Measurement of Toxic and Related Air Pollutants

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Investigations of Factors Determining the Occurrence of Ozone and Fine Particles in Northeastern USA

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ABSTRACT

An investigation which concentrates on the atmospheric pollution in an urban region, where the fine particle concentrations and ozone concentration levels are frequently coupled during pollution events, has been undertaken by a consortium of university investigators and research institutes. These investigations in the northeast region are focused on the urban area of Philadelphia, PA. The major goal of these air pollution studies is to understand the emission sources and atmospheric processes which cause elevated air pollutant concentrations by carefully planned measurement programs using the most advanced techniques at ground based sites and on aircraft platforms to consider both local and regional scales. Many pollutants exhibit similar spatial and temporal concentration patterns, due to photochemical, meteorological, and other factors, including the fact that they have similar precursors which have similar emissions sources. The links between ozone and fine particle mass are especially striking, but there are several significant gaps in understanding which can limit effective decision making and control strategies. The initial research efforts and plans for the three year program are described. A first year pilot study has been conducted in July/August 1998 to prepare the primary measurement site in Philadelphia PA and conduct a series of measurements that can be used to evaluate key instrument performance and assess accuracies of data products.

KEY WORDS: air pollution, ozone, particle matter, atmospheric chemistry, urban environment, atmospheric dynamics

INTRODUCTION

This paper reports on a newly formed consortium of universities and laboratories in Northeast US that has come together to provide the best and most up-to-date measurement techniques in an urban environment to measure the detailed chemistry, aerosol properties, profiles of ozone, profiles of aerosols, and profiles of meteorological conditions. Aircraft measurement techniques (conducted by University of Maryland and Brookhaven National Laboratory) are used to provide regional and local variations of the chemistry (precursors of ozone and aerosols) and particulate matter. Four regional ground sites are used to define the regional conditions, these are located at Harvard Forest MA, Whiteface Mountain NY, Mount Washington NH and Shenandoah National Park VA. The effort includes the development and testing of models and calculations to interpret and understand the source in the evolution of pollution events in the urban atmosphere, with a particular emphasis on the vertical and horizontal transport of the air parcels. The goal is to investigate the meteorological influences upon the evolution and distribution of ozone and fine particle concentrations in the urban environment within the context of important regional scale factors. This three year project was initiated in April 1998 and the first pilot study was conducted during July/ August 1998 to prepare the prime urban site and obtain a set of data that can be used to evaluate the measurement techniques and provide information for planning subsequent intensive campaigns during the periods June/August 1999, December 1999 and June/August 2000.

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The site for the primary urban measurements is the City of Philadelphia's Baxter Water Treatment Plant in northeast Philadelphia. The site is approximately 8 miles northeast of the city center, 4 miles southeast of the Northeast Philadelphia Airport and 14 miles north-northeast from the Philadelphia International Airport, and is located between US Interstate 95 and the Delaware River. The site location was chosen because it provides an optimal scientific location for performing the measurements and it is located such that the lidar and tether balloon instruments can be most safely used in a congested urban environment The site preparation has included the clearing and preparation for surface instrument hardware as well as installation of an electrical power transformer and telephone lines. During the three intensive campaigns to follow, additional smaller measuring sites will be established 20-30 km toward the southwest and northeast to provide upwind and downwind surface monitoring of the chemical species and particle matter.

The consortium includes a rather large group of co-investigators who bring expertise in the several important areas needed to successfully achieve the goals of the project. The researchers involved in this effort, along with an indication of their contributions, are listed in Table 1. The project has been given the name NEC-OPS, meaning NorthEast Corridor - Oxidant and Particle Study. The project is being submitted as a participant for the national NARSTO (North American Research Strategy for Tropospheric Ozone) program and has adopted the guidelines of the national program for data quality testing (QA/QC of the data) and distribution of data products so that the results will have maximum utility for the scientific community. Table 2 provides an outline of the planned project activities during the next three years.

