APPENDIX H

Regional Transport and 8-Hour Ozone

Regional Transport of Pollutants and Implications for 8-Hour Ozone Non-Attainment Areas in Maryland:

Report for the National Governor's Conference

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For:

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Introduction

The purpose of this report is to provide a review of the current understanding of the size of the region that may contribute to exceedances of the 8-hour ozone (O_3) and PM_{fine} National Ambient Air Quality Standard (NAAQS) in the mid-Atlantic metropolitan areas.

Observations and analyses carried out during the last 5 years have shown that, during high O_3 episodes in the mid-Atlantic, significant concentrations of O_3 and O_3 precursors are transported on regional scales (hundreds of km). This scale of transport is far in excess of the size of current 1-hour non-attainment regions and, as the regional load is on the order of 80 parts per billion by volume (ppbv) O_3 or more, can contribute significantly to exceedances of the both the 1-hour and 8-hour NAAQS locally. The direction of transport during these events is primarily west to east though transport along the I-95 Corridor from southwest to northeast is also observed.

As a result, the region whose emissions impact O_3 concentrations in the mid-Atlantic is much larger than previously thought. Control of O_3 precursors should be region-wide to have maximum effectiveness. Although data on regional haze and fine particulate matter are sparser, available observations suggest a similarly large region of influence.

O_3 Chemistry, Local O_3 Concentrations, and Regional Transport

O_3 is a secondary pollutant formed by photochemical processes. Formation of O_3 requires sunlight, oxides of nitrogen (NO_x) and reactive volatile organic compounds (VOCs).[1] For the majority of the mid-Atlantic region, VOCs are in large supply; recent observations with advanced analytical techniques reveals even higher concentrations of VOC's in urban areas than previously estimated.[2] This is due primarily to large natural sources of VOCs although local motor vehicle emissions and industrial activities are also important.[3] The natural source of VOC precursors is primarily emissions from deciduous trees; the natural source of secondary aerosol forming VOC is coniferous trees. These emissions are strongly temperature dependent.[4] Even in the urban core region of Baltimore, natural VOCs are a significant factor.[5] NO_x emissions are concentrated in urban areas and near elevated point sources associated with power plants and large industrial combustion sources. The latter are concentrated along the I-95 Corridor and across the Ohio River Valley.[6]

For the majority of the mid-Atlantic region, O_3 production is regulated by NO_x concentrations.[7] There is a non-linear relationship between concentrations of NO_x and O_3 production.[8] At very low or very high concentrations of NO_x, O_3 formation is retarded. In the mid-range, which includes most of the mid-Atlantic region, with the exception of the center cities where NO_x concentrations can be very high, O_3 production is proportional to NO_x concentrations.[9]

The lifetimes of O_3 and its precursors in the atmosphere control the distances over which transport is significant. These lifetimes are highly variable. In general, lifetimes of trace gases are much longer above the surface layer. For example, O_3 well above the surface has a lifetime of several days or weeks.[10] However, O_3 trapped near the surface overnight has a lifetime of less than one day. As will be noted in more detail below, this has important effects when coupled with meteorological factors. In general, O_3 can be expected to have a lifetime of several days. VOCs important to ozone production vary widely in concentration and reactivity, and have lifetimes ranging from hours to weeks.[11] As noted above, however, they are ubiquitous in the mid-Atlantic region. NO_x has a more complicated lifetime. In general, the lifetime of NO_x near the surface is approximately 1 day; NO_x in the cooler upper atmosphere can have a lifetime of several days.[11] In addition, some portion of total NO_x can be transformed to so-called "reservoir" species such as PAN ($CH_3C(O)OONO_2$)) which can exist for long periods at cooler temperatures above the surface and then decompose to reform NO_x when mixed downward to warmer temperatures. In any case, O_3 and its key precursors have lifetimes that are consistent with transport over several days.

Aerosol Chemistry and Physics

Fine aerosol particles with a diameter less than 2.5μm (PM_{fine}) are removed only slowly by sedimentation and dry deposition. These particles, central to both health effects and reduced visibility, are lost from the atmosphere primarily by wet deposition, and consequently have a lifetime similar to that of atmospheric water vapor – about a week. The chemistry and physics of fine particles is consistent with transport over several days. The atmospheric Chemistry and physics of aerosol particles has been reviewed recently.[11, 12]

Meteorology and Regional Transport

On the local scale, meteorological factors on which O_3 concentrations depend are the amount of available sunlight (ultra violet range), temperature and the volume into which the precursor emissions are mixed. Sunlight drives the key photochemical reactions both for O_3 and key precursors and the emissions rates of many precursors (isoprene for example) are temperature dependent. Confining these emissions to a smaller volume results in higher ppbv concentrations of O_3. Horizontal mixing is driven mainly by winds in the lowest 2 km of the atmosphere (planetary boundary layer or PBL) and vertical mixing by the vertical temperature and moisture profiles. High O_3 is typically associated with weather conditions of few clouds, strong temperature inversion and light winds.[13]

The large-scale weather pattern that combines meteorological factors conducive to high O_3 is the presence of a region of upper air high pressure (an upper air ridge) with its central axis located west of the region of interest. The region to the east of the axis of

the high-pressure ridge is characterized by subsiding (downward moving) air. This reduces upward motion necessary for cloud formation, increases temperature and supports a stronger lower level inversion. While the upper air ridge is located west of the region (Figure 1), surface high pressure is typically quite diffuse across the region (Figure 2). This pattern occurs throughout the year but is most common, and longer lived in the summer months.

The large, or synoptic, scale, weather pattern sketched above has important implications for transport. These will be shown in more detail below but are discussed in general terms here. First, the persistence of an upper air ridge west of the region drives generally west to northwest wind in the O_3-conducive region to the east. It should be noted that stagnant air is not always a factor in high O_3 episodes. The average wind speed at this level for high O_3 events is 5.3 ms^{-1} (~ 10^+ knots). Second, the region in the vicinity of the ridge axis, being generally cloud free, will experience significant radiational cooling after sunset and therefore a strong nocturnal inversion will form. This inversion, typically only a few hundred meters deep, prevents O_3 and its precursors from mixing downward overnight. This increases their lifetime aloft (no opportunity for destruction by surface deposition) and thus their transport distance. Third, with diffuse surface high pressure (weak synoptic scale pressure gradient), smaller scale (meso-scale) effects can become dominant in the lowest layers of the atmosphere. These include bay and land breeze and the development of the nocturnal low-level jet. The latter is an orographically forced phenomenon characteristic of regions downstream of a mountain barrier. Low-level jets are most often observed in the Great Plains but have also been observed along the Atlantic coastal plain.[14] Low-level jets are commonly observed during high ozone events.[15] With a representative time scale of 4-8 hours at depths of 200-900 m above ground and speeds of 10 ms^{-1}, the low-level jet can account for significant transport (144-360 km) during only a portion of the overnight hours.

Climatology of High Ozone in the Mid-Atlantic

Ozone concentrations in the mid-Atlantic region reach a maximum during the mid-summer months.[16] Highest concentrations typically occur during the period from mid-June to late July when sun angle and temperature are at their yearly maximum. Over the course of the summer months, daily mean peak O_3 concentrations in Baltimore are 95 ppbv for 1-hour concentrations and 80 ppbv for 8-hour average concentrations (1987-1998 data). Thus, only moderate changes in either meteorological conditions, local emissions or regional scale concentrations can result in O_3 exceeding the NAAQS particularly for the 8-hour standard.

The most severe high O_3 cases in the mid-Atlantic occur in multi-day episodes.[17] This is consistent with the "synoptic scale" weather pattern of 2-5 days. For the most severe cases (peak $O_3 \geq 125$ ppbv at 3 or more monitors) in the Baltimore area during the period 1983-1998, 82% occurred within multi-day episodes.

Observations of Ozone and Precursors during Multi-Day Ozone Events

Beginning in the mid-1990's a number of observational studies were carried out to seek to determine the regional "load" of O_3 during multi-day high O_3 episodes. The most comprehensive such study was carried out in 1995 as part of the NARSTO-Northeast project.[18] This project included aircraft and enhanced surface observations at a number of locations from the mid-Atlantic to New England.[18] During this study, several high O_3 events occurred with the most severe occurring during the period July 12-15, 1995.

The July 12-15 episode was one of the strongest on record since the drought (and high O_3) summer of 1988. Peak 1-hour O_3 reached 185 ppbv during this episode and exceedances of the 1-hour NAAQS were observed across New England and the mid-Atlantic states. The weather conditions during this event were of the standard or "classic" high O_3 type.[20] A strong ridge developed to the west of the region (Figure 1) with surface high pressure over the region (Figure 2). Westerly and northwesterly winds were present above the surface and back trajectories showed a general west to east transport (Figure 3). In addition to these synoptic scale features, many of the meso-scale features commonly associated with high O_3 were present. On the mornings of July 13 and 14, a nocturnal low-level jet with peak speeds of 11-16 ms^{-1} was observed. A weak area of lower pressure (Appalachian lee trough) developed along the I-95 Corridor which resulted in a strong low-level inversion along and east of the trough axis.[21]

Regional aerosol loads were also elevated. Aerosol optical depths in the UV exceeded 2.0, and sulfate concentrations of up to 23 $\mu g/m^3$ were observed in Shenandoah National Park and Washington, DC (See http://alta_vista.cira.colostate.edu/ and Dickerson et al., *Science*, 1997). AOD and PM_{fine} are generally strongly correlated in the Eastern US. (Doddridge et al., AGU 2000)

Aircraft Observations

Research aircraft flights were undertaken throughout this episode. Of most interest to consideration of the regional O_3 load were a series of flights made upwind (west) of the I-95 Corridor during the early morning hours of July 12-15. Vertical profiles of O_3 (Figure 4) over south-central PA show O_3 concentrations of 80-110 ppbv through a deep layer. These profiles were made at or just after sunrise and so represent the previous day's photochemical activity at some distance further west of the region. As Arendtsville, the location of the profiles is approximately 140 km upwind of the I-95 Corridor, this represents the minimum distance of regional effects. As these profiles were made around sunrise, before any new photo-chemical production of O_3 had occurred, the O_3 observed was actually formed during the preceding day(s) at a much larger distance upwind. With observed winds on the order of 5 ms^{-1} (or 18 kmh^{-1}), this distance could be several hundred further km upwind.

The layer of high O_3 is co-located with secondary maxima in both SO_2 and NO_x (Figure 5). As noted above, NO_x is a key limited factor for O_3 in much of the mid-

Atlantic region. Therefore, transport of precursor pollutants impacting local air quality (as well as O_3 itself) also occurs on a regional scale. The high concentrations of SO_2 also show that the air mass in this episode contains by-products of coal combustion – most likely from elevated point sources. A very similar profile is seen in western VA where profiles were made slightly later in the day (Figure 6). It is worth noting that while regional scale modeling (such as the Ozone Transport and Assessment Group) shows fairly good skill in O_3 modeling, it cannot resolve these high O_3 loads (Figure 7).[22]

Observations of regional scale O_3 continued as part of the NARSTO-Northeast project in 1996 though that summer proved to be cooler than average and low in O_3. In 1997, however, a high O_3 event equal in magnitude to the 1995 event occurred. Once again, this episode followed the standard pattern and, as such, was forecast accurately 72 hours in advance. Once again, an upper air ridge developed west of the region (Figure 8) and back trajectories showed general west to east transport (Figure 9). Peak O_3 concentrations reached 179 ppbv in Maryland during the episode. Numerous exceedances of the 1-hour O_3 and 8-hour standard were reported across the mid-Atlantic.[23]

Ozone profiles were obtained from research aircraft during this episode and show a regional load similar to July, 1995. For example, observations in extreme southwestern and south-central PA show concentrations of 100 ppbv or higher (Figure 10) during the onset of the episode. It should be noted that these profiles were taken during the late morning and early afternoon hours and shows that the boundary is well mixed. Morning profiles were unavailable for this period because the aircraft was grounded until mid-morning due to low visibility caused by intense haze.

Surface Observations – 1998-2000

Although aircraft observations showed high concentrations of O_3 and precursors are advected into the region during high O_3 events, these flights can be undertaken only occasionally and at great expense. Surface-based monitors could also be used to observe this phenomena on a more routine basis. However, the regulatory network of O_3 monitors are concentrated in or near urban areas. As noted above, because of high concentrations of NO_x and rapid deposition of O_3 to the surface overnight, O_3 concentrations observed at urban sites typically decrease to near zero in the overnight hours. These effects tend to mask regional scale effects. However, there are a small number of rural, higher elevation sites for which this is not the case. For example, long-term measurements have been undertaken at the Big Meadows site in Shenandoah National Park. This location, at 1100 m above sea level, is in a rural area in northwestern Virginia. As such, it is well located to provide information on O_3 entering the mid-Atlantic from the west.[24]

Measurements made over several summers at Big Meadows show that average O_3 concentrations are on the order of 55-60 ppbv. However, sustained higher concentrations

are observed coincident with high regional O_3 levels. An example of this effect is seen in the August 21-25, 1998 episode. While the summer of 1998 was relatively warm overall, weather patterns conducive to high O_3 did not develop until late in the season. During late August, a strong upper air ridge developed over the eastern and central United States. Northwest winds, becoming west, prevailed throughout this period. Observations of O_3 made at Big Meadows show an abrupt rise in O_3 levels during the morning of August 21 (Figure 11). O_3 concentrations at or above 80 ppbv continued through August 25 at Big Meadows and concentrations exceeded the 8-hour standard throughout the mid-Atlantic region during this period with numerous exceedances of the 1-hour standard as well. As seen in the aircraft data, the regional load appears to be on the order of 80-110 ppbv. Of additional interest, a strong signal in SO_2 also accompanies the enhanced O_3 as seen in the aircraft data from previous summers (Figure 12).

During the onset of this high O_3 episode, an intensive observational program for fine particles was ongoing just north of the city of Philadelphia. This program, Northeast Oxidant and Particle Study (NE-OPS), is an adjunct of the larger North Atlantic Research Strategies for Tropospheric Ozone (NARSTO) program and seeks to measure fine particle concentrations as well as O_3 (see, http://lidar1.ee.psu.edu). Observations during the late August period showed a large increase in O_3 levels as air from aloft was mixed downward during the day.[25] A similar effect can be observed when a set of local monitors west (upwind) of the I-95 Corridor is considered. These monitors which range from south-central Pennsylvania (Little Buffalo State Park) to extreme western Virginia (Roanoke) – a distance of approximately 400 km – show nearly identical O_3 concentrations as the nocturnal inversion breaks down and air from aloft is mixed downward (Figure 13). While peak O_3 at these locations varies later in the day in response to local effects (Loudoun and Ashburn for example are far suburbs of Washington DC while Little Buffalo is in a rural state park), the regional signal of 80 or more ppbv O_3 is apparent in late morning. This effect is common during high O_3 episodes and gives a hint of the spatial extent of enhanced O_3. For example, the last event of 1998 (September 12-16, 1998) showed a similar pattern (Figure 14). The strongest episode of 1999 also showed a similar signal (Figure 15). A similar pattern was seen in the first high O_3 event of 2000 (June 9-11) (not shown).

Regional Scale Transport

The manner in which air parcels containing high O_3 are transported through the region depends both on synoptic and meso-scale factors. For example, during the extreme O_3 event of July 12-15, 1995, research grade back trajectory models showed that air parcel arriving in the mid-Atlantic originated at considerable distances to the west.[26] If 24-hour back trajectories are analyzed for the highest O_3 events in 1995-1996, a common pattern of westerly transport is again seen (Figure 3) with similar pattern for 1998 (Figure 16). Transport from this direction, which includes the Ohio River Valley where emissions of NO_x and SO_2 are enhanced, are consistent with aircraft and surface measurements.

However, nearer the surface, and in the smaller scale, transport within the mid-Atlantic region also occurs. For example, transport along the length of the I-95 Corridor in the overnight hours has been observed and modeled during high O_3 episodes.[27] One major mechanism for this effect is, as noted above, the low-level jet which occurs quite frequently during high O_3 episodes. With the addition of radar wind profilers along the eastern seaboard, the low-level jet is more frequently observed. For example, during the July 17, 1999 high O_3 day shown in Figure 15, a radar profiler at Fort Meade, Maryland, near BWI Airport, observed a strong west-southwest low-level jet from just after sunset until dawn the following day (Figure 17). Winds of 8-10 ms^{-1} persisted just above the nocturnal inversion for approximately 10 hours. This implies transport on the order of 200-400 km overnight. Thus, late afternoon emissions from well upwind can impact another site within the mid-Atlantic the following day.

Longer Range Transport

While this report is concerned with transport of O_3 and precursors within a region of the United States, research over the past decade has shown that the region influenced by transported O_3 and precursor occurs on a much larger scale. For example, observations from Nova Scotia show a strong impact of continental emissions.[28] Similarly, observation from Bermuda[29] and over the western Atlantic show a strong impact of continental emissions at long ranges.[30] In fact, satellite images can often identify plumes of polluted leaving the eastern United States (see, e.g., http://www.meto.umd.edu/~ryan /summary99/08june/UNIhaze159_G8.jpg).

Conclusions

Research over the past decade has shown that high O_3 episodes in mid-Atlantic tend to organize in multi-day episodes. These episodes are consistent with a synoptic scale weather pattern that features west to east transport on the order of several hundred kilometers at a minimum. Meso-scale phenomena also occur during these high O_3 episodes that feature nocturnal low-level jets along the eastern seaboard that can transport O_3 and precursors several hundred kilometers overnight. As trace gases such as O_3, and many of its precursors, have long lifetimes away from the surface layer, local emissions can affect the next day's O_3 concentrations at long distances. Observations have shown that O_3 concentrations upwind of the major metropolitan areas of the mid-Atlantic are on the order of 80-100 ppbv during high O_3 episodes. Concentrations at this level are found even in remote rural areas and are associated with increased concentrations of NO_x and SO_2. As the 8-hour standard is 85 ppbv, concentrations of O_3 near the standard are often present and in the presence of additional O_3-forming precursors means that exceedances of the 8-hour standard in urban areas are inevitable during regional events. Therefore, pollution controls affecting O_3 levels at large distances, of the order of hundred of km, are necessary to attain the 8-hour standard in the metropolitan areas of the mid-Atlantic.

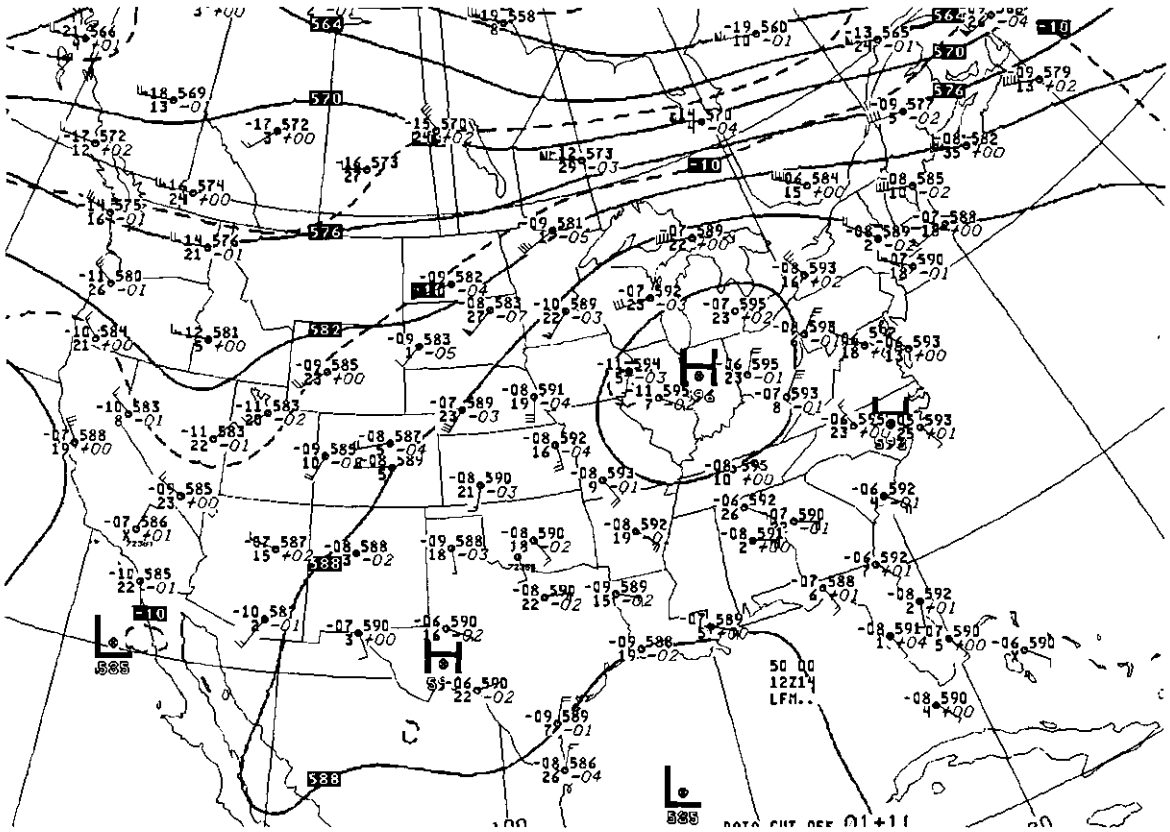

Figure 1. National Centers for Environmental Prediction (NCEP) 500 mb analysis for 1200 UTC, July 14, 1995 during a widespread ozone episode with regional haze. Contours are in decameters; station reports use standard form.

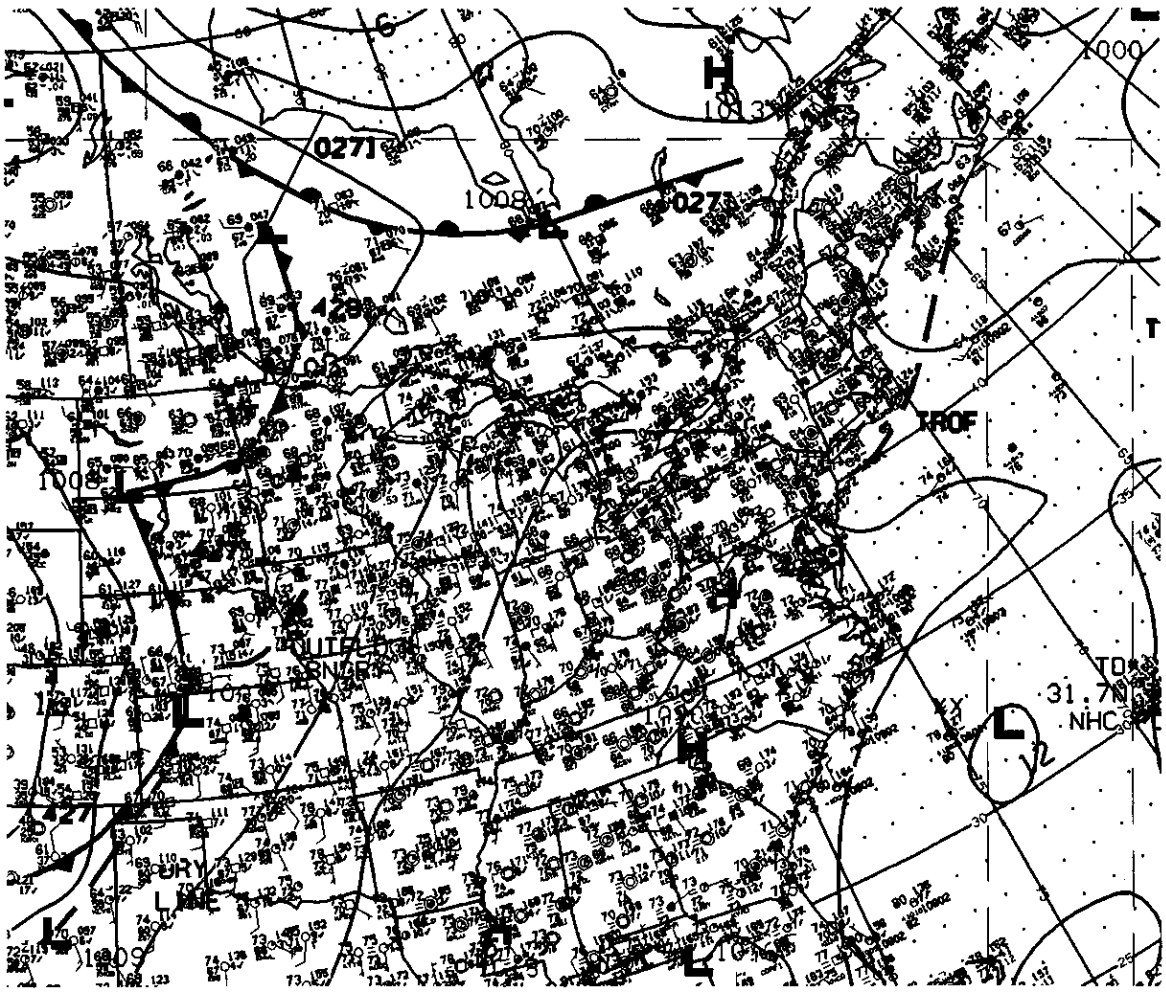

Figure 2. National Centers for Environmental Prediction (NCEP) surface analysis for 1200 UTC July 13, 1995.

