The Nature of the Ozone Air Quality Problem in the Ozone Transport Region: A Conceptual Description

Prepared for the Ozone Transport Commission

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Executive Summary

The Ozone Transport Region (OTR) of the eastern United States covers a large area that is home to over 62 million people living in Connecticut, Delaware, the District of Columbia, Maine, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, Vermont, and northern Virginia. Each summer, the people who live within the OTR are subject to episodes of poor air quality resulting from ground-level ozone pollution that affects much of the region. During severe ozone events, the scale of the problem can extend beyond the OTR's borders and include over 200,000 square miles across the eastern United States. Contributing to the problem are local sources of air pollution as well as air pollution transported hundreds of miles from distant sources outside the OTR.

To address the ozone problem, the Clean Air Act Amendments require states to develop State Implementation Plans (SIPs) detailing their approaches for reducing ozone pollution. As part of this process, states are urged by the U.S. Environmental Protection Agency (USEPA) to include in their SIPs a conceptual description of the pollution problem in their nonattainment areas. This document provides the conceptual description of the ozone problem in the OTR states, consistent with the USEPA's guidance.

Since the late 1970s, a wealth of information has been collected concerning the regional nature of the OTR's ground-level ozone air quality problem. Scientific studies have uncovered a rich complexity in the interaction of meteorology and topography with ozone formation and transport.

The evolution of severe ozone episodes in the eastern U.S. often begins with the passage of a large high pressure area from the Midwest to the middle or southern Atlantic states, where it assimilates into and becomes an extension of the Atlantic (Bermuda) high pressure system. During its passage east, the air mass accumulates air pollutants emitted by a number of sources in upwind states, including large coal-fired power plants and mobile and area sources. Later, sources within the OTR make their own contributions to the air pollution burden. These expansive weather systems favor the formation of ozone by creating a vast area of clear skies and high temperatures. These two prerequisites for abundant ozone formation are further compounded by a circulation pattern favorable for pollution transport over large distances. In the worst cases, the high pressure systems stall over the eastern United States for days, creating ozone episodes of strong intensity and long duration.

One transport mechanism that can play a key role in moving pollution long distances is the nocturnal low level jet. The jet is a regional scale phenomenon of higher wind speeds a few hundred meters above the ground just above the stable nocturnal boundary layer. The jet has been observed just before or during ozone events. It can convey air pollution several hundreds of miles overnight from the southwest to the northeast, directly in line with the major population centers of the Northeast Corridor stretching from Washington, DC to Boston, Massachusetts. The nocturnal low level jet can extend the entire length of the corridor from Virginia to Maine, and has been observed as far south as Georgia. It can also act to bring pollutants from different directions compared to the prevailing airflow outside the low level jet. It can thus be a

transport mechanism for bringing ozone and other air pollutants into the OTR from outside the region, as well as move locally formed air pollution from one part of the OTR to another.

Other transport mechanisms occur over smaller scales. These include land, sea, mountain, and valley breezes that can selectively affect relatively local areas. For example, sea breezes can differ in wind direction, thereby bringing air masses trapped in a thin layer over the cooler water back onto shore. Such mechanisms play a vital role in drawing ozone-laden air into some areas, such as coastal Maine, that are far removed from major source regions.

With the knowledge of the different transport scales into and within the OTR, a conceptual picture of bad ozone days emerges. After sunset, the ground cools faster than the air above it, creating a nocturnal temperature inversion. This stable boundary layer extends from the ground to only a few hundred meters in altitude. Above this layer, a nocturnal low level jet can form with higher velocity winds relative to the surrounding air. It forms from the fairly abrupt removal of frictional forces induced by the ground that would otherwise slow the wind. Absent this friction, winds at this height are free to accelerate, forming the nocturnal low level jet. Ozone above the stable nocturnal inversion layer is likewise cut off from the ground, and thus it is not subject to removal on surfaces or chemical destruction from low level emissions, the two most important ozone removal processes. Ozone in high concentrations can be entrained in the nocturnal low level jet and transported several hundred kilometers downwind overnight. The next morning as the sun heats the Earth's surface, the nocturnal boundary layer begins to break up, and the ozone transported aloft overnight mixes down to the surface where concentrations rise rapidly, partly from mixing and partly from ozone generated locally. By the afternoon, abundant sunshine combined with warm temperatures promotes additional photochemical production of ozone from local emissions. As a result, ozone concentrations reach their maximum levels through the combined effects of local and transported pollution. This combined air mass will then continue to blow along with the wind, carrying elevated concentrations of ozone to areas farther downwind, causing late afternoon and even overnight ozone peaks.

Ozone moving over water is, like ozone aloft, relatively isolated from destructive forces. This air pollution is also protected from vertical mixing and dilution by a relatively shallow mixing layer that occurs when the water is cooler than the air above it. When ozone is transported into coastal regions by bay, lake, and sea breezes arising from afternoon temperature contrasts between the land and water, it can arrive highly concentrated.

During severe ozone episodes associated with high pressure systems, these multiple transport features are embedded within a large ozone reservoir arriving from source regions to the south and west of the OTR. Thus a severe ozone episode can contain elements of long-range air pollution transport from outside the OTR, including nocturnal low level jets, regional scale transport within the OTR, and local transport along coastal shores due to bay, lake, and sea breezes.

From this conceptual description of ozone formation and transport into and within the OTR, air quality planners need to develop an understanding of what it will take to

clean the air in the OTR. There are distinct regional and local components that would best be addressed by implementing national, regional, and local controls, respectively. Observed ozone levels in the elevated reservoir often are close to or exceed 0.060 - 0.070 ppm averaged over 8 hours, which is the range that EPA has proposed for the revised National Ambient Air Quality Standard (NAAQS) for ozone. Given that the regional and national load will continue to play a major role in ozone episodes as the ozone NAAQS is lowered, further strengthening of national rules will be critical in mitigating the ozone problem.

Because weather is always changing, every ozone episode is unique in its specific details. The relative influences of the transport pathways and local emissions vary by hour and day during the course of an ozone episode and between episodes. The smaller scale weather patterns that affect pollution accumulation and its transport underscore the importance of local (in-state) controls for emissions of nitrogen oxides (NO_X) and volatile organic compounds (VOCs), the main precursors of ozone formation in the atmosphere. Larger synoptic scale weather patterns, and pollution patterns associated with them, support the need for NO_X controls across the broader eastern United States.

Studies and characterizations of nocturnal low level jets also support the need for local and regional controls on NO_X and VOC sources as locally generated and transported pollution can both be entrained in nocturnal low level jets formed during nighttime hours. The presence of land, sea, mountain, and valley breezes indicate that there are diverse aspects of pollution accumulation and transport that are area-specific and will warrant policy responses at the local and regional levels beyond a one-size-fits-all approach. In addition, over the course of a day, ozone can be NO_X-sensitive during some hours, and VOC-sensitive during others, indicating temporally varying regional and local influences on ozone formation and transport. This further underscores the need for air quality regulators to adopt a combination of national, regional, and local emission controls to address the problem.

The type of emission controls is important. Regional ozone formation is primarily due to NO_X, but VOCs are also important because they influence how efficiently ozone is produced by NO_X, particularly within urban centers. While reductions in anthropogenic VOCs will typically have less of an impact on long-range ozone transport, they can be effective in reducing ozone in urban areas where ozone production may be limited by the availability of VOCs. Therefore, a combination of localized VOC reductions with additional regional NO_X reductions will help to reduce ozone and precursors in nonattainment areas as well as downwind transport across the entire region.

Photochemical air quality modeling is a powerful yet limited planning tool. While it has undergone considerable improvement over the past decade, it is far from perfect in its ability to replicate ozone transport. There can be large uncertainties in various inputs and processes used by the model, such as precursor emissions inventories, meteorology, and atmospheric chemistry, yet the models can provide useful directionally correct guidance. Given the more recent understanding of the myriad complexities of ozone transport events, it is important that decision-makers use a variety of data sources to characterize the problem and assess possible solutions.

The recognition that ground-level ozone in the eastern United States is a regional problem requiring a regional solution marks one of the greatest advances in air quality management in the United States. During the 1990s, air quality planners began developing and implementing coordinated regional and local control strategies for NO_X and VOC emissions that went beyond the previous emphasis on urban-only measures. These measures have resulted in significant improvements in air quality across the OTR. Measured NO_X emissions and ambient concentrations have dropped between 1997 and 2005, and the frequency and magnitude of ozone exceedances have declined within the OTR. With the National Ambient Air Quality Standards likely continuing to be lowered over time, inter-regional transport will play an even larger role in the future. To maintain the current momentum for improving air quality so that the OTR states can meet their attainment deadlines, there continues to be a need for additional regional NO_X reductions coupled with appropriate local NO_X and VOC controls.

1. INTRODUCTION

1.1. Background

Ground-level ozone is a persistent public health problem in the Ozone Transport Region (OTR), a large geographical area that is home to over 62 million people living in Connecticut, Delaware, the District of Columbia, Maine, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, Vermont, and northern Virginia. Breathing ozone in the air harms lung tissue, and creates the risk of permanently damaging the lungs. It reduces lung function, making breathing more difficult and causing shortness of breath. It aggravates existing asthmatic conditions, thus potentially triggering asthma attacks that send children and others suffering from the disease to hospital emergency rooms. Ozone poses a significant risk to people with preexisting respiratory illnesses, such as emphysema and bronchitis, and it may reduce the body's ability to fight off bacterial infections in the respiratory system. Ground-level ozone also affects otherwise healthy children and adults who are very active outdoors, either at work or at play, during times of high ozone levels (USEPA, 1999). In addition, evidence suggests that short-term ozone exposure has potential cardiovascular effects that may increase the risk of heart attack, stroke, or even death (USEPA, 2006; Bell *et al*., 2006).

The Clean Air Act requires states that have areas designated "nonattainment" of the ozone National Ambient Air Quality Standard (NAAQS) to submit State Implementation Plans (SIPs) demonstrating how they plan to attain the ozone NAAQS. The SIPs must also include regulations that will yield the necessary emission reductions to attain the national ozone health standard. As part of the SIP process, the U.S. Environmental Protection Agency (USEPA) urges states to include a conceptual description of the pollution problem in their nonattainment areas. The USEPA has provided guidance on developing a conceptual description, which is contained in Section 11 of the document "Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze" (USEPA, 2007) (Appendix A of this report reproduces Section 11 of the USEPA guidance document). This document provides the conceptual description of the ozone problem in the OTR states, consistent with the USEPA's guidance. In the guidance, the USEPA presents a list of questions to help define the ozone problem in a nonattainment area. This report addresses these questions, as well as provides some in-depth data and analyses that can assist states in developing conceptual descriptions tailored to their specific areas, where appropriate.

1.2. Ozone formation

Ground-level ozone is formed in the atmosphere through a series of complex chemical reactions involving sunlight, warm temperatures, nitrogen oxides (NO_X) and volatile organic compounds (VOCs). Figure 1-1 is a conceptual picture of the emission sources and conditions contributing to ozone formation in the atmosphere. There are natural (biogenic) sources of NO_X, such as formation by soil microbes, lightning, and forest fires, but the dominant NO_X sources in the eastern United States arise from human

activities, particularly the burning of fossil fuels in cars, trucks, power plants, and other combustion sources (MARAMA, 2005).

In contrast to NO_X sources, there are significant biogenic sources of VOCs in the eastern United States that can play an important contributing role in ozone formation. Isoprene, a highly reactive natural VOC emitted typically by deciduous trees such as oak, is an important ozone precursor across large parts of the East. Isoprene emissions typically increase with temperature up to a point before high temperatures tend to shut off emissions as leaf stomata (pores) close to reduce water loss. The temperature and sunlight conditions favorable for increased ozone production also tend to be favorable for isoprene emissions (MARAMA, 2005).

Human-caused (anthropogenic) VOC emissions are important and may dominate the VOC emissions by mass (weight) in an urban area, even though natural sources dominate in the overall region. Some anthropogenic VOCs, such as benzene, are toxic, and may increase risks of cancer or lead to other adverse health effects in addition to helping form ozone (MARAMA, 2005).

Figure 1-1. Conceptual picture of ozone formation in the atmosphere

Picture provided by the Maryland Department of the Environment.

The relationship between the relative importance of NO_X and VOC emissions in producing ozone is complex. The relative ratio of NO_X and VOC levels in the local atmosphere can affect the efficiency of local urban ozone production, and this can vary by time (hour or day) at the same urban location, as well as across locations within the same urban area. For example, high NO_X concentrations relative to VOC levels may hinder ozone production through the destruction of ozone by NO_X (sometimes called

"NO_X scavenging"). The same NO_X, however, when diluted relative to VOCs through the downwind transport and dispersal of a pollution plume, will promote ozone formation elsewhere.

1.3. Spatial pattern of ozone episodes in the OTR

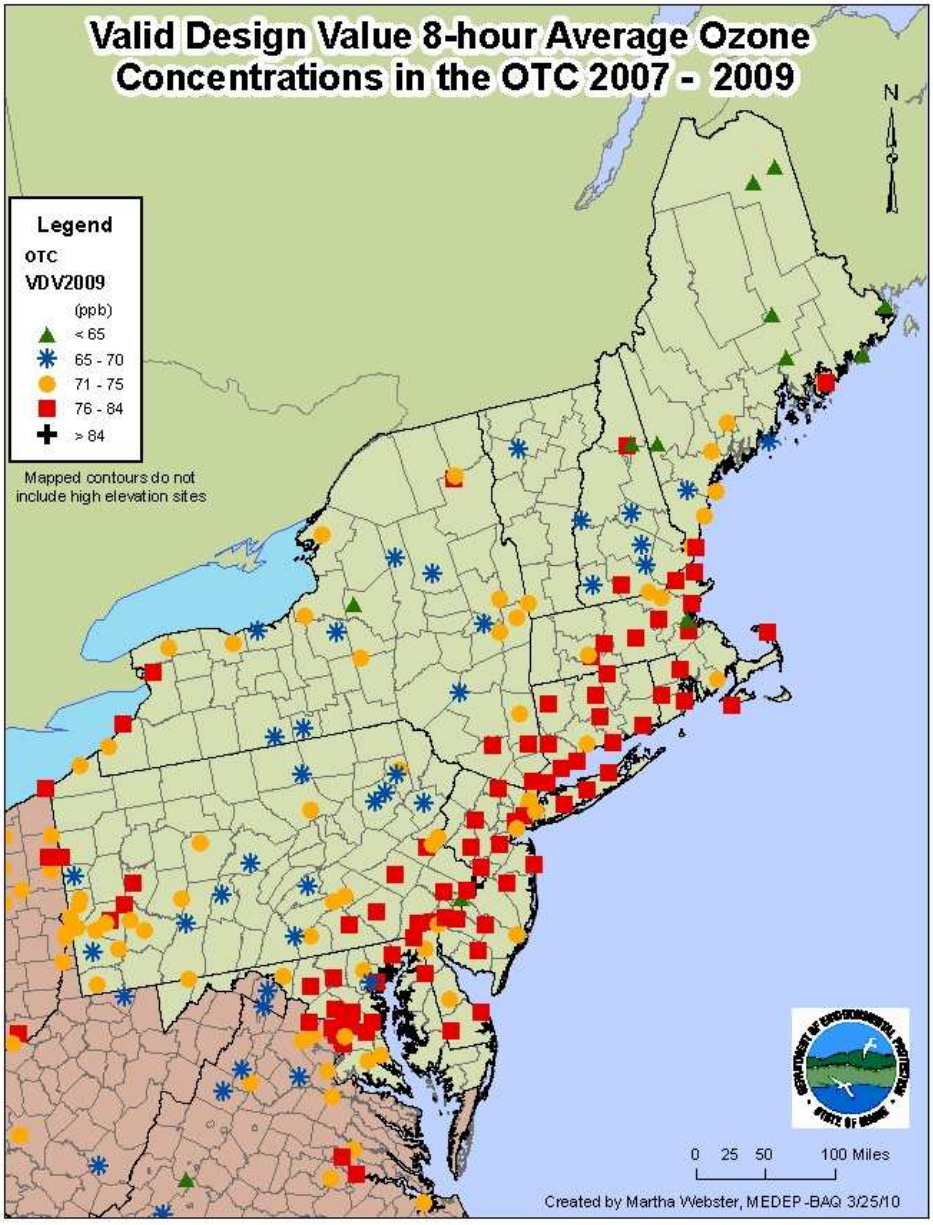
The day-to-day pattern of ground-level ozone varies according to meteorological variables that include, but are not limited to, clouds, air temperature, wind speed, and wind direction. This variability of the daily ozone pattern is tied to variations in the atmosphere's circulations over a range of scales, and how geographic features influence these circulations. These features can include boundaries between land and sea, and the influence of the Appalachian Mountains on winds to their east over the Atlantic Coastal Plain.

For the OTR, Stoeckenius and Kemball-Cook (2005) have identified five general ozone patterns: (1) high ozone throughout the OTR; (2) high ozone confined to the extreme southeastern OTR; (3) high ozone along the I-95 corridor and northern New England; (4) high ozone in the western OTR; and (5) generally low ozone throughout the OTR. Based on a clustering analysis of historical data, Stoeckenius and Kemball-Cook estimated the frequency of occurrence (number of days) for each general ozone pattern in the OTR as 37 percent Type A, 15 percent Type B, 20 percent Type C, 13 percent Type D, and 14 percent Type E. However, not all ozone episodes necessarily neatly fit into one of the five general patterns as daily conditions will vary and a given ozone episode may exhibit characteristics from different class types as the episode evolves. These five general patterns, however, are a useful classification scheme for understanding ozone chemistry and dynamics in the OTR. Appendix B presents the descriptions of the five general ozone patterns and their meteorological attributes as developed by Stoeckenius and Kemball-Cook (2005).

1.4. The regional extent of the ozone problem in the OTR

Air monitoring demonstrates that areas with ozone problems in the OTR do not exist in isolation. The map in Figure 1-2 shows an extensive pattern of closely adjacent ozone monitors throughout the OTR recording elevated levels of ozone during 2007-2009. The figure shows extensive areas throughout the OTR with ozone concentrations exceeding 70 parts per billion (ppb), which is above the upper limit of the USEPA's recently proposed revision for the 8-hour ozone primary NAAQS. The broad spatial reach of the observed high ozone is a strong indication of the regional nature of the OTR's ozone problem.