URBAN SITE SELECTION

During the period between May and July 1998, the many activities associated with picking out and establishing the urban site in Philadelphia were carried out. The approvals and coordination of the FAA offices was given special attention because of the lidar, tether balloon, meteorological balloon and aircraft measurements included in the program. The property selected for the site belongs to the City of Philadelphia and is known as the Baxter Water Treatment Plant (N40°02.5',W74°59.8'). We selected Philadelphia as our target city for several reasons:

(1) Philadelphia is centrally located in this northeast corridor region where the high pollution levels are encountered,

(2) the Philadelphia area has been carefully studied with respect to its local pollution sources,

(3) cooperation with the City of Philadelphia Air Management Services Laboratory and measurements from their PAMS site are important contributions to our measurement program,

(4) Philadelphia is sufficiently far from the marine atmosphere influence so we will not need to deal with that additional complication in our data analysis,

(5) the location was deemed to be of highest scientific interest by all of the members of the investigation team.

The criteria for selection of the site for the measurements included the following points:

Northeast of city center of Philadelphia
Federal, State or City authority for the site
Clear area of several acres
Power available
Communications - phone - data access
Clearance for tether-balloon and storage for balloon
Clearance for lidar operation
Clearance for free fly balloon
24 hour access and area security

Measurements on the atmospheric pollution and meteorological control factors are planned using lidar remote sensing techniques, sensors on two tethered balloons and standard radiosonde balloons, as well as

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many types of ground level air samplers and sensors. In addition, two instrumented aircraft, one from the University of Maryland and one from Brookhaven National Laboratory (the BNL aircraft will only participate in the 1999 and 2000 experiments), will be used to gather data in the local area and on the scale of regional effects.

During the weeks of 27 July and 3 August, the site was prepared and equipment was shipped in and setup for the measurements. Electrical power, telephones, and all other arrangements needed for the personnel were brought in and setup for the campaign. The intensive measurement period extended from 9 through 22 August. During the last 4 days of the period, the instruments were run on the continuous basis that will be employed for the major ozone events during the campaigns planned for 1999 and 2000.

The primary goal of our research consortium is to investigate and characterize factors contributing to air pollution, particularly ozone and fine particles in the Philadelphia metropolitan urban environment. Since most of the air pollution events occur during the summer, we have planned to make the measurements during the summers of 1998, 1999 and 2000, with one set of winter time measurements during 1999. The meteorological conditions associated with pollution events cause us to select the primary site near the northeast side of the Philadelphia metropolitan area. This investigation places an emphasis upon the vertical distribution of the pollution components, particularly profiles of ozone and aerosols, and seeks to understand the meteorological control processes which influence the surface concentration levels as represented by the surface chemistry measurements performed by Harvard University and the City of Philadelphia PAMS site. The Penn State University lidar (see Table 3) provides the measurements of profiles from the surface to 3 km in day and 10 km at night of density, temperature (night only), ozone, water vapor, and 3-wavelength optical extinction (one wavelength during the day). The Millersville University tethersonde (see Table 4) provides profiles of meteorological parameters plus ozone and fine particles from the surface to 300 meters. In future campaigns, the RASS sounder will provide profiles of wind and temperature in the lower at atmosphere. From the combination of sensors brought together in the NEC-OPS project, the vertical distribution of the atmospheric properties will be given primary emphasis. A major focus of the project is on the measurement of the fine particles in the atmosphere. The Harvard University School of Public Health set up a suite of particle and chemical sensors at the urban site for the August 1998 measurement campaign. They will provide additional ground-based sampling during the subsequent campaigns at both the urban site and two new sites to be established upwind and downwind from the urban site. Several sets of sensors measure the fine particles, including many of the PM10 and PM2.5 filter sensors, at several ground sites, as well as on top of the 5 story building at Baxter Water Treatment Plant, and at 50 meter intervals along the tether balloon. In addition, a light scattering instrument can be used to provide a high spatial resolution profile of the concentration of scattering particles together with the meteorological package on the smaller balloon during low wind conditions. The larger balloon was deployed to 300 m carrying several of the PM2.5 samplers with their air pump and battery power at 50 meter intervals along the tether line. The normal operation for this measurement included 10 hour samples between 10 AM and 8 PM and 10 PM and 8 AM local time to provide data sets divided into day and night samples. The optical extinction from the lidar and the optical scattering data from the tether will be compared with the fine particle results by using information on relative humidity to relate the size/number of the dry nuclei compared to moist aerosol components. In addition, two instrumented aircraft, one from the University of Maryland and one from Brookhaven National Laboratory (the BNL aircraft will only participate in the 1999 and 2000 experiments), are be used to gather data in the local area and data on the scale of regional effects. The two instrumented aircraft will measure vertical structure from local spirals and will develop cross-section maps of the properties of the polluted air mass from regional transects. Only one of the aircraft was involved in the pilot measurement program, researchers from the University of Maryland operated an instrumented Cessna 172 that staged out of Bay Bridge Airport MD and used the North Philadelphia Airport for refueling and crew breaks during August 1998. During the subsequent measurement periods, the University of Maryland researchers will fly a Piper Aztec and the Brookhaven National Laboratory will be flying the highly instrumented DOE G2 aircraft to obtain the regional view of the air mass.

