### THE EFFECTS OF LOCAL AND REGIONAL SCALE CIRCULATIONS ON AIR POLLUTANTS DURING NARSTO-NE-OPS 1999-2001

Richard D. Clark Millersville University of Pennsylvania, Millersville, Pennsylvania

C. Russell Philbrick William F. Ryan The Pennsylvania State University, University Park, Pennsylvania

> Bruce G. Doddridge Jeffrey W. Stehr The University of Maryland, College Park, MD

### 1. INTRODUCTION

Anthropogenic emissions from urban sprawl, traffic, and industrialization along the northeast corridor of the United States should have an increasingly profound effect on urban and regional air quality. Surface air quality over populated areas is an important issue given persuasive data linking high levels of atmospheric oxidants and particulate matter to deleterious human health effects (Doddridge, 2000).

Predictions of air quality episodes along the Northeast corridor can be confounded by the frequent occurrence of local and regional scale circulations that can influence the magnitude, timing, and spatial extent of air pollution events (Seaman and Michelson, 1998). While there may be good understanding of the synoptic scale patterns associated with severe air pollution episodes in the mid-Atlantic region (Ryan, 2001), local and regional gradients generated by land-sea discontinuities, topographic features. urban environments, and complicated by a convoluted coastline can give rise to sub-synoptic circulations that are difficult to predict. Forecast skill is often compromised due to the complex scale interactions between the surface layer, boundary layer, and free troposphere. An improved understanding of the influence of local and regional circulations on sources. sinks, transport, mixing, and photochemical transformations controlling the observed abundances of photochemical oxidants and fine particle haze over the mid-Atlantic region is key to developing any capability in the future to forecast such pollution events reliably.

During a nine-week period in July-August 1999 and a four-week period in July 2001 a consortium of investigators from several institutions and government laboratories conducted an intensive field campaign about 18 km ENE of Philadelphia (40.04° N, 75.00° W). The objectives of this EPAsponsored NARSTO–NE-OPS (North American Research Strategy for Tropospheric Ozone – Northeast Oxidant and Particle Study) are to investigate the conditions within the urban polluted environment to find relationships between the meteorological conditions and high  $O_3$  concentrations, increased levels of fine particles (PM<sub>2.5</sub>), and contributions from local and distant sources (Philbrick et al, 2000). During the course of the 1999-2001 campaigns, these investigations made apparent the importance of the influence that sub-synoptic scale circulations have on the variability of trace gas and particle concentrations.

This paper presents *in-situ* meteorological observations of select meteorological events and their influence on the trace gas and fine particle concentrations. It includes a brief overview that establishes the synoptic settings of 1999 and 2001.

### 2. DATA COLLECTION

The bulk of the aloft data shown in this paper were collected using two Millersville University tethered blimps. Instruments were deployed on a small 7 m<sup>3</sup> blimp to obtain vertical profiles of T, p, RH, wind speed and direction, and O<sub>3</sub> concentration to 300 m AGL with a vertical resolution of 1-3 m. Each profile took approximately 30 minutes to complete. During July-August 1999, 449 vertical profiles were completed; in July 2001 alone, 536 profiles were obtained. Fig 1. (a, b) shows the frequency of vertical profiles obtained for both 1999 and 2001.

A large (100 m<sup>3</sup>) tethered blimp with a free-lift capacity of 50 kg was also deployed in 1999 and 2001 to collect impaction samples for analysis of accumulated  $PM_{2.5}$  dry mass, canister samples for analysis of 55 toxics, and measurements of  $PM_{2.5}$  concentrations using laser scatterometry. This large blimp resided at an altitude of 300 m AGL for two 10-hour periods per day (10:00 – 20:00 LT; 22:00 – 08:00 LT). Times were chosen to isolate the daytime and nighttime boundary layer regimes. The samplers and scatterometers were attached at four levels (75, 150, 225, and 300 m AGL) in 1999 and three levels (100, 200, 300 m AGL) in 2001.