Figure 1-2. Map of 8-hour ozone design values in the OTR for 2007-2009

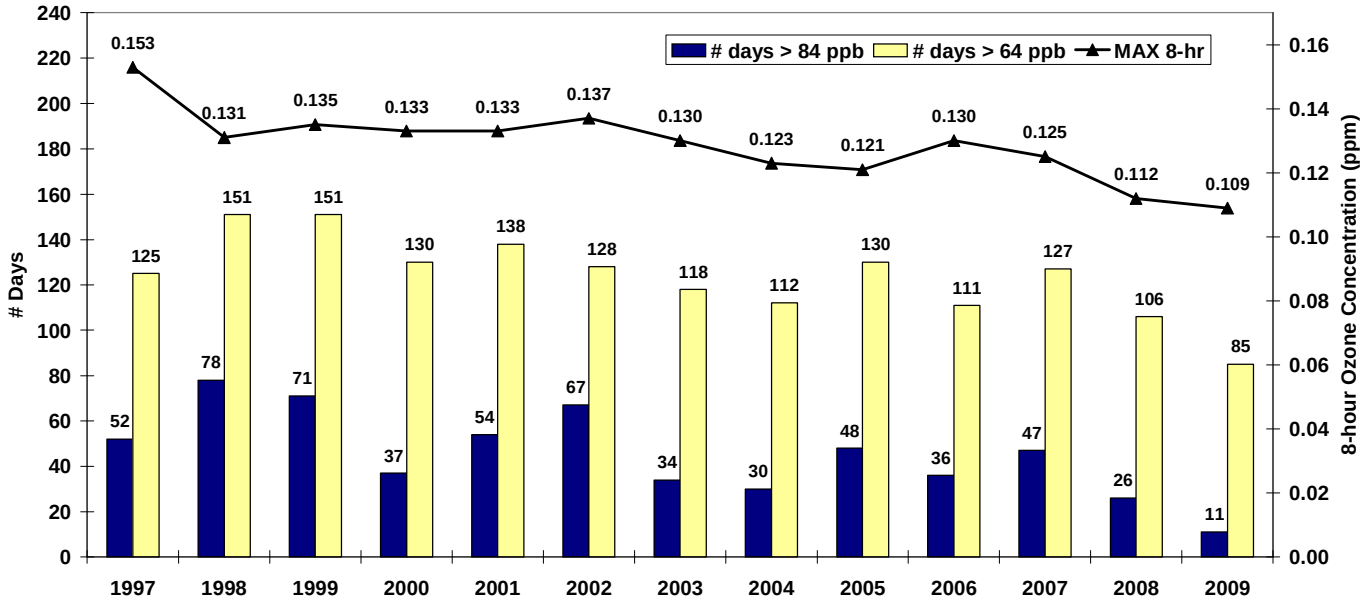


Note: A monitor's design value is the 3-year average of the annual 4th maximum 8-hour ozone level for the years 2007-2009. The figure shows the regional nature of ozone levels in the OTR, with clusters of nonattainment areas (design values ≥ 71 ppb) along with a broader region of elevated regional ozone (e.g., baseline design values ≥ 65 ppb)

1.5. Ozone trends in the OTR

The number of 8-hour ozone exceedance days vary year-to-year in the OTR, not only due to implementing control measures but also driven by variations in meteorology. During warmer summers conducive for ozone formation, the number of exceedance days at individual monitors in nonattainment areas of the OTR has been frequent, typically with 10 or more days above the 1997 8-hour ozone NAAQS of 0.08 parts per million (ppm) during the course of the summer. Figure 1-3 displays the variation in 8-hour 0.08 ppm (>84 ppb) exceedance days when collectively considering all monitoring sites across the OTR since 1997 (left hand bars in each pair). Also shown are the number of days at or above the 8-hour 0.065 ppm level (right hand bars in each pair), which is the middle of the range from the USEPA's recently proposed revision of the primary ozone NAAQS (0.060 ppm – 0.070 ppm). The figure also includes a line indicating the trend in the maximum 8-hour ozone concentrations observed in the OTR each year.

Figure 1-3. Trends in 8-hour ozone in the OTR 1997-2009



Note: The bars correspond to the number of 8-hour ozone exceedance days per year in the OTR. The left hand bar in each pair of bars gives the annual number of days exceeding 84 ppb (1997 ozone NAAQS level) and the right hand bar gives the annual number of days above 64 ppb. The upper line indicates the trend in maximum 8-hour ozone concentrations in the OTR during 1997-2009. (Figure created by Tom Downs, Maine Dept. of Environmental Protection.)

The variation in high ozone days from year-to-year due to meteorology in addition to changes in ozone precursor emissions makes it difficult to discern a clear trend in Figure 1-3, although there is some indication that the number of exceedance days has declined in recent years. Nguyen and Woodman (2008) have noted a 3-5 year cycle in Maryland during which the number of hot days (≥ 90° F) gradually increases across the cycle period. The number of ozone exceedance days in Maryland (> 84 ppb) followed the same cycle as hot days until 2002, when a break occurred with annual ozone exceedance

days consistently falling below normal regardless of the number of hot days. The departure of the ozone exceedance days from tracking with the hot days cycle coincided with implementation of the NO_X SIP Call in the eastern U.S. as power plants completed installations of controls to reduce NO_X emissions contributing to regional ozone formation and transport. Higher elevation ozone at Shenandoah National Park, an indicator of transport from the Ohio River Valley into the OTR, also dropped during the period from 1993 to 2007 by about an average of 10 ppb at the height of the ozone season in July (Nguyen & Woodman, 2008).

In a trends analysis by Bloomer *et al.*, rural surface ozone levels in the eastern United States decreased by about 6 ppb per decade over the period of 1989 to 2007. The greatest ozone decreases occurred during the periods of highest ozone (i.e., mid-day during the summer months). Over this same time span, summer temperatures increased by about 0.5 °C per decade. Recognizing that ozone generally increases with increasing temperature, Bloomer *et al.* (2010) cited this as additional evidence that emission reductions, including the NO_X SIP Call, rather than changes in weather or the climate were responsible for the ozone decrease.

Chan (2009) has also evaluated regional ground-level ozone trends in Canada and the eastern United States from 1997 to 2006 that takes into account the influence of meteorology. His analysis found a statistically significant decreasing trend in daily maximum 8-hour averaged ozone across much of southeastern Canada and the eastern U.S. The meteorologically-adjusted trend for 8-hour ozone at eastern U.S. monitoring sites was a reduction of about one ppb per year (Chan, 2009). Chan also observed that the trend in lower ozone concentrations did not fall as quickly as the trends for higher ozone concentrations. This could indicate a rising background ozone. Cooper *et al.* (2010) also documented this trend and attributed it to increasing global emissions, particularly in Asia. Rising global temperatures may also play a role by enhancing photochemical production of ozone (see discussion in Section 4.6).

The trends in ozone have important implications for strategies to achieve a future revised ozone NAAQS lower than 0.08 ppm. They may indicate that current ozone control strategies, while relatively effective at reducing higher peak ozone levels, are less effective at reducing lower levels of ozone. In addressing "background" ozone levels, researchers have identified methane reductions as a potentially effective approach (Fiore *et al.*, 2002; 2003). For higher peak ozone concentrations, reducing methane is not typically considered due to its relatively low atmospheric chemical reactivity on the time-scale of multi-day ozone episodes, but methane reductions would have a relatively larger influence on longer-term background ozone levels. Since the beginning of the industrial revolution, global atmospheric methane concentrations have more than doubled (Blunier *et al.*, 1993).

1.6. History of ozone transport science

1.6.1. From the 1970s to the National Research Council report, 1991

Research studies conducted in the 1970s gave some of the earliest indications that pollution transport plays an important role in contributing to air pollution problems in the OTR. One of the first studies to recognize the regional nature of the ozone problem in the OTR (Wolff *et al*., 1977) observed high ozone levels during the summer of 1975 in air entering the Northeast Corridor from the Midwest (up to 130 ppb), affecting the Northeast from Washington, DC to Boston, MA. Southwest flow along the Corridor appeared to amplify the ozone levels, with levels reaching 315 ppb in southern Connecticut. The combination of long-range transport from the Midwest coupled with complex movements of air parcels within the Corridor led to the early proposition that a multiregional control strategy for ozone precursors would be needed to address the ozone problem in the Northeast Corridor.

In a follow-up to this study, Wolff and Lioy (1980) described their observations of a "river of ozone" extending from the Gulf Coast through the Midwest and into New England during a series of ozone episodes during July 1978. Aircraft flights during the summer of 1979 tracked a mass of air containing ozone and its precursors leaving central Ohio, crossing the length of Pennsylvania, and entering the Northeast Corridor, where it was estimated to contribute upwards of 90 ppb on top of early morning ozone concentrations prior to local ozone formation from local emissions (Clarke & Ching, 1983).

A number of early studies documented the role of large coal-fired power plants in forming significant amounts of ozone pollution that traveled far downwind from the power plant source and contributed to a large elevated reservoir of regional ozone (Davis *et al*., 1974; Miller *et al*., 1978; Gillani & Wilson, 1980; Gillani *et al*., 1981; White *et al*., 1983). Section 2 below describes in more depth the observed meteorological processes identified as the ozone transport mechanisms important for the OTR.

A number of studies have established that regional ozone formation over the eastern United States is limited primarily by the supply of anthropogenic NO_X, with anthropogenic VOCs having less regional influence compared to their potential urban influence. This is due to the presence of significant amounts of natural VOCs across broad areas of the eastern United States (Trainer *et al*., 1987; Chameides *et al*., 1988; Sillman *et al*., 1990; McKeen *et al*., 1991; Chameides *et al*., 1992; Trainer *et al*., 1993; Jacob *et al*., 1993).

The presence of dispersed NO_X emissions sources, such as coal-fired power plants, in rural regions rich in isoprene and other natural VOC emissions from trees and other vegetation often leads to elevated regional ozone during the summer months. This ozone can be mixed vertically upward through convection as the atmosphere warms during the day, and become entrained in upper level winds. When captured above the surface boundary layer overnight as the atmosphere cools after sunset, the ozone can be

transported downwind long distances. The following morning, mixing of the atmosphere can occur again as the sun warms the earth's surface, bringing the ozone aloft back down to the ground. The transported ozone then contributes to high background concentrations entering urban areas during the early morning hours before local production of ozone occurs from local precursor emissions (both NO_X and VOCs) (see further discussion in Section 2.3.4).

In 1991, a National Research Council (NRC) committee, synthesizing the best available information at the time on ozone formation and transport in the eastern United States, reported (NRC, 1991):

> High ozone episodes last from 3-4 days on average, occur as many as 7-10 times a year, and are of large spatial scale: >600,000 km^2. Maximum values of non-urban ozone commonly exceed 90 ppb during these episodes, compared with average daily maximum values of 60 ppb in summer. An urban area need contribute an increment of only 30 ppb over the regional background during a high ozone episode to cause a violation of the National Ambient Air Quality Standard (NAAQS) in a downwind area. … Given the regional nature of the ozone problem in the eastern United States, a regional model is needed to develop control strategies for individual urban areas.
>
> [Note: The NRC discussion was in the context of the ozone NAAQS at the time of the NRC report, which was 0.12 ppm (120 ppb) averaged over one hour.]

The observed ozone spatial scale of >600,000 km^2 (>200,000 square miles) is comparable to the combined size of Kentucky, Ohio, West Virginia, Pennsylvania, Maryland, New York, and New Jersey. Additional field studies and modeling efforts (described below) since the NRC report have reinforced its basic findings and provide a consistent and coherent body of evidence for the prominence of transport throughout the eastern United States.

1.6.2. Ozone Transport Assessment Group: 1995-1997

The increasing regulatory focus on broader regional approaches to ozone control beyond the OTR began with the Ozone Transport Assessment Group (OTAG) in 1995. OTAG was a unique partnership between the USEPA, the Environmental Council of the States (ECOS), state and federal government officials, industry organizations, and environmental groups. OTAG's goal was "to develop an assessment of and consensus agreement for strategies to reduce ground-level ozone and its precursors in the eastern United States" (OTAG, 1997a). The group assessed transport of ground-level ozone across state boundaries in the 37-state region and culminated in a set of recommendations to the USEPA in 1997.

OTAG supported a significant modeling effort of four regional ozone episodes across the eastern United States. OTAG's Regional and Urban Scale Modeling Workgroup found that on a regional scale, modeled NO_X reductions produced widespread ozone decreases across the eastern United States with limited ozone increases generally confined to some urban areas. Also on a regional scale, VOC reductions resulted in limited ozone decreases generally confined to urban areas (OTAG, 1997b).

The OTAG Air Quality Analysis Workgroup provided additional observational and other analytical results to inform model interpretation and the development of OTAG

recommendations. Among its many findings, this Workgroup observed the following about the range of ozone:

> Low wind speeds (< 3 m/sec) enable the accumulation of ozone near local source areas. High winds (> 6 m/sec) reduce the concentrations but contribute to the long-range transport of ozone. The average range of ozone transport implied from an array of diverse methods is between 150 miles and 500 miles. However, the perceived range depends on whether one considers the average concentrations (300–500 miles) or peak concentrations (tens of miles at 120 ppb). (OTAG, 1997c)

In addition, it discussed the increasingly important role of transport as the NAAQS is lowered:

> The relative importance of ozone transport for the attainment of the new 80 ppb 8-hour standard is likely to be higher due to the closer proximity of nonattainment areas. (OTAG, 1997c)

Based on the variety of technical work performed by multiple stakeholders during the process, OTAG reached a number of major conclusions (OTAG, 1997d), including:

- Regional NO_X reductions are effective in producing ozone benefits; the more NO_X reduced, the greater the benefit.
- Ozone benefits are greatest in the subregions where emissions reductions are made; the benefits decrease with distance.
- Both elevated (from tall stacks) and low-level NO_X reductions are effective.
- VOC controls are effective in reducing ozone locally and are most advantageous to urban nonattainment areas.
- Air quality data indicate that ozone is pervasive, that ozone is transported, and that ozone aloft is carried over and transported from one day to the next.

The technical findings of OTAG workgroups were consistent with the modeling and observational studies of regional ozone in the eastern United States already appearing in the scientific literature at that time.

Through its work, OTAG engaged a broad group outside of the scientific community in the discussion of ozone transport. This brought a greater understanding of the role of ozone transport across the eastern United States that was then translated into air quality policy with the creation of a regional ozone control strategy focusing on the reduction of NO_X emissions from power plants.

1.6.3. Northeast Oxidant and Particle Study (NE-OPS) 1998-2002

The Northeast Oxidant and Particle Study (NE-OPS) began in 1998 as a USEPA sponsored project to study air quality issues in the Northeast. The study undertook four major field programs at a field site in northeastern Philadelphia during the summers of 1998, 1999, 2001, and 2002. It involved a collaborative effort among research groups from a number of universities, government laboratories, and representatives of the electric power industry in an investigation of the interplay between the meteorological and chemical processes that lead to air pollution events in the Northeast. A suite of measurement techniques at and above the earth's surface gave a three-dimensional regional scale picture of the atmosphere. The studies found that horizontal transport aloft and vertical mixing to the surface are key factors in controlling the evolution and severity of air pollution episodes in the Northeast (Philbrick *et al*., 2003a).

At the conclusion of the 2002 summer field study, the NE-OPS researchers were able to draw several conclusions about air pollution episodes in Philadelphia and draw

inferences from this to the conditions in the broader region. These include (Philbrick *et al.*, 2003b):

- Transported air pollution from distant sources was a major contributor to all of the major summer air pollution episodes observed in the Philadelphia area.
- Regional scale meteorology is the major factor controlling the magnitude and timing of air pollution episodes.
- Knowledge of how the planetary boundary layer evolves over the course of a day is a critical input for modeling air pollutant concentrations because it establishes the mixing volume.
- Remote sensing and vertical profiling techniques are critical for understanding the processes governing air pollution episodes.
- Ground-based sensors do not detect high levels of ozone that are frequently trapped and transported in layers above the surface.
- Horizontal and vertical transport processes, such as the nocturnal low level jets and early morning breakdown of the nocturnal boundary layer, are frequent contributors of pollutants during the major episodes.
- Specific meteorological conditions are important in catalyzing the region for development of major air pollution episodes.
- Tethered balloon and lidar measurements suggest a very rapid down mixing of species from the residual boundary layer during the early morning hours that is too large to be accounted for on the basis of NO_X reactions alone.

The findings on nocturnal low level jets occurring in concert with ozone pollution episodes are particularly salient for air quality planning for the OTR. In 19 of 21 cases where researchers observed nocturnal low level jets during the NE-OPS 2002 summer campaign in the Philadelphia area, they also saw peak 1-hour ozone levels exceeding 100 ppbv. The nocturnal low level jets were capable of transporting pollutants in air parcels over distances of 200 to 400 km. The field measurements indicating that these jets often occur during periods of large scale stagnation in the region demonstrate the important role nocturnal low level jets can play in effectively transporting air pollutants during air pollution episodes (Philbrick *et al*., 2003b).

The upper air observations using tethered balloons and lidar indicated the presence of high pollutant concentrations trapped in a residual layer above the surface, thus preserving the pollutants from destruction closer to the surface. Ozone, for example, when trapped in an upper layer during nighttime hours is not subject to destruction by NO_X scavenging from low-level emission sources (i.e., cars and trucks) or deposition to surfaces like vegetation, hence it is available for horizontal transport by nocturnal low level jets. The following day, it can vertically transport back down to the surface through vertical mixing as the nocturnal boundary layer breaks down and daytime convection. When an upper layer of ozone-laden air is horizontally transported overnight by a nocturnal low level jet, downward mixing can increase surface ozone concentrations in the morning that is not the result of local ozone production (Philbrick *et al*., 2003b).

1.6.4. NARSTO 2000

NARSTO (formerly known as the North American Research Strategy for Tropospheric Ozone) produced "An Assessment of Tropospheric Ozone Pollution – A North American Perspective" in 2000 to provide a policy-relevant research assessment of ozone issues in North America (NARSTO, 2000). While the NARSTO Assessment is

continental in scope, it encompasses issues relevant to the OTR, including results from a NARSTO-Northeast (NARSTO-NE) field campaign.

A key policy-relevant finding from the NARSTO Assessment of relevance to the OTR (NARSTO, 2000) relates to ozone transport and mitigation:

- Available information indicates that ozone accumulation is strongly influenced by extended periods of limited mixing, recirculation of polluted air between the ground and aloft, and the long-range transport of ozone and its precursors. As a result, air quality management strategies require accounting for emissions from distant as well as local sources.

The NARSTO Assessment identified the stagnation of synoptic scale (>1000 km^2) high pressure systems as a commonly occurring weather event leading to ozone pollution episodes. These systems are warm air masses associated with weak winds, subsiding air from above, and strong inversions capping the planetary boundary level in the central region of the high. The warm air mass can settle into place for days to more than a week, and slowly track from the Midwest to the East Coast. During the summer, these conditions result in the build up of pollution from local sources with reduced dispersion out of the region. In terms of air quality, these systems coincide with numerous local or urban-scale ozone pollution episodes embedded within a broader regional background of elevated ozone concentrations (NARSTO, 2000 at p. 3-34).

The NARSTO Assessment found that a variety of processes can lead to long-range transport of air pollutants that initially accumulated in these large-scale stagnation events. Over time, pollution plumes meander, merge, and circulate within the high pressure system. Because of the difference in pressures, pollutant plumes that eventually migrate to the edges of a high pressure system get caught in increasing winds at the edge regions, creating more homogeneous regional pollution patterns. Stronger winds aloft capture the regional pollutant load, and can transport it for hundreds of kilometers downwind of the stagnated air mass's center (NARSTO, 2000 at p. 3-34). For example, air flow from west to east over the Appalachian Mountains can move air pollution originating within the Ohio River Valley into the OTR.

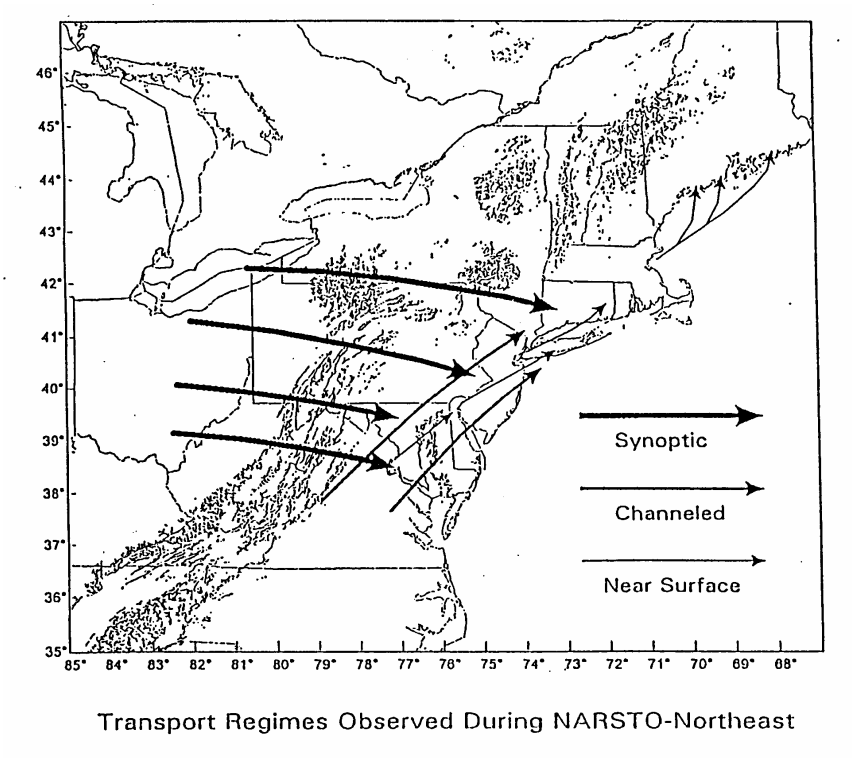
Studies undertaken by the NARSTO-NE field program also observed several regional scale meteorological features arising from geographical features in the eastern U.S. that affect pollutant transport. The NARSTO-NE field program observed nocturnal low level jets on most nights preceding regional high ozone episode day in the OTR (NARSTO, 2000 at p. 3-34), consistent with the observations of the NE-OPS campaign.