PRELIMINARY RESULTS DURING SUMMER 1998

As an introduction to the effort, a few examples of the data collected during the August 1998 pilot study will be presented here. During the coming months, the results obtained will be examined in detail to determine the accuracy of the measuring techniques. The measurement period of August 1998 will provide a foundation for planning the intensive measurement periods in the primary investigations of 1999 and 2000.

The PSU LAPS lidar instrument simultaneously measures several profiles of interest, temperature, water vapor, ozone and optical extinction. An example of the water vapor measurements obtained from the instrument is shown in Figure 1. The time sequence of preliminary measurements shows one minute updates of the specific humidity during a period of five hours on 20 August. The boundary layer exhibited a sharp transition on it's top with a change in the water vapor from greater than 6 gm/kg to less than 1 gm/kg in about 100 meters altitude. A single profile from 30 minute integration at 0200 UTC is shown with $\pm$ 1σ statistical error bars indicated. Each point in altitude is a unique measurement and no vertical smoothing has been applied to this data. This profile corresponds to the time period just before the middle of the accompanying time sequence plot. The time sequence was prepared with 4 minute integration of the signal at a time step of 1 minute. Time sequences are most useful in following the changes that occur in the lower atmosphere. The results shown here are but one of several parameters that were measured nearly continuously during the period of 16 - 22 August. In addition to the LAPS lidar instrument, the PSU team also operated a micro-pulse backscatter lidar (commercially produced by Science & Engineering Services Inc.) to measure the aerosol and cloud backscatter for comparison with the Raman extinction measurements.

The tethersonde balloons provide an important capability to profile the atmosphere at heights below 300 meters with high resolution. Most important characteristic features of polluted air masses occur in the first 300 meters where the interaction rates between the air mass and the surface material are governed by the local meteorological conditions. Figure 2 shows a set of simultaneous preliminary data profiles obtained during one ascent, these include temperature, relative humidity, ozone and wind speed. A similar set of profiles can be obtained about each 30 minutes during 24 hour periods that correspond to intensive measurement periods.

An example of the preliminary data on PM10 and PM2.5 particles obtained by the Harvard School of Public Health is shown in Figure 3. The measurements include determination of the particle concentrations and analysis of the chemical species from which they are formed. Local sources, such as diesel trucks, sometimes create spikes on the record. A significant particle event occurred in the area near the end of the record.

The measurements from the Philadelphia Air Management Services Laboratory for O_3, NO and NO_2 are shown in Figure 4. The ozone measurements were conducted at the downtown laboratory and at the Northeast Philadelphia Airport. Continuous measurements of the surface layer concentrations provide an important component of the air mass characterization. While this record confirms that no major pollution events occurred during the period of the pilot study, it does show several interesting features when special emphasis will be placed on the analysis of the results.