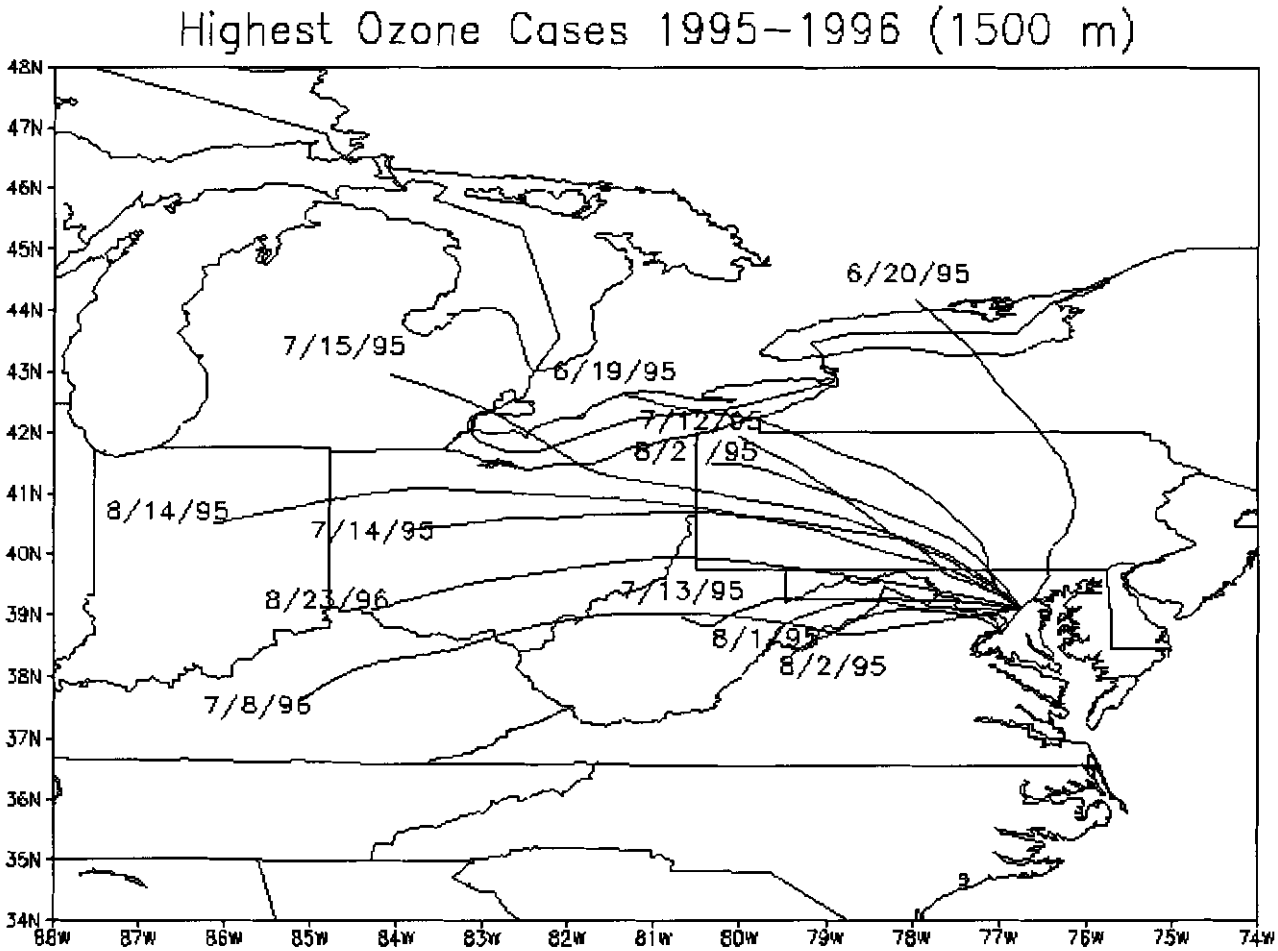

Figure 3. HYSPLIT back trajectories for Fort Meade, Maryland for the highest O_3 cases of 1995. Trajectories are initialized at 1500 m above ground level each day at 1200 UTC and carried 24 hours. The date of termination of each trajectory is given near its initialization point. Details on the HYSPLIT model can be found at the citations given in Footnote 26.

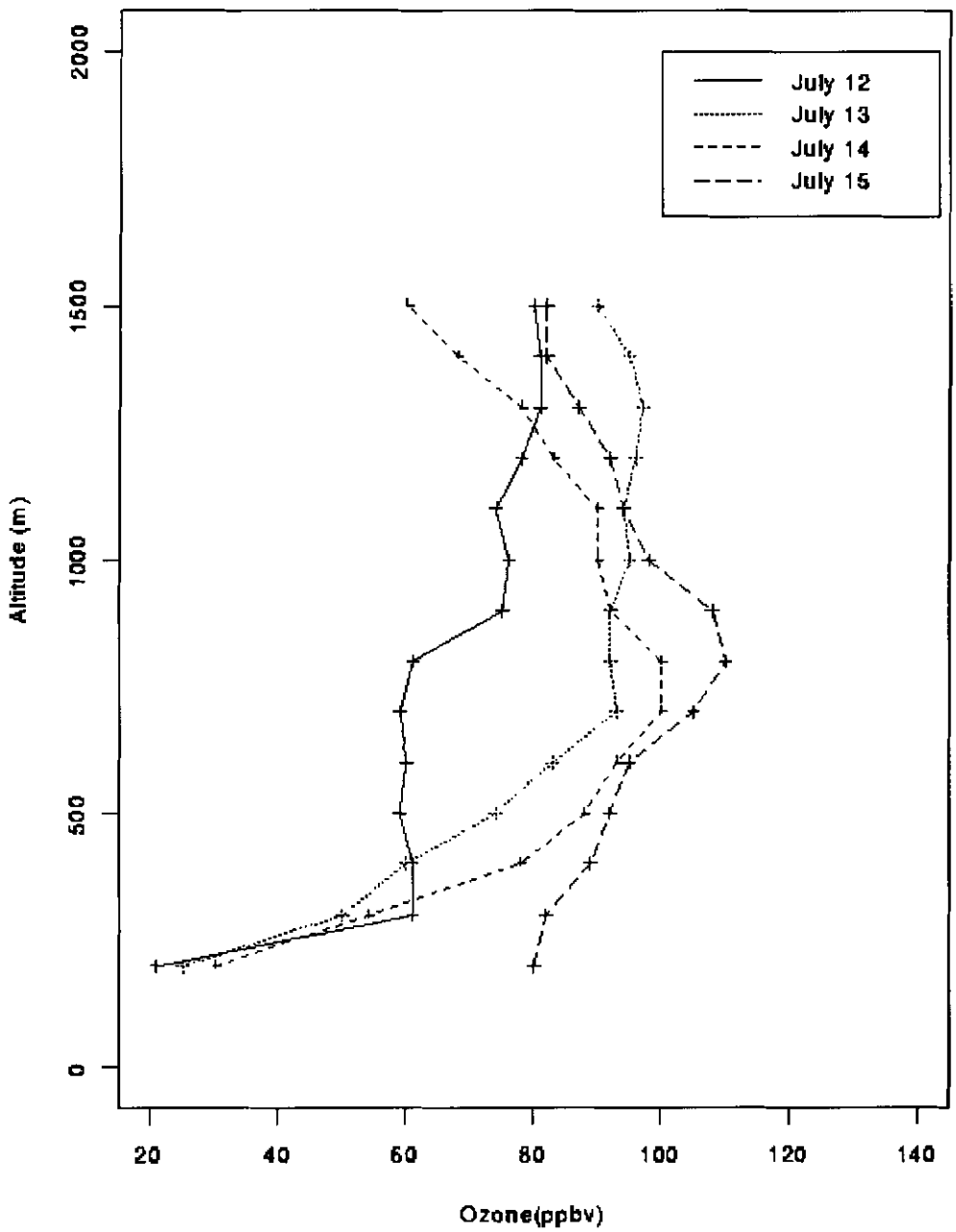

Figure 4. Vertical profiles of O_3 concentrations measured near Gettysburg, PA just after sunrise during the high O_3 episode of July 12-15,1995. Data collected by Sonoma Technologies as part of the NARSTO-Northeast research program. Ozone is given in parts per billion by volume (ppbv) and height as meters above ground level (agl).

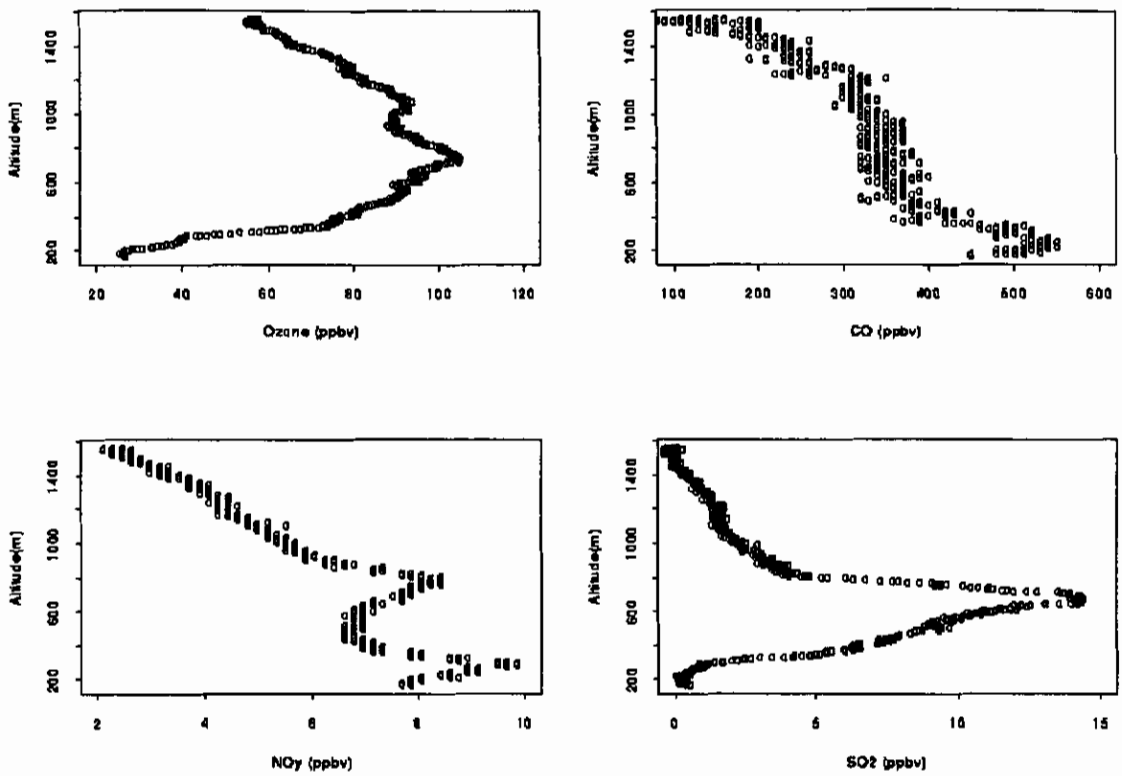

Figure 5. Vertical profiles of O_3, CO, NO_x and SO_2 concentrations measured near Gettysburg, PA just after sunrise on July 14,1995. Data collected by Sonoma Technologies as part of the NARSTO-Northeast research program. Trace gas concentrations are given in parts per billion by volume (ppbv) and height as meters above ground level (agl).

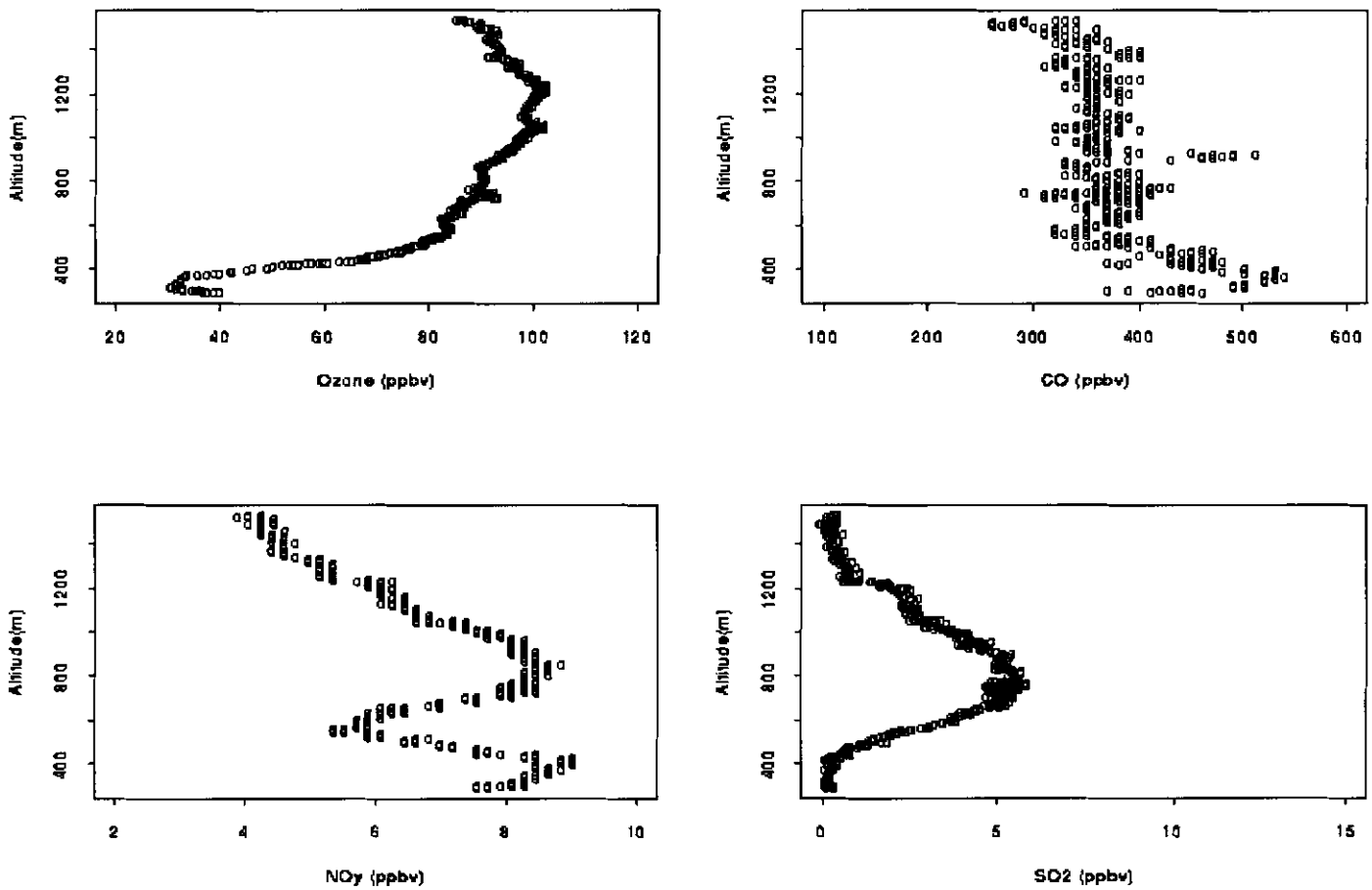

Figure 6. As in Figure 5, but for western Virginia near Shenandoah National Park. July 14, 1995.

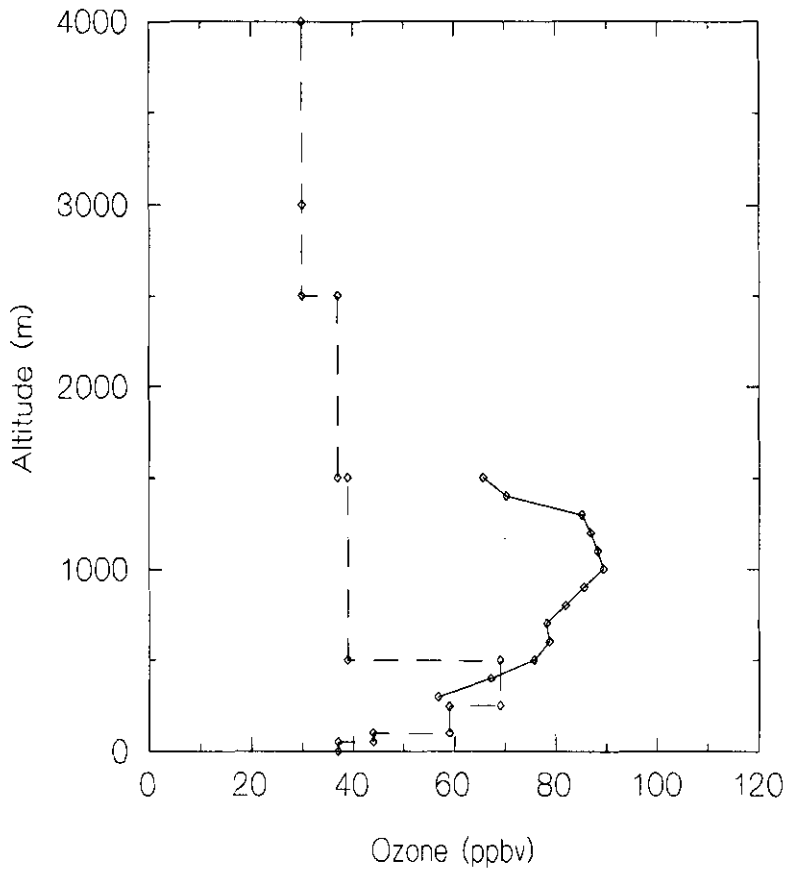

Figure 7. A comparison of observed O_3 in western Virginia on July 15, 1995 with modeled concentrations from the Urban Airshed Model (Version 5) from the Ozone Transport and Assessment Group (OTAG) modeling exercise. Concentrations from the model are given as layer averages, observed O_3 is expressed in 200-m averages.

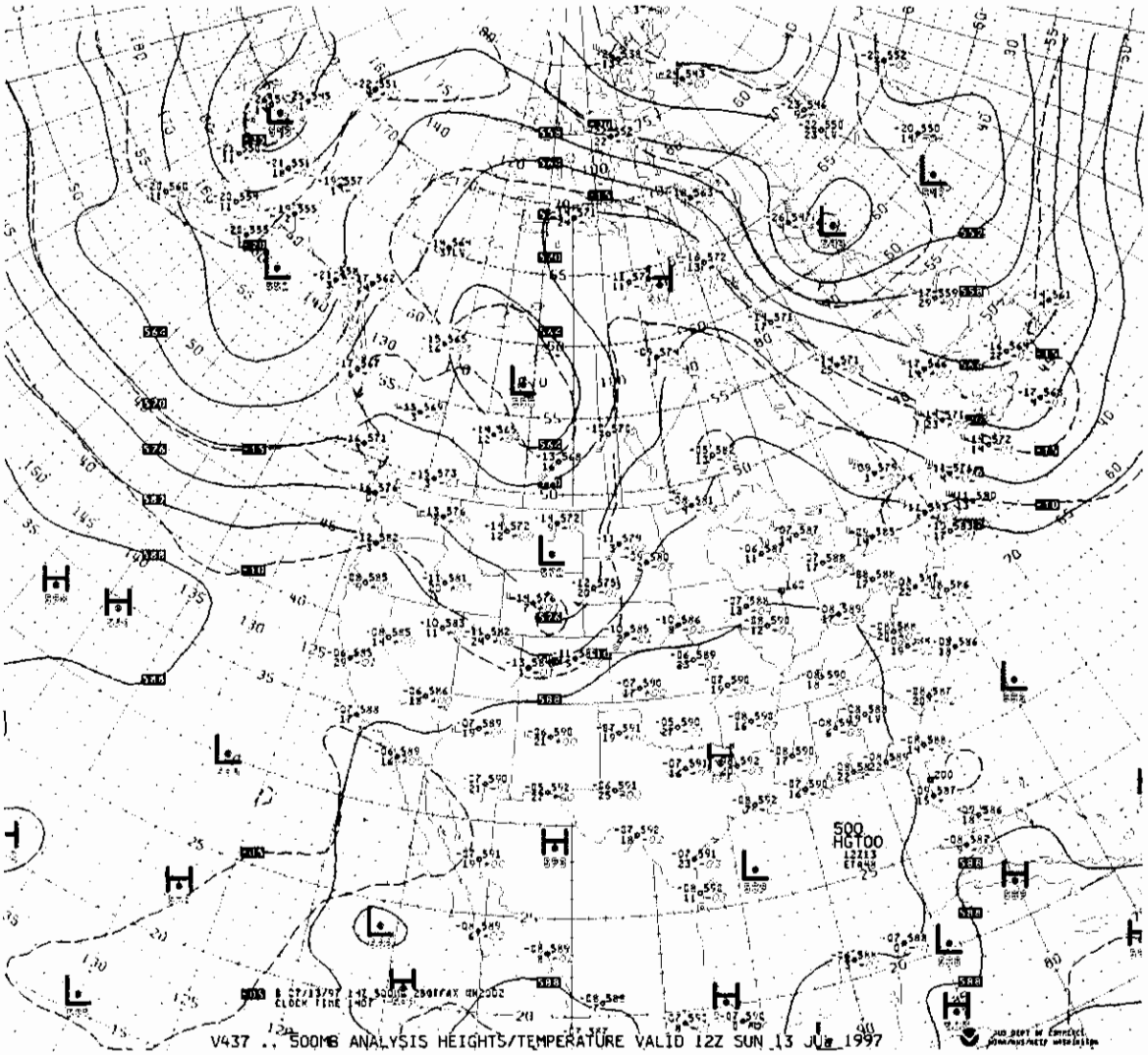

Figure 8. As in Figure 1 but for July 13. 1997.

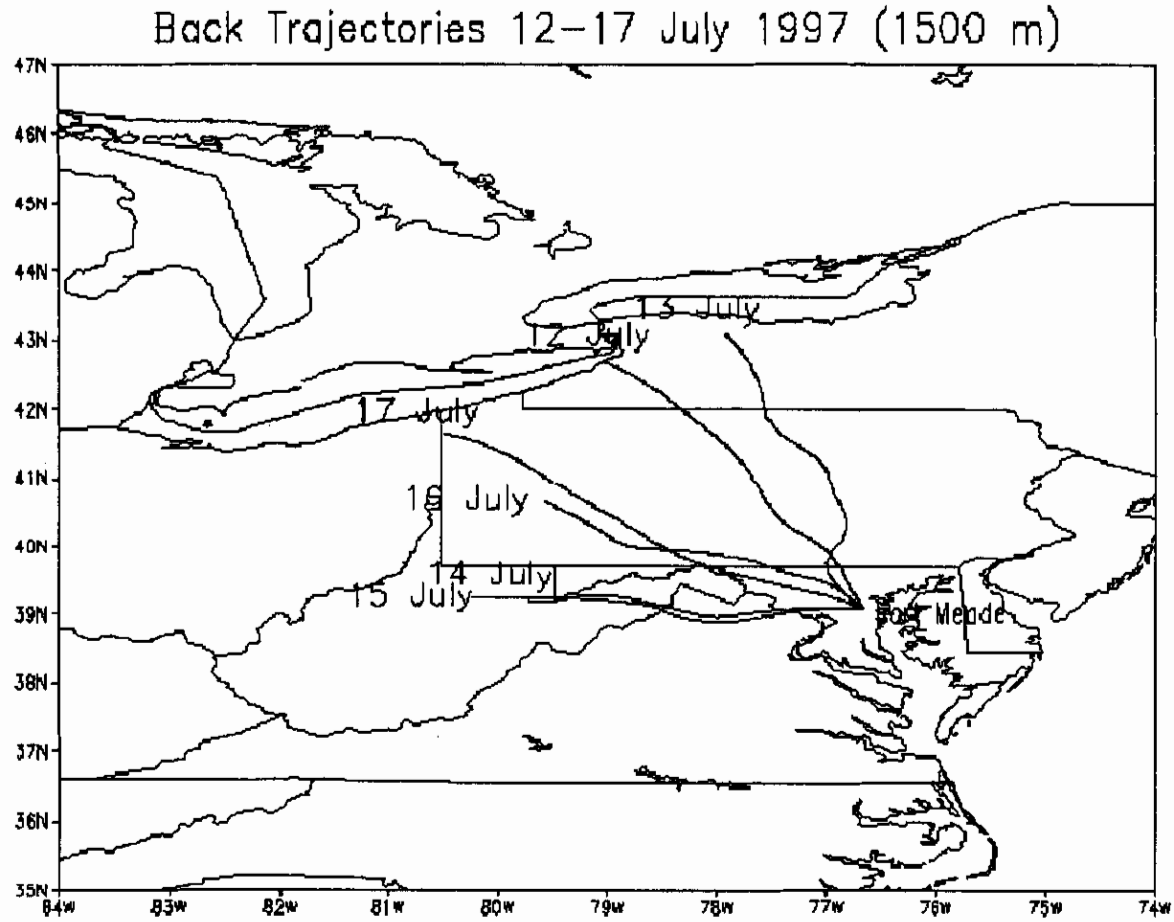

Figure 9. As in Figure 3 but for the high O_3 episode of July 12-17, 1997.

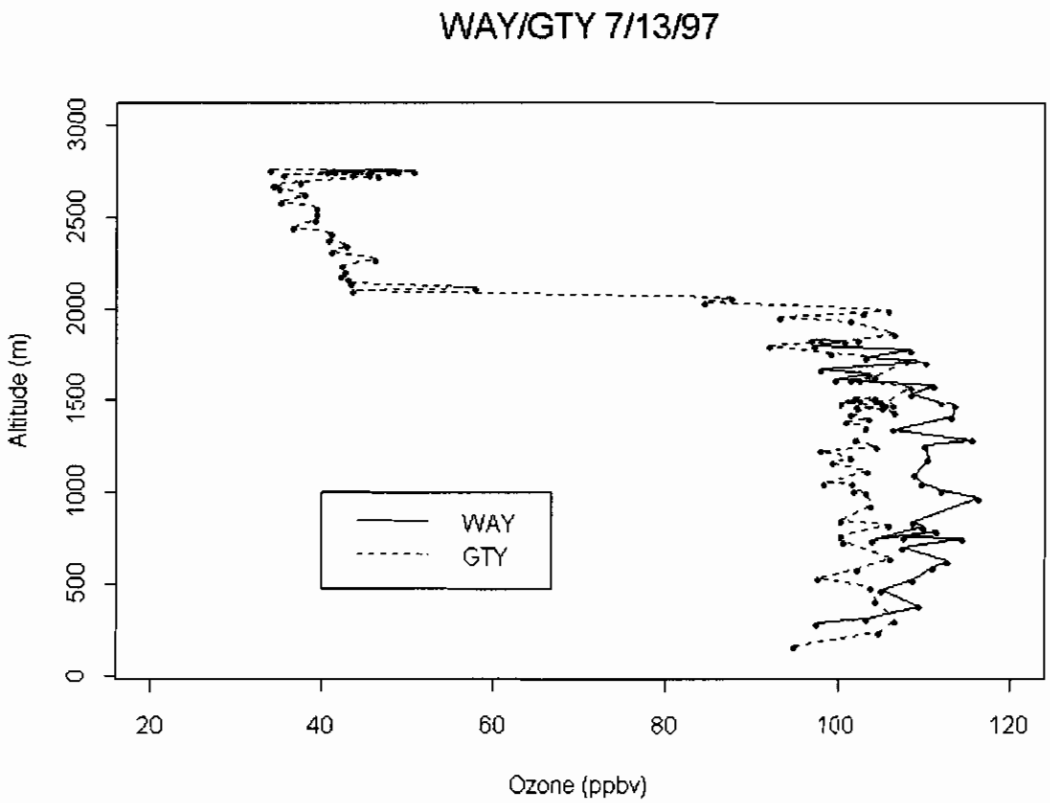

Figure 10. As in Figure 4 but for July 13, 1997. "WAY" refers to Waynesboro, PA in southwestern PA and "GTY" to Gettyburg, PA. Profiles at WAY made at approximately 1500 UTC and at GTY at 1700 UTC.