Another important smaller scale transport mechanism is the coastal sea breeze that can sweep ashore pollutants originally transported over the ocean parallel to the coastline. An example of this is the high ozone levels seen at times along coastal Maine that move in from the Gulf of Maine after having been transported in pollution plumes from Boston, New York City, and other Northeast Corridor locations (NARSTO, 2000 at pp. 3-34 through 3-37).

As a result of the NARSTO-NE field program, a conceptual picture of pollution transport into and within the OTR is possible. It consists of a combination of large-scale synoptic flow from the Midwest interacting with various regional and smaller-scale transport and meteorological features within the OTR, as illustrated in Figure 1-4.

Synoptic-scale transport from west to east across the Appalachian Mountains occurs with the slow-moving stagnant high pressure systems that foster large regional ozone episodes across eastern U.S. Regional-scale nocturnal low level jets from the southwest to the northeast along the Atlantic Coastal Plain, can occur within the synoptic system. In addition, daytime sea breezes can significantly affect bay and coast line air pollution levels within the OTR (NARSTO, 2000 at 3-36 and 3-37, citing Blumenthal *et al*., 1997).

Figure 1-4. Conceptual picture of different transport regimes contributing to ozone episodes in the OTR



Long-range (synoptic scale) transport occurs from west to east across the Appalachian Mountains. Regional scale transport also occurs from west to east through gaps in the Appalachian Mountains and in nocturnal low level jets from southwest to northeast over the Northeast Corridor. Daytime sea breezes can affect local coastal areas by bringing in air pollution originally transported near the surface across water parallel to the coast (e.g., along the Maine coastline). Figure from NARSTO, 2000, citing Blumenthal *et al.*, 1997.

Other key findings of the 2000 NARSTO Assessment include:

- Local VOC emission reductions may be effective in reducing ozone in urban centers, while NO_X emission reductions become more effective at distances removed from urban centers and other major precursor emissions.
- The presence of biogenic emissions complicates the management of controllable precursor emissions and influences the relative importance of VOC and NO_X controls.
- The effectiveness of VOC and NO_X control strategies is not uniquely defined by the location or nature of emissions. It is now recognized that the relative effectiveness of

VOC and NO_X controls may change from one location to another and even from episode to episode at the same location.

The NARSTO program also produced a comparative summary of the results of major ozone field studies undertaken in Europe and North America prior to 2000 (Solomon *et al*., 2000). The field studies occurred under a wide range of geographical and climatological conditions, and their results generally affirmed the conclusions of the earlier National Research Council ozone report (NRC, 1991). Some of the key findings of the comparative summary relevant to regional ozone and ozone control strategies in the eastern U.S. include:

(1) reaffirmation that tropospheric ozone is a multi-scale phenomenon extending to continental boundaries;

(2) aerometric conditions aloft are important to ground-level ozone;

(3) biogenic sources make important contributions to VOC and NO_X emissions in parts of eastern North America;

(4) recirculating flow over complex terrain and large water bodies are universally important factors affecting accumulation of ozone at the ground; and

(5) nonlinearities in ozone response to precursor changes create important degrees of freedom in management strategies – VOC and NO_X sensitivities vary extensively in urban and rural areas, making decisions about emissions management complicated.

1.6.5. New England Air Quality Study: 2002-2004

The New England Air Quality Study (NEAQS) has conducted field campaigns during the summers of 2002 and 2004 to investigate air quality on the Eastern Seaboard and transport of North American emissions into the North Atlantic (NEAQS, 2002).

High ozone levels in northern New England occur with light to moderate winds from source regions in the Northeast urban corridor, rather than under locally stagnant conditions. The most important transport pathway leading to high ozone in coastal New Hampshire and Maine is over water rather than over land. Transport over water is particularly important in this northern region of the OTR for several reasons. First, there is a persistent pool of cooler water in the northern and eastern Gulf of Maine and Bay of Fundy. This creates a smoother transport surface for air pollutants relative to land transport, with a decrease in convective (vertical) mixing. Second, deposition of pollutants to the water surface is very small compared to the more rapid deposition occurring on land. Third, the lack of convective mixing allows pollution to be transported in different directions and at different heights in the atmosphere (Angevine *et al*., 2004).

During the summer of 2002, researchers observed two transport events into coastal northern New England. The first occurring on July 22 through July 23 involved large-scale synoptic transport in a 400-600 m layer over the Gulf of Maine that was in contact with the water's surface. The southwesterly flow brought ozone pollution up from the New York City, Boston and other northeastern urban locations into coastal northern New England. Ozone monitors on Maine's coast extending from the New Hampshire border to Acadia National Park recorded elevated 1-hour average ozone levels between

88 and 120 ppb during this period. In a later episode during August 11-14, ozone and wind observations indicated the role of local-scale transport via a sea breeze (southeasterly flow) bringing higher ozone levels into coastal New Hampshire from a polluted layer originally transported off shore in the Gulf of Maine in a southwesterly flow arising out of the Northeast's urban corridor. Transport in an elevated layer also occurred with higher ozone recorded at a monitor on Cadillac Mountain in Acadia National Park relative to two monitors located at lower elevations in the park (Angevine *et al*., 2004; 2006).

A study of ozone at two monitors separated by only 30 km (one on an island 10 km off the New Hampshire coast, the other 20 km inland at a rural location) indicated very different ozone transport influences. The coastal site experienced consistently higher ozone concentrations than the inland site, which was attributed to different transport regimes. The coastal site was influenced by southwesterly flow along the New England coast that brought ozone and precursors from more populated areas to the southwest (e.g., Boston), while the inland site often had a more westerly wind component with fewer upwind pollution sources. At times, however, the sea breeze was a factor, and nearly equalized the ozone concentrations at the two sites by transporting ozone-rich air from the coast to the rural inland location (Darby *et al*., 2007). Investigation of ozone formation dynamics and precursor levels at the rural site were consistent with a transport mechanism bringing ozone-rich air to the location rather than the observed peak ozone being formed locally (Griffin *et al*., 2004).

The results of NEAQS indicate the important conditions contributing to ozone transport along the northern New England coast. The cool waters of the Gulf of Maine allow for transport of concentrated air pollutants over distances of 20-200 km in stable layers at the water's surface with little pollutant deposition or dilution. Sea breezes can modify large-scale synoptic transport over the ocean and bring high ozone levels into particular sites located on the coast. Transport within higher layers above the Gulf of Maine can carry pollutants over much greater distances, 200-2000 km.

1.6.6. Regional Atmospheric Measurement, Modeling, and Prediction Program (RAMMPP) 2003

The Regional Atmospheric Measurement, Modeling, and Prediction Program (RAMMPP) is a program led by researchers at the University of Maryland. Its focus is developing a state-of-the-art scientific research tool to improve understanding of air quality in the Mid-Atlantic region of the United States. It has a number of facets, including ozone and $PM_{2.5}$ pollutant level forecasting, aircraft, and surface measurements, real-time weather forecasting, and chemical transport modeling.

During the August 2003 electrical blackout in the eastern United States, one of the largest in North American history, scientists with RAMMPP obtained aircraft measurements in an unplanned "real-world" experiment investigating changes in air pollution due to the virtual shutdown of numerous coal-fired power plants across a large part of this region (Marufu *et al*., 2004). Initially, aircraft measurements were collected early in the day on August 15, 2003 above western Maryland, which was outside the blackout region. These measurements were compared with aircraft measurements taken later that day over central Pennsylvania, about 24 hours into the blackout, and to

measurements obtained over central Pennsylvania the previous year during a period of similar synoptic patterns as occurred during the blackout. Based on the comparisons, the RAMMPP scientists inferred a decrease in ozone concentrations of ~50 percent within the blackout region (as well as >90 percent decrease in SO_2 and ~70 percent reduction in light scattered by particles).

The air pollution reductions attributed to the 2003 power plant shutdowns based on the aircraft measurements and other supporting elements were subsequently questioned on several grounds (see comment by Hansen *et al.*, 2005, and reply to comment by Marufu *et al.*, 2005). A modeling reassessment of the 2003 blackout incorporating pollution reductions from power plants and other sources at that time concluded that mobile source reductions in the blackout region had a relatively greater impact on ozone in urban areas than the power plant reductions (Hu *et al.*, 2006). Power plants did have a larger modeled impact on air quality than mobile sources in some of the affected regions, but the ozone impacts were limited and much less (~4 percent) than that initially inferred by Marufu *et al.* (2004) (~50 percent). Despite the diminished projection of ozone reductions during the blackout, the reassessment by Hu *et al.* (2006) suggested that efforts to lower NO_X from power plants would continue to lower ozone in the eastern United States, and that regional and national on-road NO_X control had greater benefits than local control alone.

1.7. Summary

The chemistry of ozone formation in the atmosphere involves reactions of NO_X and VOC emissions from numerous sources during periods of warm temperatures and abundant sunshine. The day-to-day pattern of ground-level ozone in the OTR varies according to a number of meteorological variables, such as sunlight, temperature, wind speed, and wind direction. High levels of ozone within the OTR do not occur in isolation, indicating a broad regional air quality problem. Trends in 8-hour ozone levels since 1997 indicate improvement in air quality, a reflection of numerous control strategies implemented locally, regionally, and nationally to reduce emissions of the pollutants that contribute to ozone formation. For example, with the implementation of the NO_X SIP Call in the eastern U.S. after 2002, a marked departure of ozone exceedance days tracking with high temperature days was observed, with the number of high ozone days falling relative to the number of high temperature days. This has been attributed to power plants completing installations of controls to reduce NO_X emissions that contributed to regional ozone formation and transport.

The scientific literature contains a number of peer reviewed papers describing observed episodes of ozone and precursor pollutant transport. In 1991, a National Research Council report summarized the state-of-the-science, which further highlighted the broad regional nature of the ozone problem in the eastern U.S. Since then, multiple collaborative efforts and field campaigns have further investigated specific aspects of the regional ozone problem affecting the OTR, and these provide a significant foundational basis for informed policy decisions to improve air quality.

In general, the collaborative efforts and field campaigns described in this introductory chapter provide a consistent picture of the main features of ozone formation and transport affecting the OTR. On a regional scale, ozone is most sensitive to

anthropogenic NO_X, while anthropogenic VOCs can play a role in local urban areas. The sensitivity of ozone to NO_X and VOC emissions can vary across different locations, as well as at the same location at different times. Therefore, a combination of NO_X and VOC controls are likely needed for many places to meet ozone air quality standards.

Across a large part of the eastern United States, ozone can be formed by NO_X emissions from power plants, motor vehicles, and other combustion sources in the presence of highly reactive biogenic VOCs (e.g., isoprene). The regional ozone burden can mix vertically upward during the day through convection, and then be transported long distances downwind overnight. Later, it can mix back down to the surface and contribute to an elevated ozone background that downwind local emission sources add their own ozone-forming emissions to, leading to a buildup of even higher ozone levels.

Transport of ozone and its precursors occurs on multiple scales. Large-scale transport can occur over hundreds of miles and over several days through the movement of high pressure systems. These systems bring elevated levels of ozone into the OTR from areas to the west and southwest into the region. On a more moderate scale, , overnight pollutant transport can also occur through the formation of nocturnal low level jets that stretch from southwest to the northeast along the Atlantic Coastal Plain east of the Appalachian Mountains. Nocturnal low level jets have been observed in the OTR on many nights preceding high ozone episode days.

Overnight transport via either the movement of high pressure systems or low level jets occurs above the nocturnal boundary layer that forms at the surface, thus decoupling ozone aloft from ozone-destroying processes occurring at the surface. In addition, because the transport occurs aloft, it is not readily detected by ground-level ozone monitors.

An additional shorter scale transport pathway is also present in the OTR via surface transport over water. Due to greater stability of atmospheric layers over water, vertical mixing is reduced, which also diminishes the likelihood of ozone destruction at the surface. Differences in temperatures between land and water along the coast during the day can create sea breezes that bring surface transport ozone into shorelines. An example of this mechanism is pollution transported over the Gulf of Maine from emission sources in the Northeast Corridor that enter into the coasts of New Hampshire and Maine.

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2. METEOROLOGY AND EVOLUTION OF OZONE EPISODES IN THE OZONE TRANSPORT REGION

The following sections describe current knowledge of the factors contributing to ozone episodes in the OTR. The general description of weather patterns comes mainly from the work of Ryan and Dickerson (2000) done for the Maryland Department of the Environment. Further information is drawn from work by Hudson (2005) done for the Ozone Transport Commission and from a mid-Atlantic regional air quality guide by MARAMA (2005). The regional nature of the observed ozone episodes in the OTR is reinforced in modeling studies by the USEPA for the Clean Air Interstate Rule.

2.1. Large-scale weather patterns

Ryan and Dickerson (2000) have described the general meteorological features conducive to ozone formation and transport that are pertinent to the OTR. On the local scale, meteorological factors on which ozone concentrations depend are the amount of available sunlight (ultraviolet range), temperature, and the amount of space (volume) in which precursor emissions mix. Sunlight drives the key photochemical reactions for ozone and its key precursors and the emissions rates of many precursors (isoprene for example) are temperature dependent. Emissions confined within a smaller volume result in higher concentrations of ozone. Winds in the lowest 2 km of the atmosphere cause horizontal mixing while vertical temperature and moisture profiles drive vertical mixing. High ozone is typically associated with weather conditions of few clouds, strong temperature inversions, and light winds.

The large-scale weather pattern that combines meteorological factors conducive to high ozone is the presence of a region of upper air high pressure (an upper air ridge) with its central axis located west of the OTR. The OTR 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 OTR, surface high pressure is typically quite diffuse across the region. This pattern occurs throughout the year but is most common and longer lived in the summer months (Ryan and Dickerson, 2000).

The large, or synoptic, scale, weather pattern sketched above has important implications for transport into and within the OTR. First, the persistence of an upper air ridge west of the OTR drives generally west to northwest winds that can carry ozone generated outside the OTR into the OTR. A key point from this wind-driven transport mode is that stagnant air is not always a factor for high ozone episodes in the OTR. 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 ozone and its precursors from mixing downward overnight. Above the inversion layer, there is no opportunity for destruction of the pollutants by surface deposition, thus increasing the pollutants' lifetimes aloft and consequently their transport distances. Third, with diffuse surface high pressure, smaller scale effects can become dominant in the

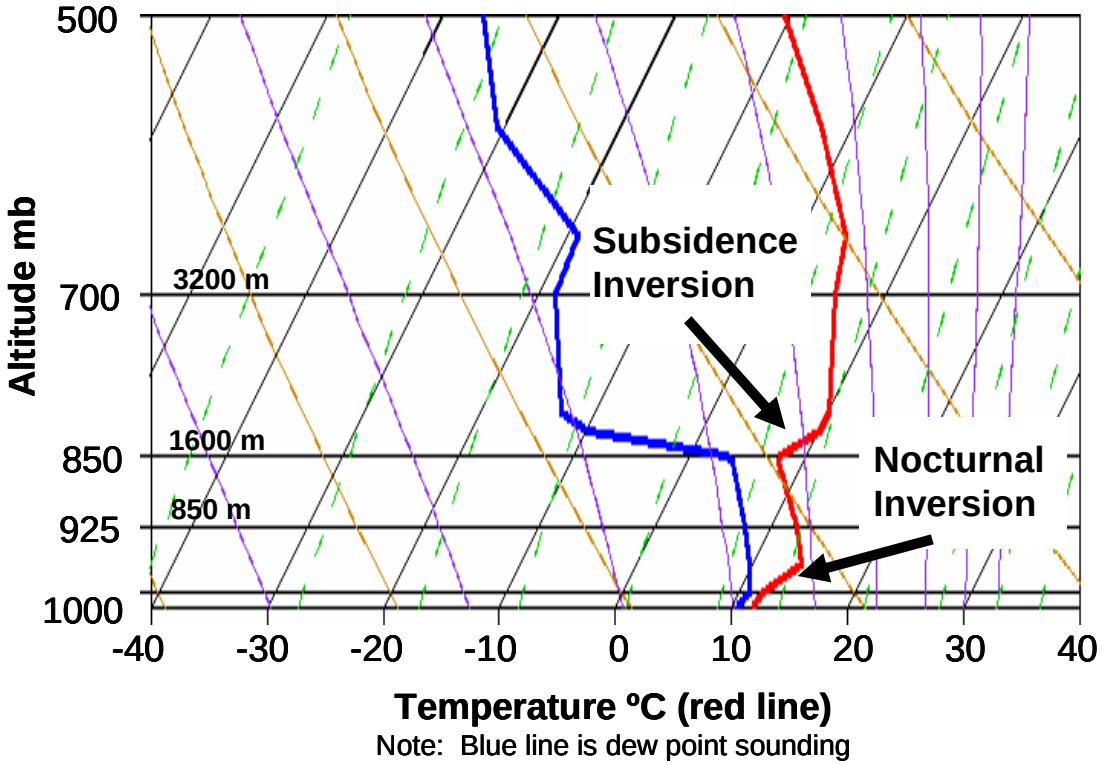
lowest layers of the atmosphere. These include bay and land breezes, the Appalachian lee side trough, and the development of the nocturnal low level jet. Nocturnal low level jets are commonly observed during high ozone events in the OTR (Ryan and Dickerson, 2000).

2.2. Meteorological mixing processes

An important element in the production of severe ozone events is the ability of the atmosphere through temperature inversions to inhibit the vertical mixing processes that under normal conditions would lead to dilution of the emitted pollutants. For the purposes of this discussion, we focus on two major classes of temperature inversions, (1) nocturnal (radiative) and (2) subsidence.

Figure 2-1 shows an example of nocturnal and subsidence inversions in a temperature profile taken over Albany, NY, on September 1, 2006 at 7 a.m. eastern standard time. The figure shows two distinct temperature inversions – the ground-based nocturnal inversion and an inversion at about 1600 meters caused by the sinking motion (subsidence) of the atmosphere in a high pressure system.

Figure 2-1. Temperature profile taken over Albany, NY, on September 1, 2006 at 7 a.m. eastern standard time



2.2.1. Nocturnal inversions

At night, the Earth's surface cools more rapidly than the air. The surface also cools the lowest hundred meters of the atmosphere. The air above this layer cools more slowly, and a temperature inversion forms. The inversion divides the atmosphere into two

layers that do not mix. Below the nocturnal surface inversion, the surface winds are weak and air pollutants can be reduced through dry deposition. Above the inversion, the pollutants are trapped above the surface. Winds can transport pollutants in the upper-level ozone reservoir through the night. Transport can be enhanced by a nocturnal low level jet as the inversion isolates the winds from the friction of the rough surface.