Examples of the preliminary data profiles from the University of Maryland aircraft spirals are shown in Figure 5. The ozone, temperature, water vapor, and carbon monoxide were measured during the August 1998 pilot study, an several additional properties will be measured during the subsequent intensives using a larger aircraft with additional sensors. Note that the profiles here show the same steep gradient on the top of the boundary layer as shown in the lidar results in Figure 1.

CONCLUSIONS

The purpose of this paper is to introduce the major effort that has been undertaken in the north-east by a consortium of researchers. The program goal is to investigate air pollution events with emphasis on the urban areas of the industrial corridor that runs along the eastern states. The NEC-OPS study was begun with the setup of the primary urban site in Philadelphia PA and use of the site to gather

data during a two-week period in August 1998. The data set will be used to evaluate the instrument performance and prepare an analysis of data quality. In this paper, an example of the measurements performed by the five research groups in the pilot study has been presented. The results indicate the strong technical capability of the group and indicate the type of data that will be available from the intensive campaigns.

The NEC-OPS project has the goal of developing an understanding of the important factors in describing the atmospheric pollution events in the urban environment and providing a critical data set for modelers to use in testing and improving their ability to accurately model the periods when unhealthy conditions are caused by the physical and chemical processes acting upon the chemical soup which is generated by traffic and industrial activity within an urban environment. Any future efforts to regulate the activities which generate air pollution will require that we can describe the local and regional levels of critical pollutants with certainty.

ACKNOWLEDGMENTS

The primary support for this effort is EPA Grant Number R826373 and the efforts of Project Officer Deran Pashyan are gratefully acknowledged. This large consortium effort would not be possible without the cheerful cooperation and technical skills of the many individuals involved. Special appreciation goes to Richard Clark (Millersville University), George Allen (Harvard University School of Public Health), William Miller (Philadelphia Air Management Services Laboratory) and Bruce Doddridge (University of Maryland) for the preliminary results included here.

Table 1. Consortium Members and Expertise

Penn State University - C.R. Philbrick (PI), D. B. Lysak
Advanced lidar remote sensing, optical scattering, atmospheric structure and dynamics
Millersville University - R. D. Clark
Boundary layer meteorology, tethersonde measurement experience
Harvard University
Harvard School of Public Health
P. Koutrakis, G. Allen, J. Lawrence, J. M. Wolfson, V. Hatch
Atmospheric gas and aerosol chemistry measurements and analysis
Engineering and Applied Science
S. C. Wofsy, J. W. Munger
Measurement verification using regional reference site at Harvard Forest
University of Maryland - R. Dickerson, B. Doddridge
Instrumentation and use of small aircraft , Shenandoah National Forest regional site
Philadelphia Air Services Management Laboratory - W. C. Miller
Philadelphia PAMS and data from several sites, PM monitor at prime site
State University of New York - S. T. Rao, V. Mohnen, I. Zurbenko, S. Porter
Analysis and model calculations of polluted air masses, Whiteface Mountain regional site
Rutgers University Environmental and Occupational Health Center - P. Georgopoulos and M. Lazaridis
Emissions inventories and chemistry modeling, particular experience in Philadelphia area
Brookhaven National Laboratory - L. Newman, P. Daum
Highly instrumented large aircraft with measurements of chemistry and aerosol properties

Table 2. Outline of activities during the project.

Task	Year 1	Year 2	Year 3
Field Monitoring: Preparation Monitoring site installation Air pollutant measurements Monitoring site removal	Verification Experiment (August 1998)	Summer Intensive #1 (June-Aug 1999)	Winter study (Dec 1999) Summer Intensive #2 (June-Aug2000)
Sample Preparation and Analysis: Gravimetric analysis Inorganic pollutant analysis Trace element analysis Organic speciation Single particle analysis	Verification Experiment	Summer Intensive #1	Winter Intensive Summer Intensive #2
Continuous Data Processing: Fine mass and particle size Elemental and organic carbon Nitrate, Sulfate, other data Local meteorology description Develop regional meteorology	Verification Experiment	Summer Intensive #1	Winter Intensive Summer Intensive #2
Data Analysis: Summary statistics Method comparisons Temporal-spatial characterization Source apportionment Transport/dynamics analysis Campaign comparisons Analysis with previous data sets Model calculations and comparison	Verification Experiment	Summer Intensive #1	Winter Intensive Summer Intensive #2
Preliminary report Final report	Data Summary Report	Data Summary Report	Final Report Publication of Results