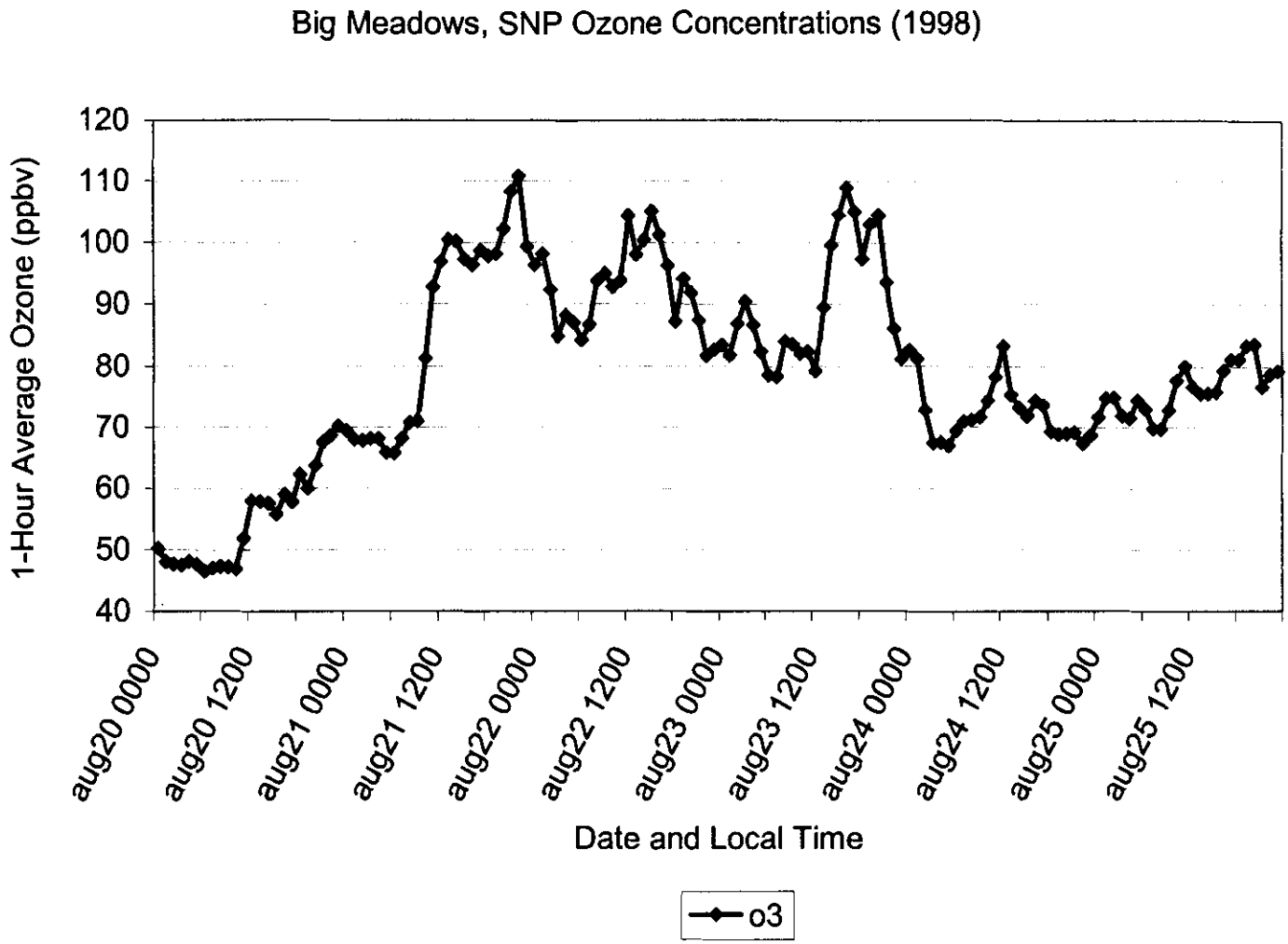

Figure 11. Hourly average O_3 concentrations from Big Meadows in Shenandoah National Park during the high O_3 episode of August 21-25, 1998.

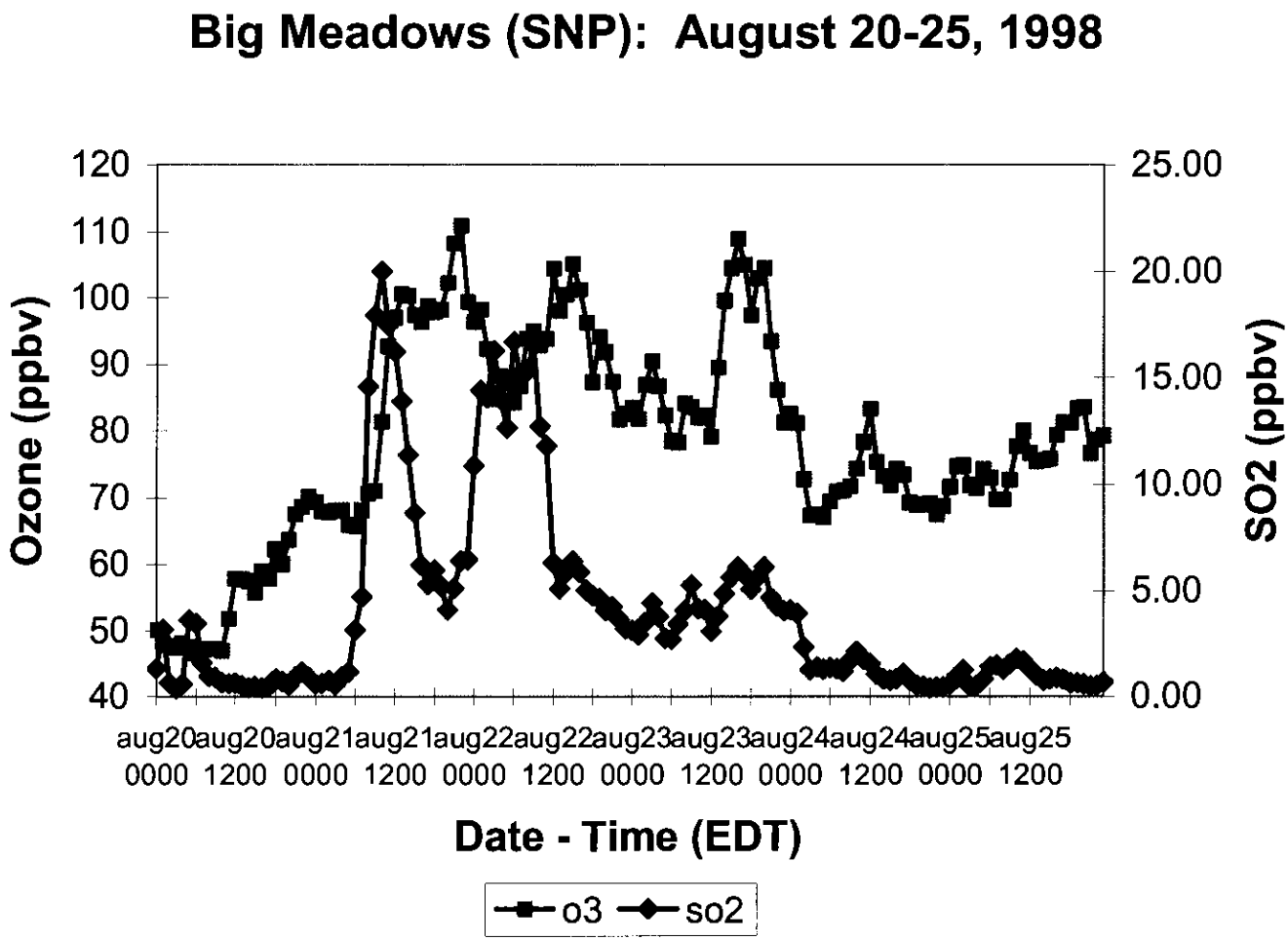

Figure 12. As in Figure 11 but for O3 (left y-axis) and SO_2 (right y-axis). For reference, mean SO2 for the summer of 1998 was approximately 1 ppbv.

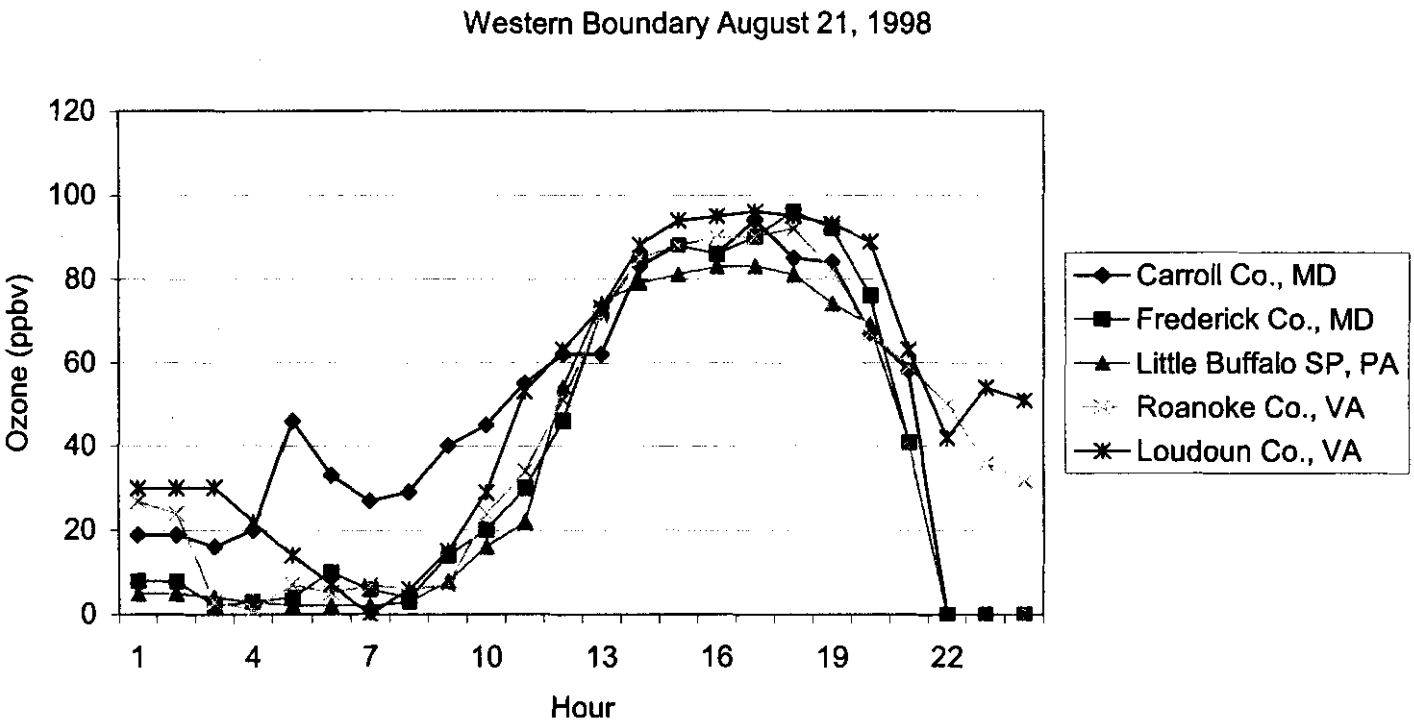

Figure 13. Hourly O_3 concentrations for August 21, 1998 at a group of state-operated monitors west of the I-95 Corridor ranging from south central PA (Little Buffalo) to western Virginia (Roanoke). Missing data for hours 22-24 are set to zero.

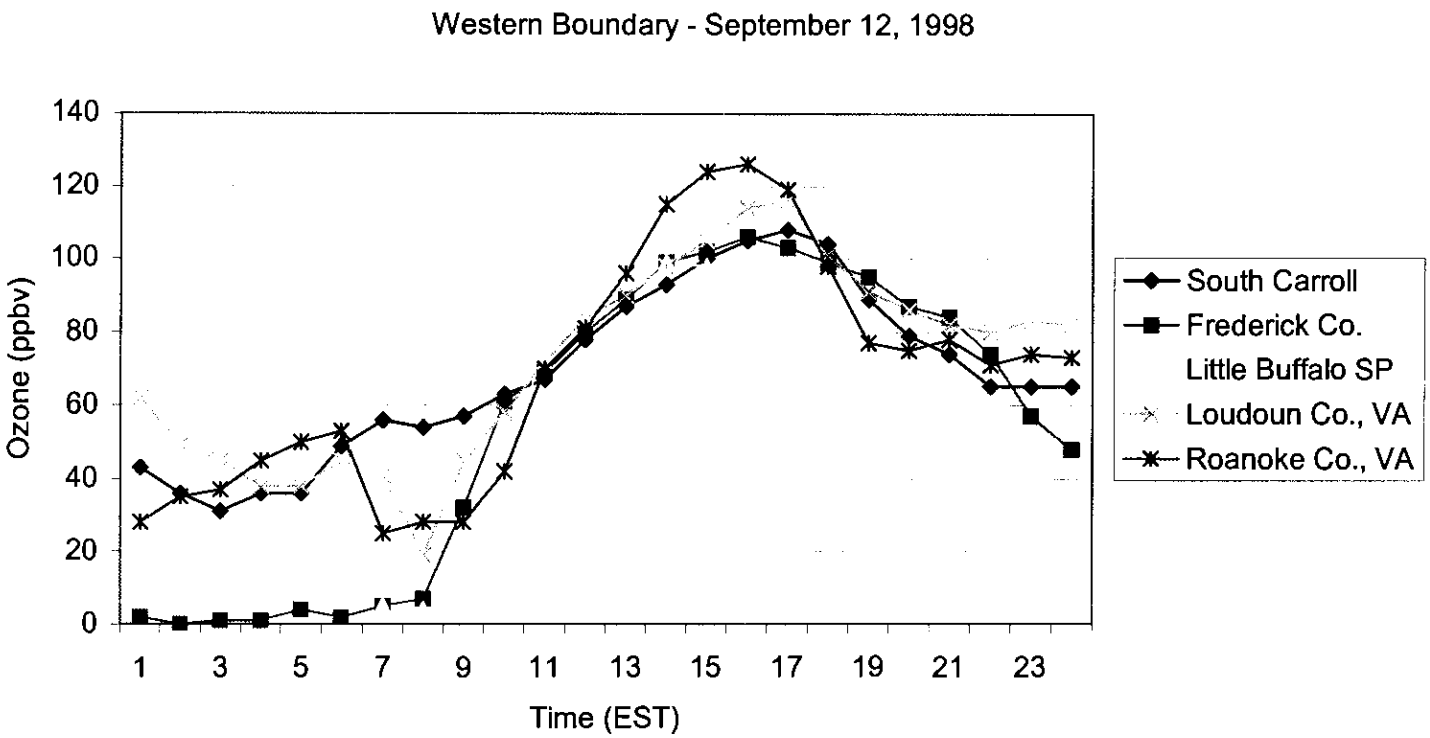

Figure 14. As in Figure 13 but for September 12, 1998.

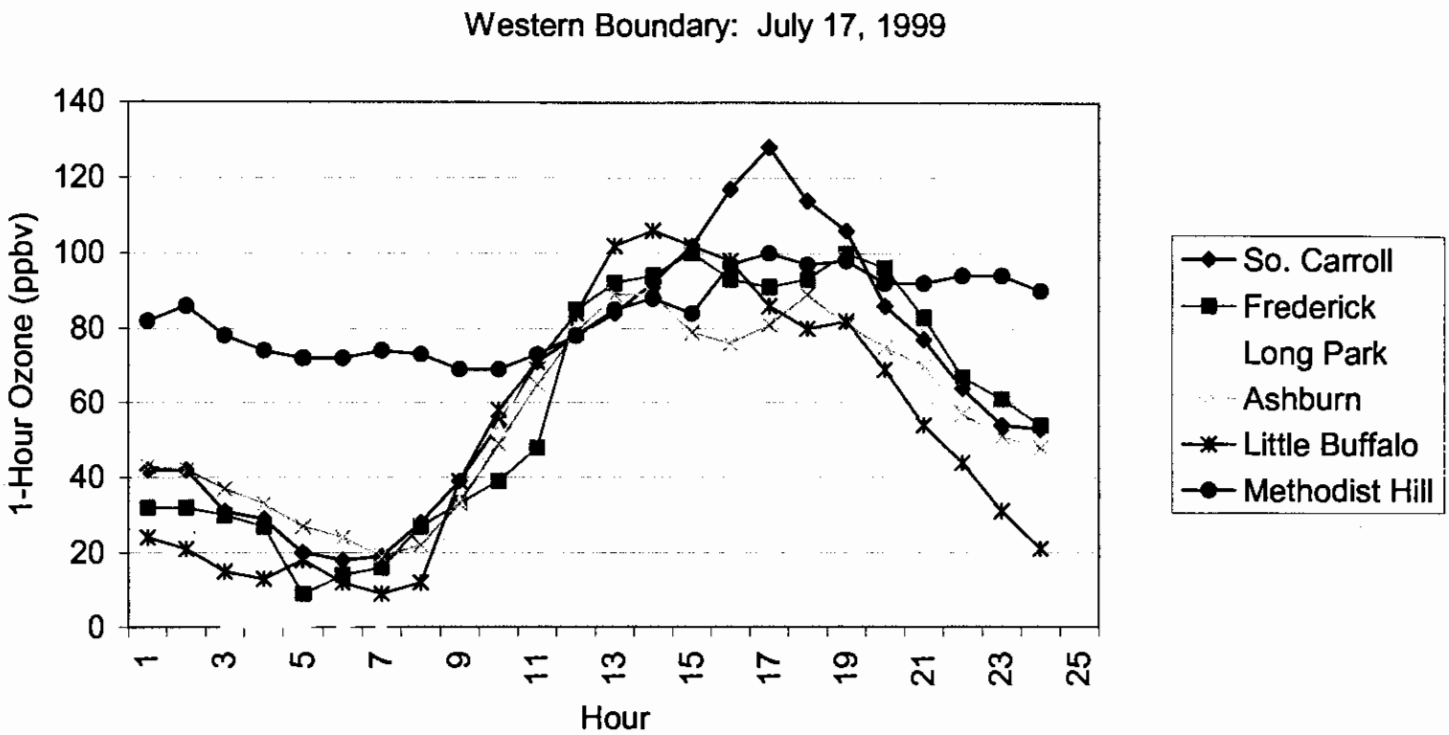

Figure 15. As in Figure 13 but for July 17, 1999.

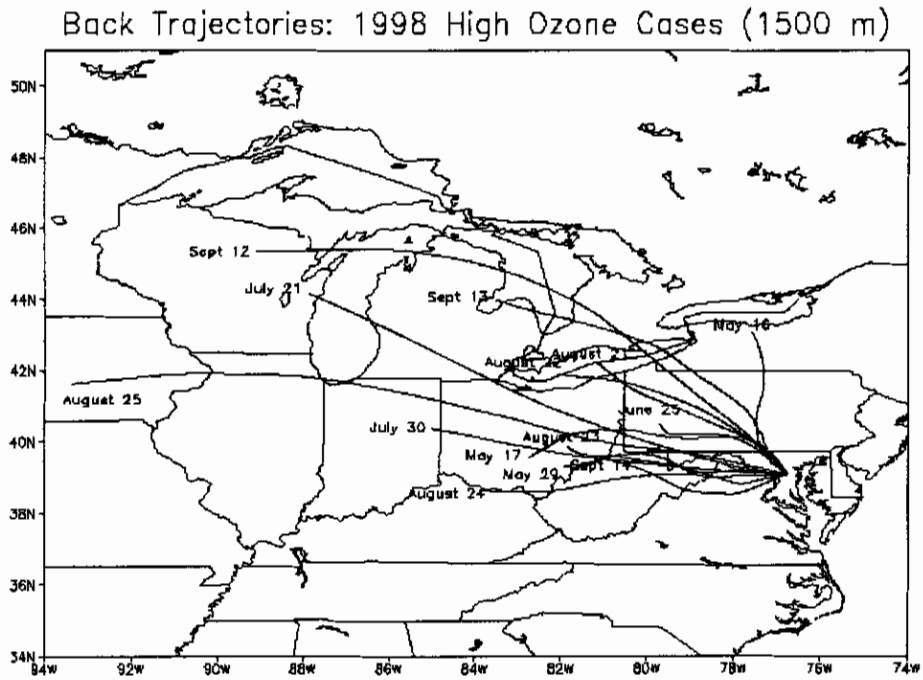

Figure 16. As in Figure 3 but for the highest O_3 cases in the Baltimore region during 1998.

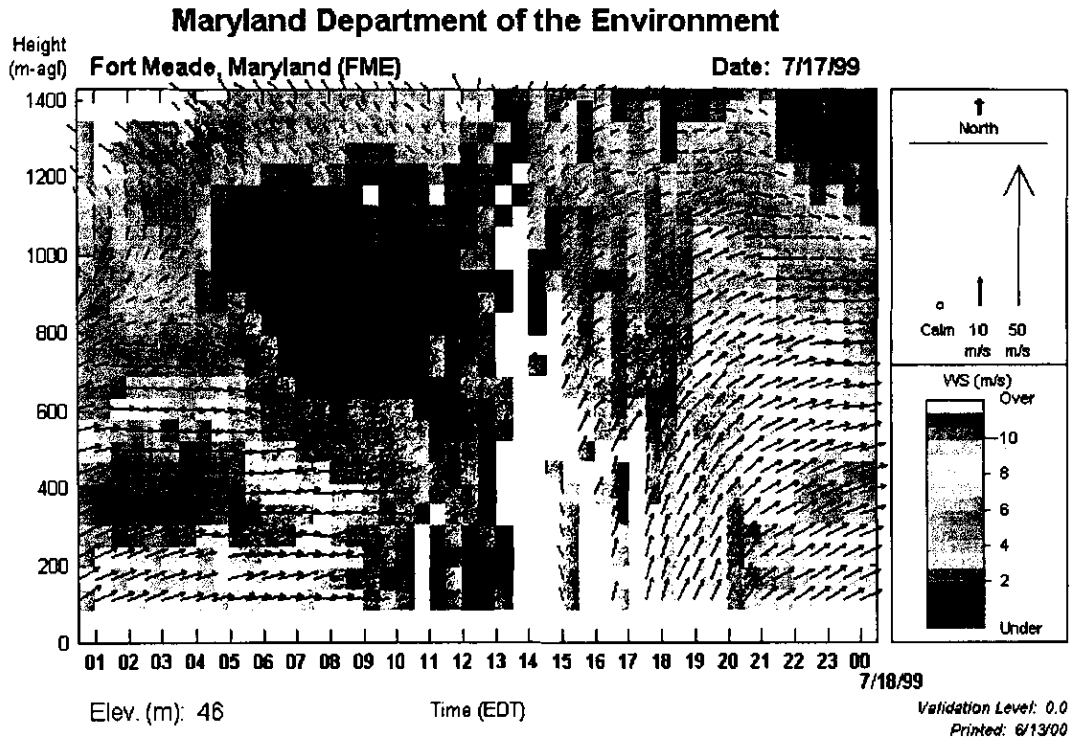

Figure 17. Vertical wind profile from the radar wind profiler operated by the State of Maryland Department of the Environment near Fort Meade, Maryland on July 17, 1999. Winds are given as a function of height in meters.

ATTACHMENT 1

Overwhelming Transport and Maryland's Ground-Level Ozone Problem

Maryland's air pollution problem is very serious and complex. State monitors record very high concentrations of ground-level ozone and high concentrations of fine particulate matter. Emissions in Maryland are relatively small when compared to the emissions in other states that contribute significantly to Maryland's air pollution problem. Maryland's air pollution problem is clearly influenced by a wide area of emissions (most of the East) and the very unique meteorology of the Mid-Atlantic region.

Maryland has been analyzing ozone transport since the early 1990's. This research includes ground-level and aircraft based measurement campaigns and photochemical modeling. In partnership with the University of Maryland's Department of Meteorology, EPA and many other states, Maryland has developed a simplified conceptual model of how and why high ozone concentrations end up in Maryland.

This simplified conceptual model has four basic components:

1. Local, Maryland emissions (Maryland's contribution)
2. Smaller scale, local transport (contributions from areas directly to the southwest of Maryland)
3. Large scale, westerly "aloft" transport (contributions from areas to the west, northwest and southwest of Maryland)
4. Medium scale, southerly "low level, night-time jet" transport (contributions from the south and southwest of Maryland)

On Maryland's worst ozone days (often associated with a Bermuda High setting up south of Maryland along the Atlantic coast) all four of these components play an important role in creating unhealthy ozone levels. On these days, emissions from other states overwhelm Maryland's own contribution and make it virtually impossible for Maryland to solve it's own ozone problem.

Mid-Atlantic Meteorology and Ozone Episodes

Many of Maryland's worst ozone days are associated with a Bermuda High setting up over southern Virginia or northern North Carolina. During this kind of weather pattern there is abundant sunshine and ground-level winds are generally light and from the southwest. This type of weather is perfect for producing ozone from "local" emissions that slowly move to the northeast over the course of the day. These "local" emissions clearly include Maryland, but also include the emission rich areas to the south such as Washington D.C. and central Virginia. The primary sources in this emission rich "local contribution" area are cars and other mobile sources and area sources associated with densely populated areas (painting, consumer products, etc.).

As a result of the Bermuda High, aloft winds move in a clockwise direction around the high. This wind pattern captures emissions and pollution from the emissions rich area in and around the Ohio River Valley. This area's power plants are the primary sources of pollutant emissions that form the "aloft contribution". These emissions and the pollution that they create can be transported for hundreds of miles aloft into Maryland where they "mix down" in the late morning or early afternoon as the atmosphere heats up.

During this same period, southerly night time winds that form approximately 1,000 feet above the earth's surface can become quite important to the ozone that forms over Maryland the next day. This phenomena is commonly referred to as the low level night-time or low level nocturnal jet. The jet forms east of the Appalachian mountains and pushes emissions and pollution for hundreds of miles from the south into Maryland. The sources that create this pollution include your typical mix of sources associated with areas experiencing growth: cars and mobile sources, area sources, power plants and manufacturers. This pollution travels towards the northeast approximately 1000 feet above the earth's surface trapped above something called the "nocturnal inversion". Again, when the earth's surface heats up in the late morning of the next day, this trapped pollution mixes down and creates an urban ozone soup as it combines with the "local" pollution and the "westerly transport" pollution. When all of these components are present, Maryland experiences extremely high ozone levels.

