

OVERVIEW OF THE NARSTO-NE-OPS PROGRAM

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ABSTRACT

The NARSTO-NE-OPS (NorthEast 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 three major field programs carried out at a field site in northeast Philadelphia during the summers of 1998, 1999 and 2001. The activity brings together the research groups from 13 universities, 5 government laboratories and representatives of the electric power industry to apply the most advanced measurement techniques to understanding the physical and chemical processes contributing to air quality issues. Results have been obtained from three ground sites, two instrumented aircraft, many different instrumented balloon platforms, and several remote sensing techniques including satellites. In addition, the database used includes the ground-based measurements conducted at several surrounding state and city operated sites in Pennsylvania, New Jersey, Delaware, New York and Maryland. The results have shown the importance of developing the 3-dimensional regional scale picture of the atmosphere to understand and properly model air pollution events. It has been shown that only from such a perspective, which includes the vertical distribution and a regional context, can one hope to properly model and predict ozone and particulate pollution. A combination of photochemical and dynamical processes transport, both horizontal and vertical, accumulate pollutants that then mix with the locally produced chemical species to cause the more severe episodes of air pollution. Efforts have also focused on developmental testing of several new approaches to improved measuring techniques for better understanding of the physical and chemical properties of the airborne particulate matter.

1. INTRODUCTION

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). 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

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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 name adopted for our project, NARSTO-NE-OPS, signifies that our project is part of the North American Research Strategy for Tropospheric Ozone (NARSTO) and is conducted in the Northeast region and focused on Oxidant and Particle Studies (NE-OPS). The affiliation of our program with the NARSTO program provides an established focus for the data quality control and data archiving. Measurements that are conducted as part of the NARSTO-NE-OPS project during the three summer intensive periods are listed in Tables 1-3. The research groups performing the measurements, the measurement parameters, and the methods used are indicated in these tables. In addition to the field measurement activity, the NARSTO-NE-OPS project also includes several modeling and theoretical calculation activities. The major modeling activities are being carried out by the groups listed in Table 4.

Table 1. Measurements obtained in August 1998 NARSTO-NE-OPS Campaign

Penn State University - Russell Philbrick

Raman Lidar - Profiles of Specific Humidity, Temperature, Ozone, Optical Extinction (285, 530 and 607 nm)

Millersville University - Richard Clark

Tethered Balloon - 100 m³ - 10 hr aloft with sensors at surface, 100 m, 200 m, and 300 m AGL

1) Personal Environmental Monitors (PEMS) 4 each - 4 L/min dry PM 10 hr integrated sample

2) Diode laser scatterometer (DustTraks) 1.7 L/min continuous data

Tethered Balloon - 7 m³ - up/down scan to 300 m each hour

Meteorological properties: T, D, RH, wind speed and direction 1 m vertical resolution and O₃ by KI oxidation method

Surface Measurements - O₃ and meteorological data

Meteorological Data Archive - Radar, Satellite Images, Surface Observations, Upper air data, ETA/RUC model output

Harvard School of Public Health - Petros Koutrakis and George Allen

Mass density of particulates: PM₁, PM_{2.5}, PM₁₀, aerosol-size, EC/OC, sulfate, nitrate, toxics

Harvard University - Bill Munger

NO_y concentrations and fluxes are used to infer the rates for NO_x oxidation and deposition.

University of Maryland - Bruce Doddridge and Bill Ryan

Instrumented Aircraft Cessna 170: Global Positioning System, Ozone, Carbon Monoxide, and a temperature/relative humidity probe
Ozone and PM event forecasting, description of evolution of interesting episodes and meteorological modeling

Drexel University - Steve McDow

1) Organics in PM_{2.5} with GCMS analysis: non-polar components (alkanes, PAH), acids and diacids

2) Polar Organics for GCMS with derivatization using PM-10 with composite samples

Table 2. List of measurements obtained during the June-August 1999 NARSTO-NE-OPS Campaign.