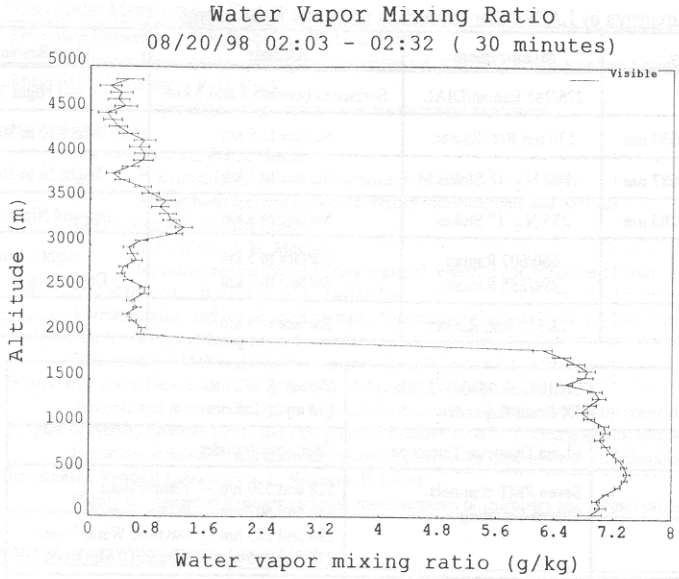
Table 3. Measurements by LAPS Lidar Instrument and Lidar Sub-systems

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

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

Table 4. Measurements by Millersville University Tethersonde (0-300 m using 38 ft and 18 ft balloons)

Property	Sensor Type	Accuracy	Resolution
Dry-bulb Temperature	Thermistor	± 0.5 C	0.01 C
Wet-bulb Temperature	Aspirated thermistor	± 0.5 C	0.01 C
Relative Humidity	Computed	± 5 %	0.1 %
Pressure	Aneroid capacitor	± 0.1 kPa	0.01 kPa
Wind Speed	3-cup anemometer	± 0.25 m/s	0.1 m/s
Wind Direction	Magnetic compass	± 5 degrees	1 degree
O_3 concentration	KI/oxidant	± 5 %	
PM10 and PM2.5	Filter		



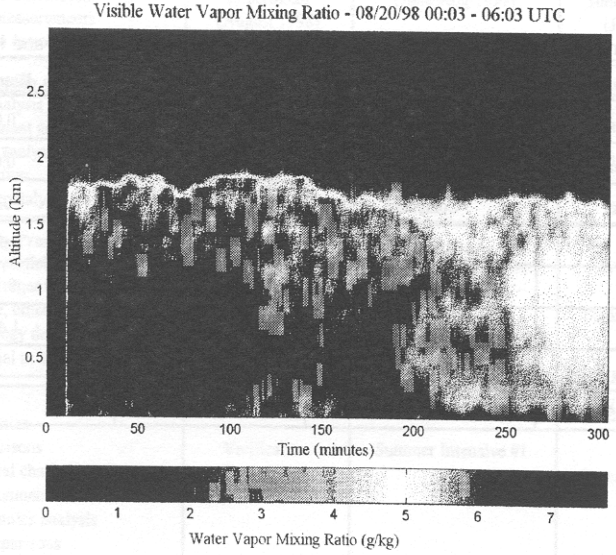


Figure 1. The water vapor measurements for the PSU LAPS lidar are shown for a time snapshot at 0200 UTC on 20 August and for a five hour time sequence on 20 August.