To reduce ozone to levels below the 8-hour standard, there will need to be aggressive strategies to reduce local emissions and equally aggressive strategies to lower the emissions being transported into the State from the south and west.

Selected papers and reports from the scientific literature that support the conclusions in this paper are listed below.

Selected Papers and Reports on Mid-Atlantic Transport

1. Ryan, W.F. and Dickerson, R.R., Regional Transport of Pollutants and Implications for 8-Hour Ozone Non-Attainment Areas in Maryland, July 12, 2000.
2. Ryan, W.F., et. al., Transport and Meteorological Regimes During High Ozone Episodes in the Mid-Atlantic Region: Observations and Regional Modeling, 10th Joint Conference of the Applications of Air Pollution Meteorology with the Air and Waste Management Association (AWMA), 11-16 January 1998.
3. Ryan, W.F., Forecasting Severe Ozone Events in the Baltimore Metropolitan Area, Atmos. Environ., 29, 2387-2398, 1995.
4. Ryan, W.F., et.al., Pollutant Transport During a Regional Ozone Episode in the Mid-Atlantic Region, J. Air & Waste Management, 48, 786-797, 1998.
5. U.S. Environmental Protection Agency Office of Air and Radiation, Air Quality Modeling Technical Support Document for the NOx SIP Call, September 23, 1998.

6. Virginia Department of Environmental Quality, Attainment Demonstration Modeling Report for the Washington DC-MD-VA Ozone Nonattainment Area (Draft), January 15, 1998.

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Pollutant Transport During a Regional O_3 Episode in the Mid-Atlantic States

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ABSTRACT

Ozone (O_3) concentrations in the Baltimore-Washington (B-W) metropolitan area frequently exceed the National Ambient Air Quality Standard (NAAQS) in the summer months. The most extreme O_3 events occur in multi-day high O_3 episodes.[1] These events can be regional in scale, with O_3 concentrations exceeding the NAAQS at numerous locations along the eastern U.S. seaboard, and are typically associated with slow-moving or stagnant high pressure systems.[2-5] In the B-W region, the most extreme events typically occur with surface high pressure overhead or just west of the region and an upper air high-pressure area (ridge) to the west or northwest.[1] Besides providing conditions conducive to local O_3 production (subsidence and strong low-level inversions, weak horizontal winds, little cloud cover), this weather pattern may also result in transport of O_3 and its precursors from heavily industrialized areas west and north of the B-W region. In this paper, observations and back trajectories made during the severe regional O_3 event of July 12–15, 1995, are used to confirm the hypothesis that significant regional-scale transport of O_3 and its precursors occur during extreme O_3 events of the standard type in the B-W area.

IMPLICATIONS

During a strong ozone (O_3) event in the mid-Atlantic region, O_3 concentrations along the upwind boundary reached 80–110 ppbv. These high regional O_3 concentrations suggest that regional-scale pollution controls may have an impact on urban-scale peak O_3 in the mid-Atlantic. Layers of high O_3 above the surface are found in association with local maxima in concentrations of sulfur dioxide and total reactive nitrogen. Concentrations of nitric oxide are very low. This suggests that the O_3-enriched air is photochemically aged, has been transported some distance, and has a source, at least in part, in coal combustion. Current regional modeling of this episode should be able to account for observations of enhanced O_3 along the upwind boundary during this episode.

DATA

The data used in this study were collected during the summer of 1995 as part of continuing observational studies carried out by the University of Maryland (UM) under the aegis of the Maryland Department of the Environment–Air and Radiation Management Administration (MDE-ARMA) and the North American Research Strategy for Tropospheric Ozone–Northeast (NARSTO-NE) field program.[3] The NARSTO-NE program included observations throughout the eastern United States. In this study, we focus on the mid-Atlantic region and, in particular, on rural surface chemistry sites and aircraft observations.

Surface Sites

Several regional-scale surface chemistry sites in the mid-Atlantic region were operated as part of the NARSTO-NE program (Figure 1 and Table 2). Data were collected in rural Virginia at Big Meadows, Shenandoah National Park (SNP), by UM. The site at SNP is located in an isolated rural area within the park bounds and has been in operation for several years. Mean summer O_3 concentrations at SNP vary year to year. The mean summer season concentrations for the period 1983–1989 were 55 ± 12 ppbv.[6,7] Unlike urban-scale sites, the high elevation rural site at SNP has little or no diurnal O_3 concentration variation.[6] During the summer of 1995, observations of carbon monoxide (CO), O_3,

Table 1. Glossary of terms.

Abbreviation	Term
ASL	Above Sea Level
B-W	Baltimore-Washington
HY-SPLIT	Hybrid Single-Particle LaGrangian Integrated Trajectory
LLJ	Low-level nocturnal jet
MDE-ARMA	Maryland Department of Environment-Air and Radiation Management Administration
NAAQS	National Ambient Air Quality Standard
NARSTO-NE	North American Research Strategy for Tropospheric Ozone
NWS	National Weather Service
UM	University of Maryland
UTC	Univeral Coordinated Time, formerly Greenwich Mean Time

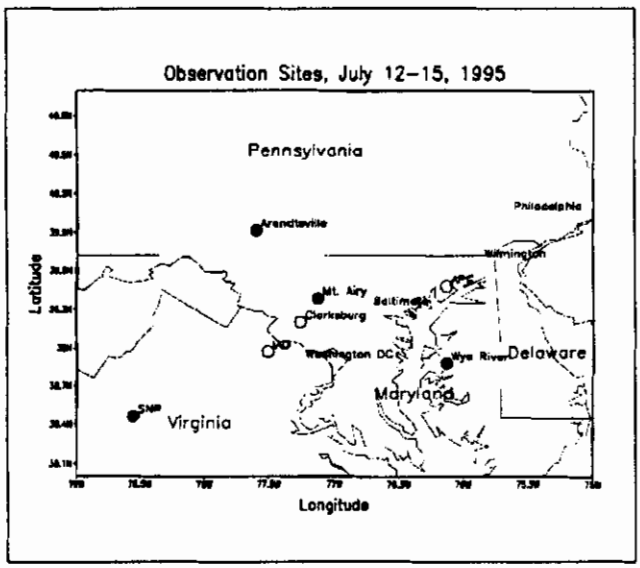

Figure 1. Map of the B-W region showing surface air quality sites and locations of aircraft profiles (solid circles) and upper air meteorological sounding sites (open circles) referred to in this paper.

Table 2. Locations of stations used for meteorological and chemical analysis. Latitude and longitude are given in decimal degrees, elevation in meters.

Station	Abbreviation	Lat	Lon	Elevation (m)
Aberdeen, MD	APG	38.8	76.1	4
Andrews AFB, MD	ADW	38.8	76.9	86
Arendtsville, PA	-	39.9	77.3	269
Baltimore-Washington Airport	BWI	39.2	76.7	45
Clarksburg, MD	-	39.2	76.2	195
Dulles Airport, VA	IAD	38.9	77.4	94
Martin State Airport	MTN	39.3	76.4	17
Shenandoah National Park	SNP	38.5	78.5	1100
Wilmington, DE	ILG	39.7	75.4	24
Wye River, MD	-	38.9	76.2	5

sulfur dioxide (SO_2), and oxides of nitrogen (NO, NO_y) were routinely collected.

A NARSTO-NE surface site operated by Environmental Science and Engineering, Inc., as part of the NARSTO-NE program was located at Arendtsville, PA. In addition to standard meteorological parameters, CO, O_3, SO_2, NO, and NO_y were measured. In addition to the NARSTO-NE site at SNP, UM also operated a station near Queenstown, MD, at the Wye Research and Education Center. The Wye River site is located in an agricultural area approximately 50 km east of the Baltimore-Washington (B-W) area.[8]

Upper Air Data

During forecasted O_3 episodes, supplemental soundings were taken at the National Weather Service radiosonde station at Sterling, VA (IAD), and Aberdeen Proving Grounds in Maryland (APG) at 4-hr intervals. In addition, UM operated a temperature and humidity sonde near Clarksburg, MD, during these events.

Aircraft Data

In order to measure vertical trace gas profiles in the vicinity of the B-W area, instrumented research flights were flown during the episode. Flights by UM were carried out by a single-engine Cessna 172 (Aerosource, Inc., Bay Bridge, MD) measuring O_3, relative humidity, and temperature (10-sec measurements) from near the surface to approximately 3 km above the surface. Research speed of the aircraft was approximately 50 $msec^{-1}$ with a climb rate of 10–25 $msec^{-1}$. Morning flights were taken upwind of the B-W region from July 13–15, and afternoon flights were taken during the same period downwind of the urban areas.

A Piper Aztec aircraft was operated by Sonoma Technology, Inc., as part of the NARSTO-NE program.[3] This aircraft measured temperature, relative humidity, and concentrations of O_3, SO_2, NO, and NO_y along the eastern seaboard. The trace gas profiles reported here were made in the early morning hours of July 12–15 in the vicinity of the Arendtsville and SNP sites at altitudes from near the surface to 1600 m above sea level (ASL).

METEOROLOGY OF THE JULY 12–15 EPISODE

During early July 1995, a strong upper air ridge built over the Midwest. Weather related to this ridge caused an extreme heat emergency situation in Chicago before slowly moving east.[9] While a large area of surface high pressure blanketed the eastern United States, at upper levels the weather pattern was characterized by a ridge centered over the Ohio and Tennessee Valleys with the axis of the ridge west of the B-W region. In Figure 2, the geopotential height field at 850 mb for 0000 UTC (Universal Coordinated Time, formerly Greenwich Mean Time) on July

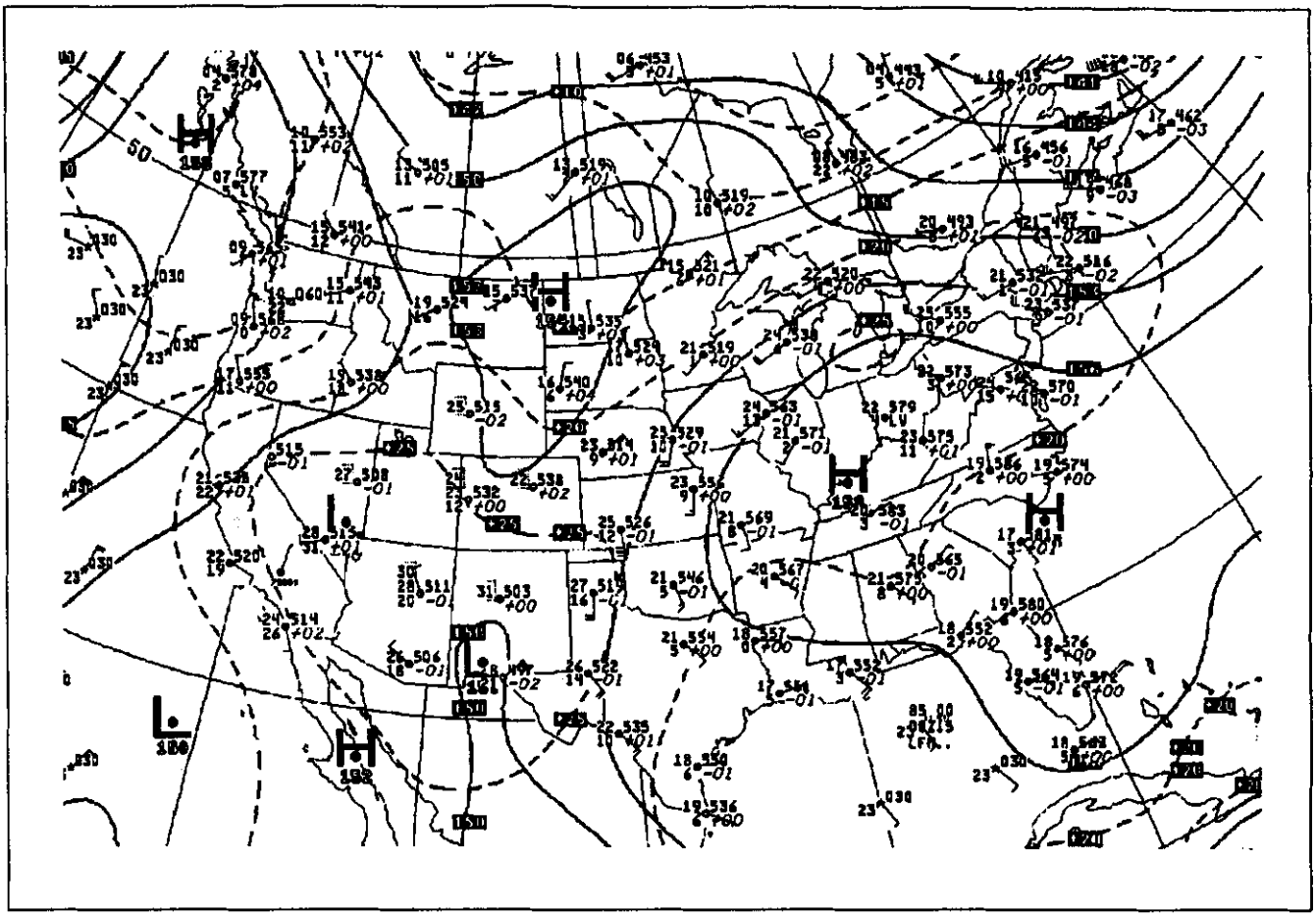

Figure 2. National Weather Service analysis of geopotential height field at 850 mb for 0000 UTC July 15, 1995.

15 is displayed showing the location of the ridge. This pattern is similar to historic high O_3 events in the mid-Atlantic region.[1,2] Eight of the 10 most severe O_3 episodes in the B-W region during the period 1983–1990, including the major O_3 events of 1988, were associated with this type of pattern. Due to the similarity of this weather pattern to historic O_3 episodes and the consistency of the medium-range forecast models in predicting this pattern over several model runs, warnings of unhealthy concentrations of O_3 were first issued by MDE-ARMA five days prior to the start of the event in the B-W area.[10]

A ridge to the west or northwest of the mid-Atlantic region maximizes the local potential for O_3 formation in several ways. First, dynamics associated with a synoptic-scale (1,000–2,500 km) ridge causes subsidence (downward motion) downstream (east) of the ridge axis. Subsidence inhibits cloud formation and strengthens the low-level inversion so that photochemistry is maximized and vertical mixing of low-level emissions minimized. Along the East Coast of the United States, synoptic-scale subsidence can be enhanced by interactions with orography. Winds from the west or northwest flow downslope normal to the eastern slope of the Appalachians. This descending air is warmed adiabatically, resulting in a layer of warm air aloft, which increases stability. An upper air ridge to the west of the region also leads to transport of air from the northwest or north at these levels. For 25 of the 32 most severe O_3 days in 1987–1994 for which upper air data are available, local observed winds (at IAD) at 850 mb (~1.5 km) were from the northwest, north, or northeast. In the remaining seven cases, the observed winds were from the west-southwest (233°–256°).

Meteorological conditions at the Baltimore-Washington International Airport (BWI) reflected the persistent high pressure regime during the episode. Daily maximum temperature rose steadily from 33.2 °C on July 12 to 38.9 °C on July 15. Though haze was reported at many sites, skies remained mainly cloud free throughout the episode, with little convective activity. Along the B-W Corridor at BWI and IAD, light westerly surface winds were measured on July 12, became southwest winds on July 13, and then reverted to west-northwest winds during the most severe O_3 days (July 14–15). However, considerable local variation in surface wind direction occurred each day. At Martin State Airport northeast of Baltimore, bay breeze (southeasterly) circulations developed each afternoon, as is common in anticyclonic conditions. Further inland, stations at Andrews Air Force Base and Wilmington, DE, observed

generally west to northwest winds overnight and into the morning hours, with southwest winds developing during the day.

During the July episode, upper level winds in the B-W region were generally west and northwest with intermittent periods of low-level southwest winds. At IAD and APG, west or northwest winds were observed at approximately 600 m and above throughout the episode. There were periods of variation from this general flow direction and additional key wind field features that bear mentioning. First, a layer of southwesterly winds from the surface to 1.0–1.5 km was observed from the evening of July 12 until the evening of July 13. Second, nocturnal low-level jets (LLJs) featuring southwesterly flow were observed during the early morning hours of July 13 and 14 at both APG and IAD. These jets contained maximum winds varying from 11–16 $msec^{-1}$, with the level of maximum winds ranging from 160–670 m. The LLJs were typically observed during the supplemental soundings taken during the period 0300–0800 UTC. There was considerable local and temporal variation in the strength of the LLJs during this episode. The LLJs were typically stronger and more persistent at APG compared to IAD. This is consistent with theoretical understanding of the LLJs in this region as primarily a coastal plain phenomena.[11-13] The juxtaposition of westerly and northwesterly flow aloft with surface winds from the southwest or south and intermittent LLJs was observed in other O_3 events during 1995.[11] On the morning of July 15, there was a transient but strong low-level wind maximum from the northwest observed at IAD and APG from 0300–1100 UTC. Although the temporal scale and location of wind speed maxima of this feature is consistent with southwesterly LLJs, the wind direction is inconsistent with standard LLJs in this region. This feature is better understood as an adjunct of a weak trough north of the region moving over the top of the ridge (Figure 2). This trough, or region of lower pressure aloft, was associated with a weak "back door," or north-to-south moving, cold front that later penetrated as far south as the Philadelphia International Airport (PHL) on July 16. The layer of strong northwesterly winds (200–1000 m) on July 15 also contained the layer of maximum upper-level concentrations of O_3, SO_2, and NO_y upwind of the region.

The presence of a strong upper air ridge and its effect on the evolution of the boundary layer was seen in successive radiosonde observations at Clarksburg during this episode (Figure 3). The radiosonde observations were taken at approximately 1800 UTC each day, near the time of maximum boundary layer depth. The lower troposphere warmed steadily each day, and the inversion height could be seen lowering and strengthening through the period, with the base of the upper-level inversion decreasing from 2.8 km on July 12 to approximately 1.0 km on July 15.

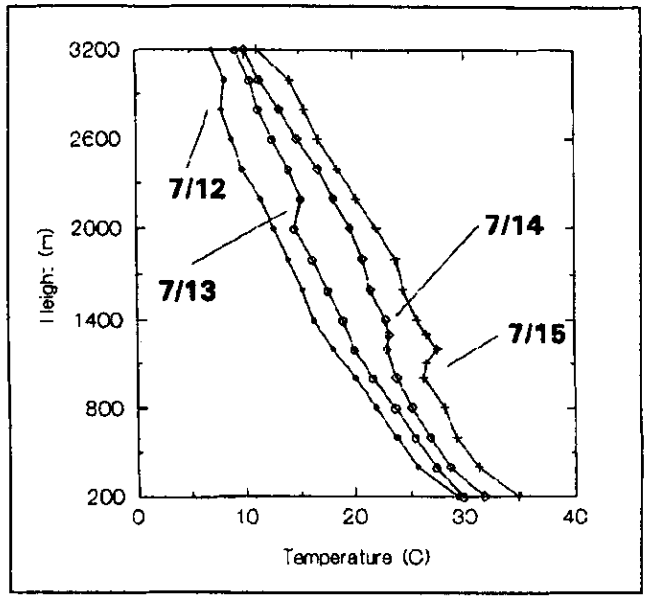

Figure 3. Temperature soundings for the lower troposphere at approximately 1800 UTC on July 12-15, 1995 near Clarksburg, MD.

SURFACE TRACE GAS OBSERVATIONS

High O_3 concentrations occurred in the B-W region and throughout the eastern United States during the July 12–15 episode. Peak O_3 concentrations in the B-W area increased from 136 ppbv on July 12 to 179 ppbv on July 15. Exceedances of the National Ambient Air Quality Standard (NAAQS) for O_3 occurred throughout the eastern seaboard of the United States beginning on July 13, with 12 monitors exceeding the NAAQS from Maryland to Connecticut.[3] On July 14, 37 exceedances were monitored from Maryland to Massachusetts, with 170 ppbv measured at Colliers Mills, NJ, and 175 ppbv at Madison, CT. On July 15, 36 exceedances occurred over an area stretching from Virginia to Connecticut, with 184 ppbv measured at Lums Pond, DE, and 179 ppbv at Aldino, MD, north of Baltimore. The exceedances ended in the northeastern states on July 16 with the passage of a back door cold front. High O_3 concentrations persisted in the B-W region until a more powerful west-to-east moving cold front crossed the area on July 18.

The observations at SNP and Arendtsville illustrate the development of the regional episode. A time series of O_3 concentrations at SNP during the July 1995 episode (Figure 4) shows a gradual increase in the concentrations from July 12 to July 15. During the final two days of the episode, mean O_3 concentrations were 69.0 ppbv and 70.5 ppbv, respectively. These concentrations were well in excess of climatological mean summer O_3 concentrations of 55 ppbv at SNP.[6] Increased O_3 concentrations late in the episode were also observed at Arendtsville (Table 3). At both sites, increases in O_3 concentrations coincided with increases in SO_2 and, at Arendtsville, a slight decrease in CO. As will be discussed in more detail below, the surface

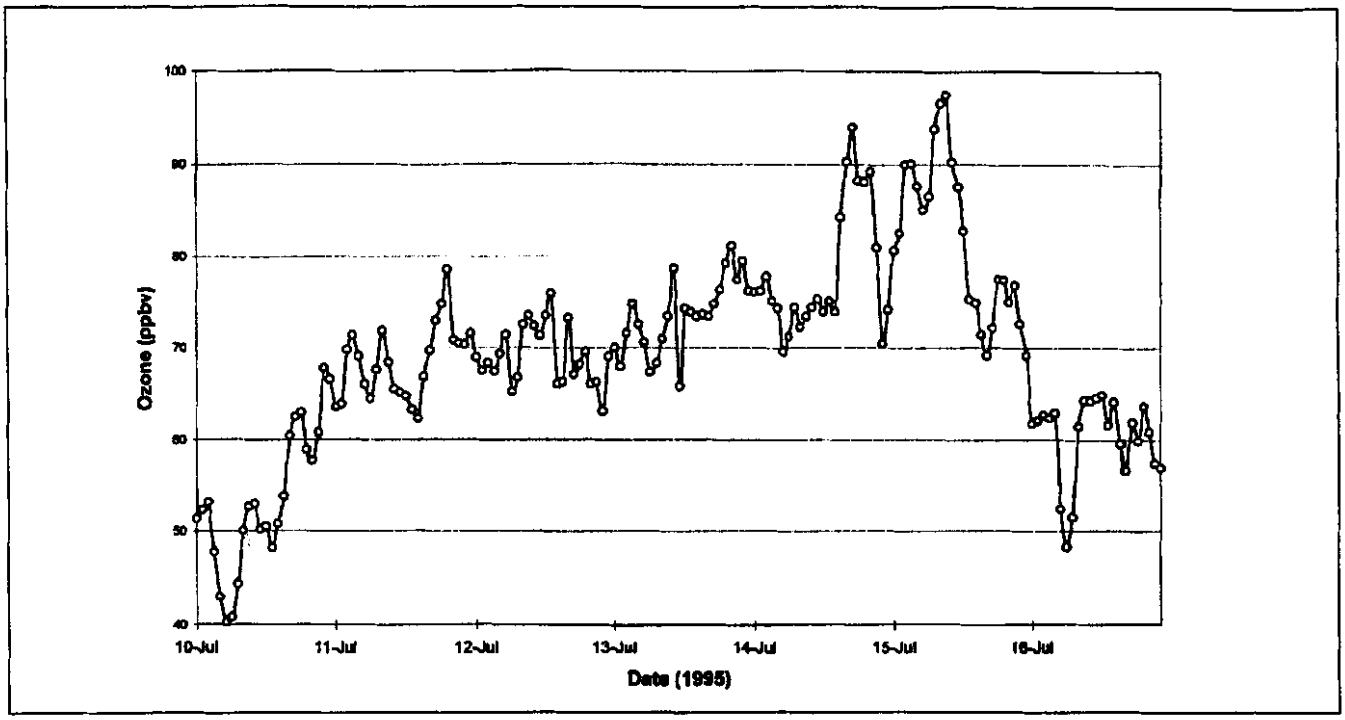

Figure 4. Hourly mean O_3 concentrations at Big Meadows, Shenandoah National Park for the period July 12-16, 1995. The Big Meadows site is located approximately 1100 m above mean sea level.

Table 3. Daily mean trace gas concentrations at the NARSTO-NE sites at Shenandoah National Park (Big Meadows) and Arendtsville.

Station	O_3 (ppbv)	SO_2 (ppbv)	CO (ppbv)
	July 12, 1995		
Shenandoah	69.0	1.5	228
Arendtsville	68.9	3.3	215
	July 13, 1995		
Shenandoah	70.5	0.6	271
Arendtsville	NA	4.5	NA
	July 14, 1995		
Shenandoah	78.1	2.0	226
Arendtsville	71.1	6.0	218
	July 15, 1995		
Shenandoah	82.8	4.3	247
Arendtsville	82.7	12.2	193

sites and aircraft profiles showed similar relationships between O_3, SO_2, and CO concentrations. High O_3 concentrations were also observed well downwind (east) of the B-W region at the Wye River site (Figure 5). Maximum O_3 concentrations at Wye River rose from 88 ppbv on July 13 to 161 ppbv on the afternoon of July 15. Observations of high O_3 concentrations at this distance (approximately 50 km) from the B-W Corridor was unexpected and indicated the extensive spatial coverage of high O_3 concentrations.

AIRCRAFT OBSERVATIONS OF O_3 AND PRECURSORS

Aircraft observations as part of NARSTO-NE began on July 12. Upwind profiles by UM aircraft began on July 13. As noted above, supplemental soundings in the region showed winds from the west and northwest throughout the episode, with the exception of bursts of low-level southwesterly flow. The consistency in flight level (500–3000 m) wind direction allowed research flights upwind of the urban areas each day and generally in the same location. Profiles of O_3 concentrations from UM research flights upwind of the B-W region near Mt. Airy, MD (Figure 6) were taken near the same location at approximately 1500 UTC on July 13 and July 15. The upwind profile on July 13 observed maximum O_3 concentrations of 90–100 ppbv in a layer approximately 900–1500 m above the surface. On July 15, maximum O_3 concentrations were found slightly lower and at approximately the same magnitude.