In the morning, the sun warms the Earth's surface, and conduction and convection transfer heat upward to warm the air near the surface. By about 10:00 – 11:00 a.m., the temperature of the surface has risen sufficiently to remove the inversion. Air from above and below the inversion can then mix freely. Depending on whether the air above the inversion is cleaner or more polluted than the air at the surface, this mixing can either lower or increase air pollution levels.

2.2.2. Subsidence inversions

Severe ozone events are usually associated with high pressure systems. In the upper atmosphere, the winds around a high pressure system move in a clockwise direction. A high pressure system is formed as upper-level air subsides, increasing the air pressure at the surface. As upper-level subsidence continues, the air at the ground is pushed away horizontally, a process called "divergence." As the subsiding air descends, it is heated by adiabatic processes. When this warmer air meets the colder air below, it forms an inversion. The height of a subsidence inversion ranges from caps pollution 1200 to 2000 meters. It is far more difficult to break down than the nocturnal inversion and can last from hours to days. The subsidence inversion limits vertical mixing in the middle of the day during an air pollution episode, keeping pollutants trapped closer to the ground.

2.3. Meteorological transport processes

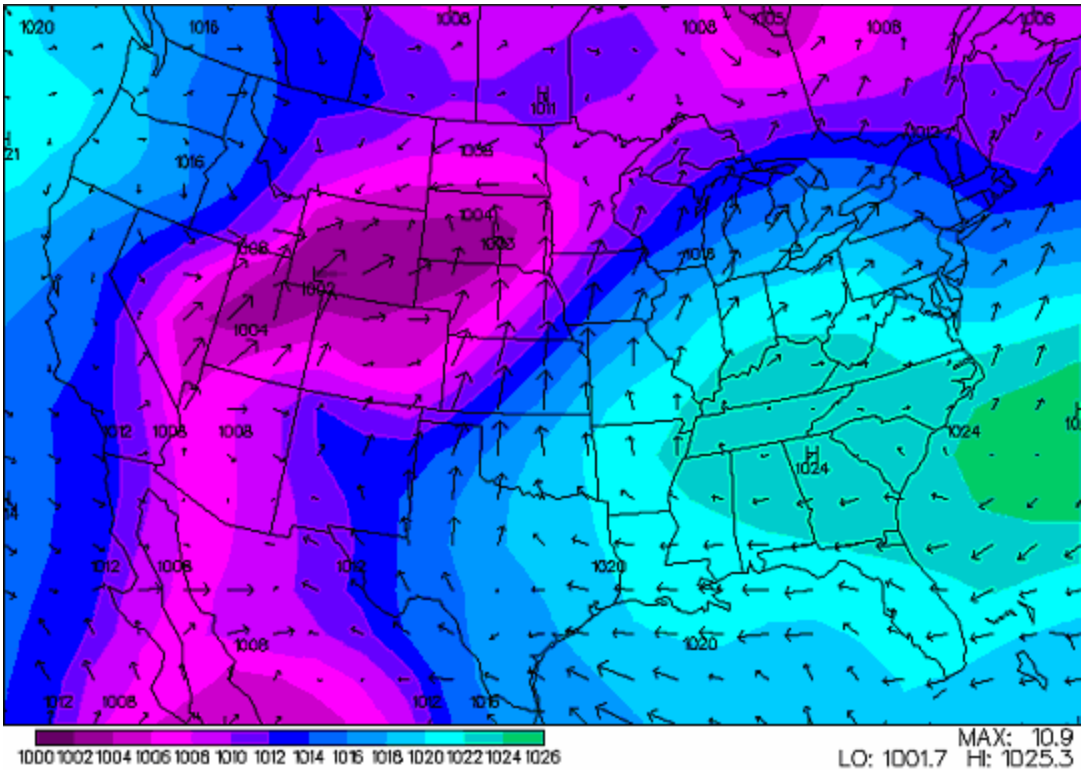
2.3.1. Introduction

Figure 2-2 shows the classic synoptic weather pattern at the Earth's surface associated with severe ozone episodes within the OTR. A quasi-stationary high pressure system (the Bermuda high) extends from the Atlantic Ocean westward into interior southeastern U.S., where a second weaker high is located. Surface winds, circulating clockwise around the high, are especially light in the vicinity of the secondary high. Farther north, a southwesterly flow strengthens toward New York and southern New England. This situation illustrates two circulation regimes often existing in OTR ozone episodes: more stagnant conditions in southern areas and a moderate transport flow in the OTR from southwest to northeast.

The Appalachian Mountains induce changes in the wind field that also play important roles in the formation and transport of ozone in the OTR. The mountains act as a physical barrier confining, to some degree, pollution to the coastal plain. They also induce local effects such as mountain and valley breezes, which, in the case of down-slope winds, can raise surface temperatures thereby increasing chemical reactivity. In addition, a lee side trough can develop on the east of mountains as a result of dynamics from a system just west of the mountains. A leeside trough helps to channel a more concentrated ozone plume, and contribute to the formation of nocturnal low level jets, the engine of rapid nighttime transport.

The Atlantic Ocean also plays a strong role during ozone episodes where sea breezes can draw either heavily ozone-laden or clean marine air into coastal areas.

Figure 2-2. Schematic of a typical weather pattern associated with severe ozone episodes in the OTR



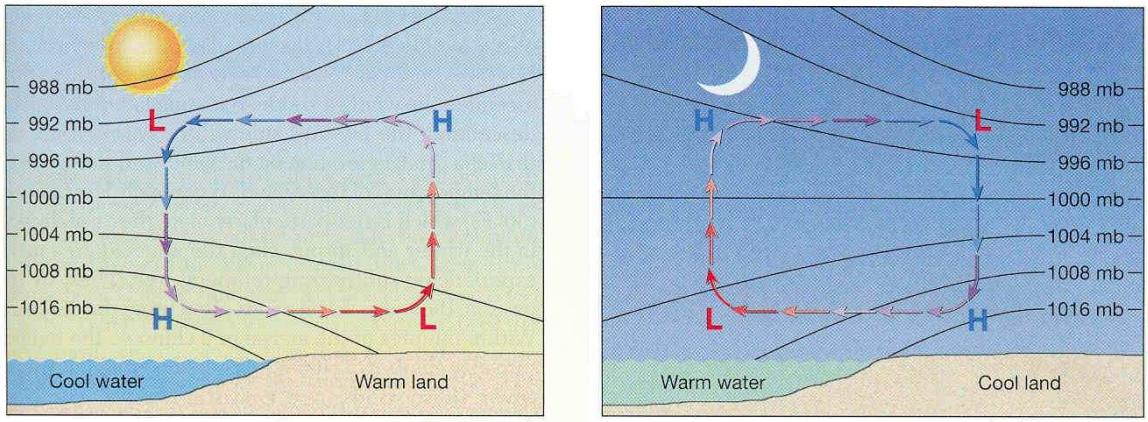
Meteorological processes that transport ozone and its precursors into and within the OTR can roughly be broken down into three levels: ground, mid and upper. The following sections discuss the three wind levels associated with meteorological transport processes in more detail.

2.3.2. Ground level winds

Land, sea, mountain, and valley breezes

In the OTR, land and sea breezes, and mountain and valley breezes can have an important influence on local air quality. These local winds are driven by a difference in temperature that produces a difference in pressure. Figure 2-3 shows a schematic of the formation of a sea breeze. The sea breeze forms in the afternoon when the land is considerably hotter than the ocean or bay, and the air flows inland from the ocean. At night, land cools more quickly than the ocean, and a land breeze blows out to sea. Because the nighttime land and water temperature differences are usually much smaller than in the day, the land breeze is generally weaker than the sea breeze. Sea breezes typically only penetrate a few kilometers inland because they are driven by temperature contrasts that disappear inland.

Figure 2-3. Illustration of a sea breeze and a land breeze



a) Sea Breeze **b) Land Breeze**

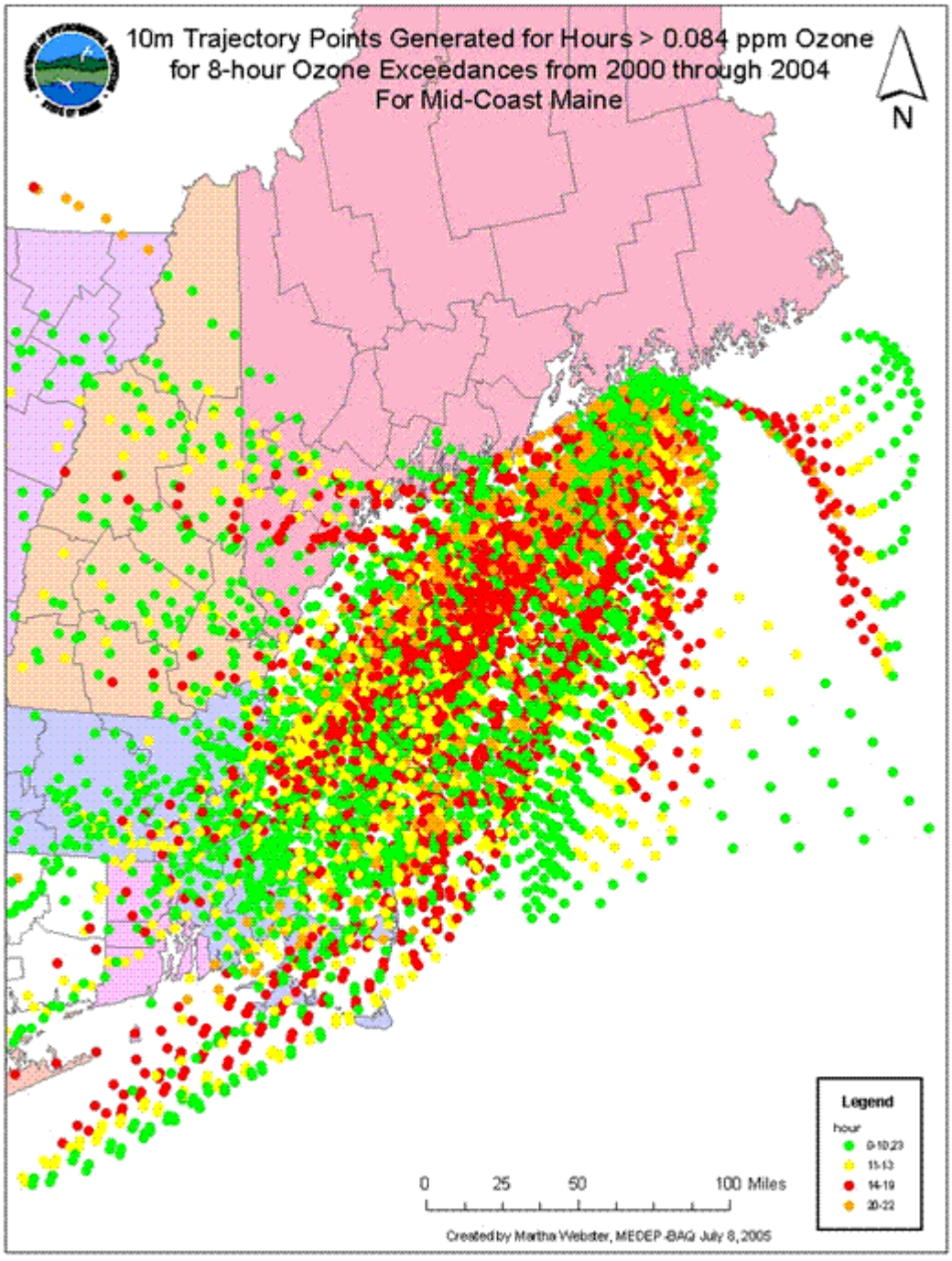
Figure from *Lutgens & Tarbuck*, 2001.

Along coastlines, such as coastal New England, sea breezes bring in air pollution transported near the surface over water from urban locations located to the southwest. Ray et al. (1996) have described high ozone levels transported over the Gulf of Maine and brought inland by sea breezes to coastal regions of Maine. Figure 2-4 shows 24-hour back trajectories of air parcels at a height of 10 m arriving at mid-coastal Maine during the 2000 – 2004 ozone seasons on days coastal ozone monitors recorded 8-hour concentrations exceeding 84 ppb. The bundle of trajectories demonstrates a clear propensity for maximum ozone levels to be transported across the Gulf of Maine before coming ashore with the sea breeze (ME DEP, 2010).

A similar situation occurs along the New Hampshire coast, where ozone events are driven by distinctively different wind directions than experienced inland. An analysis of wind directions at the time inland New Hampshire monitoring sites measured unhealthy ozone levels between 1995 and 2002 discerned an overwhelming trend of winds coming from the southwest. By contrast, high ozone levels at New Hampshire coastal sites were associated largely with a sea breeze from the southeast that brought in pollution transported over water from the Boston metropolitan area and possibly other urban areas in the Northeast Corridor (NH DES, 2010).

Sea breezes can also affect air quality in coastal cities because, under stagnant synoptic-scale winds, a city's emissions may be recirculated or pushed back over land after having drifted out over the sea before sea breeze circulation begins. In the absence of a shift in winds due to a sea breeze, the city's air pollution will be blown away. The sea breeze circulation acts like a barrier and pushes the polluted air back over the land. The sea breeze only pushes a few miles inland, which is where the barrier to mixing lies. Later in the day, the air may be quite clean on the ocean side of the city, but the air is usually quite dirty on the inland side. The city suffers from its own recirculated pollution as the sea breeze prevents pollution from the city to flow away from it. Appendix C presents more detailed information on sea breezes and flow over the ocean that contributes to ozone transport in parts of the OTR.

Figure 2-4. Average 2000 – 2002 wind direction frequency associated with elevated one-hour ozone levels in coastal Maine



The bay breeze is a shallow circulation over large inland bays, which usually extends only a couple hundred meters above the surface. For example, the northern Chesapeake Bay region suffers from the highest ozone concentrations in the state of Maryland, often due to the development of the bay breeze. The formation of this micro-

scale circulation has been known to create a sharp gradient of observed ozone concentrations across its boundary. A bay breeze event occurs when a non-onshore wind flow switches to onshore flow perpendicular to the shoreline. This circulation allows the build up of ozone along the bay breeze boundary. Landry *et al.* (2010) reviewed five years of Maryland data (2004–2009) and found that the bay breeze played a significant role in approximately one-third of all days exceeding the 8-hour ozone standard of 75 ppb. The ozone measured east of the bay breeze boundary ranged from +17 to +39 ppb higher than sites to the west.

Mountain and valley breezes are also driven by a temperature contrast. In the daytime, the side of the mountain will heat up more quickly than the valley, and hence a flow from the valley to the mountain results. At night this flow is reversed as the mountain side cools more quickly than the valley. As a result of these differences in cooling and heating, during the day, warm winds blow up toward the peaks from the valley below, while at night, cool air sinks and flows down the valley, settling in the lowest points. Local topography is very important in generating this phenomenon, making the breeze unique to a particular area.

Mountains and valleys also serve to isolate air in the valleys, while air at the mountaintops may be coming from very far away. Mountain winds, inversions, and mixing are quite complex. On a quiet night, the mountaintops may be in the free troposphere, open to long-range transport, while the valley below is usually capped by a nocturnal inversion, isolating pollution in the valley. Air quality measurements taken during plane flights in the Shenandoah River Valley have shown that the air pollutants in the valley may be rather different from the air at the nearby peaks. Cities on the western side of the mountains will find that the Appalachians are capable of damming pollution up against them (MARAMA, 2005 at pp. 42-43).

Appalachian lee side trough

The Appalachian lee side trough forms on the leeward (downwind) side of the Appalachian Mountains. In the OTR, a lee side trough forms when winds blow over the Appalachian Mountains and down the lee side of the mountain range to the coastal plain. As the column descends down the lee side, it stretches vertically and spins faster, pulling up air and creating low pressure, thus rotating the winds to the southwest. Because the air is typically rather dry, and the trough itself is rather weak, it does not usually lead to showers and thunderstorms the way a trough associated with other weather systems would. It does cause winds to shift their direction, so a wind that comes over the mountains from the west will turn and blow from the southwest along the coastal plain. Therefore, when surface winds on the coastal plain are from the southwest, if the Appalachian lee side trough is in place, it may be that the air actually came from the west, descended, and turned. The implication for air quality policy is straightforward. Pollution making its way over the mountains from the west will turn once it reaches the coastal plain and come from the southwest. Because surface winds are then from the southwest, when the Appalachian lee side trough is in place, the limits of a nonattainment area's airshed will be expanded farther south and west than they might otherwise be (MARAMA, 2005 at pp. 41-42). Studies have observed high ozone levels in the OTR

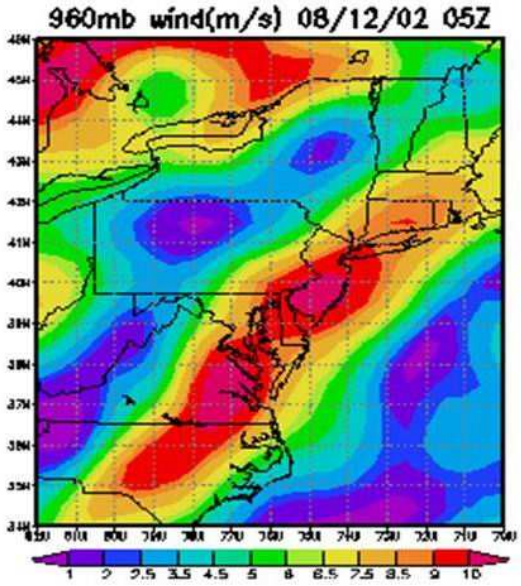
associated with a lee side trough east of the Appalachian Mountains and aligned with the Northeast Corridor (Gaza, 1998; Seaman & Michelson, 2000; Kleinman *et al*., 2004).

2.3.3. Nocturnal low level jets

The nocturnal low level[a] jet is a localized region of rapid winds in the lower atmosphere (typically 500-1500 m above the ground level) that can form at night under the same calm conditions often present in a pollution episode. Forming just above the nighttime temperature inversion, the nocturnal low level jet depends on isolation from the surface provided by the inversion. It is primarily a nocturnal phenomenon that occurs more frequently during the spring and summer seasons.

A nocturnal low level jet is generally found where a range of mountains meets a flat plain. There is a particularly strong nocturnal low level jet in the Great Plains of the central United States on the eastern side of the Rocky Mountains. On the Eastern Seaboard, a nocturnal low level jet can develop along the Atlantic Coastal Plain located to the east of the Appalachian Mountains and to the west of the Atlantic Ocean. Figure 2-5 shows an example of a nocturnal low level jet. While the typical wind speed minimum of a nocturnal low level jet is often defined as more than 12 meters per second (m s^{-1}), Ryan (2004) has proposed a weaker minimum speed criterion of 8 m s^{-1} in the East because of the expected weaker terrain-induced forcing in this region. The mid-Atlantic nocturnal low level jet has a width of 300-400 km (to its half peak value) and a length scale of more than 1500 km, following closely the orientation of the Appalachian Mountains (Zhang *et al*., 2006).

Figure 2-5. Example of a nocturnal low level jet



Note: The low level jet (shown by red, orange, and yellow) normally sets up along the eastern side of the Appalachian Mountains and blows from southwest to northeast (source: University of Maryland).

[a] "Low level" in this instance is relative to upper level jets occurring in the upper troposphere to lower stratosphere at heights of 10-15 km above the ground level. It is not a "ground level" phenomenon of the types described in the previous section.