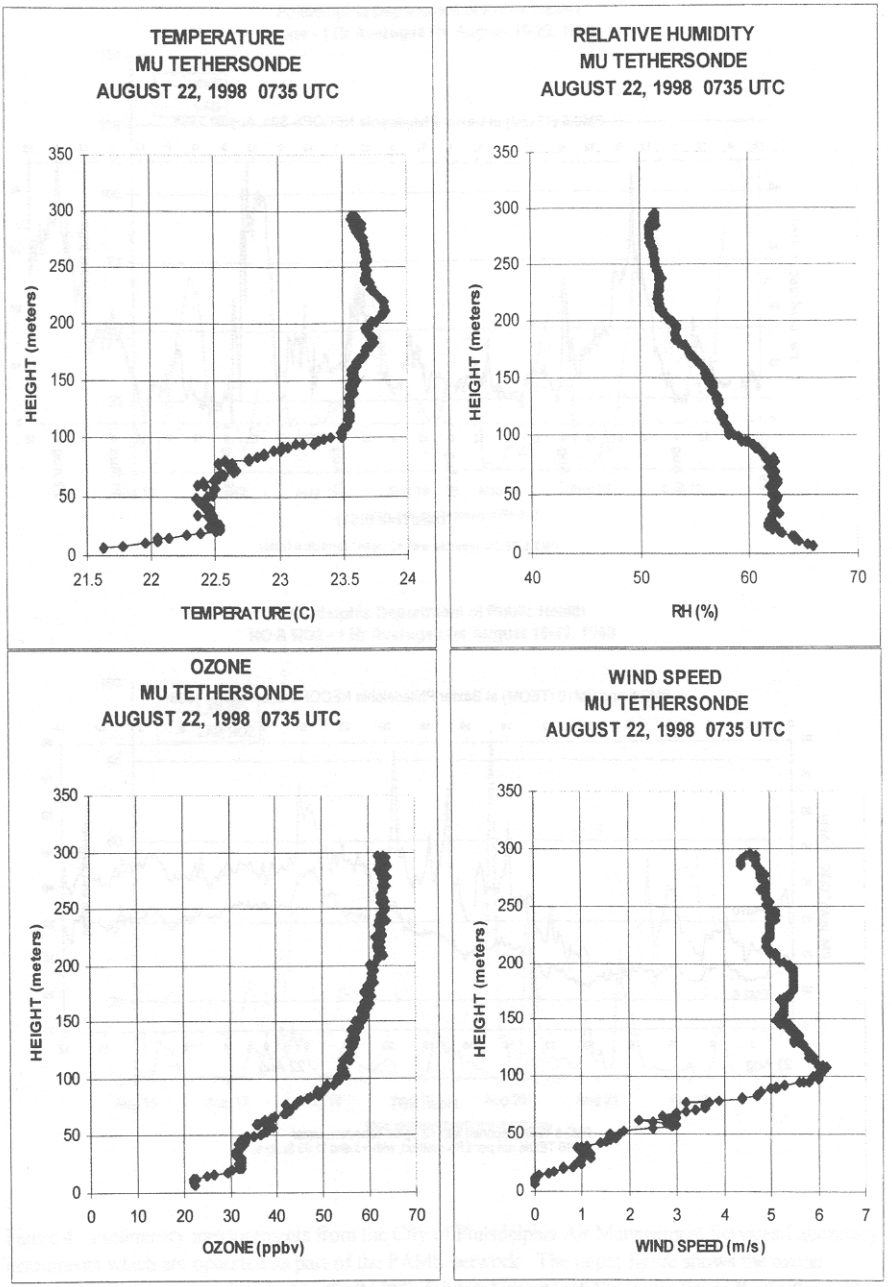
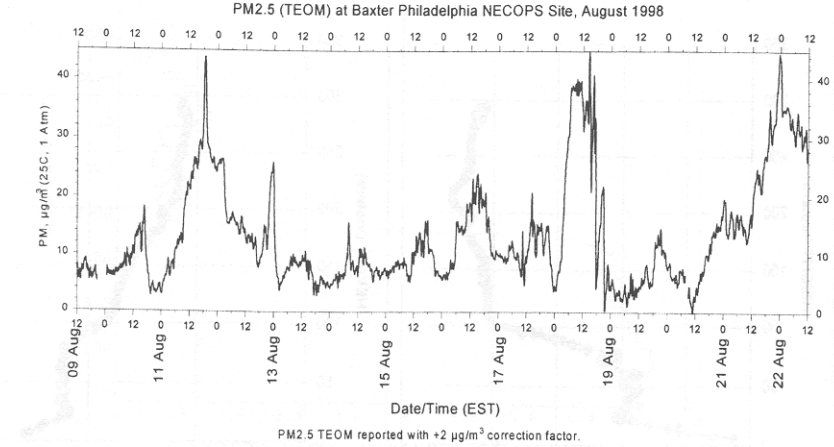