Penn State University - Russell Philbrick

1) Raman Lidar - Profiles of Specific Humidity, Temperature, Ozone, Optical Extinction (285, 530 and 607 nm)

2) Radar-RASS - Wind velocity, Virtual Temperature

3) 10 m Tower - Temperature, dew point, relative humidity, wind velocity, wind gust, solar flux, atmospheric pressure, precipitation

Millersville University - Richard Clark

Tethered Balloon - 100 m³ - 10 hr aloft with sensors at surface, 100 m, 200 m, and 300 m AGL

1) Personal Environmental Monitors (PEMS) 4 each - 4 L/min dry PM 10 hr integrated sample

2) Diode laser scatterometer (DustTraks) 1.7 L/min continuous data

3) VOC - Micro-orifice vacuum canister at surface and at 300 meters, 10 hour accumulated sample with GC/MS lab analysis

Tethered Balloon - 7 m³ - up/down scan to 300 m each hour

1) Meteorological properties: T, D, RH, wind speed and direction 1 m vertical resolution

2) O₃ by KI oxidation method, 2-3 second time resolution (1 meter altitude)

Surface Measurements - O₃ and meteorological data

Meteorological Data Archive - Radar, Satellite Images, Surface Observations, Upper air data, ETA/RUC model output

Harvard School of Public Health - Petros Koutrakis and George Allen

Mass density of particulates: PM₁, PM_{2.5}, PM₁₀, aerosol-size, EC/OC, sulfate, nitrate, toxics

Harvard University - Bill Munger

NO_y concentrations and fluxes are used to infer the rates for NO_x oxidation and deposition.

University of Maryland - Bruce Doddridge and Bill Ryan

Instrumented Aircraft Cessna 170 and Aztec: Global Positioning System, Ozone, Carbon Monoxide, and a temperature/relative humidity probe
Ozone and PM event forecasting, description of evolution of interesting episodes and meteorological modeling

Drexel University - Steve McDow

1) Organics in PM_{2.5} with GCMS analysis: non-polar components (alkanes, PAH), acids and diacids using Hi-Vol 24 hour sample

2) Polar Organics for GCMS with derivatization using PM-10 with composite samples

Brookhaven National Laboratory - Peter Daum, Larry Kleinman, Yin-Nan Lee, Stephen Springston

DOE G-1 Instrumented Aircraft - particulate and gas-phase chemistry

Brigham Young University - D. Eatough

Measurement of particle volatile mass component and identification of volatile species with RAMS and PCBOSS

Pacific Northwest National Laboratory - C. Doren, J. Allwine, J. Fast, C. Berkowitz

Radiosondes - Pressure, temperature, humidity 0-15 km at Philadelphia, Radar-RASS instrument at West Chester, 12 ozonesondes at Philadelphia

Argonne National Laboratory - R. Coulter, J. Gaffney, N.A. Marley

Radiosondes, SODAR and Chemistry Laboratory at Centerton NJ

N. Carolina A&T State University - D. Dunn

Remote sensing with lidar and SODAR

N. C. State University - H. Hallen

Laser remote sensing, particle optical scattering properties

Table 3. List of measurements obtained during the July 2001 NARSTO-NE-OPS Campaign

Penn State University - Electrical Engineering - Russell Philbrick

- 1) Raman Lidar - Profiles of Specific Humidity, Temperature, Ozone, Optical Extinction (285, 530 and 607 nm)
- 2) Radar-RASS - Wind velocity, Virtual Temperature
- 3) 10 m Tower - Temperature, dew point, relative humidity, wind velocity, wind gust, solar flux, atmospheric pressure, precipitation
- 4) Radiosondes - Pressure, temperature, humidity 0-15 km

Penn State University - Meteorology - Bill Ryan and Nelson Seaman

Ozone and PM event forecasting (with Univ. Maryland), description of evolution of interesting episodes and meteorological modeling