The O_3 profiles measured by NARSTO-NE aircraft near Arendtsville and SNP were similar in shape and magnitude and illustrate the regional nature of upper-level O_3 during this episode. Daily layer-averaged O_3 concentrations for NARSTO-NE flights near Arendtsville during the episode are shown in Figure 7. The increase in O_3 and the gradual lowering of the level of maximum O_3 as the episode progressed was similar to the UM flights to the south. The profiles near SNP (Figure 8) were similar in shape and magnitude, though maximum O_3 concentrations were 5-10 ppbv lower than the northern profiles.

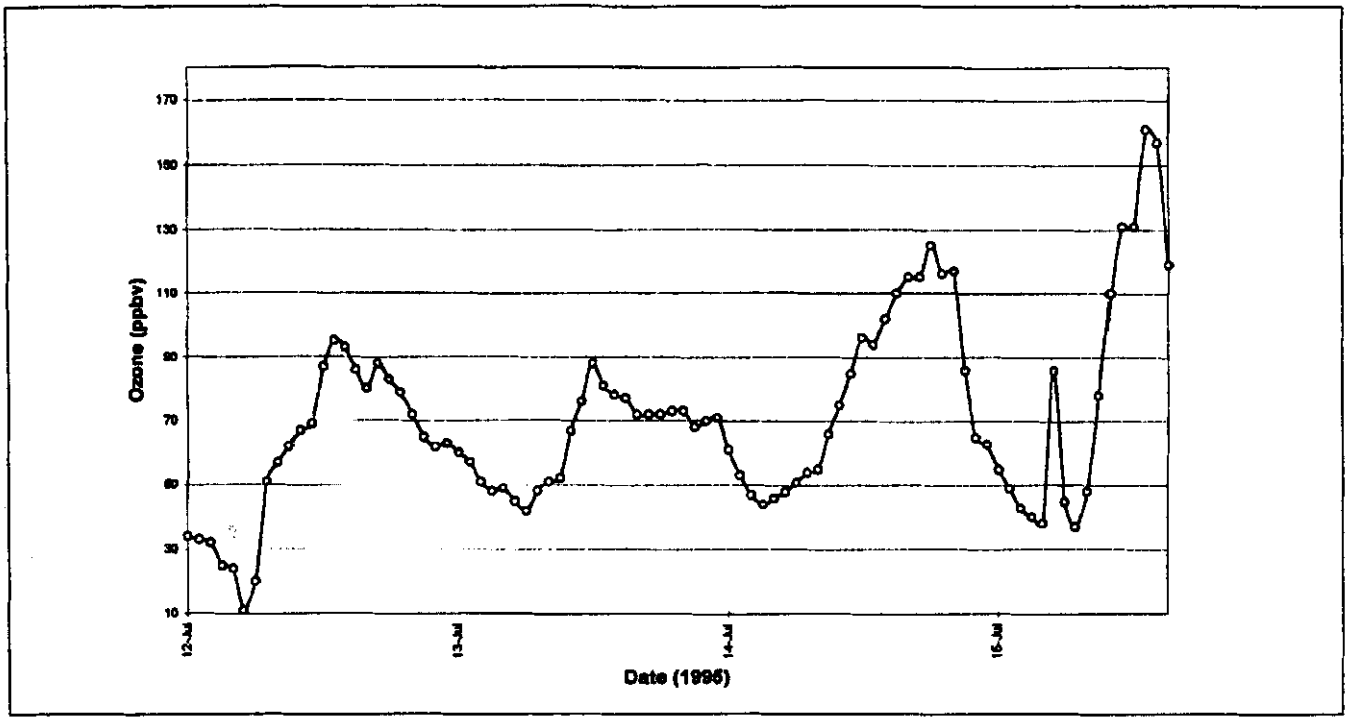

Figure 5. Hourly mean O_3 concentrations at Wye River, MD for the period July 12-15, 1995.

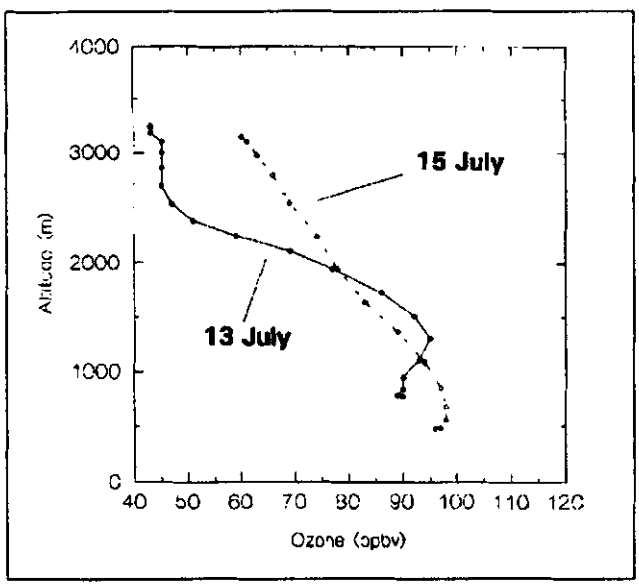

Figure 6. Vertical profiles of O_3 concentrations near Mount Airy during the mid-morning hours (approximately 1400 UTC) of July 13 (open circles, solid line) and July 15 (crosses, dashed line), 1995. Profiles are reported as 200 m layer averages with date of observation indicated on figure.

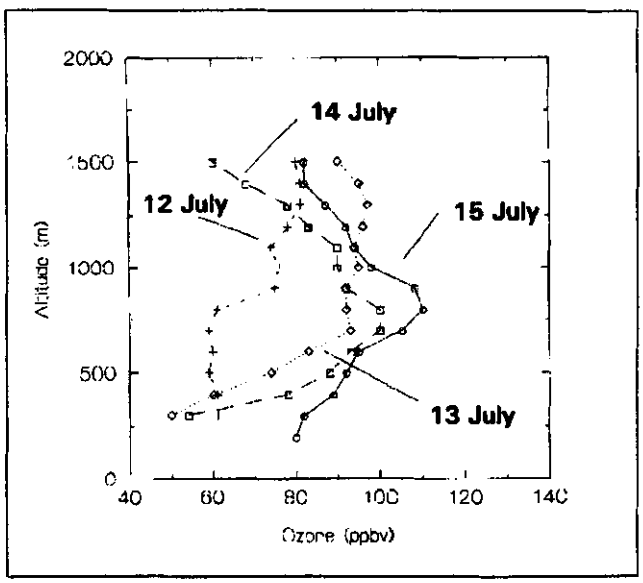

Figure 7. Vertical profiles of O_3 concentrations near Arendtsville, PA, during the early morning hours (approximately 1000 UTC) of July 12-15, 1995. Profiles are reported as 100 m layer averages with date of observation indicated on figure.

The aircraft profiles corroborate the surface signal of increased O_3 and SO_2 concentrations during the later, and more extreme, days of the episode. In Figures 8 and 9, profiles of key trace gases measured by the NARSTO-NE aircraft near SNP are given. In each case, the layer of enhanced O_3 coincides with upper-level maxima of NO_y and SO_2. This feature is also observed near Arendtsville (Figure 10). Consistent with surface observations at SNP (Table 3), aloft concentrations of SO_2 and NO_y approximately double from July 14 to July 15. Although the CO profile does not show a peak co-located with peak O_3 as the SO_2 and NO_y profiles do, the observed CO aloft increases from July 14 to July 15. The concentrations of NO (not shown) are very low throughout all the early morning upwind profiles, with concentrations in the range of 0.1–0.4 ppbv. The low ratio of NO to NO_y means that emissions of NO_x initially contained in the air mass have been processed over time to NO_y. The fact that the sampled air contains

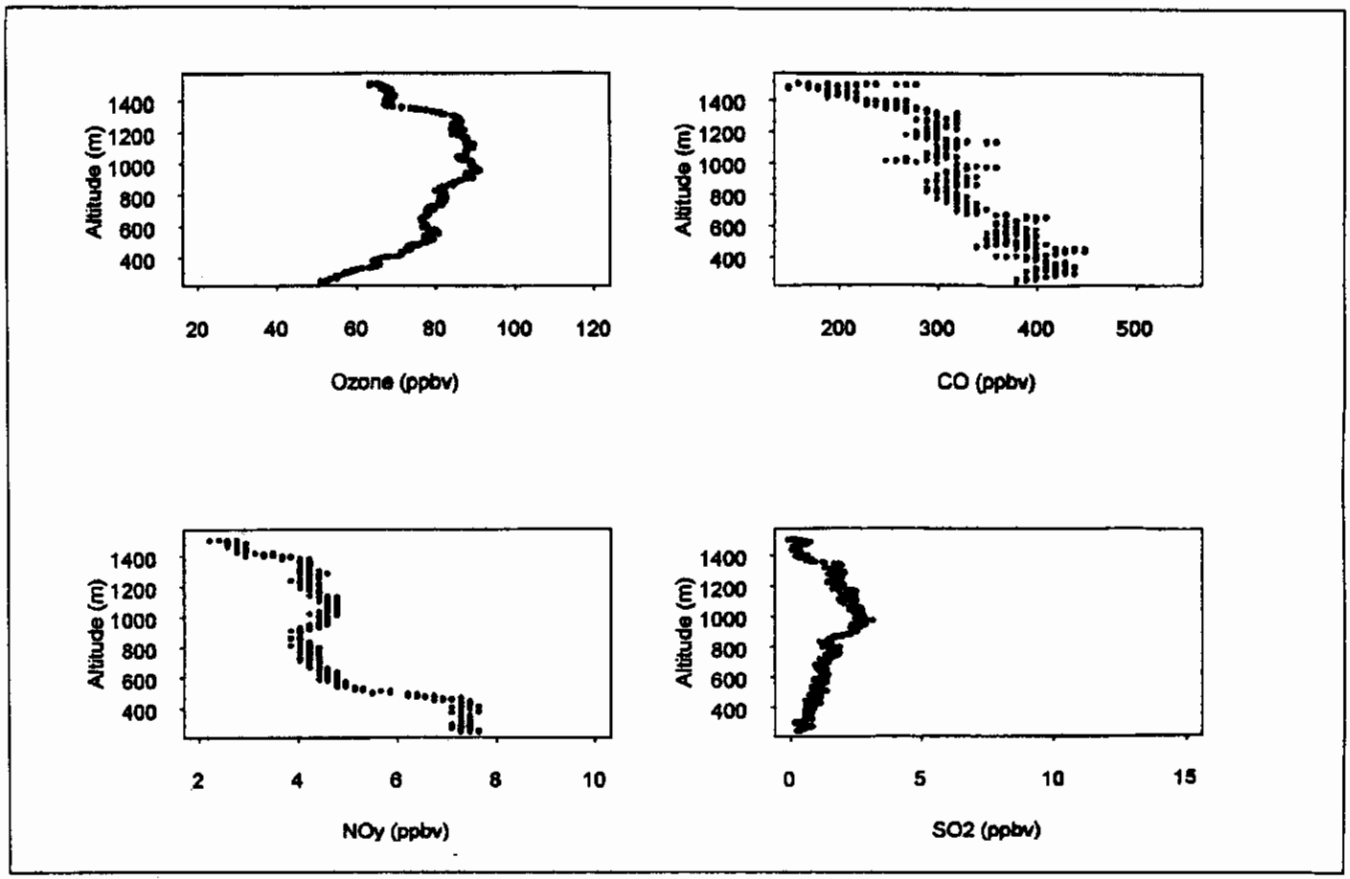

Figure 8. Vertical profiles of O_3, CO, NO_y, and SO_2 concentrations (in ppbv) near Shenandoah National Park, VA in the early morning (1000 UTC) of July 14, 1995.

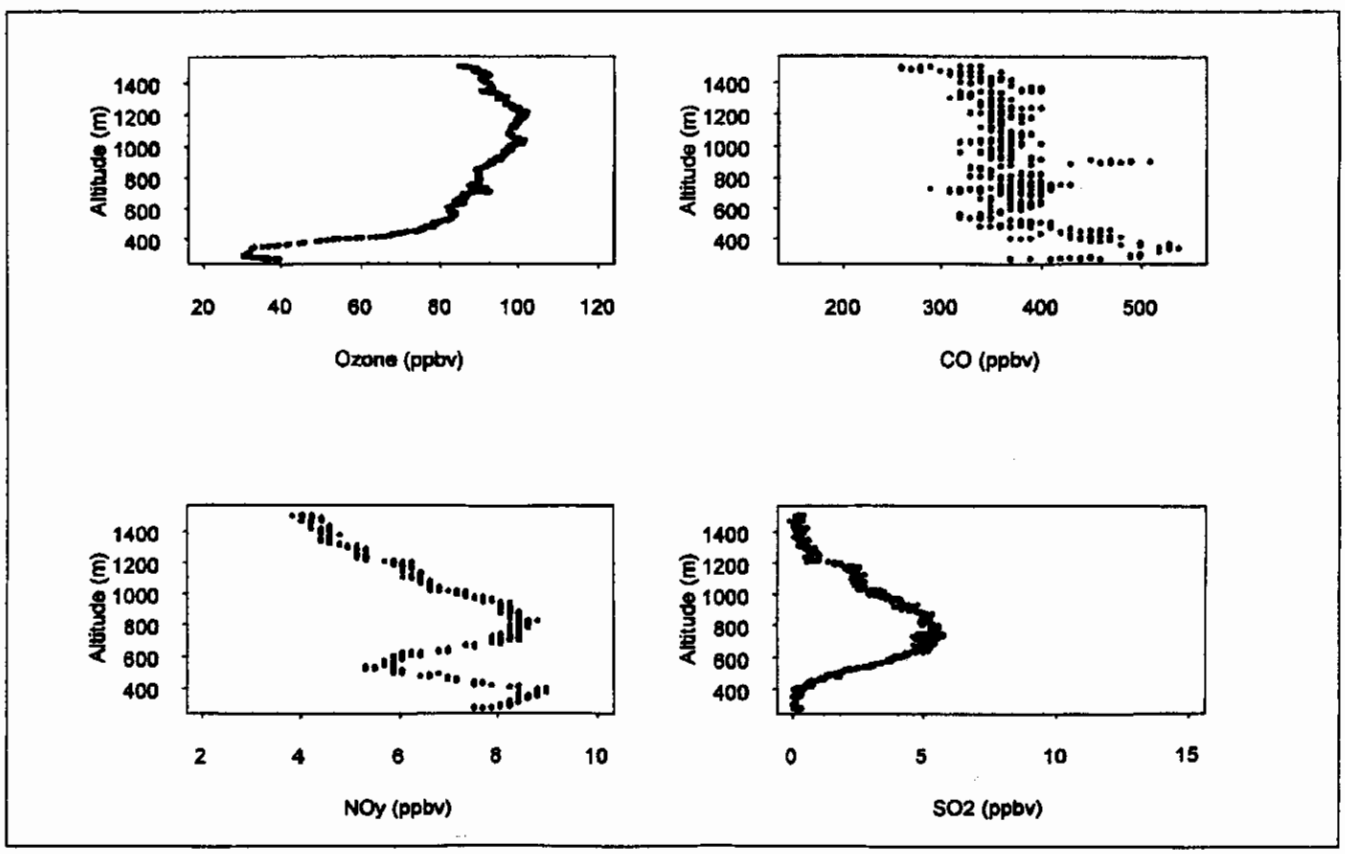

Figure 9. Vertical profiles of O_3, CO, NO_y, and SO_2 concentrations (in ppbv) near Shenandoah National Park, VA in the early morning (1000 UTC) of July 15, 1995.

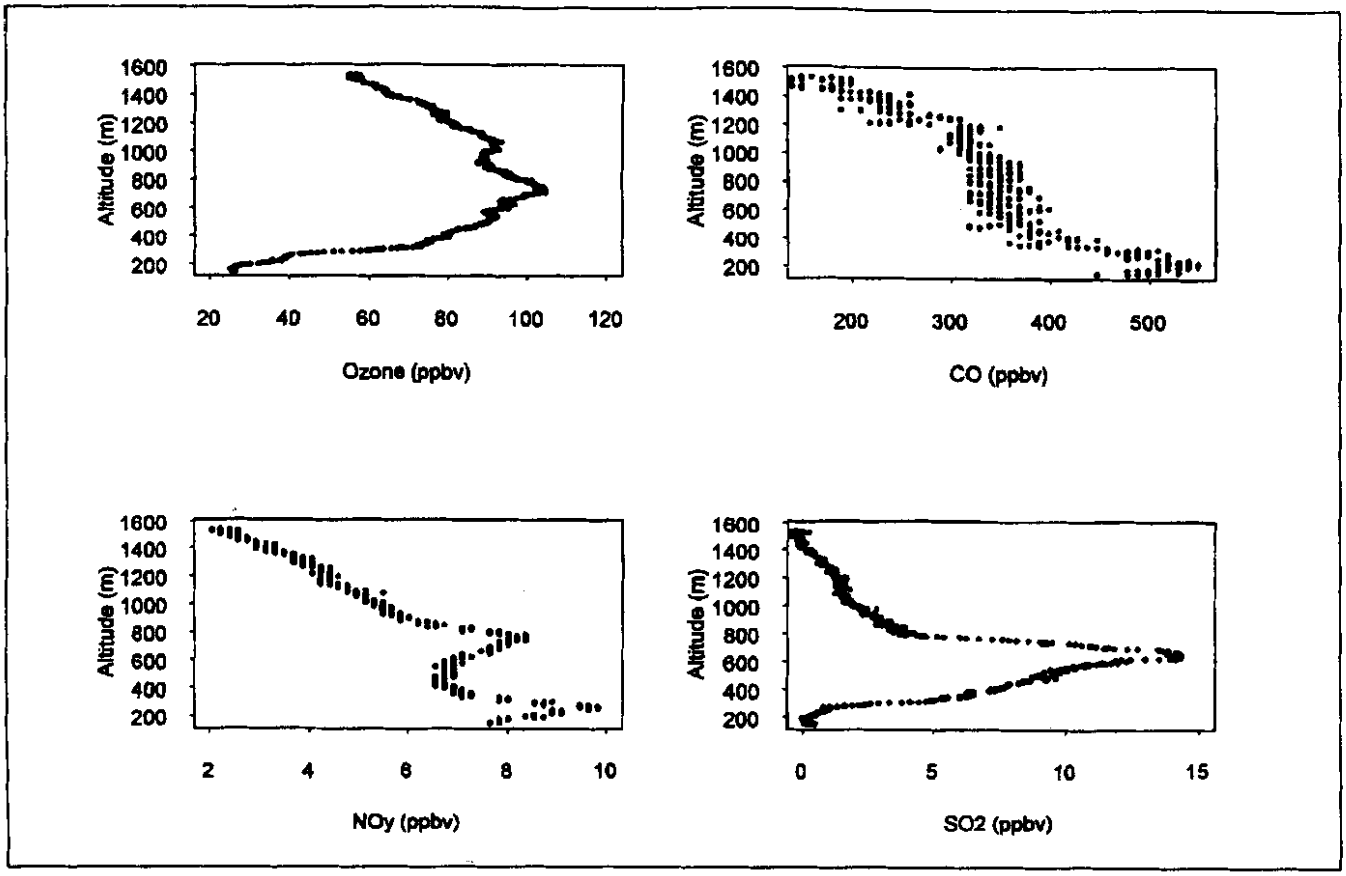

Figure 10. Vertical profiles of O_3, CO, NO_y, and SO_2 concentrations (in ppbv) near Arendtsville, PA, in the early morning (0900 UTC) of July 14, 1995.

little NO relative to overall NO_y is a signal that the air is photochemically "aged" and has traveled some time and distance from its sources.[18-20] This is particularly true in the early morning profiles, as no photochemical activity could have occurred overnight. The aircraft profiles, which show co-located layers of increased SO_2 and O_3 concentrations while CO remains constant or decrease, also corroborate the observations at the NARSTO surface sites.

The fate of the trace gases observed at upper levels during this episode is complicated because the direction of transport at the level of peak O_3 concentrations brought the air directly over the urban corridor later in the day. Thus, local, and fresh, emissions of NO_x and other O_3 precursors were mixed into transported air that was photochemically aged (very low in NO_x relative to NO_y). The outcome, in terms of new O_3 production, of mixing air from markedly different photochemical regimes is complex. What is clear is that the upwind layer of maximum O_3 was below the top of the afternoon boundary layer along the B-W corridor, and the layer was therefore mixed downward during the day. The depth of the afternoon boundary layer was approximately 1.2 km on July 14 and 1.0 km on July 15, as measured at the three upper air sites in the B-W area. Downwind aircraft profiles show the extent of the mixing. On July 14, the UM aircraft profile over the Chesapeake Bay just east of the B-W corridor showed that peak O_3 concentrations were confined to the layer below 1.2 km. This was consistent with boundary layer heights obtained from radiosonde observations (Figure 11). Above the boundary layer, concentrations were consistent with the upwind O_3 profile for altitudes above the observed maximum O_3 layer. Thus, the layer of enriched O_3 seen in Figure 8, which extended from 600–1200 m, was later contained within the boundary layer. Further downwind, the surface site at Wye River reached 125 ppbv this day, consistent with the aircraft observations. A similar profile (not shown) was observed in northern Delaware on this day. Thus, the incoming air, containing high O_3 aloft, was not decoupled from the surface layer and may contribute to local O_3 maxima.

While local peak O_3 concentrations varied widely from monitor to monitor during this episode in response to local emissions and weather conditions, the changes in mean O_3 maxima over the region as a whole followed the trend in upper-level O_3 observed at the boundary. As seen in Figure 7, peak O_3 aloft near Arendtsville increased by approximately 10 ppbv per day during the July 13–15 period. A similar signal is seen for the profiles at SNP. In

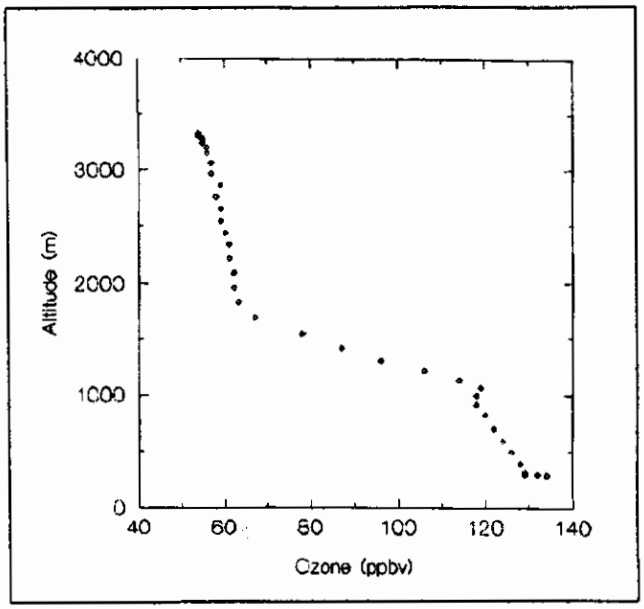

Figure 11. Vertical profile of O_3 concentrations (in ppbv) over the Chesapeake Bay east of Annapolis, MD, in the afternoon (2000 UTC) of July 14, 1995.

the Baltimore non-attainment area, mean peak O_3 concentrations for all monitors increased from 110 ppbv on July 13 to 121 ppbv on July 14 to 140 ppbv on July 15. For the Washington, DC, area, the trend is the same, from 97 ppbv on July 13 to 102 ppbv on July 14 to 124 ppbv on July 15. While the extent to which local sources contribute to the observed maximum surface O_3 cannot be determined by this study, the trend in boundary conditions and regional peak O_3 is consistent.

TRAJECTORY ANALYSIS

Observations from instrumented aircraft show a local O_3 maxima during the episode at approximately 1–2 km. The possible source regions for air parcels arriving at these levels can be estimated using back trajectory analysis. For this episode, back trajectories for air parcels entering the B-W region were modeled with the Hybrid Single-Particle LaGrangian Integrated Trajectory (HY-SPLIT) model using Nested Grid Model (NGM) 2 to 12 hour forecast fields as input.[14,15] For the period July 12–15, back trajectories were calculated beginning at mid-day (1800 UTC) at the center of the B-W corridor near Fort Meade, MD. Each back trajectory was modeled for 48 hr from an initial height of 1500 m. This was roughly the level of peak upper-level O_3 observed upwind and was sufficiently elevated to avoid impact on the Appalachians to the west and the intermittent LLJs. The trajectories that terminate near Fort Meade throughout the episode were clustered to the west and northwest of the B-W region, with the upwind source region typically in the Ohio Valley and west (Figure 12). This trajectory pattern is consistent with previous studies of sources for the regional-scale site at SNP.[16] Although the HY-SPLIT model cannot resolve specific sources for air parcels entering the region, it shows the regional source for this episode west and northwest of the B-W region.

DISCUSSION AND CONCLUSIONS

The meteorological conditions observed during the high O_3 episode of July 12–15, 1995, were consistent with historic severe O_3 events in the B-W region. A persistent ridge to the west of the region drove strong local subsidence and transported air at upper levels from the west and northwest. Although this weather pattern has been frequently observed, particularly in 1988, there have been few coincident measurements of upper air chemistry along the upwind boundary. The vertical profiles of O_3 during this episode showed upwind, or boundary, O_3 ranging from 80–110 ppbv, consistent with observations at the elevated rural sites. In addition, the correlation of high O_3 with high SO_2 and NO_y suggests that the air parcels contain emissions from coal combustion and that these emissions occurred at a significant distance from the region. Investigations with a trajectory model indicate that the general source region for the air entering the region at upper levels was west and northwest of the region.

