROGRAM

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1. INTRODUCTION

The NARSTO-NE-OPS (North East Oxidant and Particle Study) is an investigation of the coupling of the meteorological and chemical processes that control the evolution of air pollution events. The project includes four major field programs which have been carried out at a field site in northeast Philadelphia during the summers of 1998, 1999, 2001 and 2002. These activities bring together the research groups from several universities and government laboratories to apply advanced measurement techniques for understanding the physical and chemical processes contributing to air quality issues. During the 21 cumulative weeks of summer field intensives, the meteorological and chemical characteristics associated with a wide range of atmospheric processes were observed. The results provide a three-dimensional regional scale picture of the atmosphere, which is essential for understanding the physical and chemical processes that control air pollution events. Vertical profiles of atmospheric properties are required to properly model and predict ozone and particulate matter concentrations. Horizontal transport aloft and vertical mixing processes are found to be key factors controlling the development and evolution of important periods of air pollution. Efforts have also focused on development and testing of several new approaches to improve measuring techniques for better understanding of the physical and chemical properties of the airborne particulate matter. The results of the NEOPS program are providing a data base for development and testing the next generation of atmospheric air pollution models.

Concerns regarding the health effects that have been tied to air pollution events cause us to undertake major efforts to understand the physical and chemical processes that control air pollution episodes. The two principal components of the atmosphere that have been singled out as major air pollution concerns are ozone and airborne particulate matter (PM).

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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, and those with asthma and other respiratory problems.[3,4] Future requirements to forecast hazard levels for warning and protecting those most susceptible requires the development and testing of physics based models that will accurately describe the evolution of air pollution episodes. The efforts of this project are focused on developing the needed understanding of the governing physical and chemical processes and providing results for testing and developing the next generation of meteorological and air quality models.

The research conducted during summer 2002 continued the core activity of our research effort to investigate air quality issues in the northeast, as a follow-on project to the USEPA sponsored study entitled **Investigations of Factors Determining the Occurrence of Ozone and Fine Particles in Northeastern USA**. The associated measurement program has been referred to as the **NARSTO-NEOPS** project, which is from the acronym **N**orth **E**ast **O**xidant and **P**article **S**tudy and has been adopted as a part of the NARSTO Program for data archiving purposes. The 2002 campaign is referred to as NEOPS-DEP 2002, signifying that this measurement campaign was added to the project through support from the Pennsylvania Department of Environmental Protection. The special goals of the 2002 summer intensive included an opportunity to provide a data set that will overlap and have special significance to the northeast Supersites studies and provide measurements during a special investigation of airborne particulate matter in the northeast, referred to as MANE-VU. The measurements were carried out between 29 June and 7 August 2002 to obtain measurements during the most interesting summer period and fully overlap with the other measurement programs in the region. The parameters include the key meteorological properties, gas chemistry, and particulate physical/chemical properties. The major efforts include remote sensing techniques through the troposphere and insitu tethered balloon measurements of meteorology and ozone from surface to 300 m for vertical

profiling of the meteorological and air quality properties, a suite of surface instruments for measuring the concentrations of key chemical species and the particulate matter physical/chemical properties using the most recent techniques for high time resolution sampling, measurements of meteorological parameters at the surface using a 10 m tower, and measurements of minor/toxic species from filter samples using very high resolution GC/MS laboratory techniques. The analysis is extended using other data to include local and regional results. The instruments and measured properties are described in more detail in the following sections. The primary results provide an important data base to test model developments and extend our understanding of physical/chemical processes and sources of air pollution. The results will be shared through the web with other researchers, with particular emphasis on the cooperation with the northeast Supersites in an effort to help create a regional picture of air pollution episodes.

There are three primary factors which make the NEOPS-DEP 2002 activity particularly valuable. First, the NE-OPS project site is ideally located and the team strengths provide valuable instrument techniques for measurements of the air quality and meteorological context at a central regional site for coordination and collaboration with the Northeast Supersites in Baltimore, New York and Pittsburgh. Second, the NE-OPS emphasis on the measurements of particulate matter and the vertical extent of airborne PM provides key measurements for understanding the regional haze questions and useful coordinated measurements with the MANE-VU (Mid-Atlantic/Northeast Visibility Union) investigation of northeast regional haze. Third, the major intensives of the NE-OPS project to date have been conducted during the summers of 1999 and 2001 which represented the 90th percentile wet and 90th percentile dry conditions, and additional summer air pollution episodes need to be examined. By maintaining the effort of a core group of NE-OPS investigators, additional emphasis and value is given to analysis and interpretation of the results from prior intensives and setting the new results into proper context.

1.1 Overview of Research

The USEPA funded three Supersites in the northeast US to conduct a set of special studies that extend beyond the national regulatory networks for PM to elucidate source-receptor relationships and atmospheric processes in support of State Implementation Plans (SIPS). The guiding principles for this research are based on the insights provided in the PM Measurements Workshop Report, *Atmospheric Observations: Helping Build the Scientific Basis for Decisions Related to Airborne Particulate Matter* (EPA/NARSTO, October 1998). The "Supersites Conceptual Plan" outlines objectives of the Supersites Program that emphasize the need for atmospheric measurements essential for the understanding atmospheric transport and meteorology on regional to local scales. Three of the Supersites (Pittsburgh, Baltimore, and New York) are located in the mid-Atlantic region, with two along the northeast corridor.

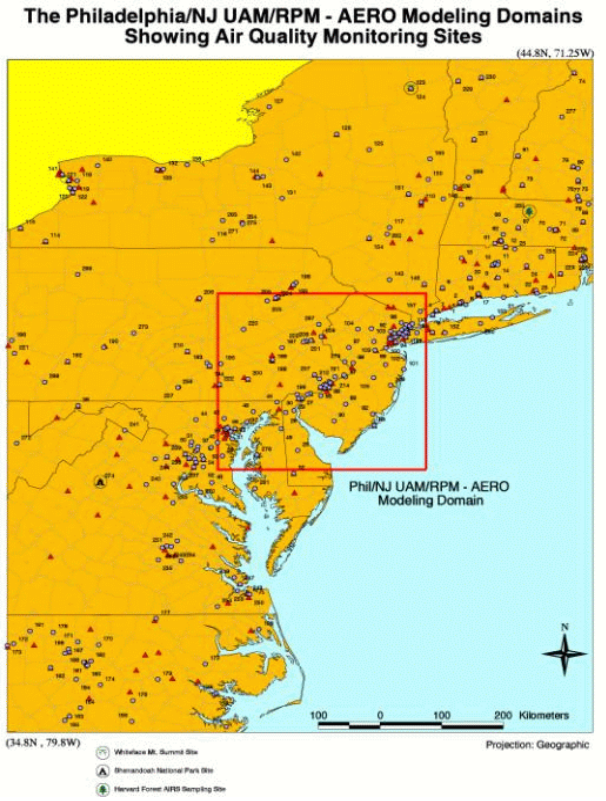

Figure 1. The map of the northeast region shows the regional emphasis of the NARSTO-NE-OPS investigation as the square centered on Philadelphia and it includes the area of the Supersites in New York and Baltimore. The Supersite in Pittsburgh is in an important upwind location for coordinated studies.