7.5

<sup>&</sup>lt;sup>\*</sup>*Corresponding author address:* Richard D. Clark, Dept. of Earth Sciences, P.O. Box 1002, Millersville University, Millersville, PA 17551; <u>Richard.clark@millersville.edu</u>

Identical and additional instruments were located at the surface for the same sampling duration.

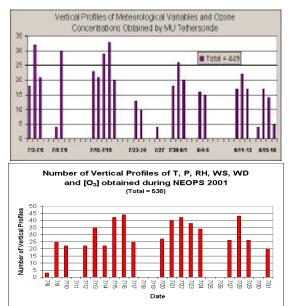


Fig. 1. A total of 985 vertical profiles of T, p, RH, WS, WD, and  $O_3$  concentration were obtained using the small blimp. (a) 449 in July-August 1999; (b) 536 in July 2001.

The blimp-borne instruments, which provided a detailed record of the development and evolution of the surface layer and lower boundary layer, were a complement to data collected by a variety of universities, national laboratories, and government agencies. They include: The Pennsylvania State University, University of Maryland, Harvard School of Public Health, Pacific Northwest, Brookhaven, and Argonne National Laboratories, Environmental Protection Agency, City of Philadelphia Air Services Management Laboratory, Drexel University, and Brigham Young University, Texas Tech University, and Clarkson University. Details of their instruments and measurement are presented in other papers in this session.

# 3. OVERVIEW OF SUMMERS 1999 AND 2001

Summer 1999 was considerably warmer and drier than normal with temperature in the 90<sup>th</sup> percentile and precipitation in the 10<sup>th</sup> percentile relative to the 1895-1998 long-term average. The weather was dominated by a 500 hPa ridge over the mid-Atlantic region and surface high pressure centered over the Carolinas. Winds at 850 hPa were westerly to northwesterly, while southerly to southwesterly winds persisted at the surface during the daytime. By contrast, July 2001 was cooler and drier than the 1895-1999 long-term average with anomalies in the 10<sup>th</sup> percentile for temperature and 20<sup>th</sup> percentile for precipitation. During July 2001 the 500 hPa ridge was located over the western states with a persistent trough in the east and

surface high pressure displaced well to the south (Ryan, 2001). Wind anomalies in July 2001 show strong northerlies over the Ohio River Valley and enhanced easterlies along the mid-Atlantic coastal plain. These differences in the overall synoptic patterns between 1999 and 2001 had a significant influence on the local and regional meteorology affecting air chemistry and fine particle concentrations in the Philadelphia area.

The regional trace gas concentrations reported for July-August 1999, such as those shown for O<sub>3</sub> in Fig. 2 (a-b), were characterized as normal overall but highly variable (Ryan, 2001). This variability can be linked to mesoscale and boundary-layer-forced circulations that were able to develop and evolve because the large-scale upper air gradients were weak. By comparison, the July 2001 period was noted for the frequent intrusion of cleaner, drier continental air masses, stronger synoptic gradients, and convective activity on the leading edge of short waves. As a consequence, not only were the mean trace gas concentrations reduced, variability was diminished in response to the suppression of conditions that favor the development of local and regional circulations (Fig. 2. (c)).

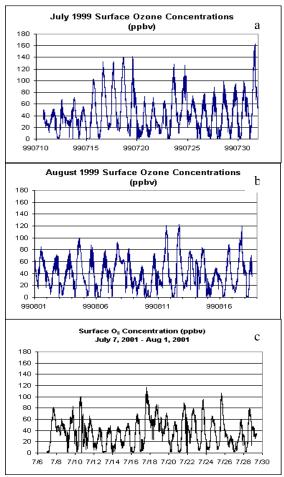


Fig. 2. Time traces of surface  $O_3$  concentration for the months of July-August 1999 (a - b) and July 2001 (c).