The association of local high O_3 episodes to a westerly transport regime can be inferred from a preliminary analysis of other episodes. The 12 strongest O_3 episodes in the B-W region during the summers of 1995 and 1996 show similar transport patterns (Figure 13). Not included in this figure is the initial day of the second strongest O_3 event during 1995, which occurred during the period June 17–20, 1995. The trajectory for this day was consistent with the rest of the higher O_3 days and was unusual in that the exceedance in the B-W region occurred on a Sunday, typically a day of much lower local emissions. Another moderately high O_3 event occurred in late July and early August with NAAQS exceedances observed on July 31–August 2. After August 2, temperatures continued to increase with maximum surface temperatures at BWI peaking on August 4–5. However, O_3 concentrations fell off markedly after August 2. In Figure 14, back trajectories initialized on the first two days of the episode (July 31 and August 1) are compared to back trajectories initialized on the warmest days of the episode (August 4–5). The source of the air entering the region early in the episode was from the west, consistent with the earlier strong events, while the air entering the region late in the period was more maritime (southeasterly transport) in nature. The latter trajectories may have provided cleaner boundary conditions and overall lower O_3 concentrations even though surface temperatures were considerably higher. This is corroborated by observations at the SNP

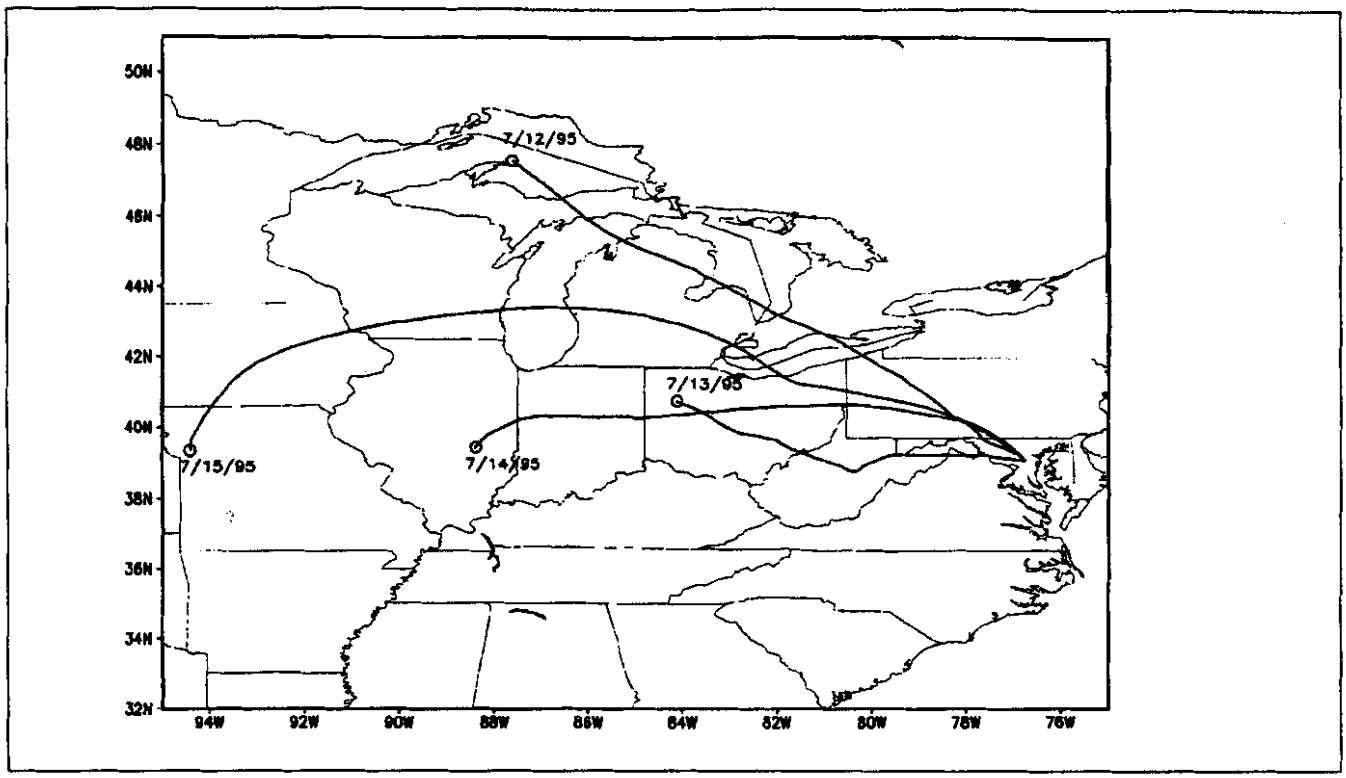

Figure 12. Modeled back trajectories for air parcels entering the B-W region during the high O_3 episode of July 12-15, 1995. The starting point for each back trajectory is 1500 m above mean sea level near Fort Meade, MD. The starting time is 1800 UTC in all cases. The back trajectories are run for 48 hr with the terminal date of the trajectory noted in the figure. For example, a parcel located over Fort Meade at 1800 UTC on July 14 has a modeled back trajectory which crosses southwestern Pennsylvania and central Ohio. The end point of the back trajectory path is in central Illinois at 1800 UTC on July 12. All back trajectories are for 48 hours with the exception of the trajectory initialized at 1800 UTC on July 15, which is run for 42 hr and terminates at 0000 UTC on July 14.

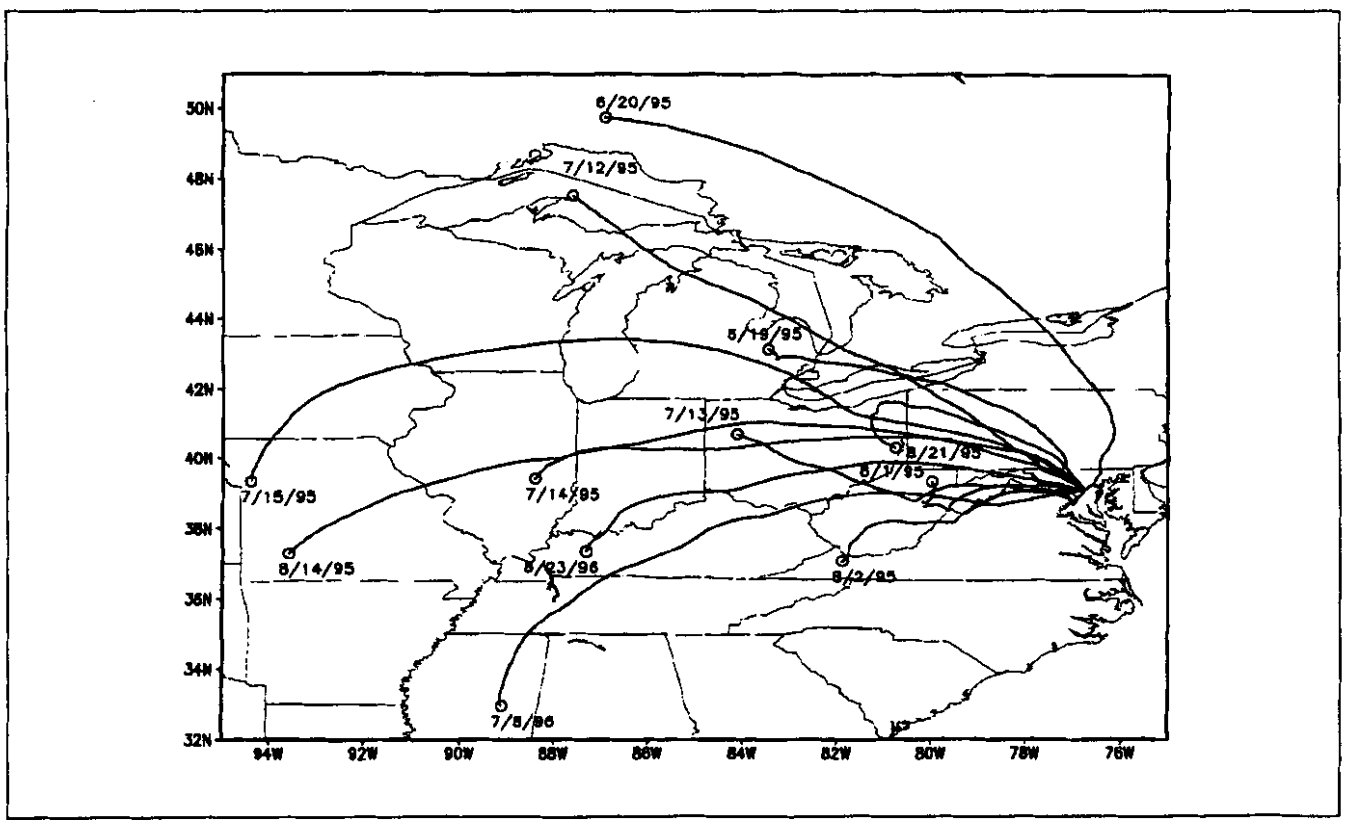

Figure 13. As in Figure 12, but for the 12 most severe O_3 events in the B-W region during the 1995-1996 summer seasons.

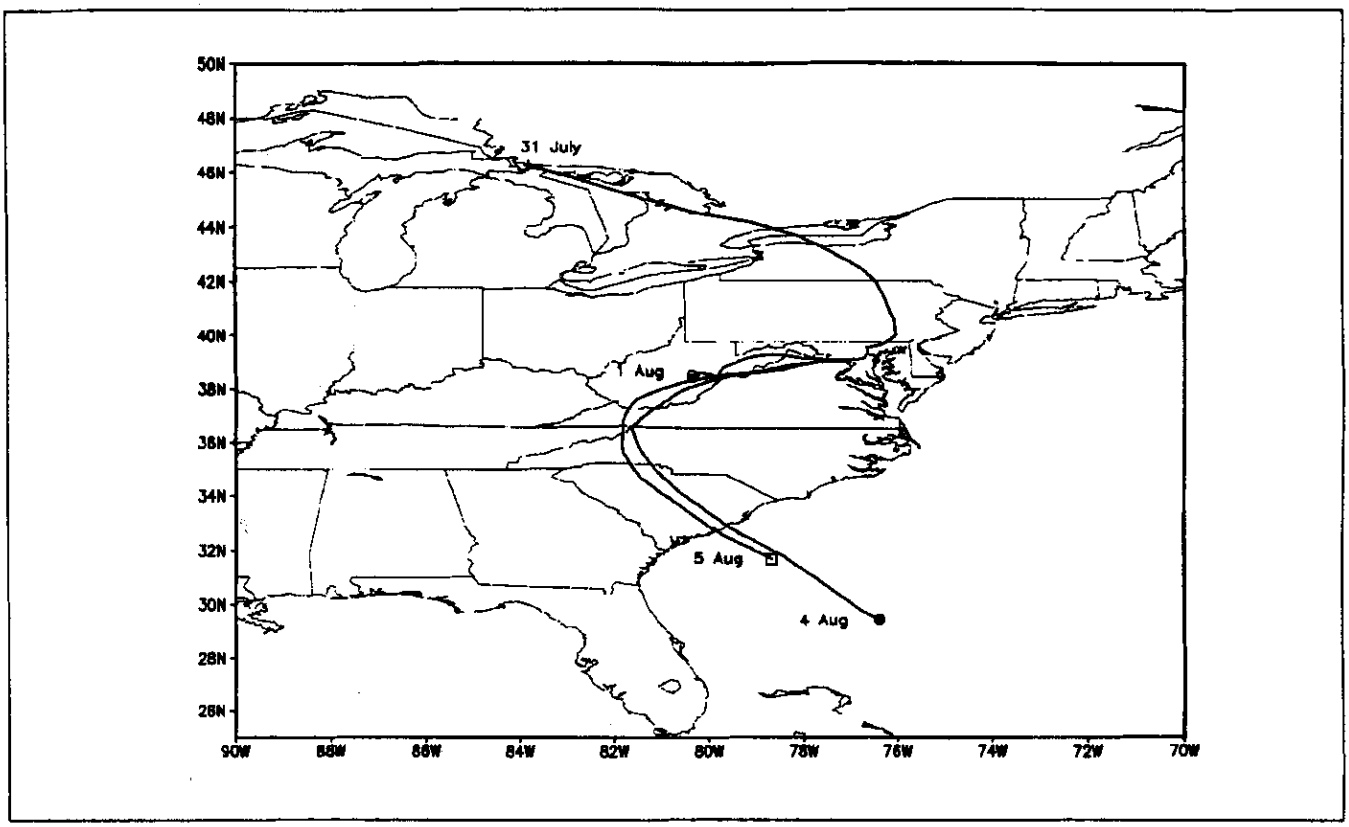

Figure 14. As in Figure 12, but for July 31-August 1 and August 4-5, 1995.

Table 4. Daily mean trace gas concentrations and daily maximum temperature at Shenandoah National Park and Arendtsville.

Location	Maximum Temperature (°C)	O_3 (ppbv)	SO_2 (ppbv)	CO (ppbv)
		July 31, 1995		
SNP	23.2	73.2	3.3	201.9
Arendtsville	30.9	75.3	4.9	164.0
		August 4, 1995		
SNP	25.5	42.8	1.1	119.3
Arendtsville	32.0	38.1	1.9	97.3

site. Concentrations of CO, O_3, and SO_2 at SNP are markedly lower on August 4 as compared to July 31 (Table 4).

Meteorological analysis and profiles of concentrations of O_3, NO_y, SO_2, and CO show that emissions from upwind elevated sources or upwind low-level sources transported vertically by turbulent mixing in the Ohio River Valley region overlaid pollutants from more local, low-level sources, and their combination led to a severe photochemical smog event. The preliminary data reported here will be augmented by the richer NARSTO database in the future. However, this first look at the data has raised a number of interesting questions. The foremost question is the effect of upper-level boundary O_3 on local O_3 maxima. The evidence from the July episode suggests that the air entering the B-W region was already rich in O_3. The fate of O_3 and its precursors upon entering a high emissions metropolitan area is a complex issue involving, among other things, the degree to which upper level O_3 is mixed downward and its interaction with hydrocarbon and NO_x-enriched urban air. This issue is further complicated by the low-level bay breeze and vertical wind shear phenomena observed during this episode. More specific source attribution is also needed. Trajectory models can provide a rough estimate of the source region, but further coincident observations of O_3, reactive nitrogen species, hydrocarbons, and SO_2 can also yield important information.

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The 2003 North American Electrical Blackout:

An Accidental Experiment in Atmospheric Chemistry

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Abstract

The August 2003 North American electrical blackout provided a unique opportunity to quantify directly the contribution of power plants to regional haze and O_3. Airborne observations over central Pennsylvania on August 15, 2003, ~24 h into the blackout, revealed large reductions in SO_2 (>90%), O_3 (~50%), and light scattered by particles (~70%), relative to measurements outside the blackout region or over the same location when power plants were operating normally. CO and light absorbing particles were unaffected. Low level O_3 decreased by ~38 ppbv and the visual range increased by > 40 km. This clean air benefit was realized over much of the eastern U.S. Reported SO_2 and NO_x emissions from upwind power plants were down to 34 and 20% of normal, respectively. The improvement in air quality provides evidence that transported emissions from power plants hundreds of km upwind play a dominant role in regional haze and O_3 production.

Introduction

The August 2003, electrical blackout that affected over 100 power plants in northeastern U.S. and southeastern Canada, provided a unique opportunity to evaluate the contribution of power plants emissions to regional haze and ozone (O_3). The impact of transported point source pollution on regional air quality depends on emissions, meteorology, and non-linear chemical responses. So far, quantification of the impacts has been based on multi-year measurement and modeling studies (e.g., Solomon *et al.*, 2000) and results of long-term emissions reduction scenarios (Malm *et al.*, 2002). This paper presents results of direct measurements of the effect of power plant emissions reductions on regional air quality with all other factors held relatively constant.

Fossil fuel burning power plants account for more than half of electrical energy production in the U.S., but also ~22% of the nitrogen oxides ($NO_x = NO + NO_2$) and ~69% of the sulfur dioxide (SO_2) emissions (USEPA, 2003(a)). NO_x combines with volatile organic compounds (VOCs) in the presence of sunlight to produce O_3, the principal component of photochemical smog. SO_2 may be oxidized to produce sulfate (SO_4^{2-}), the primary constituent of $PM_{2.5}$ (particles with diameters $\leq 2.5\ \mu m$) in the northeastern U.S. (IMPROVE, 2000). In summertime, under high pressure with westerly transport, emissions of NO_x and SO_2 in the northeastern U.S. induce severe smog and haze events, primarily comprised of O_3 and sulfate-dominated fine particles (Ryan *et al.*, 1998, Sistla *et al.*, 2001, Taubman *et al.*, 2004(a)). Both pollutants have been linked to adverse health effects, degradation of the environment, and global climate change (McClellan, 2002, Gent *et al.*, 2003, USEPA, 2003(b), IPCC, 2001).

Airborne measurements were made over Maryland and Virginia (outside the blackout area) and Pennsylvania (in the center of the blackout area) on August 15, 2003, ~24 h into the blackout. The results are compared to those from the previous summer in the same locations and under similar meteorological conditions when upwind power plants were operating normally. Emissions data are examined in conjunction with back trajectories to quantify the contribution of power plants to the observed air quality. Results help quantify the impact of reduced SO_2 and NO_x emission on air quality in the NE U.S.

Sampling Platform

A light aircraft outfitted for atmospheric research was used as the sampling platform. O_3, CO, and SO_2 mixing ratios were measured using Thermo Environmental

Instruments analyzers. Sub-micrometer particle counts were determined using a MetOne 9012 optical particle counter. Particle light scattering at 450, 550, and 700 nm was measured using a TSI 3563 integrating nephelometer. Particle light absorption at 565 nm was quantified with a Particle/Soot Absorption Photometer. For full details of instruments used see Taubman *et al.* (2004(b)).

Results and Discussion

Two flights were conducted on August 15, 2003. During the first flight, three vertical spirals (surface - 3 km) were performed over Luray (38.70°N, 78.48°W) and Winchester (39.15°N, 78.15°W) in Virginia and Cumberland, Maryland (39.60°N, 78.70°W) at ~14:00, 15:00, and 15:30 UTC, respectively. Two spirals were performed over Selinsgrove, Pennsylvania (40.82°N, 76.86°W) at ~19:00 and 20:00 UTC during the second flight.

The morning spirals (outside the blackout region) revealed trace gas mixing ratios and particle properties typical of those routinely observed on previous flights (Dickerson *et al.*, 1995, Ryan *et al.*, 1998, Taubman *et al.*, 2004(a)). Observations over Luray, for example, show maxima in SO_2 and O_3 mixing ratios in a thin layer at ~1 km MSL (Figures 1a,b). A corresponding peak in particle light scattering was also seen at this altitude; but values increased again below 500 m MSL (Figure 1c), corresponding to a maximum in CO (Figure 1d). These observations indicate a stable nocturnal boundary layer with a maximum depth of 500 m MSL. Above this altitude, NO_x and SO_2 from power plants produced O_3 and SO_4^{2-}, respectively, which were transported in the residual layer. Below 500 m, the pollution was most likely of local origin. Particles observed in the nocturnal boundary layer may have been largely organics, the products of vehicle

exhaust and home heating and cooking, which can scatter visible light efficiently (Malm *et al.*, 1994).

Observations from the afternoon flight were different. Spirals over Selinsgrove, Pennsylvania revealed very little O_3, SO_2, and PM relative to the morning flight and areas to the south (Figures 2a-c). CO concentrations were within 0.5 σ of the 1992 median August and September values over Baltimore, Maryland and vicinity (Dickerson *et al.*, 1995), and remained fairly constant throughout the afternoon, apparently only varying with altitude (Figure 2d). Linear regressions between O_3 and SO_2 measured during the flight showed that O_3 over Selinsgrove was not correlated with SO_2 ($r = 0.13$), while it was elsewhere ($r = 0.80$). Observations over Selinsgrove are consistent with reductions in power plant emissions but no corresponding changes in vehicle emissions.

To investigate whether the improvement in air quality over Selinsgrove was due to reductions in upwind power plant emissions, 24 h backward trajectories were run from Selinsgrove at 500, 1500, and 2500 m AGL using the NOAA ARL HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Version 4) (Draxler and Rolph, 2003) and EDAS meteorological fields (Figure 3a). A 100 km wide swath was then assigned to the trajectory paths to account for uncertainties. Hourly NO_x and SO_2 emissions data (USEPA, 2003, Personal Communication) for U.S. power plants falling within the swaths were integrated over the 24 h period preceding the measurements (Table 1). This enabled the pairing of upwind emissions data with wind trajectory analyses. A large source of uncertainty in this approach is the lack of emissions data from Canada.

For comparison the same back trajectory and emissions procedure was followed for Selinsgrove, PA on August 4, 2002 (prior to blackout), and Cumberland, MD on August 15, 2003 (out of blackout area). On August 4, 2002 Selinsgrove was under similar synoptic patterns as on August 15, 2003. Regional mean surface temperatures were ~33°C on both days and wind speeds and directions were similar (Figure 3b). This analysis yielded large differences in upwind power plant emissions (Table 1). SO_2 and NO_x emissions upwind of Selinsgrove on August 15, 2003 were reduced to 34% and 20% of normal and to 34% and 25% of that observed upwind of Cumberland, respectively (Table 1).

The impact of this emissions disparity on downwind air quality is illustrated in Figure 4. SO_2, O_3, and light scattered by particles measured over Selinsgrove in 2003 were reduced by >90%, ~50%, and ~70%, respectively, relative to 2002 observations (Figures 4a-c). Defining visual range as the 98% extinction point, the reduction in aerosol extinction corresponds to an increase in visual range of > 40 km. The concomitant decreases in SO_2 and particle light scattering suggest that improvements in visibility resulted directly from reduced power plant SO_2 emissions. Reductions in O_3, apparently the result of decreased NO_x emissions, were greatest near the surface (~38 ppbv) and fell off at higher altitudes where large-scale processes play a more dominant role in the O_3 budget. As with CO concentrations, however, light absorption by particles shows a less dramatic difference (Figure 4d). In fact, absorption was higher in 2003 than in 2002, suggesting little or no reduction in vehicle emissions during the blackout relative to typical values. The single scattering albedo was 0.95 on the normal day, but fell to 0.85 during the blackout. Electricity generation produces very little CO or absorbing

aerosols; instead, they are mainly emitted by vehicles that continued to operate during the blackout. No discernible changes in road vehicular traffic activity could be observed near or upwind of the study area during the blackout (Szekeres, 2004).

Forward trajectories (not shown) run from Selinsgrove reach Baltimore, Philadelphia, and New York, depending on the altitude. Thus, the improvement in air quality depicted in Figure 4 was experienced over several major eastern cities. This is corroborated by the fact that O_3 concentrations in Maryland were forecasted to be 125 ppbv but reached only 90 ppbv (Maryland Department of Environment, 2003). Because the RMS forecast error is 10 ppbv, we attribute the bulk of this overestimation to reduced power plant emissions.

Conclusions

Airborne measurements made over central Pennsylvania on August 15, 2003, ~24 hours into one of the largest electrical blackouts in North American history, showed large reductions in SO_2 (>90%), O_3 (~50%), and light scattered by particles (~70%) relative to observations over western Maryland earlier in the day and over the same location the year before. This translated into a reduction in low level O_3 of ~38 ppbv and an improvement in visual range of > 40 km. Forward trajectories show that these improvements in air quality benefited much of the eastern U.S. CO and particle light absorption values did not change much, however, suggesting that vehicle emissions were largely unaffected during the blackout. Reported power plant SO_2 and NO_x emissions upwind of central Pennsylvania on August 15, 2003 were 34% and 20% of normal, respectively. Thus, the decreases in SO_2, O_3, and particle light scattering appear to be predominantly due to reductions in power plant emissions hundreds of km upwind of the study area. The

observed reductions exceed expectation based on estimated relative contribution of power plants to these pollutants and their precursors (NO_x ~22%, SO_2 ~69% and PM ~ 8%) (USEPA, 2003(a)). The dramatic improvement in air quality during the blackout may result from underestimation of emissions from power plants, inaccurate representation of power plant effluent in emission models or unaccounted for atmospheric chemical reaction(s). These unique observations will provide a resource for determining whether air quality models can accurately reproduce the contributions of specific pollution sources to regional air quality.

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Regional Transport of Ozone

For the 8-hour ozone episode of August 12th and 13th, 1999 backward and forward trajectories for Hagerstown, MD (Washington County) were modeled using the NOAA Hysplit model. The August 12th backward trajectories show that the air parcels coming into Washington County were from the high pollutant emissions areas of OH. The August 12th morning and afternoon backward trajectories for Washington County are provided in Figures 1 and 2, respectively. The August 12th forward trajectories show that an air parcel leaving the low emissions area of Washington County headed north into PA and NY State. The August 12th morning and afternoon forward trajectories for Washington County are provided in Figures 3 and 4, respectively.

On August 13th, 1999 the backward trajectories show that the air parcels coming into Washington County were from the areas of WV and VA. The August 13th morning and afternoon backward trajectories for Washington County are provided in Figures 5 and 6, respectively. The August 13th forward trajectories show that an air parcel leaving the low emissions area of Washington County headed north into ME and Canada. The August 13th morning and afternoon forward trajectories for Washington County are provided in Figures 7 and 8, respectively.

The 8-hour ozone episode of August 12th and 13th, 1999 is typical of what Washington County usually experiences during the course of an ozone season. Usually back trajectories from the west or southwest allow pollutants from high emissions areas to be transported into Washington County.

Figure 2 – 20z (4 PM) 24-Hour Back Trajectory for Hagerstown on August 12, 1999.

Draxler, R.R. and Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://www.arl.noaa.gov/ready/hysplit4.html). NOAA Air Resources Laboratory, Silver Spring, MD.