Millersville University - Richard Clark

Tethered Balloon - 100 m³ - 10 hr aloft with sensors at surface, 100 m, 200 m, and 300 m AGL

- 1) Personal Environmental Monitors (PEMS) 4 each - 4 L/min dry PM 10 hr integrated sample
- 2) Diode laser scatterometer (DustTraks) 1.7 L/min continuous data
- 3) VOC - Micro-orifice vacuum canister at surface and at 300 meters, 10 hour accumulated sample with GC/MS lab analysis

Tethered Balloon - 7 m³ - up/down scan to 300 m each hour

- 1) Meteorological properties: T, D, RH, wind speed and direction 1 m vertical resolution
- 2) O₃ by KI oxidation method, 2-3 second time resolution (1 meter altitude)

Surface Gas and Particles - O₃, NO/NO₂/NO_x, SO₂, CO, 3-8 Nephelometer

Meteorological Data Archive - Radar, Satellite Images, Surface Observations, Upper air data, ETA/RUC model output

Harvard School of Public Health - Petros Koutrakis and George Allen

Particle Size and Count: 0.02 to 0.6 : m electrostatic classification, 0.7 to 15 : m time of flight, PM_{2.5} CAMM, Black carbon soot aethalometer, sulfate from HSPH thermal conversion method, EC/OC analyzer

Particulate 10-hour Day/Night Samples: HEADS for acid gases [HNO₃, HONO, SO₂], NH₃, and sulfate/nitrate/strong aerosol acidity EC/OC on quartz filters with DRI's TOR analysis, PM_{2.5} and PM₁₀ from Harvard impactors with Teflon filters and gravimetric analysis daily, Hivolume OC speciation sampler, HSPH PUF substrate collection; Drexel University selected filter analysis

Harvard University - College of Engineering - Bill Munger

NO_y concentrations and fluxes are used to infer the rates for NO_x oxidation and deposition.

University of Maryland - Bruce Doddridge and Russ Dickerson

Instrumented Aircraft Piper Aztec: Particle-Soot Absorption Photometer, Global Positioning System, Ozone, Sulfur Dioxide, Carbon Monoxide, 3-wavelength integrating nephelometer and a temperature/relative humidity probe

AERONET (Aerosol Robotic Network) data available include AOT at 1020, 870, 670, 500, 440, 380, and 340 nm plus precipitable Water (cm).

Drexel University - Steve McDow

- 1) Organics in PM_{2.5} with GCMS analysis: non-polar components (alkanes, PAH), acids and diacids using Hi-Vol 24 hour sample
- 2) Polar Organics for GCMS with derivatization using PM-10 with composite weekly samples
- 3) Metals with Inductively Coupled Plasma Mass Spectrometer (ICPMS) low volume teflon membrane filter daily 24 hour sample

Clarkson University - Phil Hopke and Alex Polissar

- 1) PM_{2.5} with 0.5 hr resolution using RAMS, TEOM and 3OC
- 2) PM_{2.5} with 1hr resolution using CAMM's instrument
- 3) Nephelometers- with and without dryer

EPA - RTP & Texas Tech University - Bill McClenny (EPA) and Sandy Dasgupta (Texas Tech)

- 1) Fluorescence Detector: H₂O₂, HCHO, MHP, NH₃
- 2) Ion Chromatography: Sulfur Dioxide, Nitric Acid, Nitrous Acid, HCl, Oxalic Acid, Oxalate, Nitrate, Nitrite, Sulfate, Chloride, Ammonium

Brookhaven National Lab - Larry Kleinman, Linda Nunnermacker, Xiao-Ying Yu, Yin-Nan Lee, Stephen Springston

IC measurements of cations: Na⁺, K⁺, NH₄⁺, Ca₂⁺ and anions: SO₄²⁻, NO₃⁻, Cl⁻, NO₂⁻, oxalate; 3-channel NO_x, CO, Ozone, SO₂

Carnegie Mellon University - Spyros Pandis

Particulate Matter 0.02 to 0.6 um, electrostatic classification, run dry

Philadelphia Air Management Services - Fred Hauptman and Lori Condon

Speciation Air Sampling System: PM_{2.5} mass, trace metals, organic and elemental carbon, sulfate, nitrate, and other ions/elements

Table 4. Modeling activities included as part of the NARSTO-NE-OPS program.