Figure 1 shows the northeast regional area investigated by the NE-OPS project, and it indicates the proximity to the locations of the Supersites in New York and Baltimore. While it is recognized that transport, mixing, and atmospheric stability play key roles in understanding source-receptor relationships, surface meteorological variables are expected to make up the bulk of "atmospheric" measurements. The Supersites do not address the meteorological processes, the importance of the thermodynamic structure in determining atmospheric stability, the transitions from daytime mixing to nocturnal stratification, and the effect of planetary boundary layer (PBL) depth on the concentrations of $PM_{2.5}$ and the co-pollutant complex. The central location of the Philadelphia site and the special capabilities of the NE-OPS instruments and investigators can be used to advantage to characterize these processes on a local scale and apply that information to help interpret the results over a regional scale.

Significant variation can occur within the PBL on spatial scales of the order of tens of meters, and time scales of the order of seconds to minutes. Synoptic scale analyses are not capable of resolving structure that plays a major role in the vertical exchange between the lower troposphere and PBL. Mesoscale models continue to be plagued by large divergences in solutions for the height of

the PBL when initialized with surface data without knowledge of the aloft field variables. The relationship between transport on the synoptic and regional scales as determined from NCEP analyses and mixing processes on the local (sub-grid) scale has not been resolved. Uncertainties can lead to significant differences between the model solutions of the height of the stable (nocturnal) PBL and the determination of the daytime PBL height, these disparities are particularly significant during the transition periods. Knowledge of the temporal evolution of the PBL is a critical input into models of the pollutant concentrations because it establishes the mixing volume. The Baltimore and New York Supersites are separated by over 300 km. Over this distance PM and trace gases are so influenced by the northeast corridor urban plumes that attempts to characterize them will fail.

With the creation of PM Supersites in Baltimore and New York the opportunity presents itself that has the potential to integrate aloft and surface measurements in Philadelphia, PA with those at the PM Supersites during a major, combined field intensives. Since 1998 Philadelphia has been the site of an intensive multi-institution/agency investigation that has focused on the factors affecting the occurrence of ozone and fine particles along the northeast corridor. Aircraft, Raman LIDAR, tethered and free balloons, profiles from RASS (Radar-Acoustic Sounding System), and measurements from surface-based PM and trace gas analyzers provided both surface and aloft measurements which are used with emission inventories and numerical models to provide a detailed and integrated characterization of the local and regional conditions accompanying high ozone and fine particle events. An extensive infrastructure has been developed that is driven by the research objectives and involves many scientists from universities, with close coordination with Federal and State agencies, national laboratories, regional planning organizations in the Northeast (MARAMA, OTC, NESCAUM), and the City of Philadelphia. The instruments and measurements deployed during the 1998 NEOPS pilot study (6-21 August 1998), the 9-week NEOPS field intensive (28 June – 18 August 1999), and the 2001 NEOPS (July 2001) are given in summary papers.[5,6] Many of these instruments and sampling techniques are the same or equivalent to those that will be deployed at the PM Supersites with the exception that the aloft measurements are not emphasized at the Supersites.

The 300 km separation between Baltimore and New York is too large a distance to adequately delineate transport from one urban center to another, except by means of analysis of the upper air data obtained twice per day (00-12 UTC) at the NWS WFOs, which are separated by an average distance of 400 km. Philadelphia offers a location approximately midway between the Supersites (182 km from Baltimore; 118 km from New York), and provides an array of surface measurement platforms that are nearly equivalent to the PM Supersites, facilitating measurement comparisons, while adding the required aloft measurement component. The measurements obtained during the 1998/1999/2001 NEOPS intensives, and the recent NEOPS-DEP 2002 intensive, have highlighted the importance of the boundary layer transition period in determining the resulting height of the daytime PBL and the concentrations of gases and fine particles. Maximum daytime concentrations were affected by the time required for the transition from a stable nocturnal PBL to a fully developed daytime mixed layer. In some cases, this transition was extremely rapid, with the PBL becoming convective within 30-60 minutes after the onset of surface heating, and leading to better fumigation and lower afternoon concentration maxima. In other cases, the transition was much slower, occurring over several hours while allowing concentrations of gases and particles to accumulate, and often leading to large exceedances by afternoon. Such assessments can only be documented using a combination of high frequency surface and aloft measurements.

Enhancements of the measurements capability of the investigators are continuously being addressed. Millersville University expanded its on-site capabilities in 2001 so that, in addition to the aloft platform capabilities, measurements and on-site calibrations of O_3, SO_2, CO, NO, NO_2, NO_X, and path-integrated total, scattering, and absorption coefficients are included. The TSI three-wavelength nephelometer also provided valuable on-site comparisons with the development of a new technique by PSU to characterize the airborne particles using a multi-static lidar. The development of the new multi-static lidar techniques are expected to eventually provide a new dimension in vertical profiling of airborne particulate matter by characterizing the size and concentration of PM from 10 m to 2 km above the surface. First tests of this new technique were obtained during the NEOPS 2001 campaign and provide the foundation for this capability.[7]

The central location of Philadelphia with respect to the two PM Supersites along the northeast corridor, its influence as an up/downwind source/receptor, proximity to the coastal regions, yet more inland than either Baltimore or New York, stands to elucidate important similarities and differences in the regional temporal and spatial distribution of the $PM_{2.5}$/co-Pollutant complex. These results could benefit the development of SIPs, the setting of NAAQSs, and the conclusions drawn from sites separated by 300 km in an attempt to improve our understanding of the complex interactions between meteorology and chemistry based largely on surface measurements alone. In addition, summer 2002 overlaps an initial program planned to study the problems of regional haze, MANE-VU. The NE-OPS measurements of particulate matter and chemical species provide key results that are need for investigations of regional haze.

The NARSTO-NE-OPS program is intended to add to our understanding of the relative importance of the various physical and chemical processes which control the evolution, development through dissipation, during air pollution episodes. The data from this program will be used to test the capability of next-generation air quality models to provide short term (1-2 day) predictions of the concentration levels of air pollution hazards over regional scales. Future needs for regulatory actions cannot be undertaken without major improvements in our detailed understanding of the processes controlling the evolution of air pollution events and a demonstrated capability to accurately model the air quality hazards over the region.