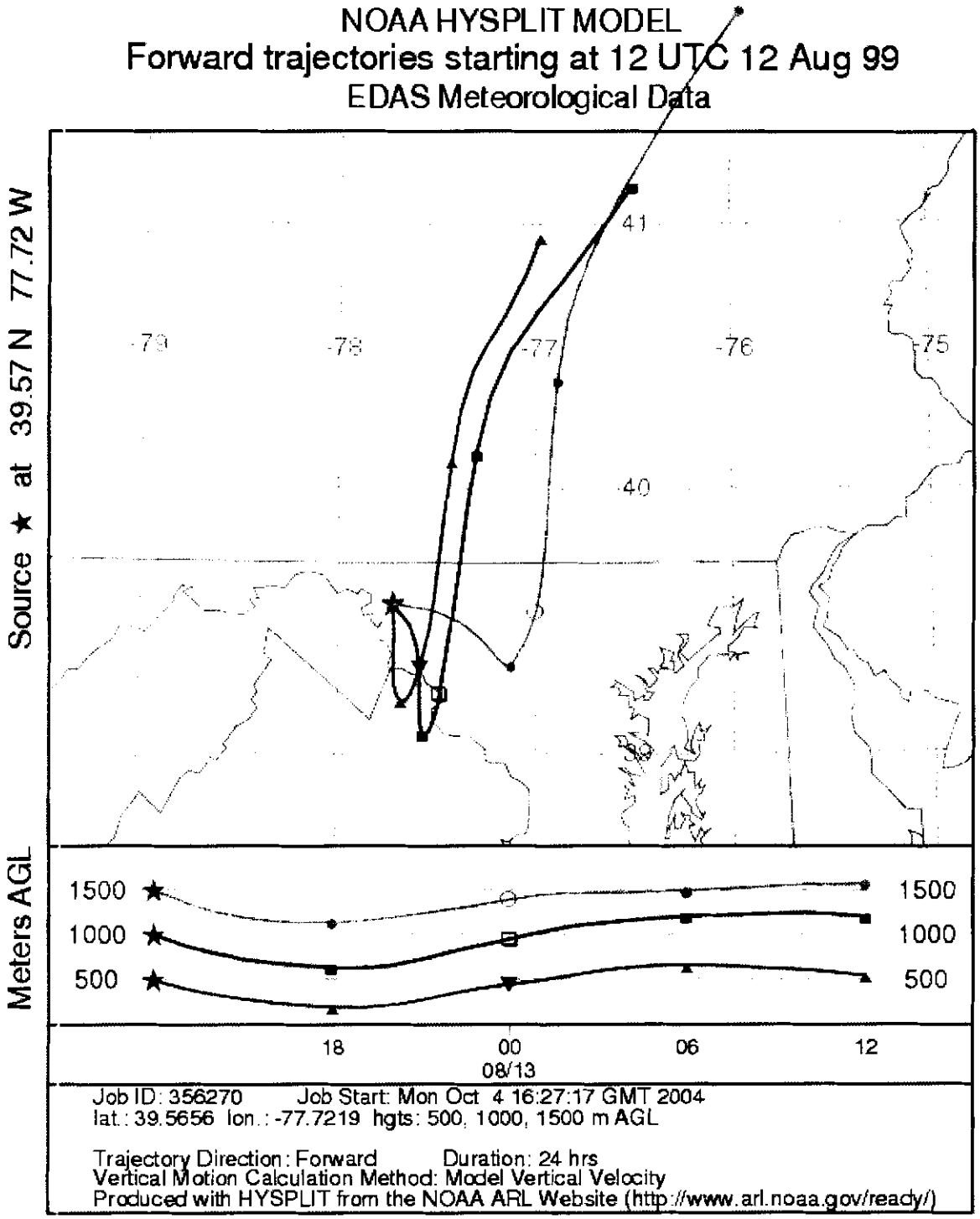
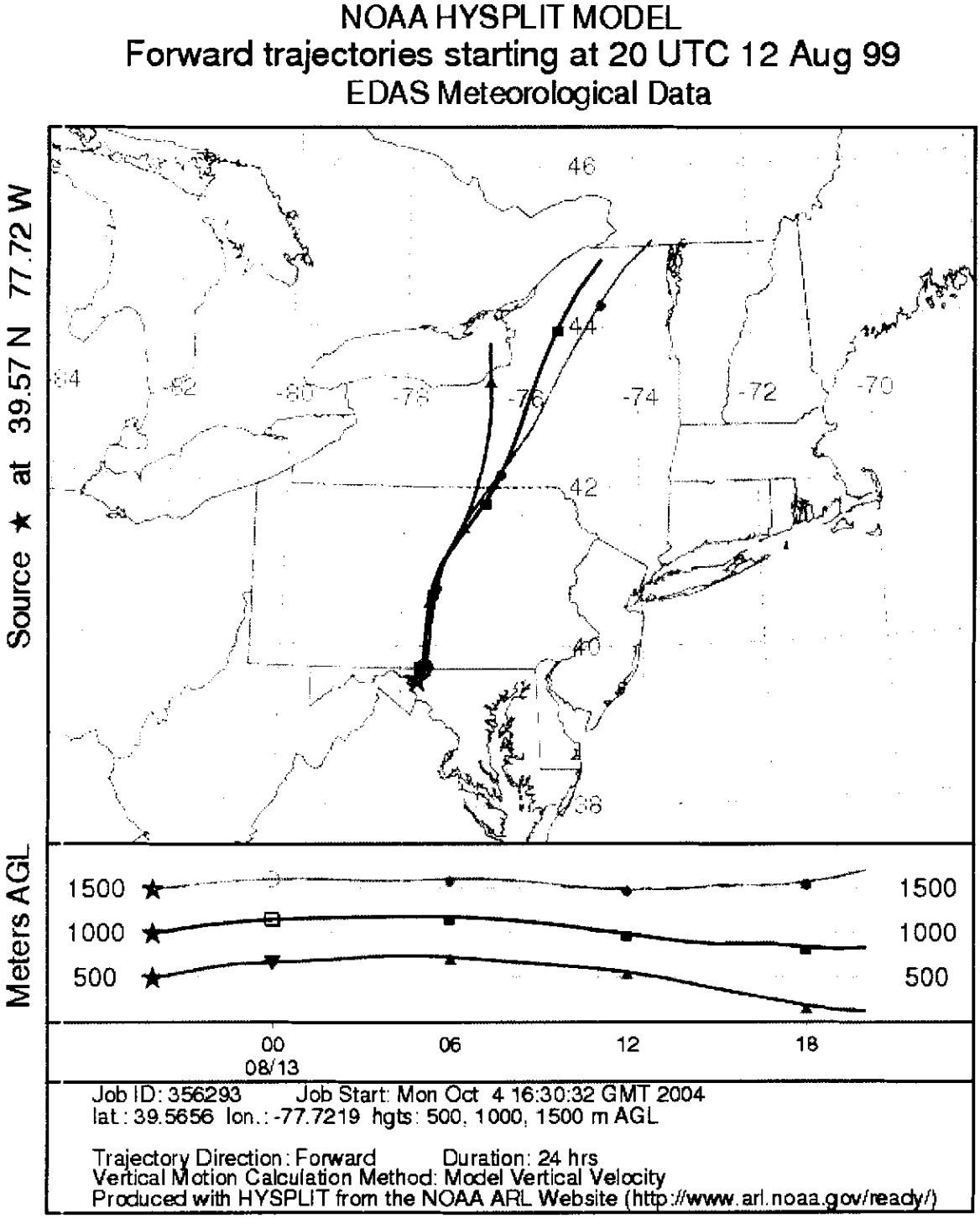


Figure 3- – 12z (8 AM) 24-Hour Forward Trajectory for Hagerstown on August 12, 1999.



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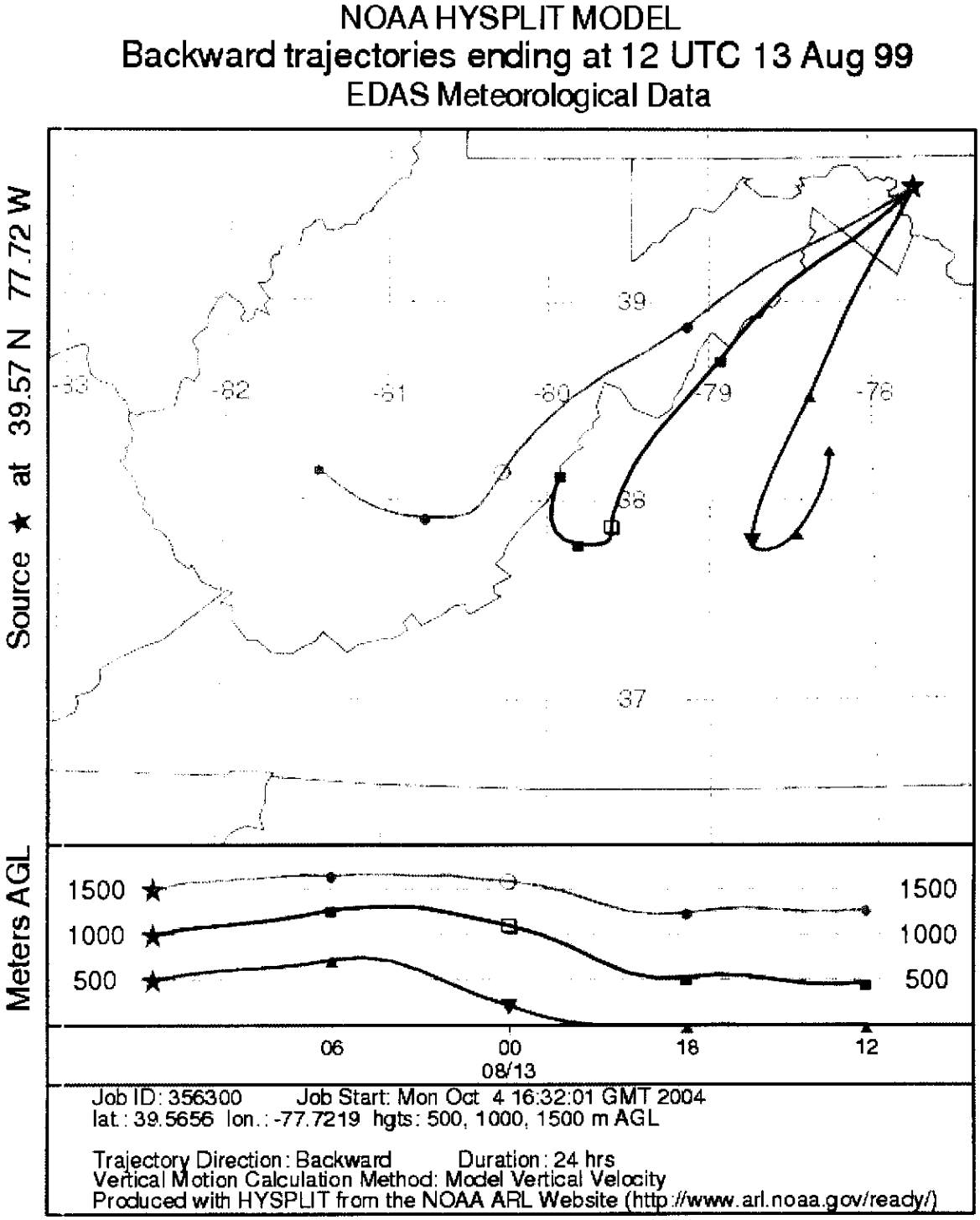

Figure 5 – 12z (8 AM) 24-Hour Backward Trajectory for Hagerstown on August 13, 1999.

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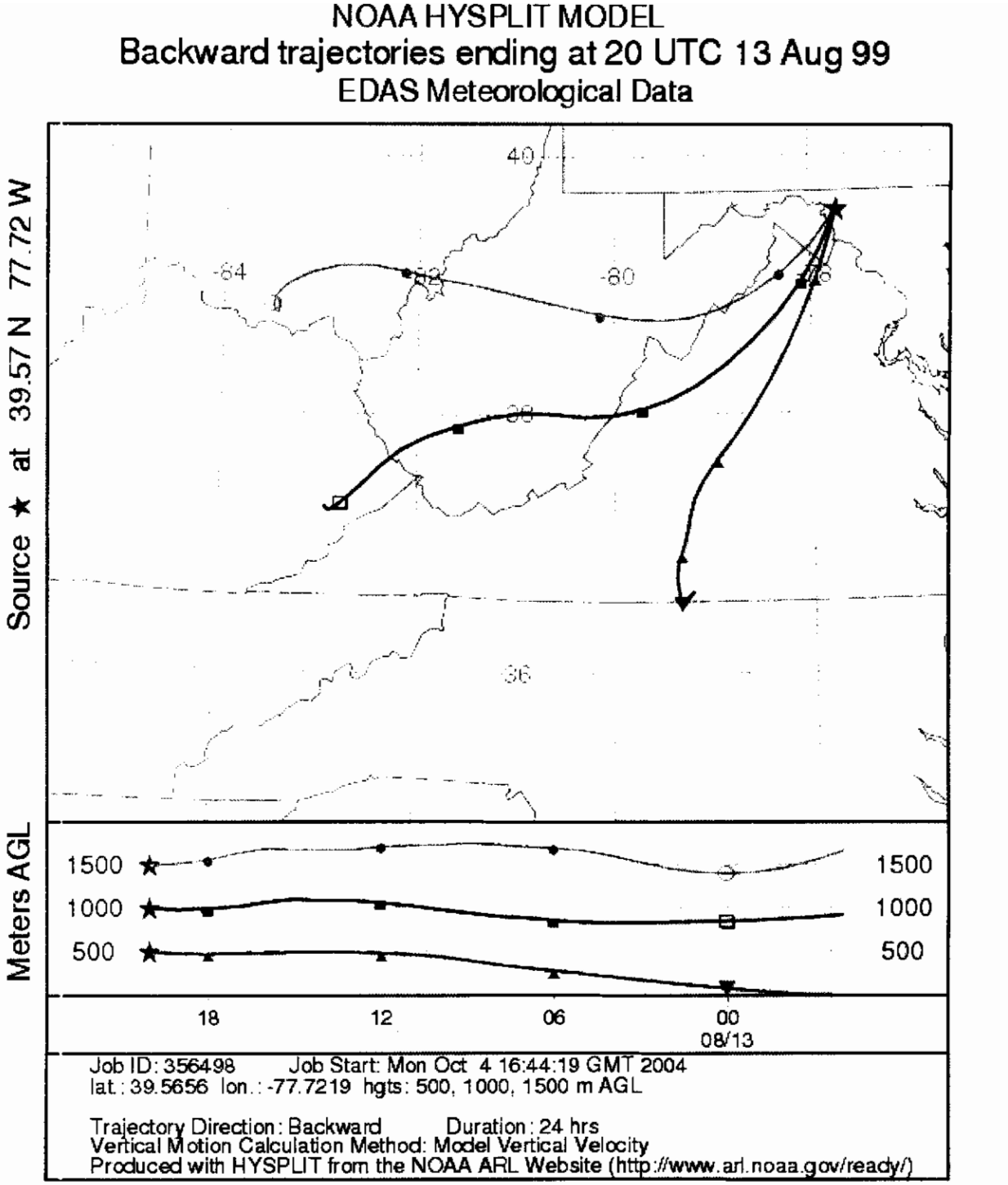

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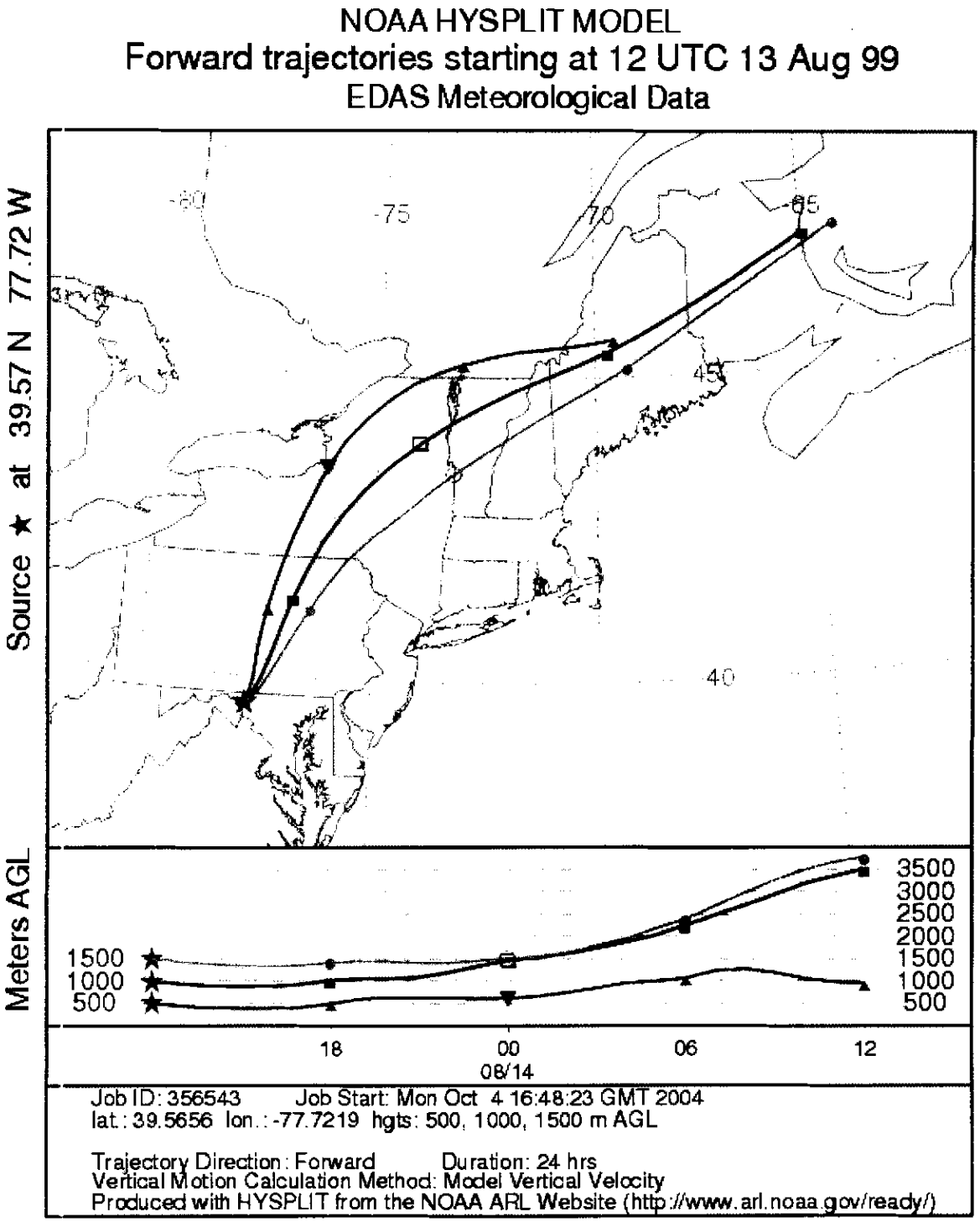

Figure 6 – 12z (8 AM) 24-Hour Forward Trajectory for Hagerstown on August 13, 1999.

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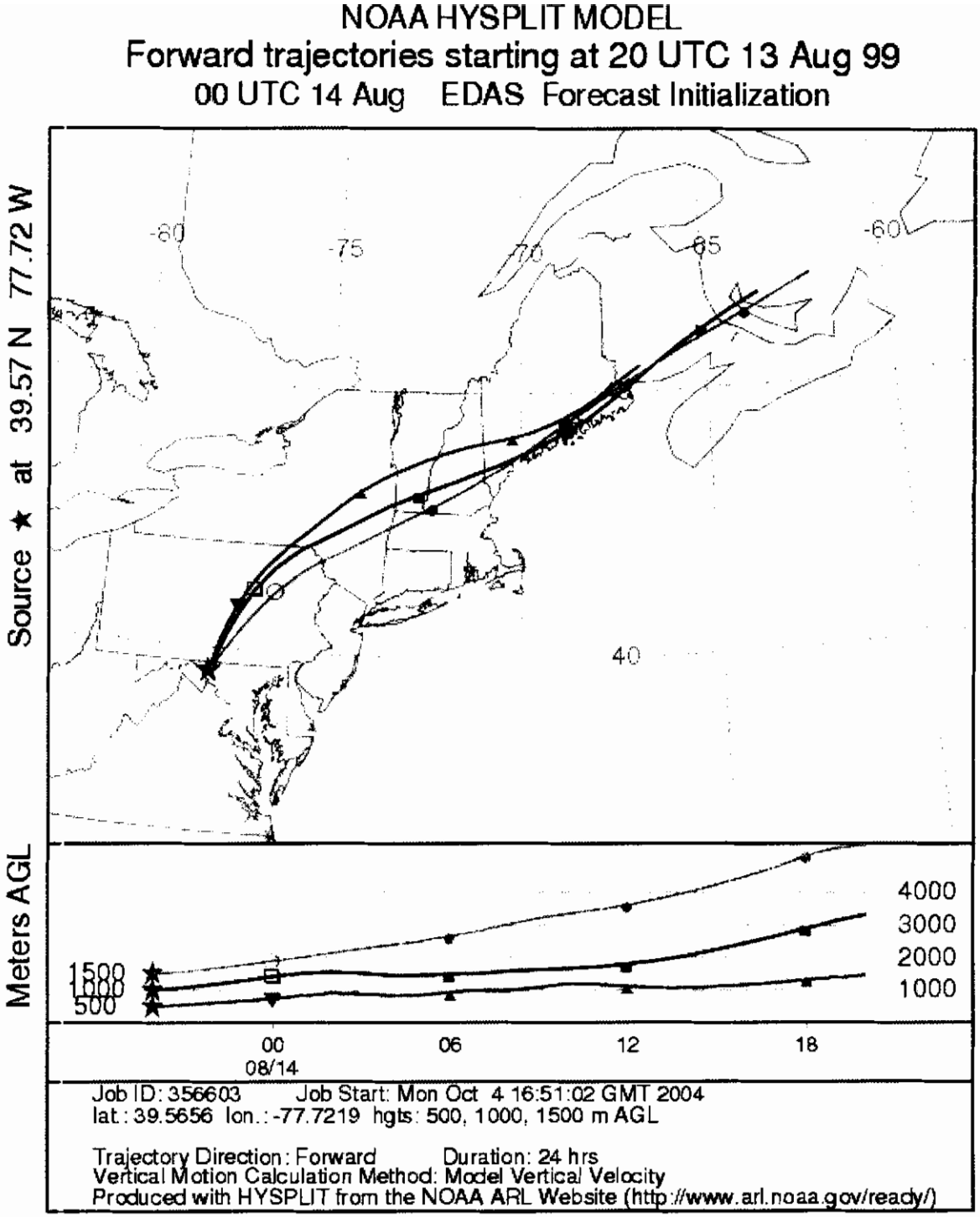

Figure 7 – 20z (4 PM) 24-Hour Forward Trajectory for Hagerstown on August 13, 1999.

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Regional Transport of Ozone in Washington County

For the 8-hour ozone episode of August 12th and 13th, 1999 backward and forward trajectories for Hagerstown, MD (Washington County) were modeled using the NOAA Hysplit model. The August 12th backward trajectories show that the air parcels coming into Washington County were from the high pollutant emissions areas of OH. The August 12th morning and afternoon backward trajectories for Washington County are provided in Figures 1 and 2, respectively. The August 12th forward trajectories show that an air parcel leaving the low emissions area of Washington County headed north into PA and NY State. The August 12th morning and afternoon forward trajectories for Washington County are provided in Figures 3 and 4, respectively.

On August 13th, 1999 the backward trajectories show that the air parcels coming into Washington County were from the areas of WV and VA. The August 13th morning and afternoon backward trajectories for Washington County are provided in Figures 5 and 6, respectively. The August 13th forward trajectories show that an air parcel leaving the low emissions area of Washington County headed north into ME and Canada. The August 13th morning and afternoon forward trajectories for Washington County are provided in Figures 7 and 8, respectively.

The 8-hour ozone episode of August 12th and 13th, 1999 is typical of what Washington County usually experiences during the course of an ozone season. Usually back trajectories from the west or southwest allow pollutants from high emissions areas to be transported into Washington County.

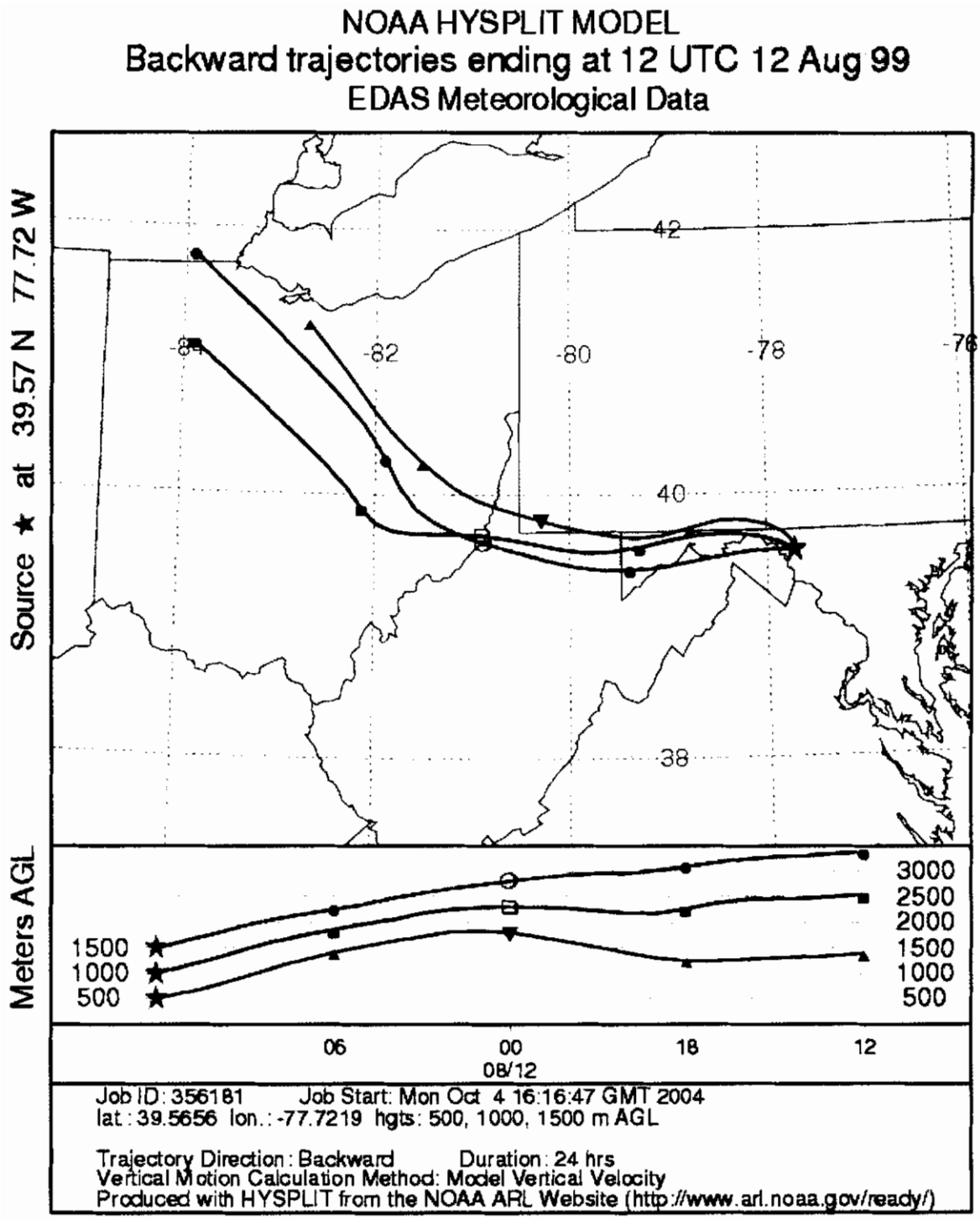

Figure 1 – 12z (8 AM) 24-Hour Back Trajectory for Hagerstown on August 12, 1999.