The nocturnal low level jet forms in the absence of strong synoptic systems. Surface winds run parallel to the mountains, which in the case of the OTR is southwest running over the Atlantic Coastal Plain in front of the Appalachian Mountains. The nocturnal low level jet forms because land cools quicker than the air above it at night. The quickly cooling land results in the air closest to the surface cooling quicker than the air higher above. This creates a temperature inversion that separates the atmosphere into layers. The warmer air above the inversion layer (~200-800 m above ground) disengages from the frictional effect of the surface and increases in speed. In the eastern United States, the nocturnal low level jet has been observed in Georgia, the Carolinas and Virginia (Weisman, 1990; Sjostedt *et al*., 1990) in addition to the OTR (NARSTO, 2000). Appendix D provides a descriptive example of the evolution of a nocturnal low level jet occurring over the length of the OTR during a period of high ozone in July 2002.

The amount of ozone within the nocturnal low level jet can be measured by balloon-launched ozone sensors called "ozonesondes". During an event on July 12-13, 2008, Howard University launched two ozonesondes from Beltsville, MD, one as a nocturnal low level jet emerged at 10:30 p.m. local time and one later at 2:30 a.m. during peak winds. As the jet increased in peak winds, ozone levels increased within the jet from just above 80 ppb to almost 95 ppb. The jet traveled over 22 mph for above 14 hours, resulting in a total transport distance of more than 300 miles. (MDE, 2009).

In southern New Hampshire, indications of a nocturnal low level jet were seen and modeled during a July 2001 episode. The core of the nocturnal low level jet occurred at around 500 m and reached maximum wind speeds of greater than 10 m/s. On one occasion, the jet extended southward across the mid-Atlantic and southern states, and westward to mid-Pennsylvania and West Virginia. The jet transported ozone and precursors that helped to produce elevated daytime ozone levels in southern New Hampshire during this period (Mao & Talbot, 2004).

2.3.4. Ozone and precursors higher aloft

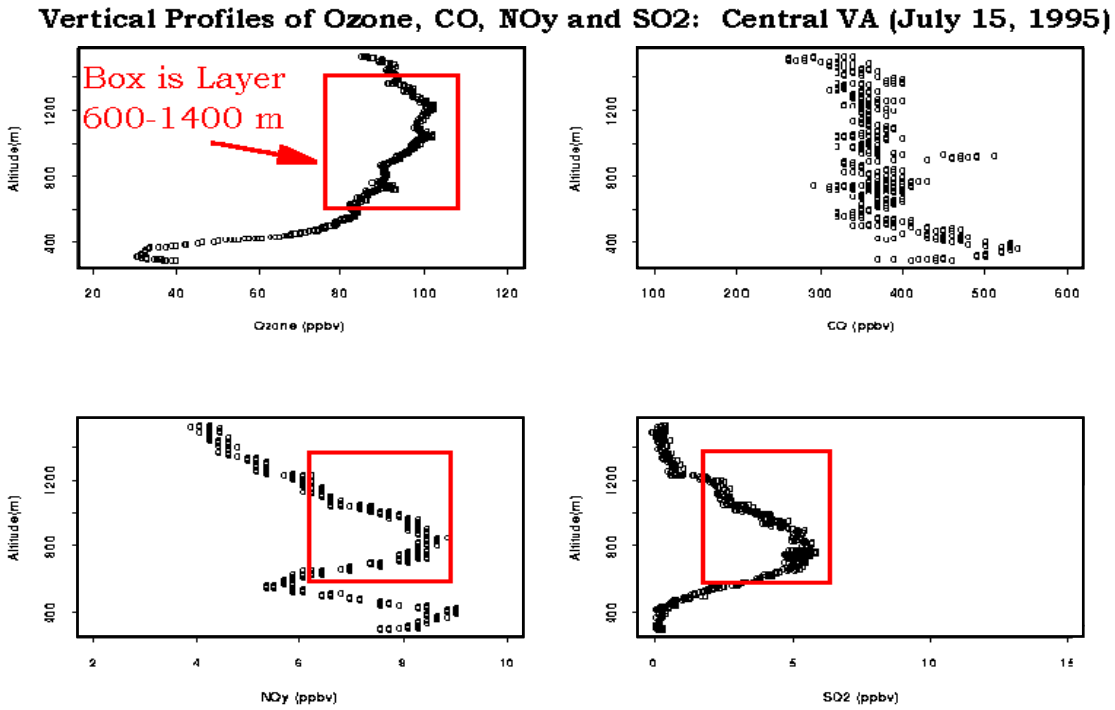
Theoretical and numerical model simulations have suggested for some time that there is a strong regional component to urban air quality in the northeastern United States (Liu *et al*., 1987; Sillman *et al*., 1990; McKeen *et al*., 1990). Since 1992, thousands of aircraft flights have been made to measure vertical profiles of ozone, the nitrogen oxides, carbon dioxide, sulfur dioxide, and aerosol particles during high ozone episodes.[b] Figure 2-6 shows the results of profiles taken over central Virginia on July 15, 1995, at about 9:00 am on the last day of a four-day severe ozone episode. During this episode, winds measured at Dulles Airport in Sterling, Virginia in the 500-3000 m layer, where ozone was at a maximum, were consistently from the west to the north. There were no periods of stagnation or reversal of wind direction during this period. Figure 2-6 shows that the ozone mixing ratio above the boundary layer is much larger than that at the ground, peaking at about 1200 meters.

[b] These measurements were made as part of the University of Maryland's RAMMPP (Regional Atmospheric Measurement, Modeling, and Prediction Program) under the sponsorship of ARMA, MARAMA (Mid-Atlantic Regional Air Management Association), VADEQ (Virginia Department of Environmental Quality), and NCDEQ (North Carolina Department of Environmental Quality).

An examination of the various pollutant data in Figure 2-6 helps to identify possible sources of the elevated ozone. It should be noted that while both automobiles and power plants emit NO_X, automobiles emit carbon monoxide (CO) but not sulfur dioxide (SO_2), while power plants emit SO_2 but not CO. The CO profile is not correlated well with the ozone data, indicating that the source of the ozone is not from local sources, i.e., automobiles. The peak in the NO_Y[c] profile at around 800 meters is an indication of "aged air" (hence transport) as a number of studies have found a strong relationship between increasing ozone and NO_Y in photochemically aged air masses (Trainer *et al*., 1993; Kleinman *et al*., 1994; Olszyna *et al*., 1994). Finally, the peak in the SO_2 profile, which occurs above the nocturnal inversion, is unlikely to come from local sources. Indeed the presence of the SO_2 leads to the conclusion that the air is coming from power plants west of the Appalachian Mountains.

[c] NO_Y = NO + NO_2 + all other oxidized nitrogen products of NO_X, excluding N_2O.

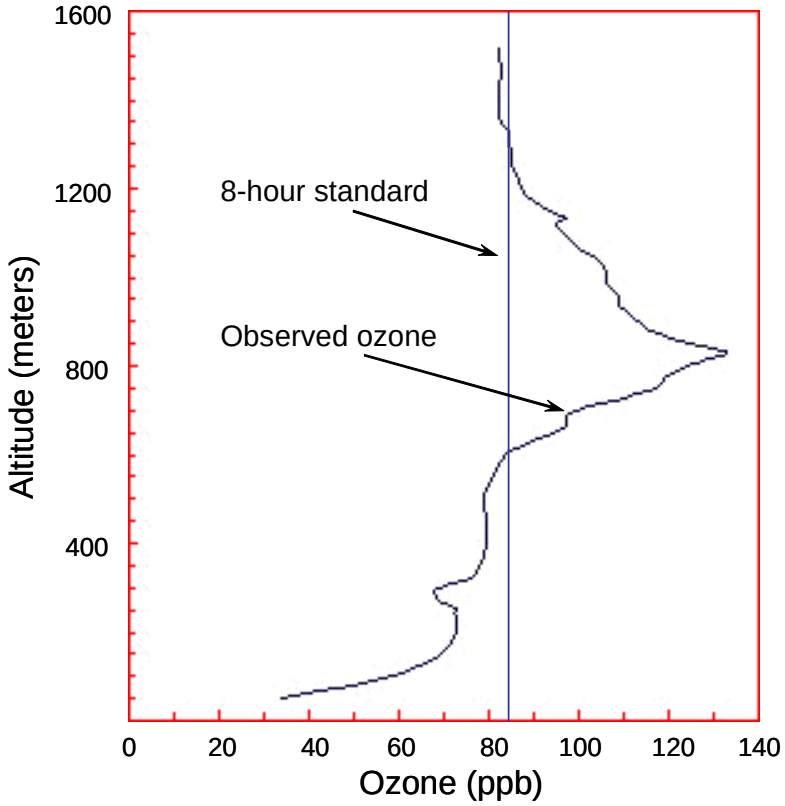
Figure 2-6. Altitude profiles for ozone, carbon monoxide, NO_Y, and SO_2 taken on July 15, 1995



During the same July 1995 period, measurements aloft in other parts of the OTR also recorded high ozone overnight in layers 500 m or higher above the surface. Ozone aloft concentrations above Poughkeepsie, NY and New Haven, CT approached levels of 120 ppb or greater on the night of July 14 (Zhang *et al*., 1998; Zhang & Rao, 1999). Figure 2-7 displays the aircraft measurements above Poughkeepsie, NY around 4 a.m. EST.

This aloft ozone at night has the potential of reaching the surface the next day as the mixing height rises in the morning due to surface warming. A study in central North Carolina obtained vertical measurements of ozone from a 610 m tall tower and found a strong correlation between nighttime and early morning ozone concentrations in the residual layer above the nocturnal boundary layer and the maximum ground level concentration the following afternoon (Aneja *et al*., 2000).

Figure 2-7. Observed vertical ozone profile measured above Poughkeepsie, NY at about 4 a.m. EST on July 14, 1995

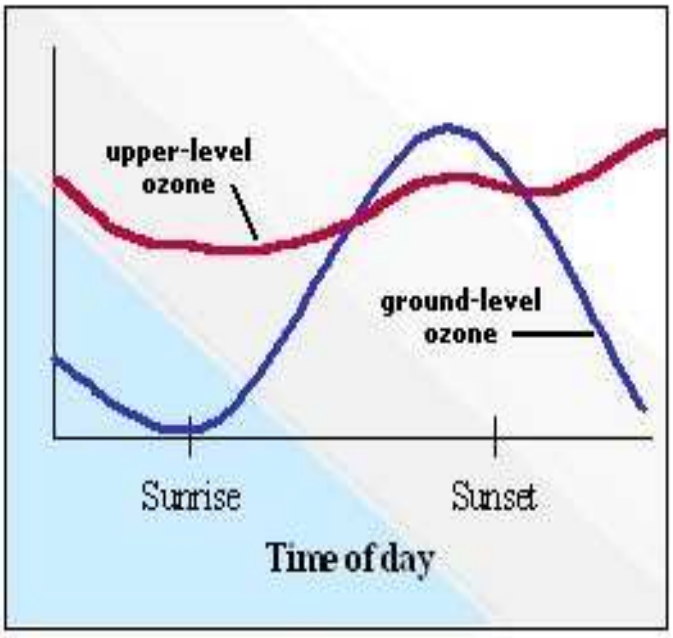


Note: The figure includes a vertical line at 85 ppb for comparing aloft measurements with the 8-hour ozone NAAQS (observed ozone data from Zhang & Rao, 1999).

The upper air measurements since 1992 reinforce the previously mentioned observations by Clarke and Ching (1983) during the summer of 1979, in which aircraft measurements recorded aloft ozone concentrations of about 90 ppb transported overnight from eastern Ohio and entering into the Northeast Corridor over a region stretching from the lower Hudson River Valley north of New York City down across eastern Pennsylvania and into Maryland just west of Baltimore. The measurements also showed NO_X aloft during the overnight hours that could contribute to additional ozone formation in the OTR as it mixed down to the surface in the morning. Further transport up the Northeast's urban corridor can occur via the formation of the Appalachian lee side trough and nocturnal low level jets (Taubman *et al*., 2004).

Absent transport, ground-level ozone concentrations typically increase during daylight hours as a result of photochemical production and decrease substantially at night when ozone removal exceeds production (see Figure 2-8). At higher elevations, however, concentrations of ozone and ozone precursors may remain high at night, as there is minimal downward mixing of the atmospheric transport layers at night. During daylight hours, when solar energy heats the ground, the resulting warm air near the ground begins to rise. Rising air creates an unstable atmospheric situation resulting in the up- and down-ward mixing of air masses (including ozone transport layers). Thus ground level ozone concentrations typically rise for several hours immediately after sunrise.

Figure 2-8. Typical day/night ozone cycle at ground level and aloft



Note: Solar ultraviolet energy helps to create ozone that is present in high quantities late in the day. At night, there is no ultraviolet sunlight to create ozone and ground level objects and gases act to remove ozone, resulting in the curve in blue, which represents ozone at ground level. Ozone at higher elevations, conversely, is often not depleted at night and may remain at elevated concentrations throughout the day (red curve).

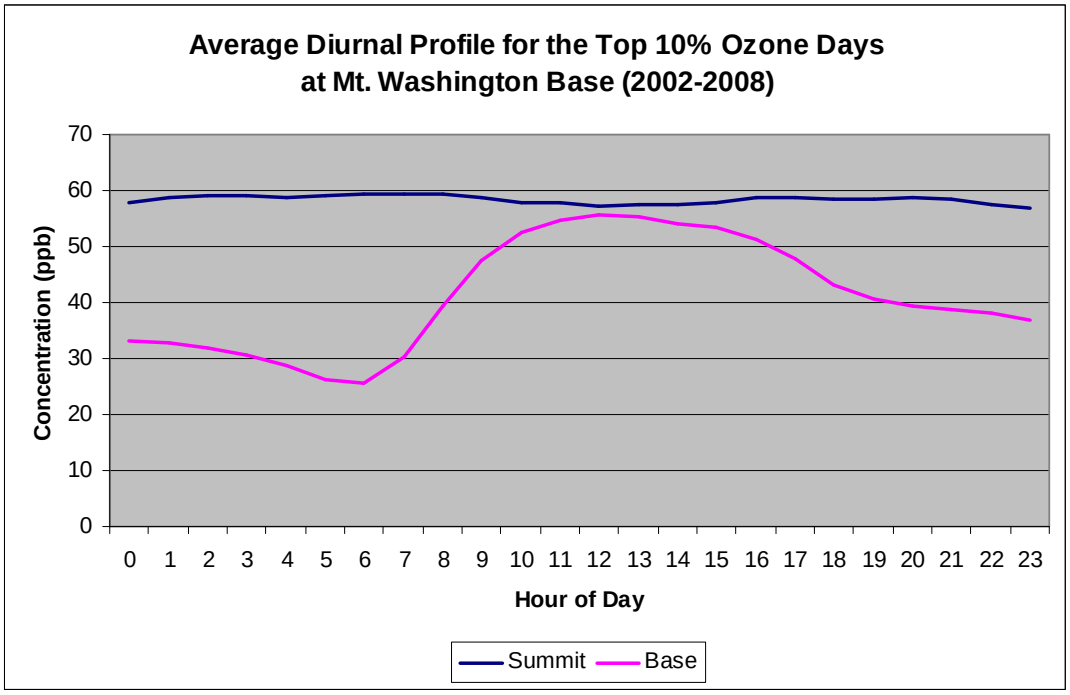
Monitoring data demonstrate this pattern. Towards the northern end of the OTR in New Hampshire stands Mt. Washington. The mountain is surrounded by hundreds of miles of forest and very few major emission sources within 75 miles. The mountain has ozone monitors at its 2,000 foot elevation base and at its 6,288 foot summit. When the wind patterns are right, significant amounts of ozone can be measured at both the base and summit. Because the summit of Mt. Washington is high enough to be exposed to high elevation transport (synoptic flows), downward mixing is not a factor in creating the peak ozone values it experiences. Transport time from upwind source areas appears to be the single largest factor in determining the time at which the maximum ozone level occurs at the summit. As suggested by Figure 2-9, the ozone at the base of the mountain is also dependent on this same long range transport that mixes downward from higher elevations once the mixing layer breaks up with the morning sun. While the highest ozone concentrations at the summit often occur overnight, there is actually a dip during the day when the mixing layer below breaks and allows vertical mixing. Relatively cleaner air from below mixes up and more polluted air from above mixes down, causing the summit measurements to dip and the base levels to rise (see Table 2-1).

Table 2-1. Night and daytime patterns of ozone at the base and summit of Mt. Washington, NH

Mt. Washington Monitor Location	Percent of Hourly Maxima During Daylight Hours (9 a.m. to 5 p.m.)	Percent of Hourly Maxima During Overnight Hours (6 p.m. to 8 a.m.)
Summit	18%	82%
Base (Camp Dodge)	80%	20%

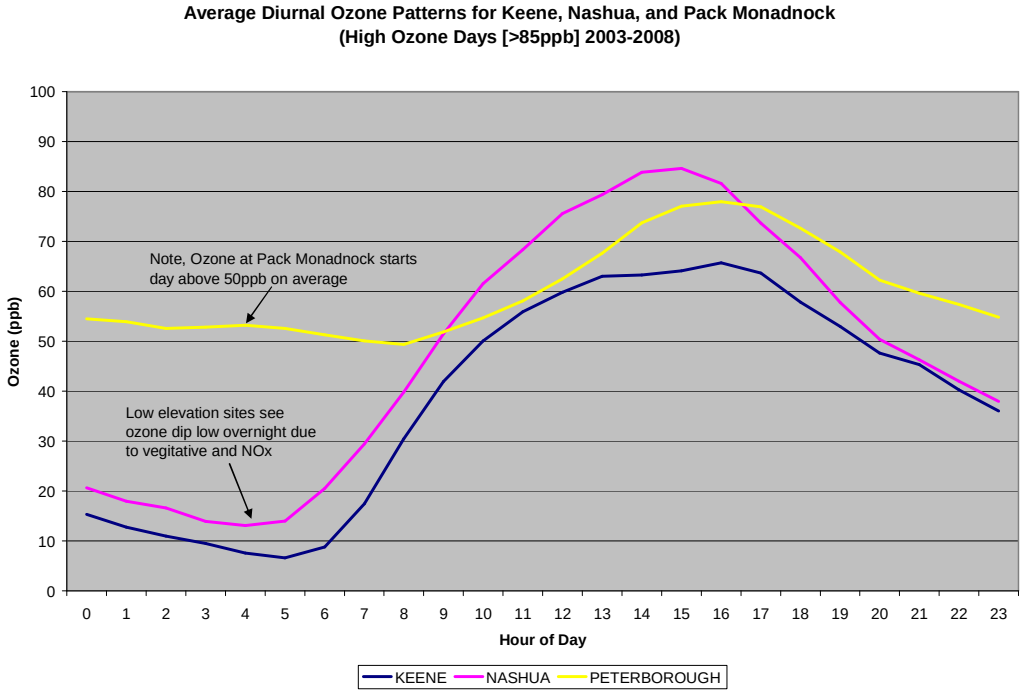
Note: Daylight heating hours are hours of the day when solar energy drives vertical mixing of transport layers

Figure 2-9. Average diurnal ozone patterns at the summit and base of Mt Washington, NH during high ozone days



A similar pattern can be found farther south in New Hampshire. Figure 2-10 compares ozone profiles from a mountaintop monitor located at Pack Monadnock Mountain (2,288 feet) with nearby low elevation monitors in Keene and Nashua, NH. In this case, the mountain site is not as far removed from emission sources. Some of the observed ozone is the result of local emissions and some comes from ozone and precursors brought into the area from synoptic scale transport patterns and the East Coast low level jet.