Eight episodes of high  $O_3$  and/or high  $PM_{2.5}$ were documented during July-August 1999 with 12 days exceeding 100 ppbv at the Baxter site, whereas only three minor episodes occurred in July 2001, with three days barely exceeding 100 ppbv. Tables 1 and 2 provide a brief description of the episodes documented in 1999 and 2001.

Table 1. 1999 NE-OPS EPISODES

Date	Description of Episode
Jul 3-5	Warm sector, high temps (37.2 C), strong low level wind, Code Orange $O_3$
Jul 8-10	Frontal passage 7/8, warm sector 7/9 with strong 850 hPa advection. Moderate wind, Code Yellow $O_3$ with Code Orange west of site.
Jul 15-20	Strongest O <sub>3</sub> episode of season. Ramp-up and recirculation event followed by stagnation. Weak W to SW wind with strong Bermuda High. 17 1-hour exceedances on 7/19. Ramp-up [PM <sub>2.5</sub> ].
Jul 23-24	Recirculation late 7/22 followed by SW wind and Code Orange $O_3$ . Upper level ridge brings warm 850 hPa temps. TRWs end the episode on 7/24.
Jul28-Aug 1	Lower $O_3$ levels 7/28-7/30 with W wind followed by lee trough on 7/31, SW wind, spike of 165 ppbv $O_3$ and passage of sea breeze front. Mobile trough on 8/1 ends the episode. High [PM <sub>2.5</sub> ] correlate with low [O <sub>3</sub> ].
Aug 4-5	High $O_3$ levels disturbed by frontal passage, NW winds and low $T_d$ keep $O_3$ in Code Orange. Reduced temps.
Aug 11-13	Warm sector, recirculation of high O <sub>3</sub> before passage of bay breeze, strong bay breeze on 8/12 cleanses.
Aug 16-17	Similar meteorology to Aug 11-13 with spike in $O_3$ on 8/17.

## Table 2. 2001 NE-OPS EPISODES

Date	Description of Episode
July 10	Recirculation brings WSW wind, temps > 30 C, leading to single-day spike in $O_3$ , followed by strong afternoon convection.
July 17	Highest $O_3$ (120 ppbv) in July at site, convergence between backdoor front in NY and disturbance in the midwest.
July 21-25	Intermittent high $O_3$ associated with northward movement of Bermuda High.

# 4. SUB-SYNOPTIC CIRCULATIONS

When synoptic forcing is strong and dominates as it did in July 2001, there is little evidence of the influence of local and regional circulations on the meteorology and chemistry of the NE corridor other than that driven by the diurnal cycle and the proximity of the site to a large urban area. However, under conditions of stagnation and weak upper level gradients over the mid-Atlantic region, as in the summer 1999, differential heating/cooling in the surface layer can emerge as the primary driver for local and regional circulations. Slight variations in solar irradiance, cloud cover, coastal topography, terrain elevation, outflow boundaries, convergence zones, and land-sea discontinuities are just a few of the features that can either enhance or diminish the development of local and regional circulations. These events can be difficult to predict or even diagnose because they are so sensitive to local forcing, yet they can dramatically alter trace gas and aerosol concentrations in a matter of minutes.

However, there are two important sub-synoptic events that appear as recurring features when the mid-Atlantic region is under the influence of a stagnant summertime air mass such as the Bermuda High. On at least three occasions during the 1999 campaign, July 31, August 12, and August 17, the intrusion of a shallow moist air mass from the southeast resulted in dramatic change in meteorological variables, rapid reduction in trace gas concentrations, and a simultaneous increase in total scattering by fine particulates. These sea breeze fronts were observed around 1700-1800 local time in the Philadelphia area, and within minutes replaced a hot, dry, polluted boundary layer with cooler, moist, clean air.