State University of New York at Albany - S. T. Rao, V. Mohnen, I. Zurbenko, S. Porter, K. Civerolo

Regional analysis and model calculations of polluted air masses

Rutgers University, Env. & Occup. Health - P. Georgopoulos and M. Lazaridis
Emissions inventories and chemistry modeling, particular experience in Philadelphia area

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 MEASUREMENTS CONDUCTED

The NARSTO-NE-OPS project has included three major measurement programs. The participants and measurements conducted during the campaigns in the summers of 1998, 1999 and 2001 are summarized in Tables 1, 2 and 3. Significant measurement periods that have been selected for specific investigations are summarized in Table 5. 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 O₃ 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 plan to delay the NE-OPS measurements from summer 2000 to summer 2001 was an effort to provide an important coordination role in relating the measurements of the three Northeastern Supersites in New York, Pittsburgh and Baltimore. The NE-OPS measurements in Philadelphia provide a central location and the important regional measurements of the meteorological properties using remote sensing and aircraft measurements. Each of the campaigns are briefly described below.

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 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 : g/m³) were observed. Results from the episode are presented in Figure 1. 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.

Table 5. Summary of the intensive observational periods during 1998, 1999 and 2001 campaigns.

Summer 98		7-22 Aug 1998
1	7-22 Aug	Pilot study to prepare site and 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 change, UMD A/C
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, very strong stable PBL, wind shift, RASS and sondes
5	27J-1 Aug	West flow brings increase in ozone on 27 th , regional convection limits ozone on 28 th , 31 st 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 12 th followed by storm on night of 13 th
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	Sudden event that is short duration, begins with interesting convective activity
2	16-17 July	Ozone and PM event
3	20-25 July	Moderate levels of ozone repeated on succession of 4 days

The primary purposes for making the push to perform the 1998 pilot study were to prepare the measurement site in Philadelphia that would be used for measurements during the next 3 years and to gather data to investigate the comparisons of the instrument techniques. During the pilot study, the most significant pollution episode of the 1998 summer occurred.

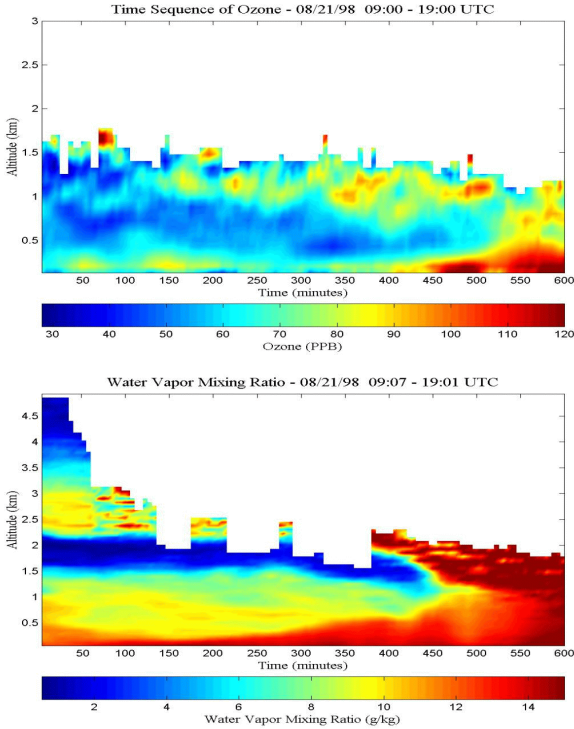


Figure 1. Raman lidar profiles show the growth of the PBL during the morning of 21 August using the water vapor as a tracer. Merging of the rising PBL with an upper layer, which originated in the mid-west, appears to initiate the ozone and PM event that occurred that afternoon.