2. FIELD MEASUREMENT CAMPAIGNS

The NARSTO-NE-OPS project included three major measurement programs. The information on participants and measurements conducted during the campaigns in the summers of 1998, 1999 and 2001 are summarized in a previous paper.[6] Significant measurement periods that have been selected for specific investigations are summarized in Table 2. The 1998 program was intended to prepare the field site and to evaluate the instrument techniques. During the summer of 1999, the first intensive measurements were carried out over a two-month period that captured data during eight periods of significant air pollution episodes, including one which recorded the highest O_3 in Philadelphia during the past decade. Techniques which were used during the investigation included a wide range of instruments that measured the surface layer and boundary layer properties. The summer 2001 campaign was conducted during a period that did not contain any major air pollution events, however three interesting periods have been identified. The NE-OPS measurements, delayed from summer 2000 to summer 2001 and those in summer 2002, provide an important role relative to the measurements of the three Northeastern Supersites in New York, Pittsburgh and Baltimore. The 2002 project has provided results on several interesting events, see Table 2. The NE-OPS measurements in Philadelphia provide a central location for analyzing the regional conditions of the meteorological properties. Each of the campaigns are briefly described below.

2.1 1998 Pilot Study

During summer 1998, the NARSTO-NE-OPS pilot study was conducted in Philadelphia.[5] It was most valuable in establishing a field site and bringing together researchers from several different universities and laboratories to develop the team for the future field measurement programs. The research is carried out by a consortium of investigators from universities and government laboratories. Several of these groups were involved in preparation of the Philadelphia field measurement site and participated in a two-week pilot measurement campaign in August 1998. The primary purposes of the pilot study were to evaluate the site and to compare instrument platforms as preparation for future intensive measurement campaigns. However, the 21-22 August 1998 measurement period included a significant pollution episode, when substantial increases in both ozone (concentration 125 ppb) and air borne particulate matter ($PM_{2.5}$ 65 $\mu g/m^3$) were observed. Results from the episode are presented in Figure 2. This case appears to be initiated by precursor material that was transported into the region and then rapidly converted when it was mixed down to the surface.

The Millersville University tethersonde instrument is able to provide high resolution profiles from the surface to 300 meter each 30 minutes using a 4 meter long (5 m^3) tethered balloon. During the summer of 1998, 176 vertical profiles were obtained. The instrument package measurers profiles of the temperature, pressure, wind velocity, relative humidity and ozone. A larger 12 m long

Table 2. Summary of the intensive observational periods during 1998, 1999, 2001 and 2002 campaigns.

	Summer 98	7-22 Aug 1998
1	7-22 Aug	Pilot study, prepare site, evaluate instruments
2	21-22 Aug	Sudden ozone and PM event with vertical mixing of transported material
	Summer 99	28 June - 20 August 1999
1	3-5 July	Ozone event limited by strong winds, depth of PBL, variation in local emissions due to traffic
2	8-10 July	Weak cold front oscillation north/south, frontal passage effects, ozone and PM enhanced in PBL as front approached
3	16-21 July	Major ozone event, UMD aircraft
4	23-24 July	Limited regional ozone, strong stable PBL, wind shift, RASS and sondes
5	27J-1 Aug	West flow brings increase in ozone on 27th, regional convection limits ozone on 28th, 31st has high ozone, DOE-G1 and UMD aircraft, ozone 162 ppb surface-180 ppb aloft
6	7-8 Aug	Particulate matter event
7	11-13 Aug	Re-circulation event, 125 ppb on 12th followed by storm on night of 13th
8	15-17 Aug	Standard ozone event but did not last to produce large build up of ozone, UMD Aztec
	Summer 01	29 June - 1 August 2001
1	10 July	Short sudden event with interesting dynamics
2	16-17 July	Ozone and PM event
3	20-25 July	Moderate levels of ozone repeated on 4 days
	Summer 02	29 June - 7 August 2002
1	30J-4 July	On 1 July ozone 105 ppb and 2 July 110 ppb
2	4 & 22 July	UMD aircraft flyby
3	6-7 July	Smoke Event from fires in Canada profiled optical scatterometer on tethered balloon.
4	8-9 July	High ozone (140 ppb range) with an abrupt rise to about 145 ppb late on 8 July (8PM local), 130 ppb on 9 July
5	17-19 July	On the 18th, ozone exceeded 100 ppb for more than 10 hours, 130 ppb on 19 July with excursions to 140 ppb
6	20 July	Sea breeze event.
7	23 July	Frontal passage and rain-out effects
8	28-29 July	Weak ozone and PM event
9	1-2 Aug	Ozone (>120 ppb) and PM Event ending with significant frontal passage
10	4 Aug	130 ppb ozone

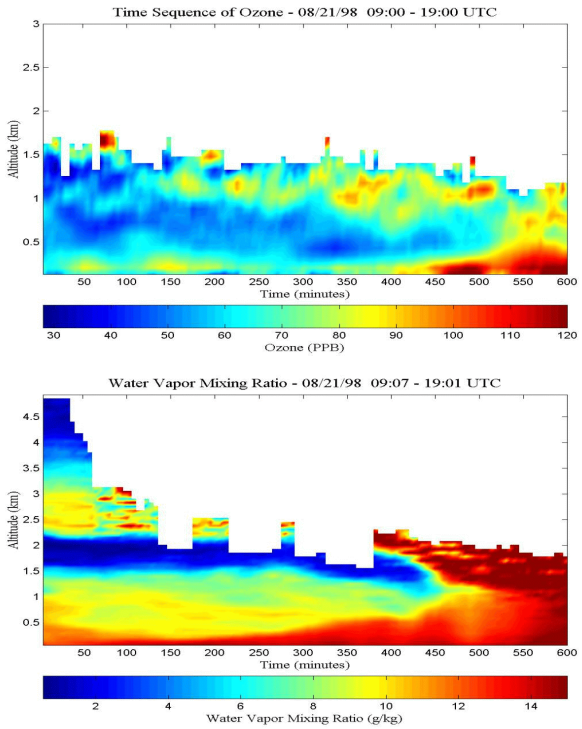

Figure 2. Raman lidar profiles show the growth of the PBL during the morning of 21 August using the water vapor as a tracer. Merging of the rising PBL with an upper layer, which originated in the mid-west, appears to initiate the ozone and PM event that occurred that afternoon.

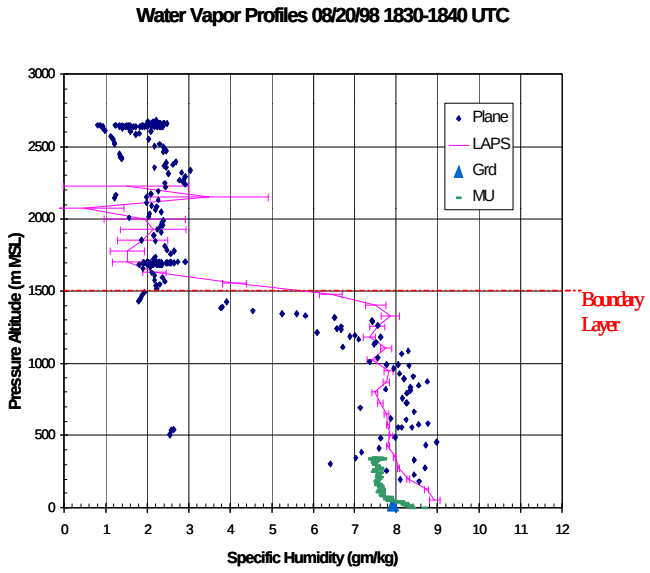

Figure 3. Water vapor profiles from lidar, tethersonde and aircraft are compared during a daytime period when the convective turbulence causes large variations.