A second recurring feature of summer 1999 is the development of nocturnal boundary layer jets (LLJ). While not as strong or persistent as the Great Plains LLJ, the formation mechanism is similar to conceptual model formulated by Blackadar (1957), and modified by many investigators (e.g. Parish et al, 1988) to explain the Great Plains LLJ. Differential surface heating between the coastal plain and the Appalachian leeside can produce substantial gradients within the boundary layer during the daytime. These gradients support a near-surface geostrophic wind, but frictional stress prevents the observed wind from achieving geostrophic balance. An agesotrophic wind is induced proportional to the frictional stress. Around sunset, with the development of the nocturnal inversion and the cessation of turbulence within the inversion, the velocity field accelerates in an attempt to adjust to the mass field. This acceleration is subject to Coriolis Force and results in a shallow layer of faster moving air residing at the top of the nocturnal boundary layer. In 1999, LLJs were observed between 400-600 m AGL. Observed wind speeds can achieve twice their geostrophic value and can transport pollutants hundreds of kilometers. LLJs can be adequately simulated with mesoscale models (Zhang et al, 2001). The remainder of this paper will focus on these two recurring sub-synoptic features.

# 5. THE CASE STUDY OF JULY 15-19 1999

The period from July 15-19 1999 represents the strongest and longest ozone episode of the season. PHL and Baltimore reach Code Red for 4 consecutive days (July 16-19). A cold front approached the region on July 10 and then stalled to the south by July 12. Weak disturbances running along the frontal zone brought intermittent clouds and showers to the region through July 13. The front finally dissipated in place on July 14 with southerly return flow and much higher ozone forecast for July 15. The transition to higher ozone began on July 15 with a slightly greater magnitude than expected. Temperatures reached 31.1 C with the highest  $O_3$  levels northeast of Philadelphia near Trenton, NJ (Ryan, 1999).

By July 16, a very complicated picture developed with westerly transport into the region overnight. A weak (12 ms<sup>-1</sup>) LLJ is seen in the profiler data and is partly responsible for the transport of 100+ ppbv O<sub>3</sub> from the Ohio Valley and western mid-Atlantic region. At the surface, essentially calm winds prevailed overnight with SE winds developing later in the day pushing the high O<sub>3</sub> plume back across the I-95 Corridor. Large areas of Code Orange and Red extended from Trenton westward to Harrisburg and south to Washington, DC, while the bay and sea breeze circulations kept O<sub>3</sub> levels moderate east of the I-95 corridor, but did not reach Philadelphia.

By July 17, a complex local recirculation occurred with a strong SW LLJ within the nocturnal residual layer (see Fig. 3). The influence of the LLJ

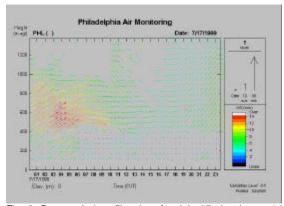


Fig. 3. Baxter wind profiler data for July 17 showing at 14 ms<sup>-1</sup> LLJ between 400-600 m AGL around midnight local time.

can be seen in the surface  $O_3$  and scatterometer traces and the time-height series of meteorological variables. When the LLJ reaches a maximum around 0400 UTC, shear instability (shear ~  $3x10^{-2}$  s<sup>-1</sup>) is sufficient to sustain downward momentum, heat, moisture, and  $O_3$  flux until the inversion strengthens and decouples the layer. The downward flux is not continuous, but appears instead as periodic bursting events at the surface.

Measured and derived meteorological variables and  $O_3$  concentration obtained with the tethered instruments indicate that the surface to 300 m layer remains fairly well-mixed until 0500 UTC (Fig. 6). This mixing helps keep  $O_3$  concentrations in the 20-30 ppbv range overnight over the remainder of the episode. This effect is seen again on July 18-19 and contributes to the spikes in  $O_3$  concentration seen on both days in the early morning hours (Fig. 4 and 5). The LLJ becomes more westerly as the episode matures, and diminishes in intensity on July 19 as the cold front approaches.