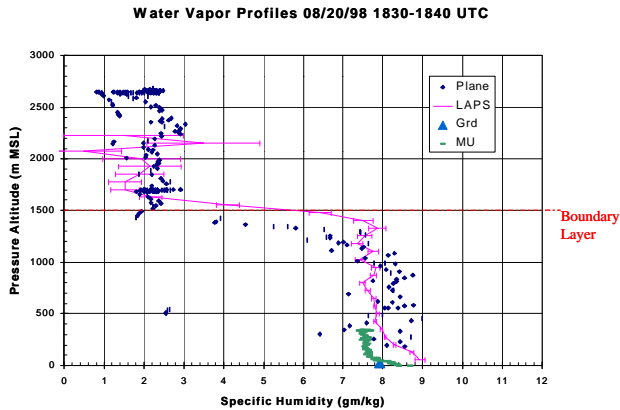


Figure 2. Water vapor profiles from lidar, tethered sonde and aircraft are compared during a daytime period when the convective turbulence causes large variations.

The Millersville University tethered sonde instrument is able to provide high resolution profiles from the surface to 300 meter each 30 minutes using a 4 meter long (5 m³) tethered balloon. During the summer of 1998, 176 vertical profiles were obtained. The instrument package measures profiles of the temperature, pressure, wind velocity, relative humidity and ozone. A larger 12 m long

(100 m³) 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.⁶ 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.⁷ Figures 2 and 3 show examples of the comparisons of the lidar, tethered sonde and aircraft profiles. Generally good agreement between the sensors has been found and detail comparisons will be presented in reports that are in preparation.

Ozone Profile 08/20/98 0245-0335 UTC

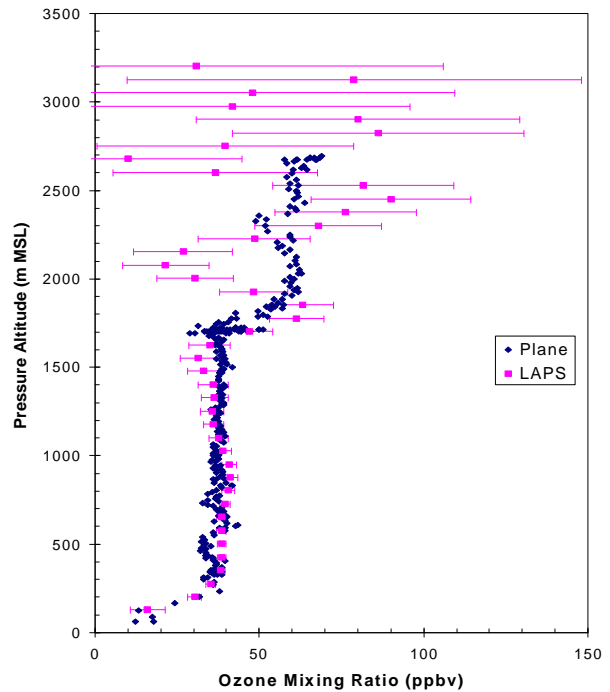


Figure 3. The aircraft and lidar profiles of ozone are compared during a spiral by the aircraft near the vertical beam of the lidar. The ±1F error associated with the photon counting is indicated on each lidar measurement.

1999 Summer Intensive

Advanced techniques used during the NARSTO-NE-OPS project included Raman lidar, tethered 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.⁸ Profiles of the properties of the surface layer to 300 meters were obtained by Millersville University using tethered sondes 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 flights⁷ 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. The contributions of the researchers participating in the project are shown in Table 2.