Figure 2-10. Average diurnal ozone patterns at the summit and near the base of Pack Monadnock Mountain, NH during high ozone days

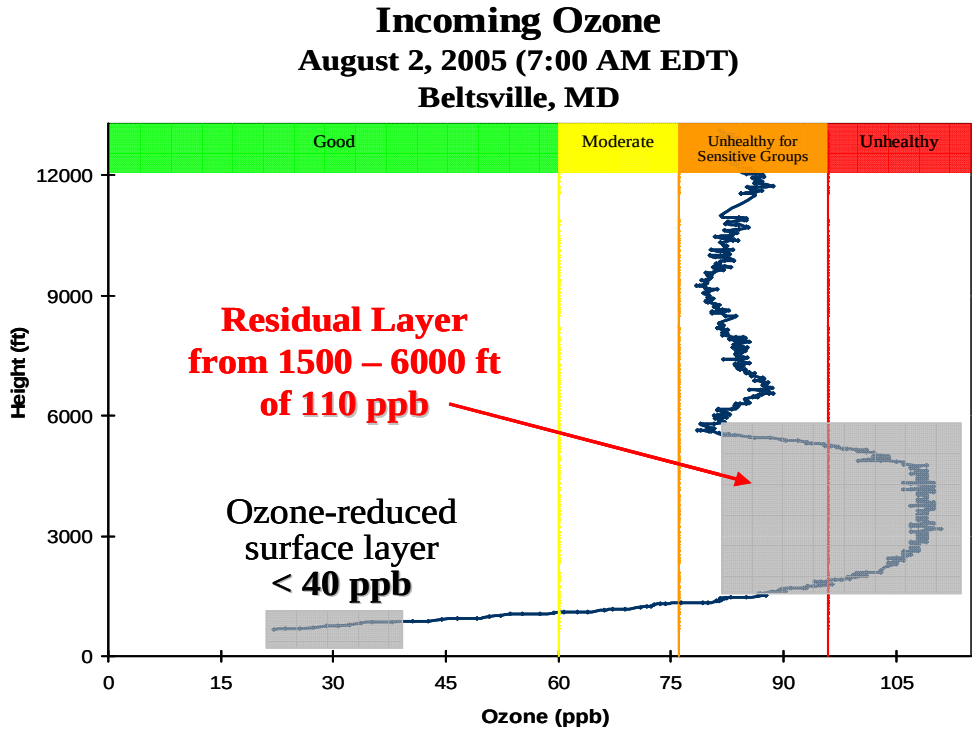


2.4. The "ozone reservoir"

The presence of high levels of ozone and precursors aloft across a large spatial region gives rise to the concept of an "ozone reservoir" forming in the eastern United States. The pollutants in this reservoir are not subject to destruction at the surface, and can be transported long distances in the wind flows created by the synoptic scale weather patterns conducive to ozone formation and transport. Transport up the Northeast's urban corridor can subsequently occur augmented by the Appalachian lee side trough and nocturnal low level jet (Taubman *et al.*, 2004).

In the ozone reservoir aloft, concentrations of ozone between 1,500 and 6,000 feet routinely can reach levels approaching and exceeding 100 ppb, while ozone at the surface can be below 40 ppb (Figure 2-11). This ozone air mass can be quickly transported long distances by strong winds like the jet stream. This effectively disconnects the location of emission sources that produce ozone from the location where ground-level concentrations of ozone are the highest.

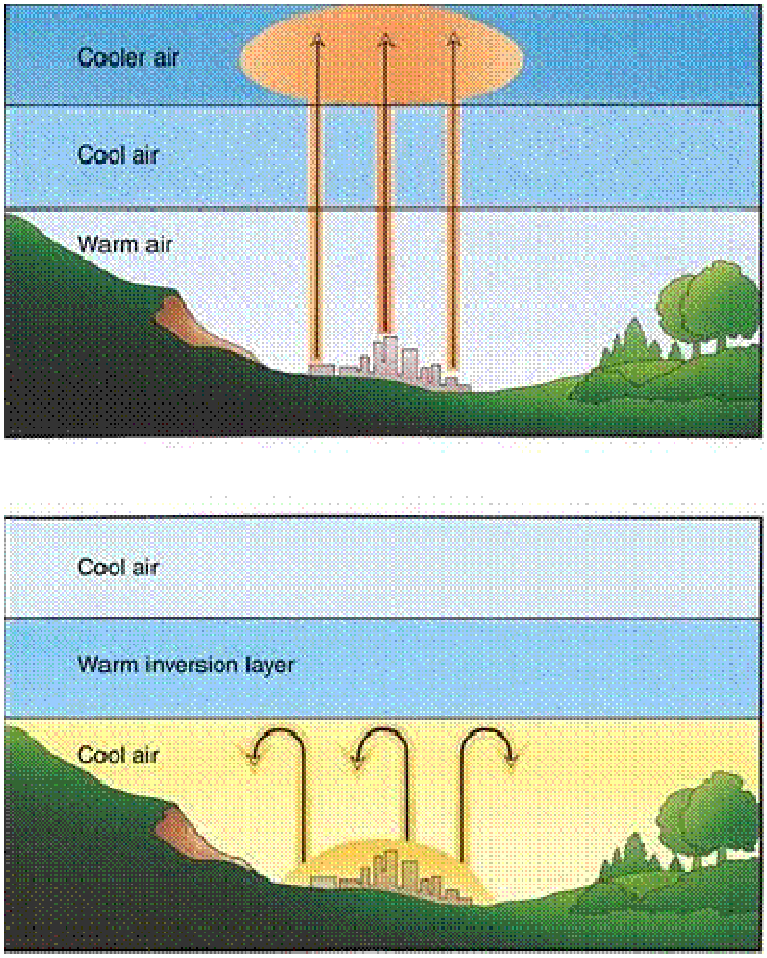
Figure 2-11. Concentration of ozone by altitude based on ozonesonde measurements



Source: Maryland Department of the Environment & Howard University

A simple conceptual picture of how the ozone reservoir forms is as follows: ozone produced during the day by sunlight acting on NO_X and VOCs is lifted high in the atmosphere by convection, mixing it within a relatively thick atmospheric boundary layer. At night, the boundary layer created by surface cooling after the sun goes down is closer to the ground than typical daytime boundary layers (Figure 2-12). As it forms, it leaves ozone from the daytime boundary layer in a residual layer aloft.

Figure 2-12. Mixing of ground-level ozone aloft during the day (above) and prevention of aloft ozone mixing down due to a nighttime inversion (below)



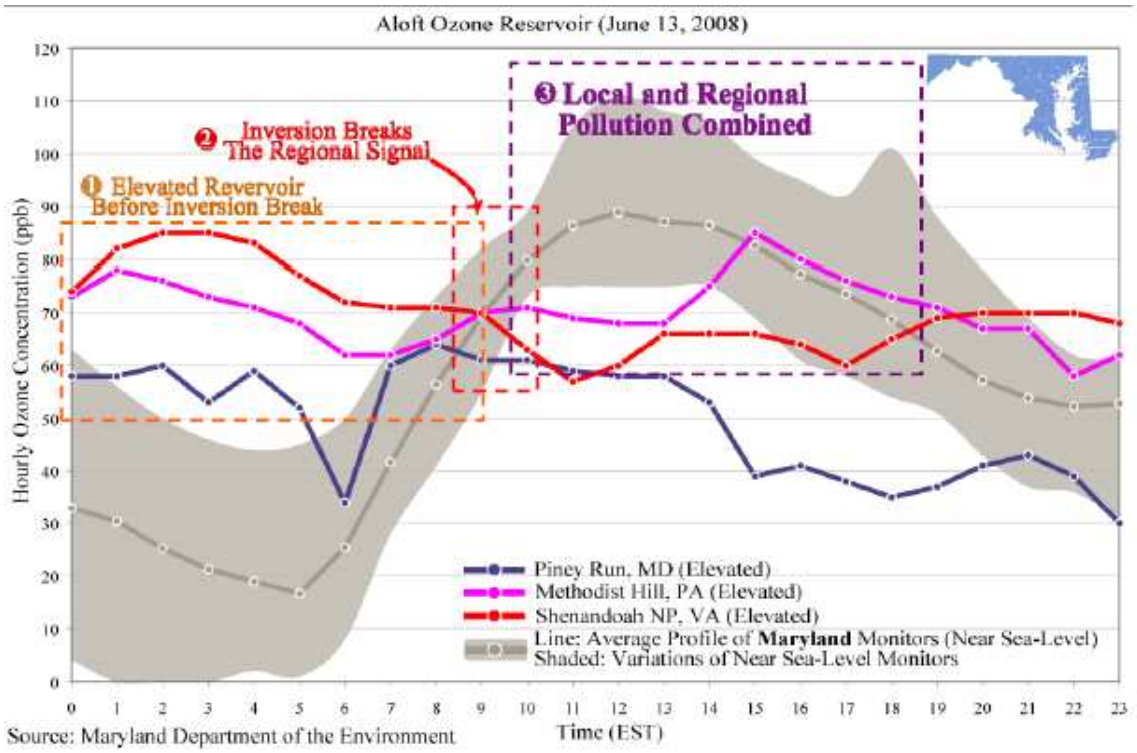
Source: Science Education Resource Center, Carleton College, MN.

In the morning, ozone within the stable and shallow nocturnal boundary layer is very low due to ozone loss mechanisms like dry deposition and NO_X titration. However the ozone cutoff from the ground in the residual layer is not destroyed and can travel for hundreds of miles in the strong winds of the free troposphere. As the nocturnal boundary layer lifts due to the sun's warming of the ground, the elevated ozone and its precursors mix down to the surface where they contribute to surface ozone concentrations.

The concentration level of this ozone reservoir can be measured by looking at the differences between elevated and ground-level ozone monitors over time. As described previously for Mt. Washington, NH, high elevation monitors are often above the top of the nocturnal boundary layer and directly measure aloft ozone concentrations. As the day progresses, mid-elevation monitors show the start of the ozone reservoir mixing down. The nocturnal inversion breaks up between 8-10 a.m. and ground-level monitors measure high levels of ozone comparable to the concentration at high elevation monitors. By

midday, the regional and local air pollutants have mixed and are indistinguishable (see Figure 2-13).

Figure 2-13. Comparison of the diurnal ozone concentrations measured by high elevation monitors (red, pink, and blue) and ground-level monitors (gray) on June 13, 2008



This pattern of "up..over..down" ozone transport via the ozone reservoir has been repeatedly observed across the OTR, as previously described in the published results of numerous field and modeling studies. A major trend over time, however, has been the magnitude of ozone concentrations within the elevated reservoir. While concentrations up to 180 ppb were common in the 1990s, current peaks are closer to 100 ppb as a result of regional NO_X reductions from power plants, motor vehicles, and other sources. Despite this reduction in transported ozone, levels are still well above the upper end of the range for the proposed revision for the ozone NAAQS (60 – 70 ppb).

2.5. Ozone background and exceptional events

With an ozone NAAQS of 0.08 ppm and the likely revision to a more stringent level in the range of 0.060 – 0.070 ppm as proposed by EPA, concerns have been raised over the ability to attain the current or a future ozone NAAQS due to ozone contributions thought to be beyond the control of air quality planners. To address these concerns, Congress amended section 319 of the Clean Air Act when it passed the Safe Accountable Flexible Efficient-Transportation Equity Act: A Legacy for Users (SAFE–TEA–LU) of 2005. Congress added to CAA section 319 language containing criteria to be applied by the USEPA for determining when air quality conditions could be attributed to "exceptional events," thus not counting as an exceedance or violation of a NAAQS. The

concept of "exceptional events" is to identify instances of degraded air quality for which the normal CAA planning and regulatory processes are not appropriate.

The amended CAA section 319 defines an exceptional event as an event that affects air quality; is an event that is not reasonably controllable or preventable; is an event caused by human activity that is unlikely to recur at a particular location or a natural event; and is determined by EPA to be an exceptional event. The statutory definition of exceptional event specifically excludes stagnation of air masses or meteorological inversions; a meteorological event involving high temperatures or lack of precipitation; or air pollution relating to source noncompliance. The USEPA has established by rule what is required by states to demonstrate the occurrence of exceptional events, thus allowing the exclusion of air quality monitoring data that would otherwise show an exceedance or violation of a NAAQS (72 Fed. Reg. 13560 (March 22, 2007)). Examples of exceptional events for ozone include stratospheric intrusions[d] and ozone generated from large forest fires.

At the time the USEPA lowered the ozone NAAQS from a 1-hour level of 0.12 ppm to an 8-hour average of 0.08 ppm, concern was expressed that the new lower level would be difficult to achieve due to frequently occurring naturally elevated hourly ozone levels of ≥0.05 and ≥0.06 ppm during winter and spring. The elevated levels were attributed to air from the upper troposphere and stratosphere being transported to the surface. Most elevated ozone occurrences were during April and May, based on air monitoring at rural sites in the northern U.S., southern Canada, and northern Europe where anthropogenic local sources of air pollution as well as photochemistry were believed to have little influence on ozone levels during these times (Lefohn *et al*., 2001). It follows that if stratospheric ozone is a relatively frequent cause of or significant contributor to exceedances of the 0.08 ppm ozone NAAQS, it will also be of concern in addressing a future ozone NAAQS set at an even lower level.

A modeling study by Fiore *et al*. (2003) has concluded that much of the observed ozone variability, including high springtime ozone events, in relatively remote areas of the U.S. are attributable to anthropogenic air pollution rather than being of predominate stratospheric origin. The researchers pointed out that one cannot assume regional photochemistry during the spring is inactive, as springtime UV radiation levels are comparable to the summer. Because the modeling was able to reproduce the ozone behavior seen at remote monitoring sites, Fiore *et al*. (2003) proposed that previous interpretations attributing a greater stratospheric component were underestimating the role of regional and hemispheric pollution. The results indicated that the natural ozone background is typically in the range of 10-25 ppb, and never exceeds 40 ppb. The contribution of hemispheric pollution above the natural background ranges from 4-12 ppb and only rarely exceeds 20 ppb. The stratospheric contribution is minor, always below 20 ppb. This is consistent with earlier findings that stratospheric ozone contributions are highest in the upper troposphere while contributions to ozone at the surface are typically small (Parrish *et al*., 1993; Dibb *et al*., 1994; Mauzerall *et al*., 1996).

[d] A stratospheric intrusion is the entrainment of ozone-rich air from the stratosphere into the troposphere, which is the lowest layer of the atmosphere at the earth's surface. A stratospheric intrusion is typically associated with the passage of strong frontal systems or severe thunderstorms, and may occur primarily in the spring (USEPA, 1986).

Even during the winter it cannot be assumed that high ozone levels are necessarily stratospheric in origin. Ozone levels at a site in rural Wyoming during February, when the ground was snow-covered and air temperatures were as low as -17 C, reached a hourly peak average of 140 ppb just after solar noon (122 ppb 8-hour average) (Schnell *et al*., 2009). A stratospheric intrusion was discounted due the diurnal ozone behavior, the presence of a high pressure system preventing stratospheric air from mixing downward, and an ozonesonde profile indicating the highest ozone was contained in a shallow surface layer with no enhancement at higher altitudes. It was suggested that a more likely source for the ozone was photochemical production from precursors associated with a developed natural gas field in the area.

In general, stratospheric intrusions leading to high surface ozone appear to be relatively rare (Logan, 1989; Cooper *et al*., 1998; Fiore *et al*., 2003). At Mt. Washington, NH, the highest point in the OTR, observed ozone levels greater than 80 ppb at night were associated with westerly (71 percent) and southwesterly (29 percent) transport, implicating pollution transport from industrialized/urban areas of the Midwest and the East Coast. Stratospheric ozone appeared to contribute to only about 5 percent of the extreme ozone events, but occurred under conditions also conducive to photochemical ozone production, making the relative contributions difficult to discern (Fischer *et al*., 2004). The use of wind profiler radars suggested a stratospheric contribution to ground level ozone in Montreal, QC, during an event in early May when nighttime ozone increased from a typical level of 15 ppb to 30 ppb. Typical daytime levels of 30 ppb increased to 50 ppb (Hocking *et al*., 2007). The increase in daytime ozone of 20 ppb is consistent with the modeling results of Fiore *et al*. (2003).

In addition to stratospheric ozone intrusions, another potential type of "exceptional event" is ozone associated with long range transport of smoke from forest fires. These would qualify as "exceptional events" to the extent they are considered by the USEPA as meeting the criteria set out in CAA section 319. Examples of observed elevated surface ozone concentrations attributed to long range transport from wildfires include impacts in the Washington, DC area from forest fires in central Quebec (Colarco *et al*., 2004), and in Houston, TX from forest fires in eastern Alaska and western Canada (Morris *et al*., 2006).

2.6. Summary

This section has summarized current knowledge of the meteorological processes that affect local ozone levels within the OTR. A conceptual description of transport processes within the OTR can be divided into three principle components: surface winds, the nocturnal low level jet, and winds in the free troposphere. All three modes of transport depend on the location of the high pressure system. Surface winds are the result of interaction between the synoptic flow and local effects, such as the sea breeze and the Appalachian lee side trough. The nocturnal low level jet results from the combination of shallow nocturnal inversions and conducive terrain. It has been observed throughout the Eastern Seaboard from Georgia to Maine. The nocturnal low level jet can transport ozone that formed within the OTR or was transported into the OTR from outside the region. Transport aloft is dominated by the anti-cyclonic flow around a high pressure system, which can lead to transport of an ozone reservoir into the OTR created by emissions in

areas that lie outside the OTR. Local emissions within the OTR add to the polluted air mixing down from above that arrived from more distant locations.

Atmospheric modeling by the USEPA underscores the observations that the OTR's ozone problem has contributions from outside and upwind of the region. Pollution sources in the Ohio River Valley and the Southeast significantly contribute to ozone nonattainment problems in various portions of the OTR.

Stratospheric sources of ozone appear to make relatively little contribution to surface ozone concentrations relevant to exceedances of the ozone NAAQS. Long distance transport from large wildfires can have an impact on local air quality and can be considered "exceptional events" for purposes of air quality planning.

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3. OZONE-FORMING POLLUTANT EMISSIONS

The pollutants that affect ozone formation are volatile organic compounds (VOCs) and nitrogen oxides (NO_X). The emissions dataset presented for the OTR in the first section below is from the 2002 MANE-VU (Mid-Atlantic/Northeast Visibility Union) Version 2 regional haze emissions inventory. MANE-VU is the regional planning organization (RPO) for the mid-Atlantic and Northeast states coordinating regional haze planning activities for the region. While the context of the MANE-VU inventory is regional haze, it includes inventories of NO_X and VOCs that also inform air quality planners on sources important to ozone formation.[e] To provide a fuller context of precursor emissions contributing to regional ozone affecting the OTR, the section following the MANE-VU information presents NO_X and VOC emissions information from the 2002 National Emissions Inventory (NEI) for states in adjacent RPOs.

Note that in future inventory efforts, the USEPA's Motor Vehicle Emission Simulator (MOVES) model will be replacing the previous mobile source inventory models – MOBILE for on-road and NONROAD for non-road emissions inventory development. On average, this change is anticipated to: (1) lower modeled CO and VOC emissions; (2) increase modeled NO_X and PM emissions; (3) lower the percent NO_X reduction from modeled control measures; and (4) raise the percent PM reduction from modeled control measures for the on-road and non-road sectors (Dolce, 2009).

3.1. Emissions inventory characteristics in the OTR

3.1.1. Volatile organic compounds (VOCs)

Existing emission inventories generally refer to VOCs as hydrocarbons whose volatility in the atmosphere makes them particularly important in enhancing ozone formation in the presence of NO_X.

As shown in Figure 3-1, the VOC inventory for the OTR is dominated by mobile and area sources. Most VOC emissions in the OTR, however, come from natural sources, which are not shown in the figure. Among the human-caused VOC emissions, on-road mobile sources of VOCs include exhaust emissions from gasoline passenger vehicles and diesel-powered heavy-duty vehicles as well as evaporative emissions from transportation fuels. VOC emissions may also originate from a variety of area sources (including solvents, architectural coatings, and dry cleaners) as well as from some point sources (e.g., industrial facilities and petroleum refineries).