(100 m^3) balloon was tethered at 300 m and carried Personal Environmental Monitors (PEMs) suspended at 75, 150, 225 and 300 m for 10 hour periods (10 AM to 8 PM and 10 PM to 8 AM local time) and the filters were analyzed by Harvard School of Public Health. A more detailed meteorological description for the events has been prepared by W. Ryan.[8] The University of Maryland used a Cessna aircraft to obtain profiles and distributions of ozone, water vapor and temperature during 12 flights as part of the August 1998 campaign. Figures 3 and 4 show examples of the comparisons of the lidar, tethersonde and aircraft profiles. The lidar results for water vapor shown in Figure 3 are limited to 2 km by the large scattering cross-section at ultraviolet wavelengths. The ozone results in Figure 4 show good agreement through the mixed region of the PBL but the layered structure of the free troposphere shows significant differences in space and time, above 1700 meters in this case. Generally good agreement between the sensors has been found and detail comparisons are in reports that are in preparation.

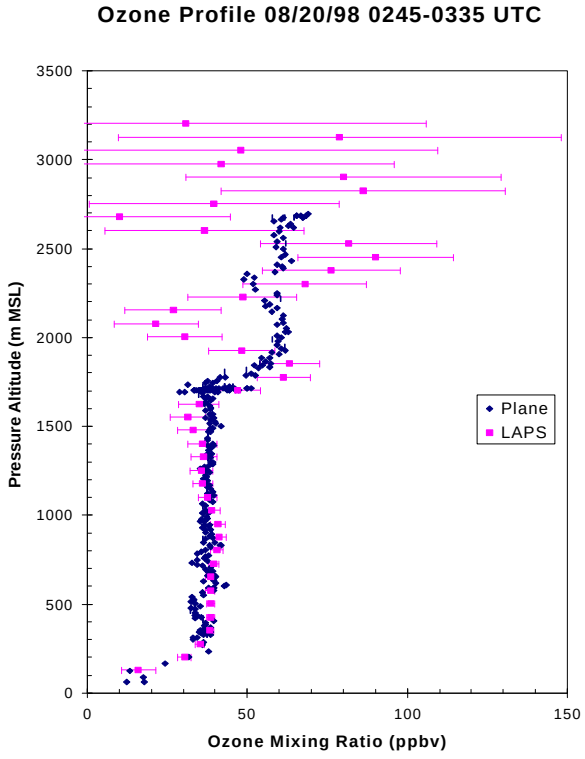

Figure 4. The aircraft and lidar profiles of ozone are compared during a spiral by the aircraft near the vertical beam of the lidar. The ±1σ error associated with the photon counting is indicated on each lidar measurement.

2.2 1999 Summer Intensive

Advanced techniques used during the NARSTO-NE-OPS project included Raman lidar, tethersonde balloons, particle/chemical samples using HSPH instruments (TEOMS, HEADS and HV-TOX), and the latest aircraft sampling techniques on the DOE-G1 and UMD aircraft.

The Raman lidar has proven to be a useful tool for providing time sequences of vertical profiles of ozone, temperature, water vapor and optical extinction.[9] Profiles of the properties of the surface layer to 300 meters were obtained by Millersville University using tethersondes for ozone, temperature, water vapor, wind velocity and fine particle concentration. The regional distributions of particulate matter, chemical species and meteorology were obtained using the University of Maryland instrumented aircraft for 24 flights and the DOE-G1 instrumented aircraft for 19 flights. The latest techniques for measuring the particulate matter properties were used by Harvard SPH, Drexel University and Brigham Young University.

Instrument setup for the summer intensive at the Baxter Water Treatment Plant began on 15 June 1999 and the site was fully operational from 28 June to 19 August, 1999. Particulate matter and gaseous samplers were operated continuously during the project by HSPH, and NOx/ NOy chemistry was measured by Harvard University. Profiles of ozone, optical extinction and meteorological parameters were obtained each day with PSU lidar, except for the period July 18-21. Measurements were made by Millersville University using the two tether balloons during intensive periods and other interesting periods. The smaller tether balloon (5 kg capacity) measured profiles of meteorological properties and ozone from surface to 300 meters and obtained a total of 430 profiles. A larger tether balloon (50 kg capacity) was used to suspend $PM_{2.5}$ filter samplers, continuous optical scattering instrument for $PM_{2.5,}$ and VOC canisters at several altitudes between the surface and 300 meters. The NE-OPS Radar/RASS sounder, obtained from EPRI with support of MARAMA and PECO, was placed into operation at the Philadelphia site on 23 July and operated through the program. Dynamical properties of the meteorological fields are investigated using the data from this Radar/RASS and two other sounders from ANL and PNNL. Particulate matter samples were collected each day by Drexel University for GC/MS analysis of minor species. The University of Maryland provided CO and UV radiation measurements at the site and conducted instrumented flights with Cessna and Aztec aircrafts.[10] Brookhaven National Lab made 19 regional flights using the instrumented DOE-G1 aircraft between 25 July and 11 August.[10] The Pacific Northwest National Lab (PNNL) released 61 radiosonde balloons and 10 ozonesonde balloons between 23 July and 10 August from the Philadelphia site. PNNL setup and operated a Radar/RASS sounder at West Chester, PA (about 30 miles west) from 23 July through 11 August. The Argonne National Lab (ANL) operated a mobile chemistry laboratory and a Radar/SODAR sounder at Centerton NJ (about 30 mi south) during the period 24 July through 11 August.[11] At the Centerton site, ANL also released 56 radiosondes balloons. During the period 2-30 July, Brigham Young University used three instruments to measure the volatile and semi-volatile mass and species of particles.