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 NO_x/NO_y chemistry by Harvard University. Profiles of ozone, optical extinction and meteorological parameters and optical extinction 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 of CO and UV radiation measurements at the site and conducted instrumented flights with Cessna and Aztec aircrafts.⁶ Brookhaven National Lab made 19 regional flights using the instrumented DOE-G1 aircraft between 25 July and 11 August. 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. 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 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 O₃ formation included O₃, CO, VOCs, NO, NO_y, SO₂, HCHO, H₂O₂, 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 O₃ production. Highest O₃ 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 O₃ 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 SO₂ 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 O₃ of 143 ppb.

Figure 4 shows the results from a calculation of ozone production efficiency. Data is from the G-1 aircraft on 3 of the days with highest O₃ observed during field campaigns in Houston (2000), Philadelphia (1999), and Phoenix (1998). The slopes in this graph show the number of O₃ molecules formed per molecule of NO_x that is used up (converted to oxidation products – denoted as NO₂). In these 3 cities 20-30 ppb of NO_x was used in forming O₃. Maximum O₃ levels differ because the efficiency of O₃ production varies from city to city. Our calculations indicate that differences in O₃ production efficiency are due largely to differences in VOC reactivity. Efficiencies are much higher in Houston because of reactive VOCs from petrochemical facilities.

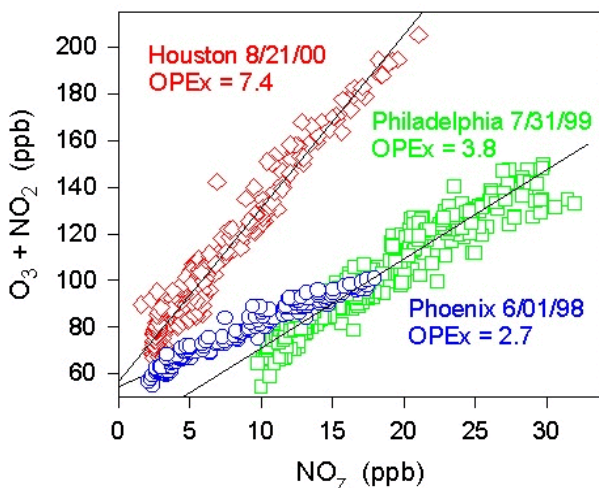


Figure 4. DOE G-1 aircraft data from high ozone days used to calculate ozone production efficiency in 3 cities.

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, see Table 3. 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 that have been carried out in NARSTO-NE-OPS. Since our Philadelphia NE-OPS site is centrally located relative to these sites, it appeared logical to move our measurement program to coincide with the Supersite activity. We were quite successful in gathering the data planned for the NE-OPS site and expect that we will be able to 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 modeling⁹⁻¹⁰ and regional forecasting description that the NE-OPS investigators (see Table 4) have provided should be extremely valuable for the analysis and interpretation of the Supersite results.

3. RAMAN LIDAR CONTRIBUTION

One of the special new instruments that has made major contributions to the NE-OPS program is the Raman lidar technique, which has been developed at Penn State University. The lidar instrument used in each of the NE-OPS campaigns is the LAPS (Lidar Atmospheric Profiles Sensor). LAPS is a fifth generation Raman lidar instrument developed by our group since 1978 and it is the first operational prototype instrument to be prepared. Raman scattering 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 specific vibrational energy states of the molecules and rotational Raman scattering provides a signal with a wavelength variation that depends directly upon the atmospheric temperature. The LAPS lidar uses Raman scattering techniques to determine profiles of meteorological and optical properties in the lower atmosphere. Table 6 lists the measurements obtained and the typical altitude range of the data products expected. 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 measured 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 O₂/N₂ 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 3 km.

Two examples of the lidar measurements from the summer 2001 campaign are shown in Figures 5 and 6. Figure 5 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 $\pm 1\sigma$ 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 6 shows the time sequence of the lidar measurements of water vapor and ozone on 10 July 2001. In Table 5, 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.