Figure 2. Preliminary results from one of the tethersonde balloon ascents by Millersville University. (Courtesy of Richard Clark)



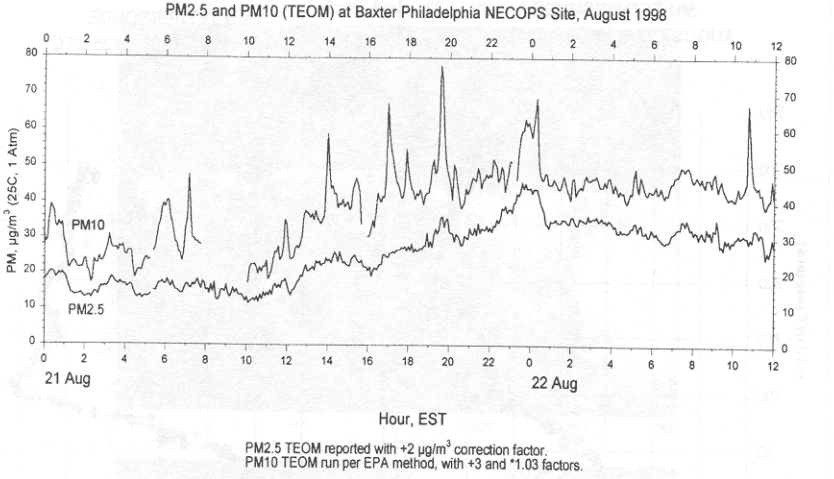
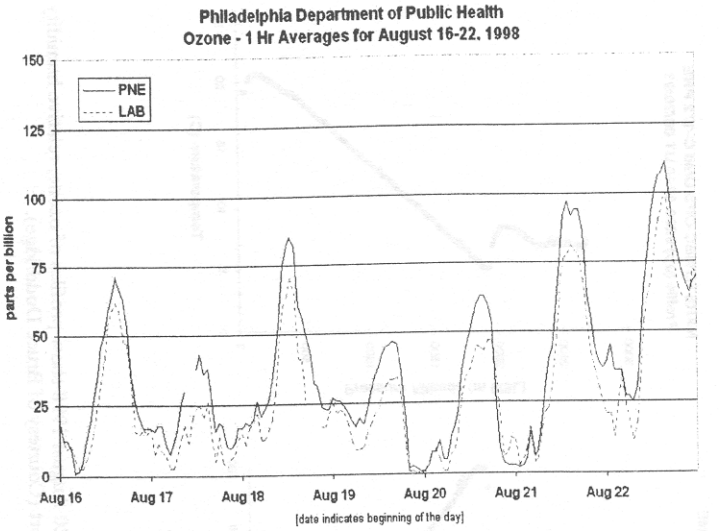


Figure 3. Preliminary results of the Harvard School of Public Health on the concentration of PM 2.5 and PM 10 particles during the August 1998 measurement program. (Courtesy of George Allen)