As part of the 1999 NARSTO Northeast Oxidant and Particulate Study (NE-OPS) field campaign, the DOE G-1 aircraft conducted 19 research flights in and around Philadelphia. Most flights consisted of boxes around the

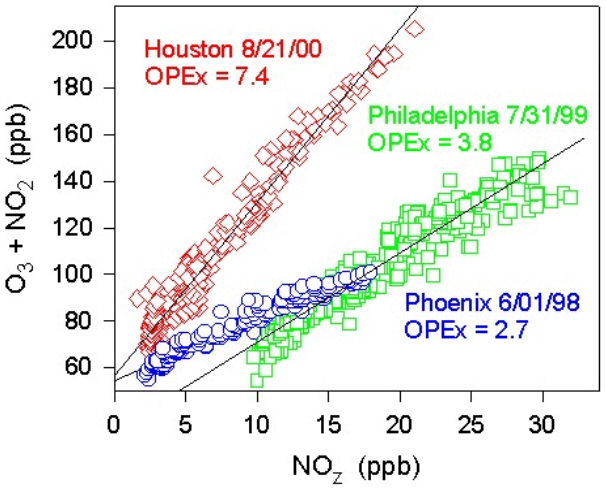

Figure 5. DOE G-1 aircraft data from high ozone days used to calculate ozone production efficiency in 3 cities.[10]

Philadelphia urban area. The NE side of the smallest box was located about 20 km from downtown Philadelphia and included vertical spirals and horizontal transects providing detailed coverage between 300 m and 2.5 to 3 km. Repeated transects at different times of the day gave information on vertical mixing. Trace gas measurements pertinent to understanding O_3 formation included O_3, CO, VOCs, NO, NO_y, SO_2, HCHO, H_2O_2, and organic peroxides. Measurements of these species were used as input to a constrained steady state box model that gave predictions for radical concentrations and the rate of O_3 production. Highest O_3 concentrations in the program were observed on July 31, a day with very light wind speeds. A mid-morning and mid-afternoon flight indicate that the O_3 increase observed from the G-1 (from about 90 to 130 ppb) can be accounted for by local production. On the same day an industrial plume with extreme values of NO, CO, VOCs, and SO_2 was traversed to the SW of Philadelphia. This plume was found at the same location in the afternoon with lower concentrations of primary pollutants and with a peak O_3 of 143 ppb.[10]

Figure 5 shows the results from a calculation of ozone production efficiency. Data is from the G-1 aircraft on 3 of the days with highest O_3 observed during field campaigns in Houston (2000), Philadelphia (1999), and Phoenix (1998). The slopes in this graph show the number of O_3 molecules formed per molecule of NO_x that is used up (converted to oxidation products – denoted as NO_z). In these 3 cities 20-30 ppb of NO_x was used in forming O_3. Maximum O_3 levels differ because the efficiency of O_3 production varies from city to city. Calculations indicate that differences in O_3 production efficiency are due largely to differences in VOC reactivity. Efficiencies are much higher in Houston because of reactive VOCs from petrochemical facilities.

2.3 2001 Summer Intensive

The summer of 2001 was the time that brought together the largest capability to investigate the particulate matter associated with air pollution investigations. The third summer intensive had originally

been planned to take place in 2000, but it was decided to delay until 2001 because of the opportunity to bring a special focus for the three Supersite activities planned for July 2001. The EPA Supersites established in New York, Pittsburgh and Baltimore had planned to operate during July 2001 and none of these efforts included the type of investigations for profiling the atmosphere like those carried out in NARSTO-NE-OPS. Since our Philadelphia NE-OPS site is centrally located relative to these sites, it was logical to move our measurement program to coincide with the Supersite activity. The results provide the regional context for the air pollution during the campaign. The NE-OPS data includes vertical sounding for the meteorological and air quality soundings and a few periods when aircraft measurements extend the regional description. The modeling[12-13] and regional forecasting description that the NE-OPS investigators have provided should be extremely valuable for the analysis and interpretation of the Supersite results.

Two examples of the lidar measurements from the summer 2001 campaign are shown in Figures 6 and 7. Figure 6 shows an example of the water vapor profiles measured by the Raman lidar together with the profile measured by a rawinsonde. The lidar profile shows the ±1σ error due to the photon counting statistics. The small differences between the profiles are due to the fact that the lidar profile averages the local vertical profile and the sonde provides the measurement at a point in space which drifts 10's of kilometers horizontally while rising to 5 km altitude. Figure 7 shows the time sequence of the lidar measurements of water vapor and ozone profiles on 10 July 2001. In Table 3, this period was identified for special study because of the sudden ozone event that occurred. The fact that the rapid onset of the ozone event was coupled to the sudden convection and rise of the PBL from 300 m to 1.5 km suggests triggering due to precursor materials aloft being transported to the surface where thermal decomposition could provide a ready source.

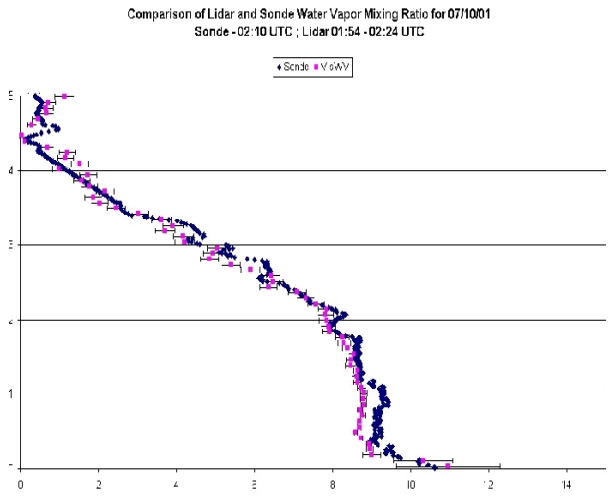

Figure 6. The profiles of water vapor measured by the rawinsonde and lidar at the same time are shown for comparison.

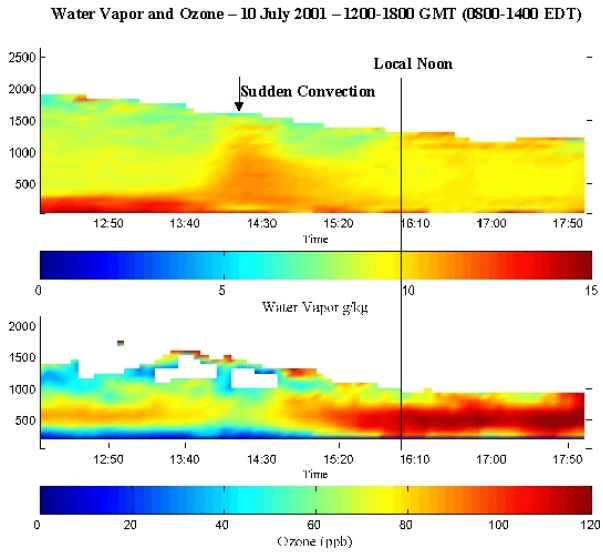

Figure 7. A time sequence of water vapor and ozone are shown for the 10 July 2001 event when a sudden increase in ozone occurred following a rapid rise in the PBL (observed in the water vapor) as the upper clouds thinned to allow heating of the surface.