Table 6. 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 - 1 st Stokes	Surface to 5 km	Night (10-30 min)
Extinction - 285 nm	285 N ₂ - 1 st 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)

4. SUMMARY

The goal of this 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. Aircraft measurements (BNL and UMD) were used to provide regional and local variations of the chemistry (precursors of ozone and aerosols) and particulate matter. Models and calculations have been developed and used to interpret and 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. The result was obtained during the pilot study of the NE-OPS project in Philadelphia in August 1998. The unique set of measurements from vertical profiles obtained with lidar, tether sondes and aircraft spirals clearly shows the incursion of processed precursor materials in an aloft layer, which was transported into the region, and then mixed downward to the surface by the rising daytime convective boundary layer. The transported material appears to be important in the initiation of an ozone and particulate matter pollution event on 21 August 1998. Vertical profiles of the ozone, aerosol extinction, water vapor and other meteorological parameters were obtained continuously over a period of three days. These time sequence profiles clearly show the advection of an aloft layer of polluted air into the region followed by entrainment into the daytime convective mixed layer and subsequent transport to the surface. The measurements of this first depiction of the meteorological control of a pollution event are providing a useful case study. Time sequences of ozone and aerosol (PM) profiles obtained during the NE-OPS project pilot study between the surface and 3 km altitude show the importance of the surface layer (first 100 meters) dynamics in determining the actual pollution hazard for the regional population. During the two week period of the pilot study, hazardous levels of ozone were observed several times 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 these several days of data 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 in measuring the vertical structure to understand the population exposure at the surface.

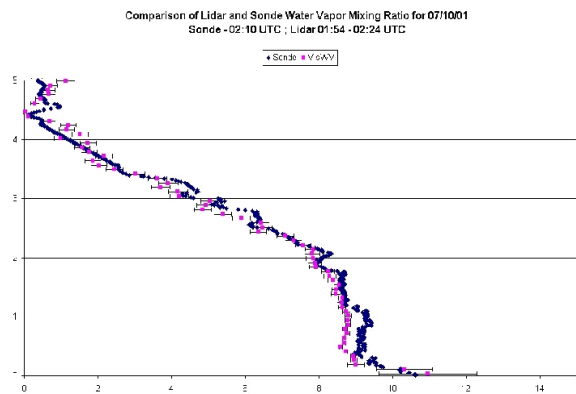


Figure 5. The profiles of water vapor measured by the rawinsonde and lidar at the same time are shown for comparison.

During 1999 there were eight periods of significance to our understanding of air pollution and during 2001 there were three more periods. The results from these studies are being presented in fourteen papers of this proceedings and in numerous papers and reports that are in progress.

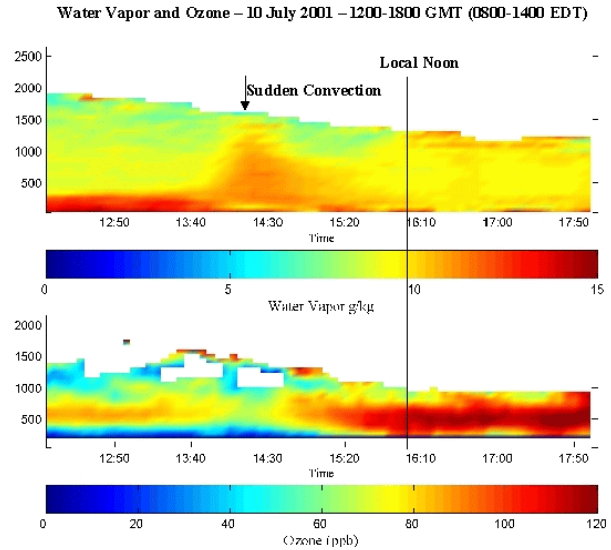


Figure 6. A time sequence of water vapor and ozone are shown for the 10 July 2001 event when a sudden increase in ozone occurred following a rapid rise in the PBL (observed in the water vapor) as the upper clouds thinned to allow heating of the surface.

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