Naturally occurring (biogenic) VOC emissions are caused by the release of natural organic compounds from plants in warm weather. Many natural VOCs that contribute to ozone formation are highly reactive. Isoprene, for example, is a highly reactive five-carbon natural VOC emitted from mostly deciduous trees (e.g., oaks) that plays an important role in enhancing regional ozone formation across the eastern U.S.

[e] The description of OTR state inventories discussed in the first section does not include the portion of Virginia in the Washington, DC metropolitan area. Information for Virginia is in the following section and comes from the 2002 National Emissions Inventory.

(Trainer *et al.*, 1987; Chameides *et al.*, 1988). Because biogenic VOC emissions are large and reactive, they are the most important part of the VOC inventory for understanding and predicting ozone formation. Biogenic VOCs are not included in Figure 3-1, but nationally, they represent roughly two-thirds of all annual VOC emissions (USEPA, 2006a). Modeling biogenic emissions can be difficult as it requires simulating biological responses to a range of environmental conditions, such as leaf temperature and the amount of sunlight reaching a leaf surface.

Figure 3-1. 2002 MANE-VU state VOC inventories in the OTR

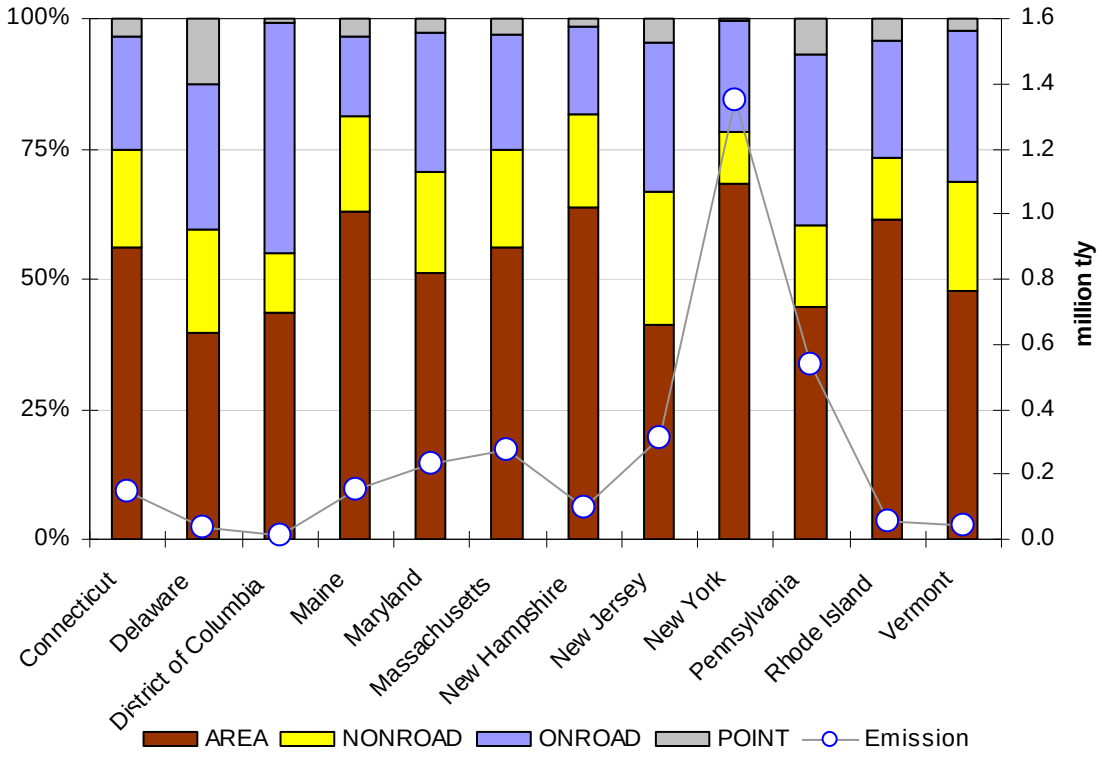


Figure key: Bars = Percentage fractions of four source categories; Circles = Annual emissions amount in 10^6 tons per year. The Virginia portion of the Washington, DC metropolitan area is not shown in the figure.

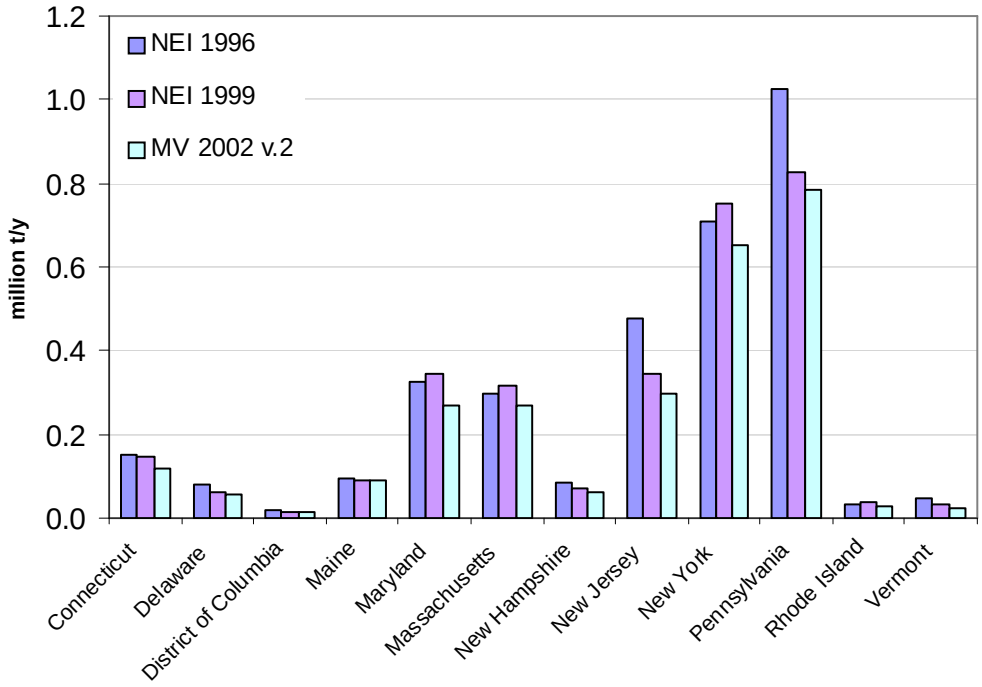
3.1.2. Oxides of nitrogen (NO_X)

NO_X emissions are a fundamental necessity for the atmospheric formation of ozone. Without NO_X, ozone formation during warm summer days would virtually cease, regardless of the amount of reactive VOCs present. By contrast, without VOCs, NO_X would still produce ozone in the presence of sunlight, albeit at a much diminished efficiency.

Figure 3-2 shows NO_X emissions in the OTR at the state level. Since 1980, nationwide emissions of NO_X from all sources have shown little change. In fact, emissions increased by 2 percent between 1989 and 1998 (USEPA, 2000). This increase is most likely due to industrial sources and the transportation sector, as power plant

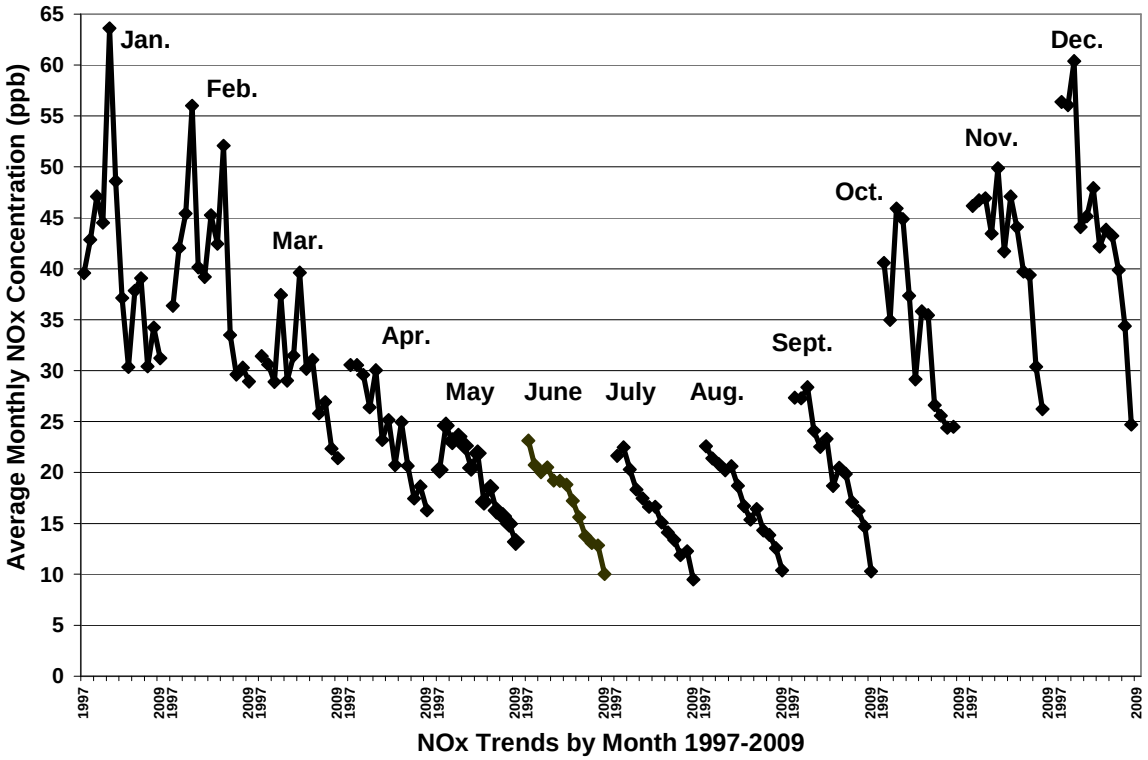
combustion sources have implemented modest emissions reductions during the same time period. Most states in the OTR experienced declining NO_X emissions from 1996 through 2002, except Massachusetts, Maryland, New York, and Rhode Island, which show an increase in NO_X emissions in 1999 before declining to levels below 1996 emissions in 2002.

Figure 3-2. State level nitrogen oxides emissions



Monitored ambient NO_X trends from 1997 to 2009 corroborate and extend the downward trend in NO_X emissions seen in the emissions inventories for the OTR. As shown in Figure 3-3, the monthly averaged NO_X concentrations indicate decreases in NO_X over this time period in the OTR. The NO_X reductions likely come from decreasing vehicle NO_X emissions due to more stringent motor vehicle standards as well as NO_X reductions from the OTR NO_X Budget Program and the NO_X SIP Call (mainly power plants). The higher NO_X levels measured during the colder months may be for several reasons, including relatively lower atmospheric mixing heights during colder months (i.e., less volume for pollutants to disperse in), less stringent NO_X requirements for power plants during months outside the April – September OTR ozone season, and increased space heating demands (e.g., NO_X from combustion of residential heating oil and wood, as well as increased generation for electric heat).

Figure 3-3. Plot of average monthly monitored NO_X trends in OTR 1997-2009



Note: The NO_X trends are 24-hour levels averaged by month during 1997-2009 (figure courtesy of Tom Downs, Maine Department of Environmental Protection).

Power plants and mobile sources generally dominate state and national NO_X emissions inventories. Nationally, power plants account for more than one-quarter of all NO_X emissions, amounting to over six million tons. The electric sector plays an even larger role, however, in parts of the industrial Midwest where high NO_X emissions have a particularly significant power plant contribution. By contrast, mobile sources dominate the NO_X inventories for more urbanized mid-Atlantic and New England states to a far greater extent, as shown in Figure 3-4. In these states, on-road mobile sources – a category that mainly includes highway vehicles – represent the most significant NO_X source category. Emissions from non-road (i.e., off-highway) mobile sources, primarily diesel-fired engines, also represent a substantial fraction of the inventory.

Figure 3-4. 2002 MANE-VU state NO_X inventories in the OTR

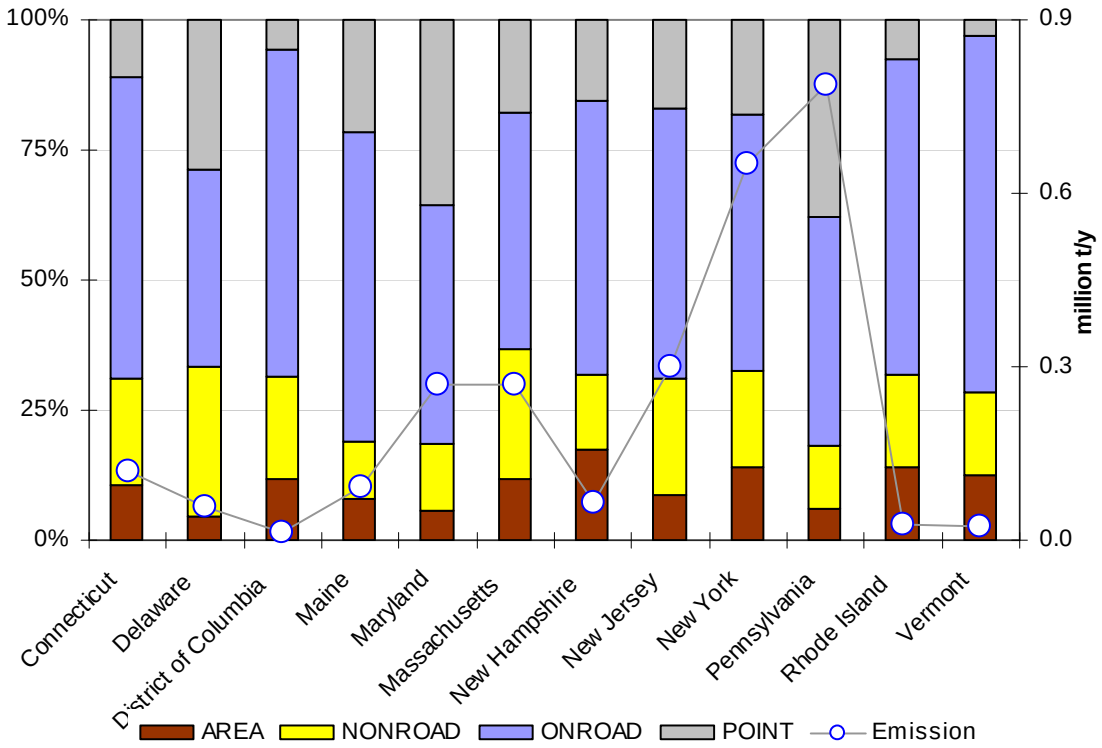


Figure key: Bars = Percentage fractions of four source categories; Circles = Annual emissions amount in 10^6 tons per year. The Virginia portion of the Washington, DC metropolitan area is not shown in the figure.

3.2. Emissions inventory characteristics outside the OTR

NO_X and VOC emissions in the OTR are only one component of the emissions contributing to ozone affecting the OTR. As regional modeling for the NO_X SIP Call and CAIR have shown, emission sources, primarily of NO_X, located outside the OTR can significantly contribute to ozone transported into the OTR. Here we present regional emissions information grouped by the three eastern RPOs – MANE-VU, VISTAS (Visibility Improvement State and Tribal Association of the Southeast), and the MWRPO (Midwest RPO). Table 3-1 lists the states in each RPO.

The inventory information is extracted from the USEPA final 2002 National Emissions Inventory (NEI). For consistency, the MANE-VU information here also comes from the 2002 NEI rather than from the MANE-VU Version 2 regional haze emissions inventory described above. The differences between the inventories are not great, as the NEI and the MANE-VU Version 2 inventory are both based on the same inventory information provided by the states.

Table 3-1. Eastern U.S. RPOs and their state members

RPO	State
MWRPO	Illinois
MWRPO	Indiana
MWRPO	Michigan
MWRPO	Ohio
MWRPO	Wisconsin
MANE-VU	Connecticut
MANE-VU	Delaware
MANE-VU	District of Columbia
MANE-VU	Maine
MANE-VU	Maryland
MANE-VU	Massachusetts
MANE-VU	New Hampshire
MANE-VU	New Jersey
MANE-VU	New York
MANE-VU	Pennsylvania
MANE-VU	Rhode Island
MANE-VU	Vermont
VISTAS	Alabama
VISTAS	Florida
VISTAS	Georgia
VISTAS	Kentucky
VISTAS	Mississippi
VISTAS	North Carolina
VISTAS	South Carolina
VISTAS	Tennessee
VISTAS	Virginia
VISTAS	West Virginia

Table 3-2 presents VOC emissions by source sector and RPO for the eastern United States. The NO_X emissions by source sector and RPO are presented in Table 3-3. Regionally, NO_X emissions are more important with respect to regional ozone formation and transport. Emissions of NO_X in combination with abundant naturally occurring VOC emissions from oaks and other vegetation have been shown to be important sources of regional ozone in the eastern U.S. (Trainer *et al*., 1987; Chameides *et al*., 1988).

Table 3-2. VOC emissions in eastern RPOs

RPO	Point	Area	On-road	Non-road	Total
MWRPO	234,938	1,182,186	660,010	492,027	2,569,160
MANE-VU	93,691	1,798,158	793,541	494,115	3,179,504
VISTAS	458,740	2,047,359	1,314,979	609,539	4,430,617

Table 3-3. NO_X emissions in eastern RPOs

RPO	Point	Area	On-road	Non-road	Total
MWRPO	1,437,284	184,790	1,290,178	723,844	3,636,096
MANE-VU	680,975	268,997	1,297,357	534,454	2,781,783
VISTAS	2,094,228	266,848	2,160,601	812,615	5,334,293

3.3. Are NO_X or VOC control strategies most effective at reducing ozone?

The effectiveness of a NO_X-focused or VOC-focused control strategy to reduce ozone is not constant by location or emissions; rather it is a changing chemical characteristic of an air parcel affecting a particular location. As a result, the effectiveness of a NO_X or VOC-focused control strategy can vary within an air parcel as it dynamically evolves over time with transport, dispersion, and photochemical aging (NARSTO, 2000 at Chapter 3.2).

On a regional basis, OTAG, CAIR and other modeling studies have consistently shown that NO_X reductions have the greatest impact on regional daily ozone concentrations, while VOC reductions have more local impacts. This is largely a result of significant naturally occurring VOC emissions (especially isoprene) in large forested regions of the eastern U.S. Observed results from regional NO_X reductions at power plants (i.e., the NO_X SIP Call) are now indicating that significant ozone reductions are occurring on a regional basis as a result of regional NO_X strategies. A USEPA report finds a strong association between areas with the greatest NO_X emission reductions due to the NO_X SIP Call and downwind sites exhibiting the greatest improvement in ozone in 2005 (USEPA, 2006b). The EPA findings are supported by a number of studies appearing in the peer-reviewed science literature. Collectively, these studies have found a strong association between the power plant NO_X reductions of the NO_X SIP Call and lower transported ozone levels in the eastern U.S. (Frost *et al*., 2006; Kim *et al*., 2006; Gégo *et al*., 2007 & 2008; Gilliland *et al*., 2008; Godowitch *et al*., 2008a & 2008b; Hudman *et al*., 2009). Of special note were the findings, based on back trajectory analyses, that ozone reductions in downwind regions east and northeast of the Ohio River Valley could be tied to NO_X reductions at power plants in the Ohio River Valley (Gégo *et al*., 2007; Godowitch *et al*., 2008b). A study based on ozone observations and modeling also suggested that air quality models underestimated the decrease in the long-range transport of ozone and its precursors after implementation of the NO_X SIP Call (Gilliland *et al*., 2008).