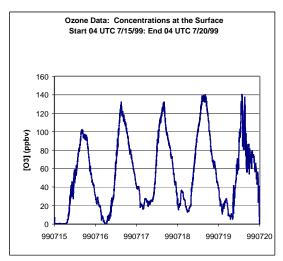


Fig. 4. Surface [O<sub>3</sub>] for the July15-19 episode.

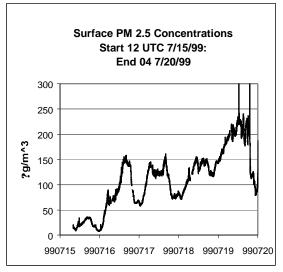


Fig. 5. One-minute averages of surface [PM<sub>2.5</sub>] for July 15-19 obtained using laser-diode scatterometry.

The effect of the LLJ on aerosol scattering is evident in the close-up of  $PM_{2.5}$  concentration obtained using the laser-diode scatterometers on the 100 m<sup>3</sup> blimp. While scattering is only a proxy indicator of particle concentration, it can be used as an effective indicator of trends and differences.

Inspection of Fig. 7 shows that scattering due to

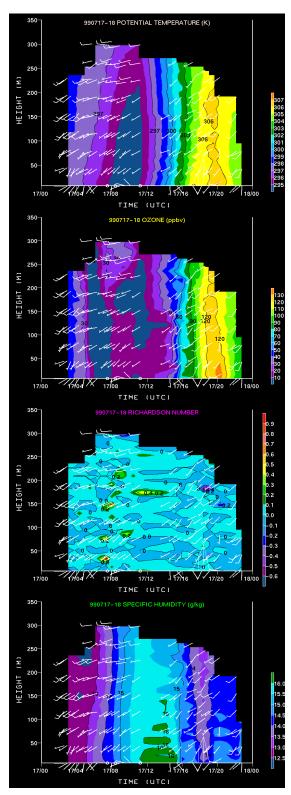


Fig. 6. (Top to bottom): Potential temp,  $[O_3]$ , Ri #, and specific humidity for 00 UTC July 17 – 00 UTC July 18.

 $PM_{2.5}$  is greatest at the 300 m level, where it is closest to the level of maximum winds and enhanced moisture transport. The co-location of the LLJ with the layer of enhanced moisture is evident in the time trace from the PSU Raman lidar. The scatterometers also show an interesting four-fold increase in [PM<sub>2.5</sub>] during the boundary layer transition, and an interesting reversal of relative concentrations between the surface and 300 m that accompanies boundary layer growth (see Fig. 8).

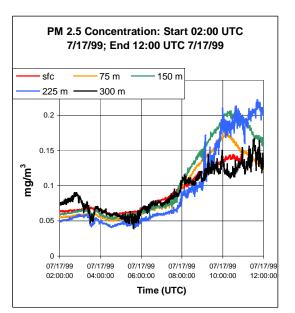


Fig. 7. PM<sub>2.5</sub> at the surface, 75, 150, 225, and 300 m.

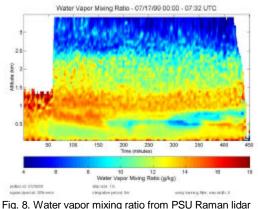


Fig. 8. Water vapor mixing ratio from PSU Raman lidar (Philbrick, 2000).

#### 6. THE CASE STUDY OF JULY 31, 1999

The July 31, 1999 episode developed rapidly beginning with Code Orange ozone levels to the west and north of Philadelphia on July 30 with westerly flow. By July 31, a lee trough had developed along the I-95 corridor, with enhanced convergence due to bay and sea breezes and numerous outflow boundaries to the southeast. The nearest cold front was still well to the west, and winds at the Baxter site were light allowing midafternoon temperatures to approach 38 C (100 F) (see Fig. 9). Ozone levels at the site soared to 165 ppbv, the highest levels recorded in 11 years in Philadelphia and Code Red conditions extended from Washington to Philadelphia along I-95 as shown in Fig. 10. By late afternoon (2100 UTC), a

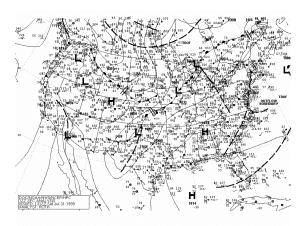


Fig. 9. Surface analysis at 12 UTC, July 31, 1999.