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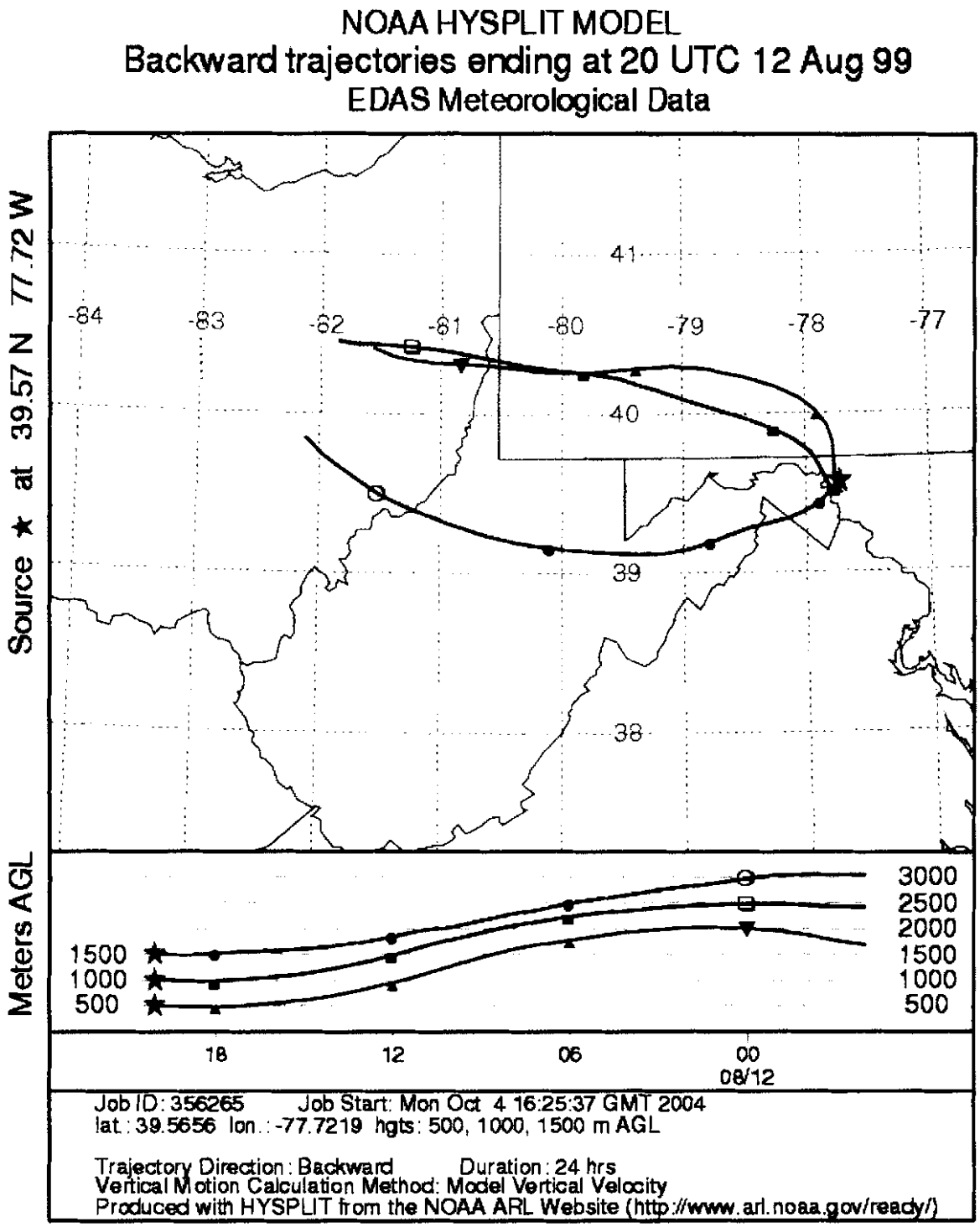

Figure 2 – 20z (4 PM) 24-Hour Back Trajectory for Hagerstown on August 12, 1999.

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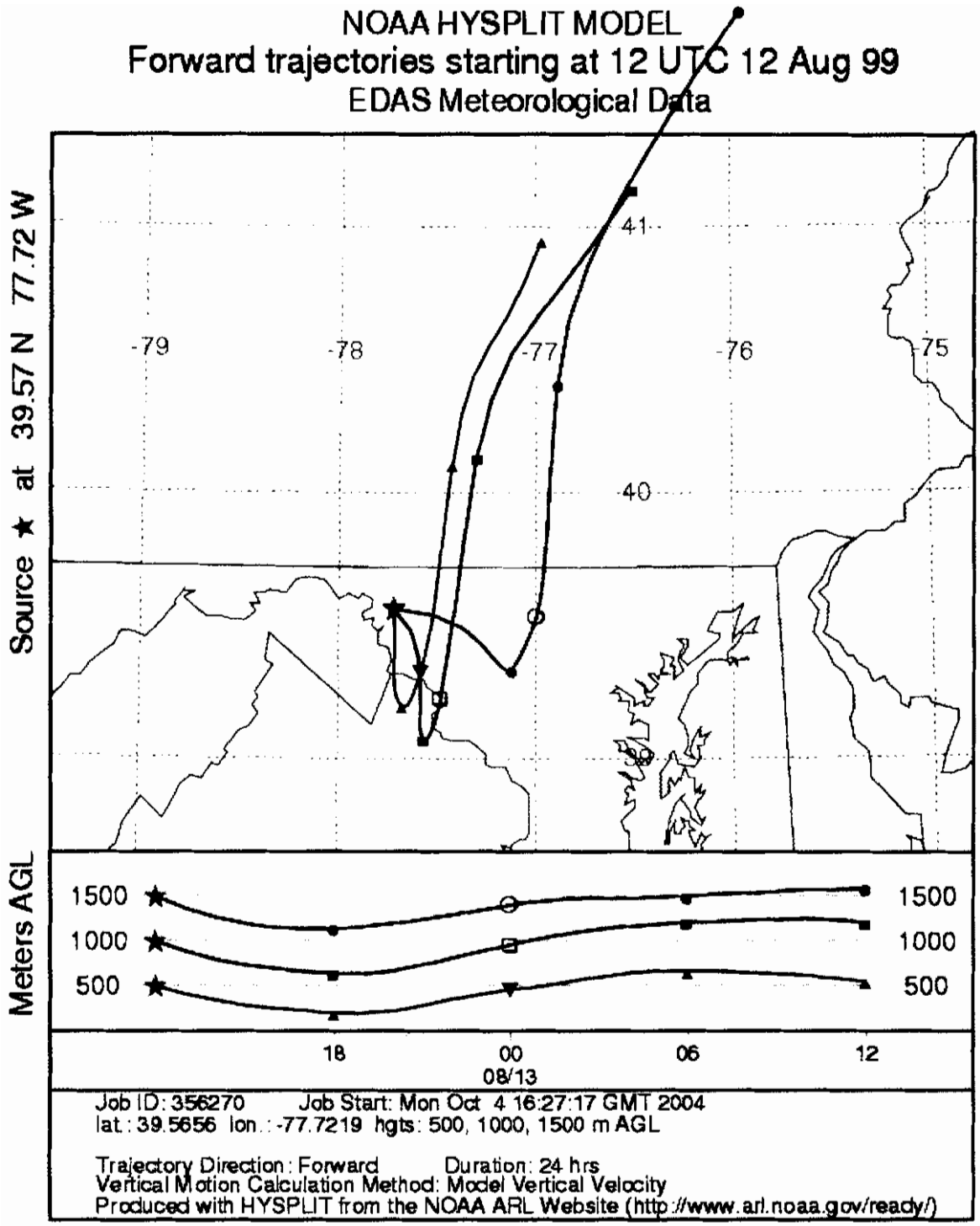

Figure 3- – 12z (8 AM) 24-Hour Forward Trajectory for Hagerstown on August 12, 1999.

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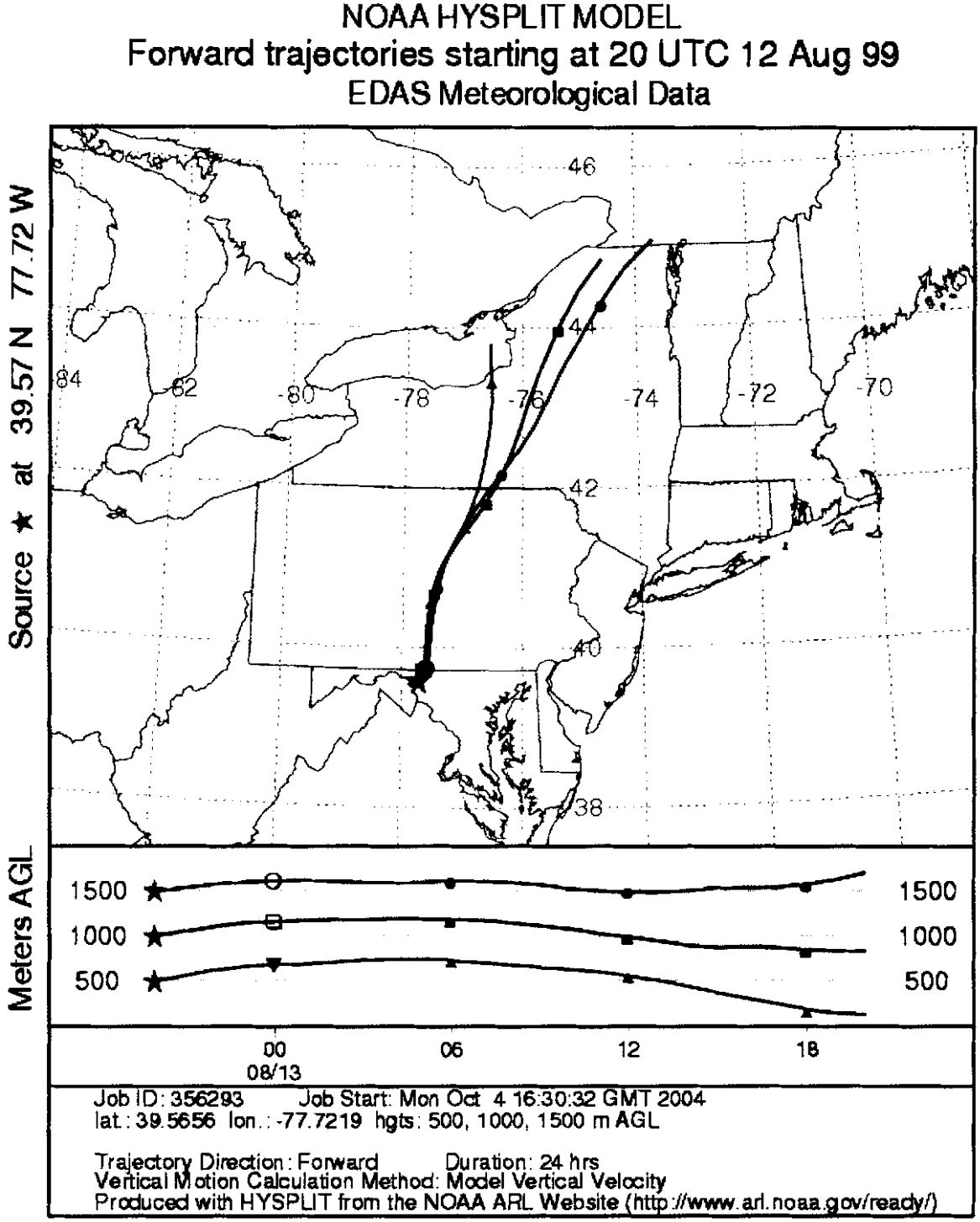

Figure 4 – 20z (4 PM) 24-Hour Forward Trajectory for Hagerstown on August 12, 1999.

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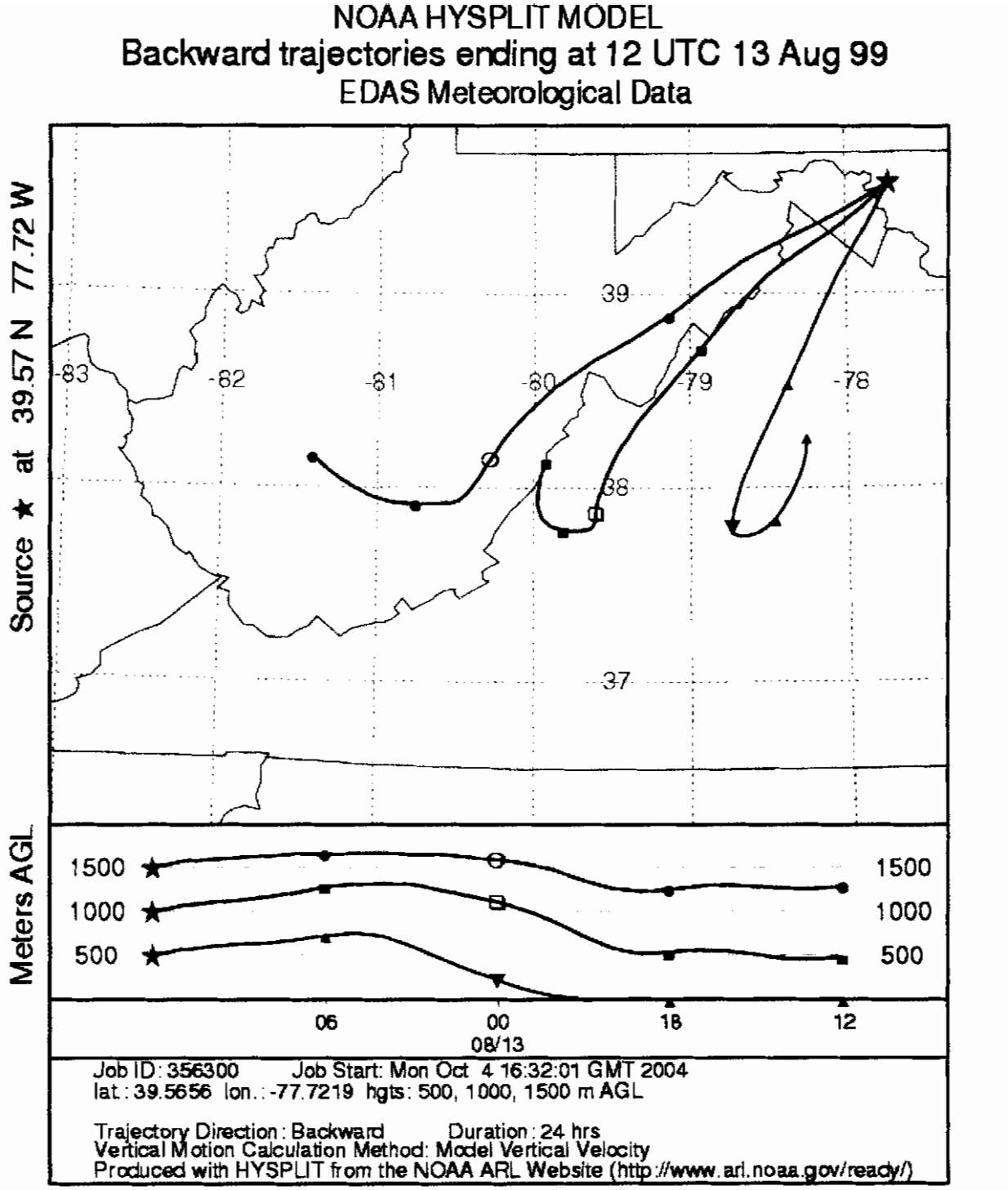

Figure 5 – 12z (8 AM) 24-Hour Backward Trajectory for Hagerstown on August 13, 1999.

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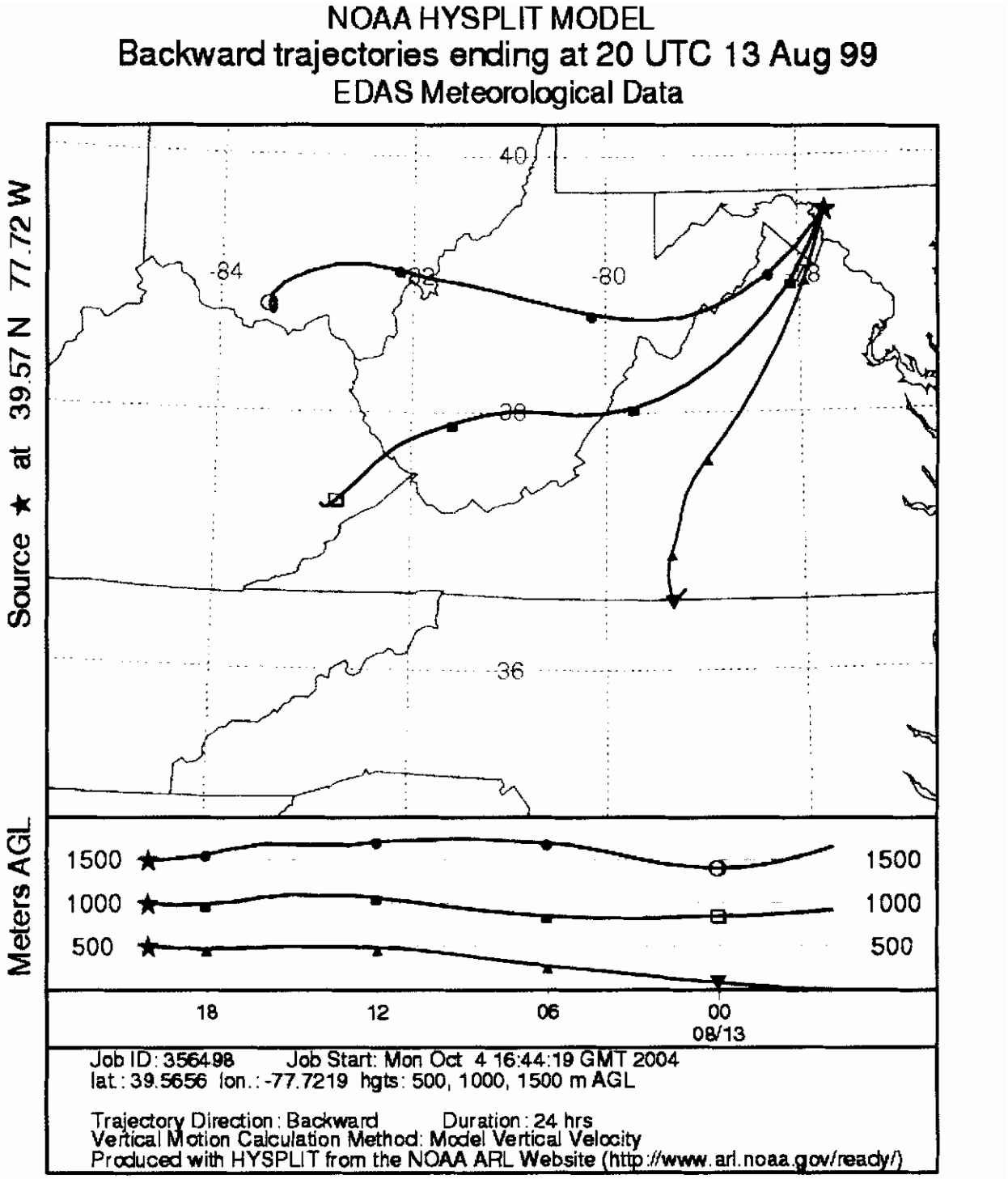

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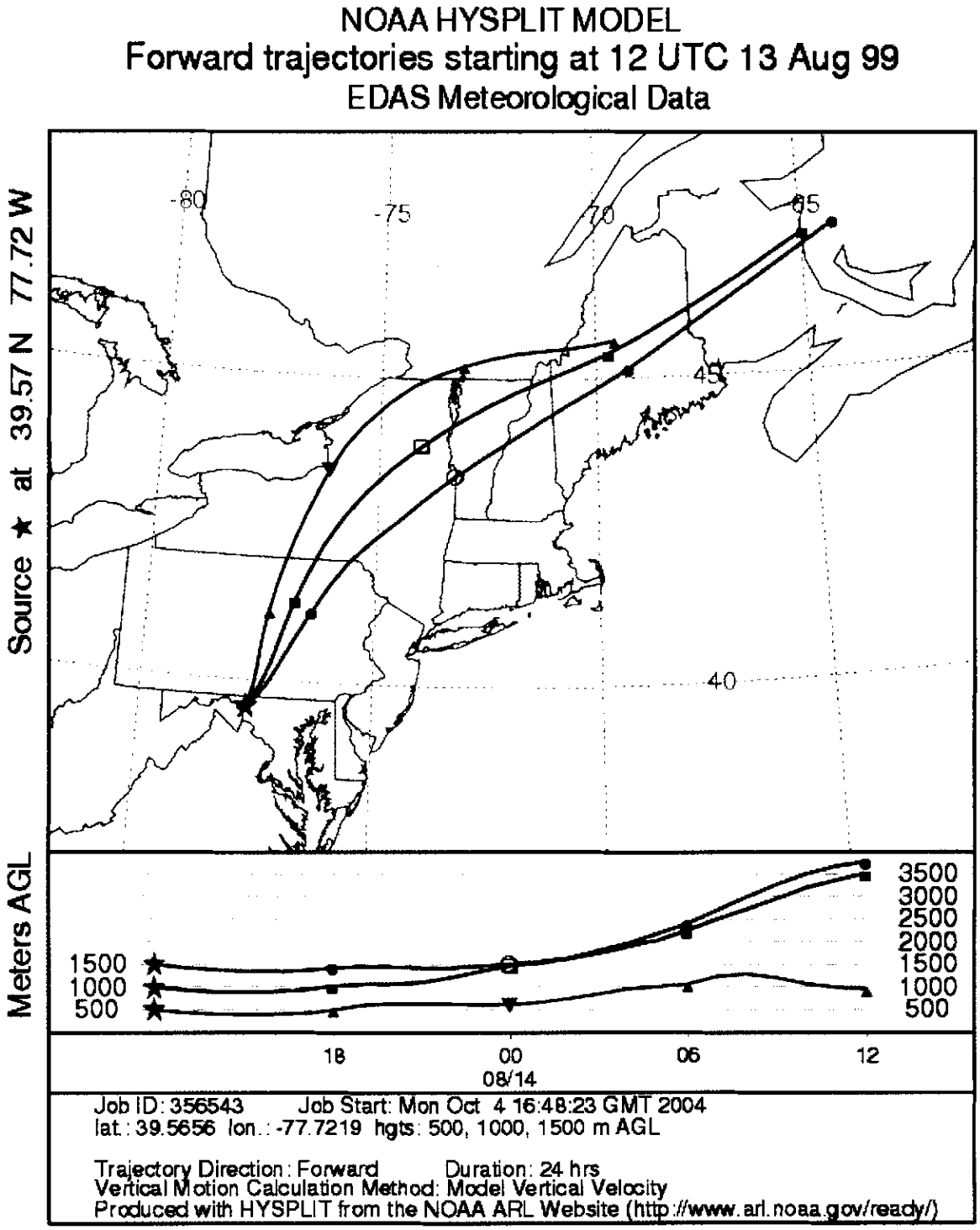

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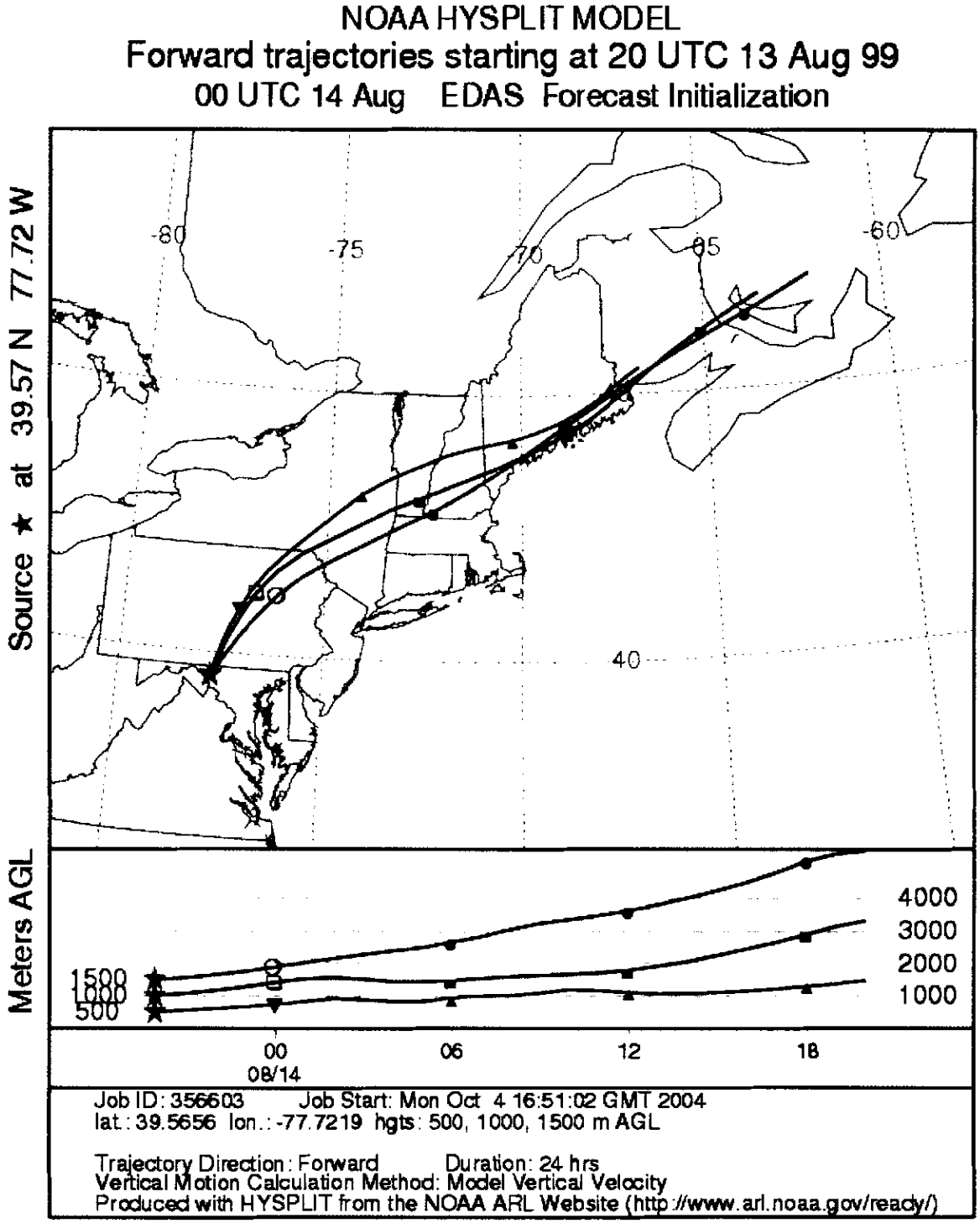

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