2.4 2002 Summer Intensive

The NEOPS-DEP 2002 measurement program was conducted between 29 June and 7 August 2002. Several special observation periods were identified during the summer when intensive efforts were made to maximize the data collection, see Table 2. There were several periods that could not be covered by all of the instruments due to various instrument problems, however a major data set was obtained which will be important for many investigations. It is interesting to note that the Millersville University tethersonde balloon was used to obtain 541 data sets, which consist of ~530 vertical profiles and several sets of data with time sequences at one altitude.

The LAPS lidar uses Raman scattering techniques to determine profiles of meteorological and optical properties in the lower atmosphere. Table 3 lists the measurements obtained and the typical altitude range of the data products expected from the lidar instrument. The ratio of the Raman scatter signals from vibrational states of water vapor and nitrogen provide a robust measurement of the specific humidity in the lower atmosphere. The temperature profile is measured using the rotational Raman scattering at 530 and 528 nm from the 2nd harmonic of the Nd:YAG laser. In addition, the optical extinction profiles are determined from the gradients in each of the measured molecular profiles compared to the molecular scale height. The 4th harmonic of the Nd:YAG laser at 266 nm produces Raman scattering at 284 nm (nitrogen), 278nm (oxygen) and 294 nm (water vapor). The 284 nm (nitrogen), 530 nm (rotational Raman) and 607 nm (nitrogen vibrational Raman) signals are used to determine profiles of optical extinction. The ozone profiles in the lower troposphere are measured from a DIAL analysis of the ratio of the vibrational Raman signals for nitrogen (284 nm) and oxygen (277 nm). These two

wavelengths are on the steep side of the Hartley band of ozone. Since the signal ratio for these species should be fixed by the constant O_2/N_2 mixing ratio in the lower atmosphere, the departures from that ratio can be interpreted to provide an ozone profile in the region between the surface and 2 km.

Table 3. Properties of the atmosphere measured using the LAPS lidar instrument.

Property	Measurement	Altitude	Time Resolution
Ozone	276/285 Raman/DIAL	Surface to 2.5 km	Day-Night (30 min)
Extinction - 530 nm	530 nm Rot. Raman	Surface to 5 km	Night (10-30 min)
Extinction - 607 nm	607 N_2 - 1st Stokes	Surface to 5 km	Night (10-30 min)
Extinction - 285 nm	285 N_2 - 1st Stokes	Surface to 3 km	Day-Night (30 min)
Water Vapor	660/607 294/285 Raman	0 to 5 km 0 to 3 km	Night Day-Night (1 min)
Temperature	528/530 Rot. Raman	Surface to 5 km	Night (30 min)

A list of the research groups participating in the NEOPS-DEP 2002 is provided in Table 4 and a summary of the measurements is given in Table 5. The measurement program for the NEOPS-DEP 2002 represents a much smaller scale effort, however those techniques that were deemed necessary were included. Figure 8 shows the surface instrument results for ozone and the particulate matter and indicates intensive periods.

Table 4. List of measurements during NEOPS-DEP 2002

Penn State University - Engineering - Russell Philbrick
1) Raman Lidar - Profiles of Specific Humidity, Temperature, Ozone, Optical Ext (285, 530 and 607 nm)
2) Radar-RASS - Wind velocity, Virtual Temperature
3) 10 m Tower- Temperature, dew point, relative humidity, wind velocity, gust, solar flux, pressure, precipitation

Penn State University - Meteorology - Bill Ryan
Ozone and PM event forecasting (with Univ. Maryland), description of evolution of episodes and met modeling

Millersville University - Richard Clark
Tethered Balloon 15 m^3 - 300 m up/down scan each hour
1) Personal Envir. Monitors (PEMS) integrated sample
2) Diode laser scatterometer (DustTraks) continuous data
3) Meteorological properties: T, ρ, RH, wind velocity
4) O_3 by KI oxidation method, 1 m altitude resolution
Surface Gas and Particles - O_3, $NO/NO_2/NO_X$, SO_2, CO, 3-λ Nephelometer
Meteorological Data Archive - Radar, Satellite Images, Upper air data, ETA/RUC model output

Drexel University - Steve McDow
1) Organics in PM2.5 with GCMS analysis: non-polar components (alkanes, PAH), acids and diacids
2) Polar Organics for GCMS, derivatization using PM_{10}
3) Metals with Inductively Coupled Plasma Mass Spectrometer (ICPMS) low volume teflon membrane filter daily 24 hour sample

Clarkson University - Phil Hopke
1) PM2.5 with 0.5 hr resolution using RAMS, TEOM (30C)
2) PM2.5 with 1hr resolution using CAMMs instrument
3) Nephelometers- with and without dryer
4) Black carbon soot aethalometer, EC/OC analyzer

Table 5. Summary of Measurements for Summer 2002

Trace Gases:
1. Vertical profiles of ozone and water vapor obtained with the Penn State University Raman Lidar
2. Millersville University uses Advanced Pollution Instrumentation (API) analyzers and a complete suite of on-site calibrators to measure CO, O_3, SO_2, and $NO/NO_2/NO_X$
3. Aloft Ozone concentration obtained using the Millersville University tethered atmospheric sounding system (TASS).
4. Air toxics and the minor species composition of particulate matter are collected with PEMS on the tethered balloon and measured using GC/MS laboratory analysis of filter material gathered at the site and measured by Drexel University.

Particulates:
1. The PSU Raman Lidar obtains vertical profiles of optical extinction at visible and ultraviolet wavelengths
2. Millersville profiles the PM between the surface and 300 m using an optical scattering instrument suspended on the 15 m^3 balloon.
3. Clarkson University measures the particulate properties using CAMMS, RAMS, Sunset Labs OC/EC, HSPH $SO_4^=$, aethalometer, and nephelometer to describe the PM concentrations and properties.
4. Millersville University uses a three-wavelength nephelometer (TSI model 3563) to measure total and backscatter at three wavelengths as proxy for particle concentration.
5. Integrated dry mass PM fine is obtained by Millersville University using impaction sampling on Personal Environmental Monitors (PEMs, SKC Inc.).
6. Laser diode particle nephelometers are used both at the surface and aloft (on the TASS) balloon for measurements of scattering due to PM fine.
7. Particle filter samples are analyzed at Drexel University using laboratory techniques to investigate the toxics and minor constituents of aerosol particles.

Meteorological Variables:
1. The PSU Radar/RASS instrument is used to monitor the vertical profiles of wind velocity between the surface and 4.5 km and the virtual temperature to 2.5 km.
2. The PSU Raman Lidar provides vertical profiles of water vapor and temperature.
3. Meteorological variables (T, p, mixing ratio, wind speed and direction) as a function of height (to ~ 300 meters AGL) and time during episodes are obtained using TASS.
4. Continuous measurements of meteorological variables at the surface and on a 10-meter meteorological tower are recorded by PSU.

Real-time Regional and Synoptic Data Archive:
U.S. upper air data; U.S. surface data; Meso-eta and RUC model data; Visible and IR satellite imagery; Base reflectivity and velocity radar imagery are archived by Millersville University and Penn State University.