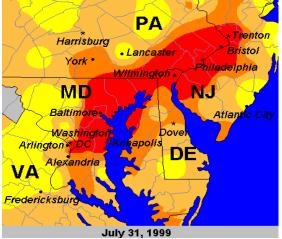


Fig. 10. Regional ozone along the I-95 corridor.

sea breeze convergence zone, enhanced by an extensive outflow boundary, propagated through the site. The modifications to the boundary layer in the wake of its passage were dramatic. In a layer only 100 m deep, wind direction at the site backed more than 90 degrees, temperature decreased by 1.5 C, wind speed doubled, and [O<sub>3</sub>] fell from 160 ppbv to 95 ppbv in 10 minutes. The PM<sub>2.5</sub> concentrations at the surface responded first with a significant increase from 140 to 210 ?g m<sup>-3</sup>, with the aloft PM<sub>2.5</sub> concentration increasing but lagging the surface signature (Fig. 11). The increase in PM is probably a result of the activation of aerosols in the shallow moist layer. The series of vertical profiles in Figs. 12 and 13 (next page) were obtained within minutes before and after the passage of the sea

breeze front and show the dramatic effect of the intruding shallow air mass.

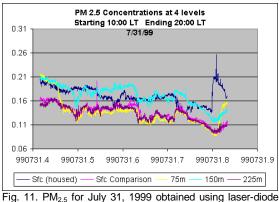


Fig. 11.  $PM_{2.5}$  for July 31, 1999 obtained using laser-diode scatterometry as a proxy indicator of concentration. The sea breeze passed the site around 990731.8.

#### 7. SUMMARY AND CONCLUSIONS

The meteorology and air chemistry at the Baxter NEOPS site exhibits overall differences that can be related to the synoptic conditions in place during the summer seasons of 1999 and 2001. The two years are on opposite extremes of percentile rank of temperature, and are similar in percentile rank for precipitation but not for the same reasons. Summer 2001 is cool and dry because of the frequent intrusion of continental air, whereas 1999 is warm and dry because of stagnation associated with a persistent maritime tropical air mass. While there may be a good understanding of the synoptic patterns associated with the general characteristics of severe air pollution events, local and regional variability can be significant when circulations are driven by sub-synoptic gradients.

Sea-bay breezes and nocturnal LLJs are recurring features of the summertime mid-Atlantic region under conditions of weak synoptic forcing. It has been shown by means of two case studies that their influence on the local and regional meteorology and chemistry can be dramatic. The daytime sea breeze can propagate through an area and replace the aged air mass with moist, cool, and clean air. On the other hand, the nocturnal LLJ can transport pollutants hundreds of kilometers without generating much of a signal at the surface. Then as the boundary layer develops the following morning, pollutant concentrations, often double the background values, mix down to the surface to produce rapid increases in pollutant concentrations that cannot be explained by local urban primary and secondary production.

The summer 1999 provides and excellent data base with which to test the sensitivity of numerical models to the magnitude, timing, and spatial extent of these flow regimes. This work is already in progress and the results appear promising.