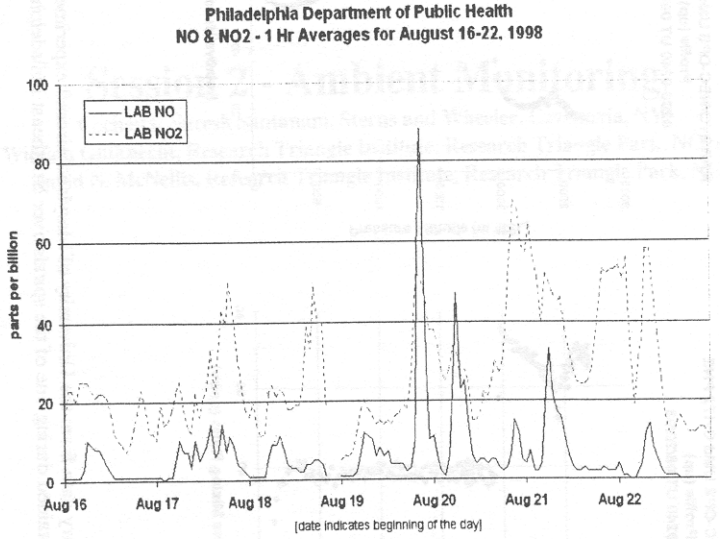


Figure 4. Preliminary measurements from the City of Philadelphia Air Management Services Laboratory instruments which are operated as part of the PAMS network. The upper figure shows the ozone measured at city center and at the Northeast Philadelphia Airport, and the lower figure shows the measurement of NO and NO_2 at the city center laboratory. (Courtesy of William Miller)

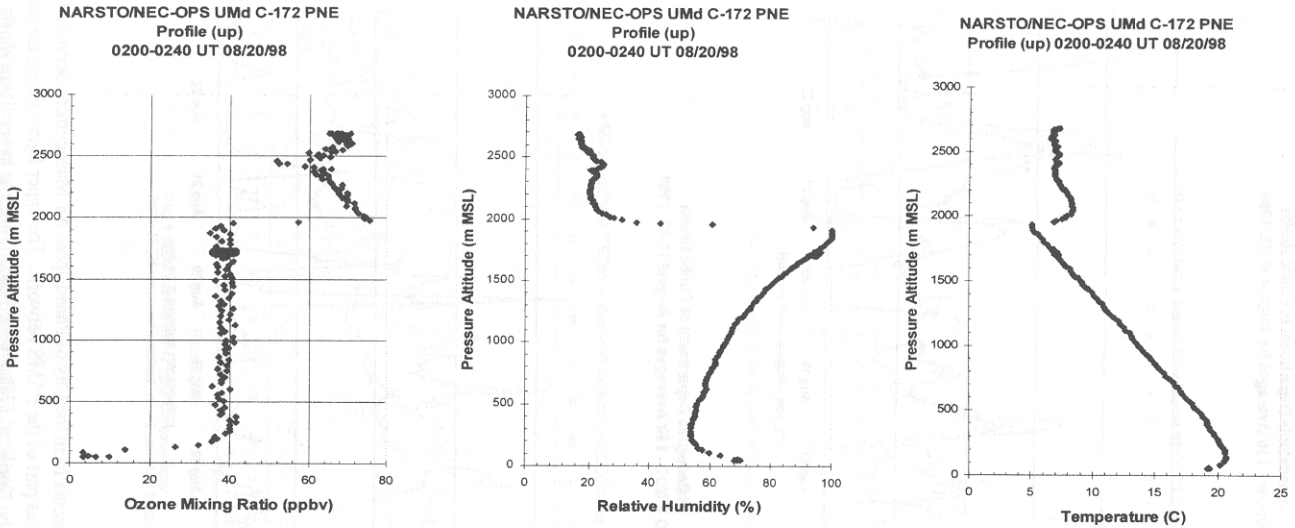


Figure 5. Preliminary data from the University of Maryland aircraft experiments on 20 August 1998 show profiles of ozone, relative humidity and temperature obtained during one of the spirals over Northeast Philadelphia Airport (Courtesy of Bruce Doddridge).