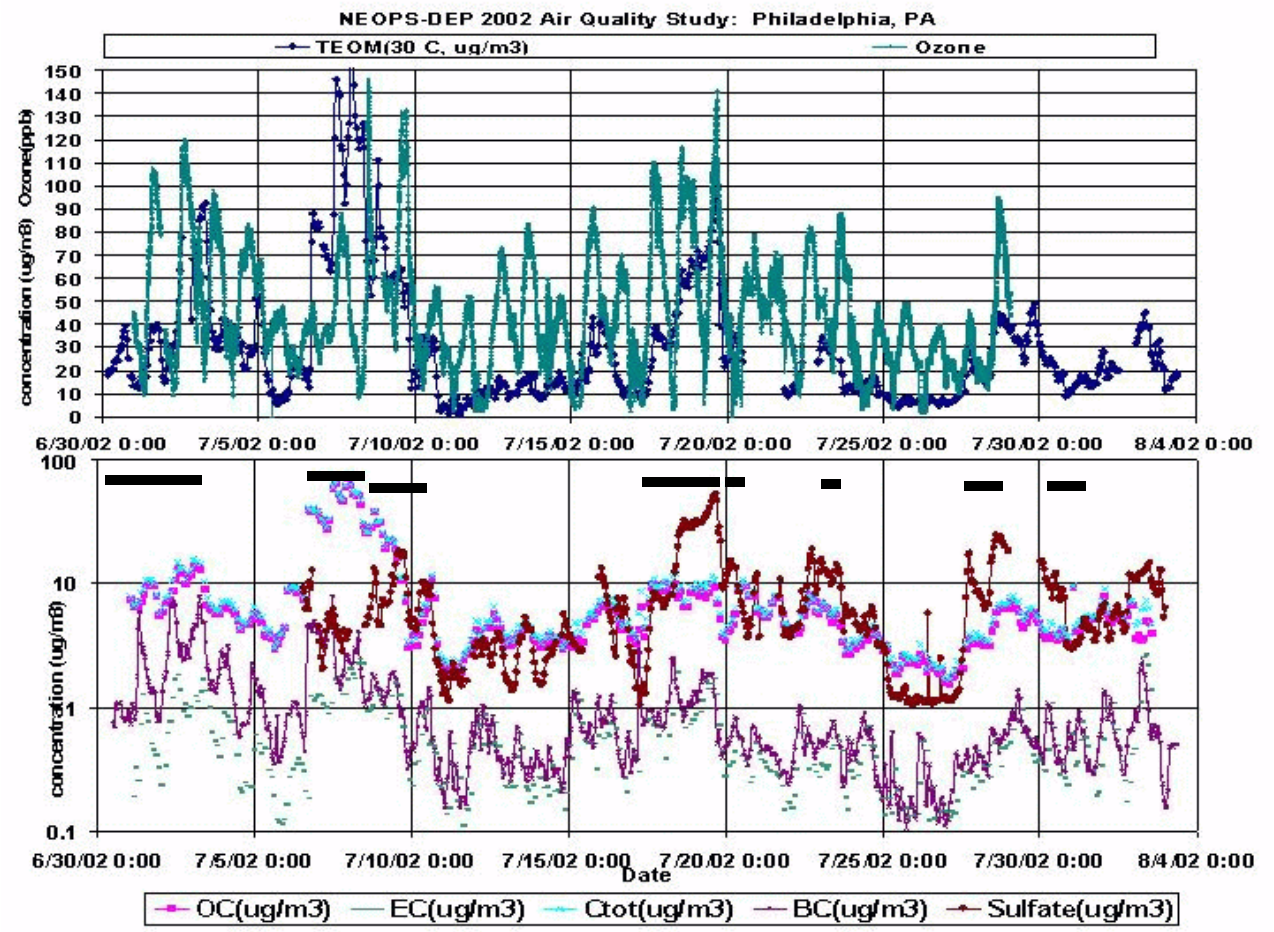

Figure 8. Results on particulate properties measured by Clarkson University and ozone measurements from Millersville University are combined to show the variations that occurred during the NEOPS-DEP 2002 campaign at the Philadelphia site. The time periods for special studies are indicated at the top of the lower panel.

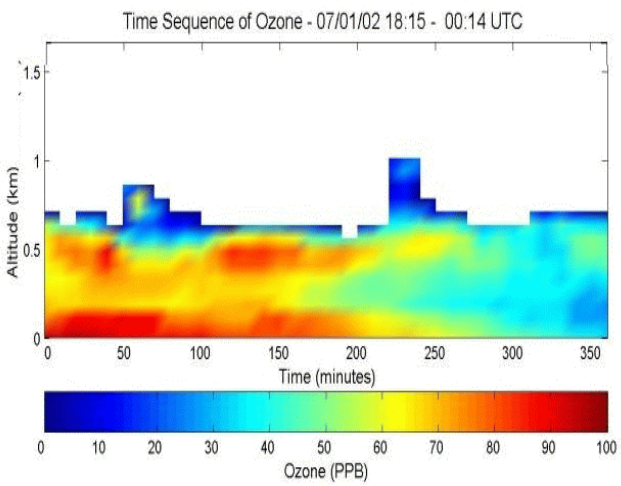

Figure 9. A time sequence of ozone on the afternoon and early evening of 1 July 2002 shows the transition to night conditions during the first stages of an ozone episode.

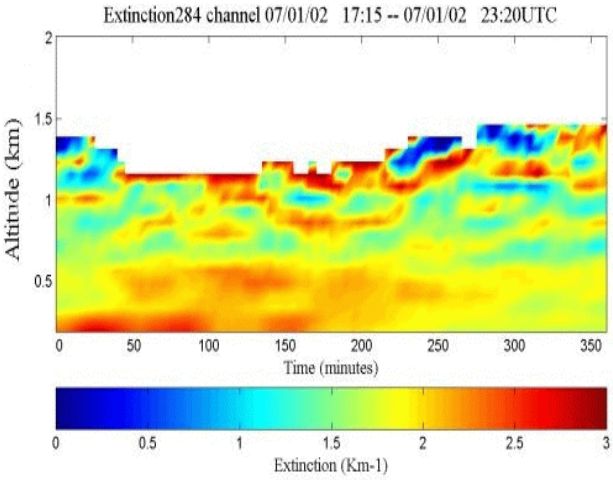

Figure 10. The optical extinction is shown for the same time as the ozone in Figure 9.