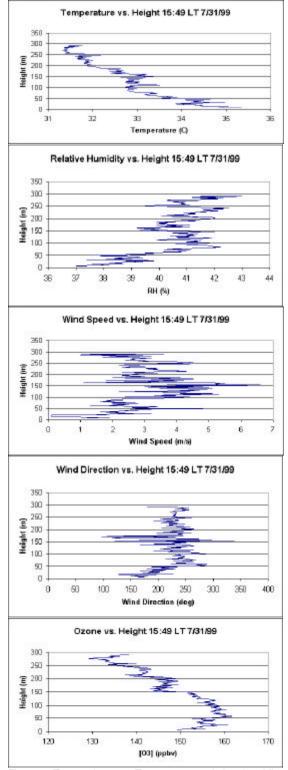


Fig. 12. Top to bottom: Temperature, relative humidity, wind speed, wind direction, and  $O_3$  concentration for 15:49 local time on July 31, 1999.

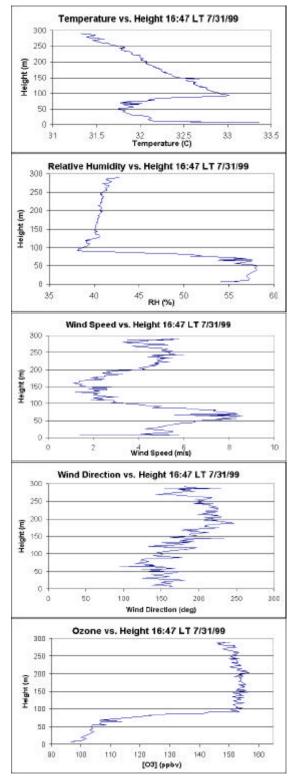


Fig. 13. Same as Fig. 12 except for 16:47 local time.

# 8. ACKNOWLEDGEMENTS

The authors extend their appreciation to the team of dedicated investigators and students who endured the intense heat, exposure to the elements, poor air quality, and long hours (often 18-20 hours/day during an episode) to collect and analyze the measurements obtained during this marathon study. A special thanks is extended to the six Millersville University undergraduate students who participated in the 1999 study: Traci Duerr, Christopher Readinger, Deborah Stein, Michael Conte, Colleen Dougherty, and Christopher Compton, and the six students who took part in the 2001 study: Sabrina Atkins, Jamie Barbush, Daniel Carre, Andrew Terry, Michelle Theis, and Richard Walker. NARSTO-NE-OPS is funded through a grant (R826373) from the Environmental Protection Agency.

#### 9. REFERENCES

- Doddridge, B. G., 2000: An airborne study of chemistry and fine particles over the U.S. mid-Atlantic region. Preprints, *PM 2000: Particulate Matter and Health – The scientific basis for regulatory decision making.* Charleston, SC, AWMA, 4AS2; pp 4-5.
- Philbrick, C. R., R. D. Clark, P. Koutrakis, J.W. Munger, B. G. Doddridge, W. C. Miller, S. T. Rao, P. Georgopoulos, and L. Newman, 2000: Investigations of ozone and particulate matter air pollution in the northeast. Preprints, *PM* 2000: Particulate Matter and Health – The scientific basis for regulatory decision making. Charleston, SC, AWMA, 4AS2; pp 1-2.
- Ryan, W. F., 1999: Discussion of the 1999 Ozone Season: Summary of Weather Conditions. <u>http://metosrv2.umd.edu/~ryan/summary99.ht</u> <u>m</u>.
- Ryan, W. F., C. R. Philbrick, and R. D. Clark, 2001: Meteorological aspects of the July 15-20, 1999 Northeast Oxidant and Particulate Study Study (NEOPS) pollution episode. (Paper 7.10, this session)
- Seaman, N. L., and S. A. Michelson, 1998: Mesoscale meteorological structure of a highozone episode during the 1995 NARSTO-Northeast study, *J. Appl. Meteor.*, **39**, 384-398.
- Zhang, K., H. Mao, K. Civerolo, S, Berman, J-Y. Ku, S. T. Rao, B. K. Doddridge, C. R. Philbrick, and R. D. Clark, 2001: Numberical investigation of boundary-layer evolution and nocturnal lowlevel jets: local versus non-local PBL schemes. *Enviro. Fluid Mech.*, 1, 171-208.