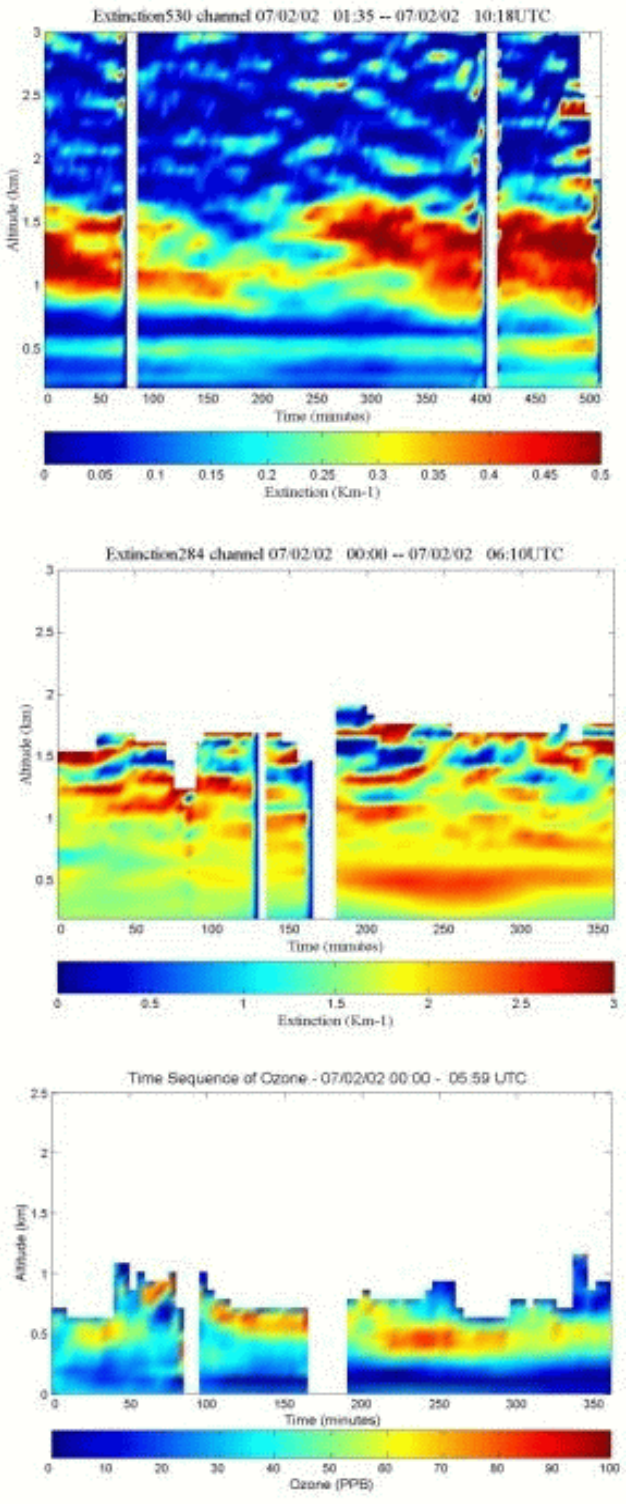

Figure 11. The optical extinction at visible and ultraviolet wavelengths are compared with the ozone measurements at the same time period. The UV extinction shows the expected response to ozone absorption and the visible extinction is most sensitive to the larger particles that are present between 1.5 and 2 km due to higher relative humidity.

The results in Figure 8 provide a useful overview of the summer 2002 conditions. The plot shows the large daily variation in the ozone concentration and identifies those periods of poor air quality. The first major episode of the 2002 summer occurred between 30 June and 4 July. Several of the examples presented here in Figures 9 - 11 are selected from this episode. Other periods of peak ozone concentration are seen to occur in 8, 10 and 19 July. Major PM events are seen to occur on 6-8 July and 18-19 July. It is interesting to compare these periods because they represent the two major processes causing PM events. The first period, 6-8 July, occurs because of major forest fire smoke from southeastern Canada and contains a large carbon component. The second event on 18-19 July is more typical of the PM component generated by smog air chemistry and contains a large contribution from sulfate aerosol.

The measurements in Figures 9 and 10 show the ozone and optical extinction measurements from the Raman lidar on 1 July 2002. The figures show the last six hours of the UTC day, approximately 2 PM to 8 PM local time. The transition periods between day and night are particularly interesting because these periods show the ozone stored aloft in the nighttime reservoir of the residual boundary layer. In this case, the extinction signature, which is partly due to ozone absorption and partly to small aerosol scattering has a very similar distribution to the ozone concentration.

Figure 11 compares the optical extinction at both visible and ultraviolet wavelengths with the ozone distribution during the first 6 hours of 2 July (8 PM on 1 July to 2 AM on 2 July local time). The time period of these measurements is immediately following the measurements shown in Figures 9 and 10. The extinction at visible wavelengths is maximum between 1 and 1.5 km because this region corresponds to high relative humidity which grows the aerosol particle size and increases the scattering. The ultraviolet extinction has a much larger component from molecular scatter and from ozone absorption and shows strong correspondence. The ozone result shows the nighttime reservoir of ozone above the nocturnal inversion. During this night a low level jet formed which transported ozone into the region from upstream formation on the previous afternoon.[11,12]

3. SUMMARY

The goal of the NEOPS project was to bring together the best and most up-to-date techniques at an urban site to measure the detailed chemistry, aerosol properties, profiles of ozone, profiles of aerosols, and meteorological conditions to archive an improved understanding of the physical and chemical processes that govern air pollution. Models and calculations have been developed based upon the results and used to interpret and better understand the polluted urban atmosphere. The purpose is to understand the meteorological influences upon the evolution and distribution of ozone and fine particle concentrations in air pollution episodes in the context of regional scale factors.

The first set of combined measurements obtained in the 1998 campaign, clearly showed the importance of transported aged materials in development of a significant pollution event. Since those first measurements, several sets of data have been obtained and many of the features of major episodes are found to be common. The important measurements of vertical profiles obtained with lidar, radar, tethersondes and aircraft spirals clearly shows the incursion of processed precursor materials in an aloft layer, which are transported into the region, and then mix downward to the surface by the daytime convective boundary layer. The transported material appears to be important in the initiation of an ozone and particulate matter pollution events during several investigations. Vertical profiles of the ozone, aerosol extinction, water vapor and other meteorological parameters provide the important results to understand the evolution of air pollution episodes. Time sequence profiles clearly show the advection of aloft layers of polluted air into the region followed by entrainment into the daytime convective mixed layer and subsequent transport to the surface. Time sequences of ozone and aerosol (PM) profiles obtained during the NE-OPS project studies between the surface and 3 km altitude show the importance of the surface layer (first 100 meters) and PBL dynamics in determining the actual pollution hazard for the regional population. Hazardous levels of ozone are frequently observed existing above the surface layer. The intensity of vertical transport, and presence of surface layer gases that can destroy ozone, can result in differences in human exposure. Examination of several episodes demonstrates that the local surface exposure depends upon the development of convective activity, wind-shear driven transport, and presence of surface gaseous species. The results show the importance of measuring the vertical structure to understand the population exposure at the surface. The results from these measurement programs are now being prepared for archiving so that the larger community of modelers and researchers will have access to this valuable data set.

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