As a general rule, VOC reductions may be effective at reducing urban-scale ozone pollution in lieu of or in combination with local NO_X reductions, while regional NO_X controls are most effective at reducing regional ozone. While a general rule can be outlined in evaluating the potential effectiveness of NO_X and VOC-focused control strategies, the optimal strategy for a specific location will depend on the particular circumstances of that location. Exceptions to a VOC-only strategy for an urban area can occur when the urban area has large natural VOC emissions, ozone is transported from

upwind, or there is recirculation of aged local pollution (e.g., sea breeze effect). Furthermore, because the conditions causing individual ozone episodes can vary, a given urban area may change in sensitivity between a NO_X and VOC-focused strategy depending on a particular episode's conditions (NARSTO, 2000). Therefore, the appropriate combination of VOC and NO_X controls at the local level depends on local circumstances with the realization that a single approach focusing on NO_X or VOC-only controls is not necessarily effective for all episode types. It is clear, however, that regional NO_X reductions provide regional ozone reductions during episodic events, and this will influence ozone levels being transported into local urban areas.

The role of longer-lived VOCs, such as methane, in contributing to the broader regional (and global) ozone background is an additional consideration in weighing the respective merits of NO_X and VOC-based strategies. As mentioned earlier in Section 1.5, the anthropogenic contribution to the ozone background has important implications for strategies to achieve a future revised ozone NAAQS lower than 0.08 ppm (e.g., 0.060 – 0.070 ppm). Current ozone control strategies, while relatively effective at reducing episodic peak ozone levels, are less effective at reducing lower chronic (i.e., background) levels of ozone. In addressing the anthropogenic "background" ozone (i.e., the portion not due to natural sources), methane can have a major impact. For higher peak ozone concentrations, reducing methane is not typically considered due to its relatively low atmospheric chemical reactivity on the time-scale of multi-day ozone episodes, but methane reductions would have a relatively larger influence on longer-term background ozone levels (Fiore *et al*., 2002; 2003). Regional and global measures to reduce methane would not only have the benefit of reducing rising background ozone levels for purposes of achieving air quality standards, but would also help address the threat of climate change, as both ozone and methane are greenhouse gases.

3.4. Summary

There are large emissions of VOCs and NO_X within and outside the OTR that contribute to local and regional ozone problems. Naturally occurring VOC emissions play an important role in combination with human-caused NO_X emissions in forming regional ozone across large sections of the eastern U.S. Regional NO_X control strategies are demonstrating success in reducing regional ozone. On a more local scale, some combination of VOC and NO_X controls may be needed, with the specific combination dependent upon local circumstances. Measures to reduce methane, on a broad regional and global scale, can help address the anthropogenic component of a rising chronic ozone background, as well as help address climate change.

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4. WHAT WILL IT TAKE TO CLEAN THE AIR? – LINKING THE SCIENCE TO POLICY

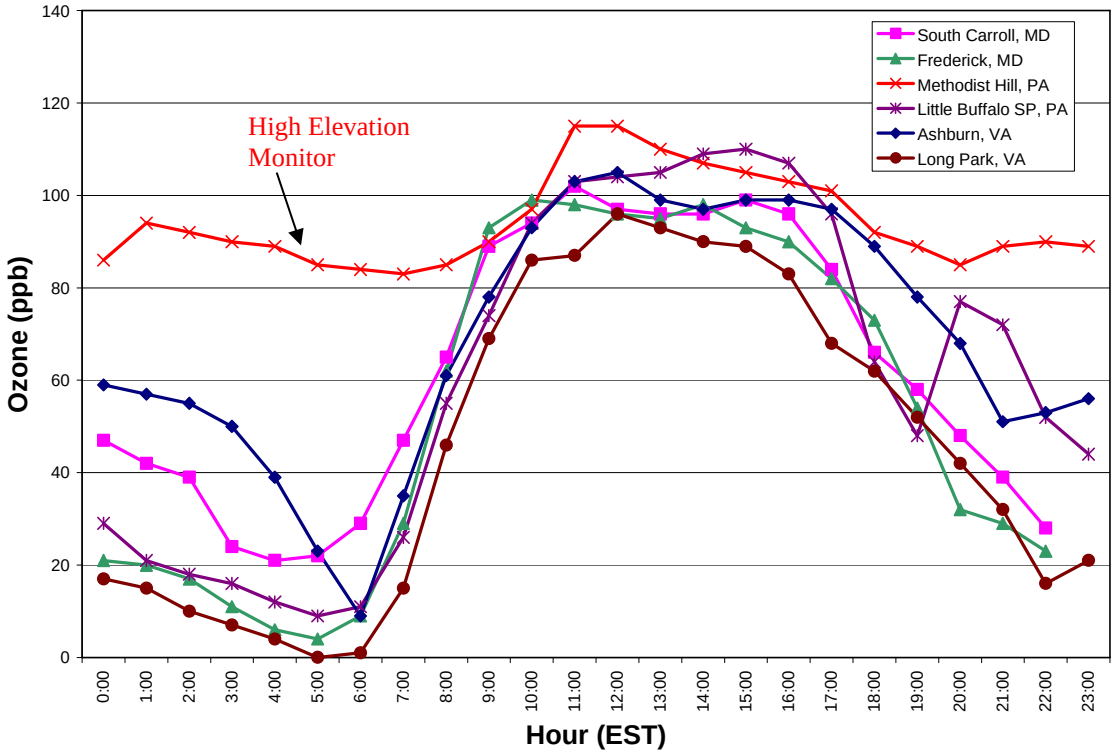
4.1. The three phases of a bad ozone day and the ozone reservoir

With the atmospheric chemistry, meteorology, and air emission inventory elements presented in the previous sections, a conceptual description emerges of ozone problem in the OTR. Consider a typical "day," defined as starting at sunset, for a severe ozone event associated with a high pressure system. Conceptually, a bad ozone day can be considered as occurring in three phases. During phase one, a nocturnal inversion forms as the temperature of the earth drops following sunset, isolating the surface from stronger winds only a few hundred feet overhead. Ozone near the surface can be destroyed by deposition. In a city, overnight, NO_X emissions can react with ozone, further reducing its concentration, so that by morning, very little ozone is left below the nocturnal inversion due to NO_X titration. At this time, the nocturnal inversion is at its strongest, and winds at the surface are typically calm. A large reservoir of ozone remains above the nocturnal inversion. Ozone and its precursors, both from the previous day's local emissions and from transport, remain largely intact. There are no surfaces and no new NO_X to remove the ozone.

During phase two of a bad ozone day, the nocturnal inversion breaks down at mid-morning, with the result that the ozone and precursors above the inversion can now mix with the air near the surface. The result of this vertical mixing is a sudden rise in ozone concentrations near the surface.

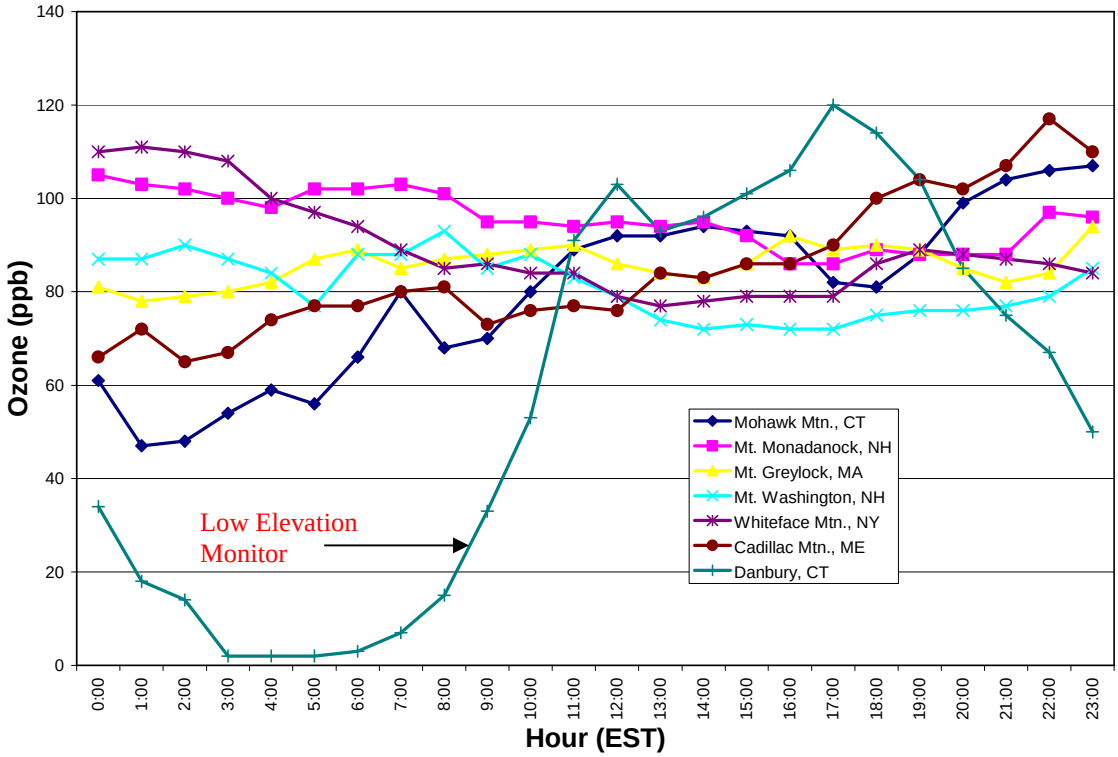
In phase three of a bad ozone day, ozone concentrations reach their highest levels in the afternoon through the combined accumulation of local pollution produced that day mixed with the transported regional pollution load brought in overnight from the ozone reservoir. Figure 4-1 shows this graphically for the southern OTR. The ozone monitor at Methodist Hill, PA is a high elevation site located at 1900 ft in altitude in south central Pennsylvania, and is above the nocturnal inversion. In the early morning hours of August 12, 2002 (e.g., 5 a.m.), it recorded ozone concentrations above 80 ppb, which was much higher than what other lower elevation monitors in the region were recording (e.g., Little Buffalo State Park, PA, South Carroll County, MD, Frederick, MD, Ashburn, VA, Long Park, VA). The high ozone levels seen at Methodist Hill, PA are above the nocturnal boundary layer and indicate the presence of significant ozone produced at some earlier point in time and transported into the region. With the break up of the nocturnal inversion after sunrise (e.g., starting about 7 a.m.), ozone concentrations at the lower elevation monitors show a rapid increase. This reflects the mixing down of the ozone reservoir from higher altitude to the surface in combination with local ozone production near the surface now that the sun has begun inducing its photochemical production.

Figure 4-1. Hourly ozone profiles in the southern OTR, August 12, 2002



The ozone reservoir extends across the OTR, as seen on the same night in high elevation ozone monitoring sites in the northern OTR. Figure 4-2 shows the hourly ozone concentrations measured on August 12, 2002 at Mohawk Mountain, CT, Cadillac Mountain, ME, Mt. Greylock, MA, Mt. Monadanock, NH, Mt. Washington, NH, and Whiteface Mountain, NY. As with Methodist Hill, PA on this day, these sites show elevated ozone concentrations during nighttime hours, as compared to lower elevation sites below the nocturnal inversion (e.g., Danbury, CT). By mid-day, however, the nocturnal boundary layer has broken down, mixing the transported ozone from the reservoir above into the locally produced ozone below. Appendix E provides more detail on contributions to the ozone reservoir within and outside the OTR.

Figure 4-2. Hourly ozone profiles of high elevation monitors in the northern OTR, August 12, 2002



Data provided by Tom Downs, Maine Department of Environmental Protection.

4.2. Chronology of an ozone episode – August 2002

The chronology of a historical ozone episode occurring in the OTR from August 8 to August 16, 2002 provides a real-world example that pieces together the elements of the ozone conceptual description given in this document. Surface maps from the period provide a synoptic overview of major weather systems that were influencing air quality across the OTR during that time. Meteorological insights combined with ozone concentration information provide a picture of the evolving ozone episode on a day-by-day basis. Figure 4-3, Figure 4-4, and Figure 4-5, respectively, show eight-panel displays of surface weather maps, back trajectories, and 8-hour maximum ozone concentrations from each day. The daily progression shows the formation of high ozone that shifts from west to east, and ultimately northward, during successive days of the episode according to local ozone formation and transport shaped by wind patterns within and outside of the OTR.

The August 2002 episode began with a slow-moving high pressure system centered over the Great Lakes initiating a northerly flow over the OTR on August 8. Over the next several days, the high drifted southeastward and became extended across a large part of the eastern U.S., bringing high temperatures to the region. Calm conditions west of the OTR on August 10 were pivotal for the formation of ozone, which first began building in the Ohio River Valley. Over the next four days, 8-hour ozone concentrations climbed well above the 85 ppb (0.08 ppm) NAAQS over a wide area of the OTR. Large parts of the heavily populated Northeast Corridor experienced 8-hour ozone levels above 100 ppb during the height of the episode, which far exceeded the 85 ppb NAAQS.

The following chronology provides a day-by-day evolution of the August 2002 ozone episode. Parts of this description are taken from Ryan (2003).

August 8: A high pressure system over the Great Lakes produces NW-N prevailing surface winds (~4-8 mph) throughout the region. Maximum daily temperatures approach or exceed 80º F.

August 9: Wind speeds fall off but the direction remains NW-N as the high moves into the Pennsylvania-New York region. Temperatures rise as cloud cover declines. Background ozone levels begin to build in the Ohio River Valley with 8-hour maximum concentrations reaching the 60-80 ppb range.

August 10: High pressure is directly over the mid-Atlantic. With dew points still in the mid-50º's F, the skies are extraordinarily clear throughout the day. Temperatures (except in northern-most areas) approach 90º F while surface-level winds turn to more southerly directions. With high pressure overhead, the back trajectories suggest very light winds and recirculation. Calm conditions through the morning hours in the lower Ohio River Valley promote increasingly higher levels of ozone noted in surface observations – now reaching above the 85 ppb 8-hour ozone NAAQS over much of Indiana, Ohio, and other states along the Ohio River, as well as states around Lake Michigan and large portions of the southeastern U.S. Ozone levels above the 8-hour NAAQS now begin appearing for the first time in the western and southern parts of the OTR.

August 11: Surface high pressure drops slowly southeastward across the mid-Atlantic with the center in western North Carolina drifting to coastal South Carolina during the day. The upper level ridge has also moved east and is located over the mid-Atlantic. Circulation around the high becomes well established. A surface-level trough descends from north of the Great Lakes during the day, passes eastward through the Ohio River Valley and stalls over the Allegheny Mountains and southward. Peak temperatures are in the low to mid-90º's F. Morning winds are low-to-calm in the area east of the Mississippi – the area of ozone now reaches from eastern Wisconsin to Tennessee and eastward to Georgia up through the Carolinas into the OTR, covering most of Pennsylvania, New York, New Jersey, Connecticut, Rhode Island and Massachusetts. Winds are generally south to southwest as is reflected in the boundary layer back trajectories. The key factor driving local ozone production appears to be a very stable boundary layer. The 8 a.m. sounding at the Washington-Dulles airport shows a very strong low-level inversion from 950-900 millibars (mb) with a deep residual layer beneath a continuing strong subsidence inversion – now based at 760 mb.

August 12: The upper level ridge remains quasi-stationary with its axis over the mid-Atlantic. The center of high pressure at 850 mb is over North Carolina and Georgia. At the surface, the characteristic Appalachian lee side trough forms. Temperatures exceed 90º F throughout the OTR except in coastal Maine. Winds are fairly strong from the northwest. This is reflected in the back trajectories that show a shift to westerly transport. Elevated upwind ozone concentrations at 11 a.m. on August 11 occur in the vicinity of the origin of the back trajectories, on the order of 78-86 ppb. Ozone concentrations fall this day west of the Appalachians but increase markedly across the mid-Atlantic. The area of highest ozone has pushed eastward and now extends from southern Maine across central Pennsylvania down through Maryland into the Carolinas, Georgia, and eastern

Tennessee. Ozone builds throughout the day as circulation forces it to the northeast between the stalled trough and a cold front approaching from the Midwest. Some of the highest 8-hour concentrations occur through the central to southern OTR on this day.

August 13: Calm conditions prevail as the trough reaches coastal New Jersey by 8 a.m. Generally clear skies allow temperatures to reach the mid-90º's F everywhere except in coastal Maine. Dew points, which had been rising since August 8, reach the upper 60º's F. A morning sounding at the Washington-Dulles airport showed a continuing strong low level inversion with a residual mixed layer to 850 mb ending just beneath a weak secondary inversion. The cap aloft has lifted to ~ 630 mb and the sounding is more unstable compared to previous day's between the two inversion layers. The Appalachian lee side trough continues in place from late on August 12. As is typically the case, the highest ozone concentrations are found in proximity to this boundary. The highest 8-hour ozone concentrations are along the eastern portions of the OTR from northeastern Virginia through New Jersey, Long Island, Connecticut, and into eastern Massachusetts. By 8 p.m., showers associated with the approaching cold front have reached into Ohio.

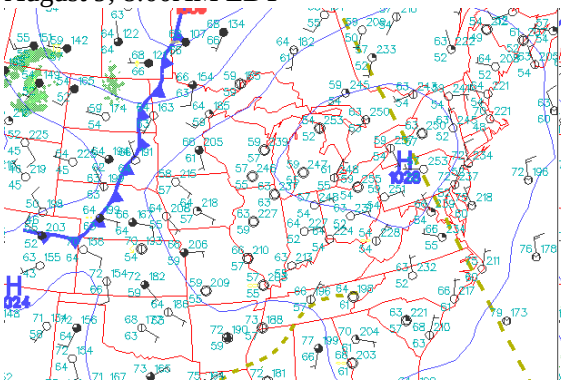
August 14: By 8 a.m., the trough has dissipated and the high is moving offshore, resulting in an increasing southerly wind component, which pushes maritime air northward. Dew points remain in the upper 60º's F and peak temperatures reach into the 90º's F everywhere and top 100º F in several locations. Ozone concentrations build again, with the highest levels concentrated in the central OTR from eastern Pennsylvania across to Massachusetts. A "hotspot" of ozone appears in upstate New York at the eastern end of Lake Ontario, and may be the result of transport from the west across the lake. Ozone concentrations decrease south and west of Baltimore and along coastal New Jersey as cleaner maritime air pushes in from the south.

August 15: This episode ends in a very different manner than the standard high ozone episode. Instead of the passage of a sharp cold front, this episode ends gradually as cleaner air sweeps north, winds increase, and the atmosphere steadily destabilizes. Ozone concentrations fall across the middle and lower OTR as low level flow becomes more southeast and the Bermuda high fills in westward. The highest levels, still exceeding the 8-hour ozone NAAQS, now occur in the northern reaches of the OTR in upstate New York, Vermont, New Hampshire, and Maine.

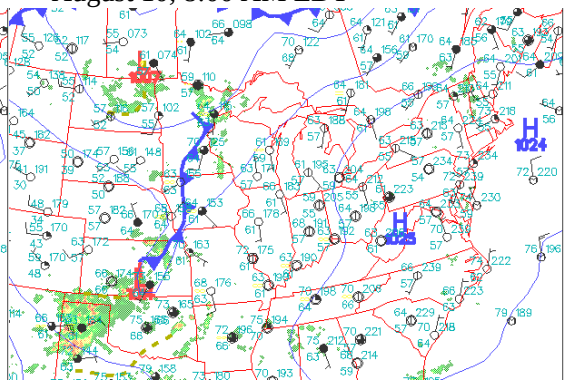
August 16: Cloud cover spreads over the region with ozone falling further. The new high building into the upper Midwest pushes the remains of the showers out of the Northeast. A spot of high ozone persists in central New Jersey. This is the last exceedance day in a string of seven exceedance days within the OTR during this extended episode.

Figure 4-3. Surface weather maps for August 9-16, 2002

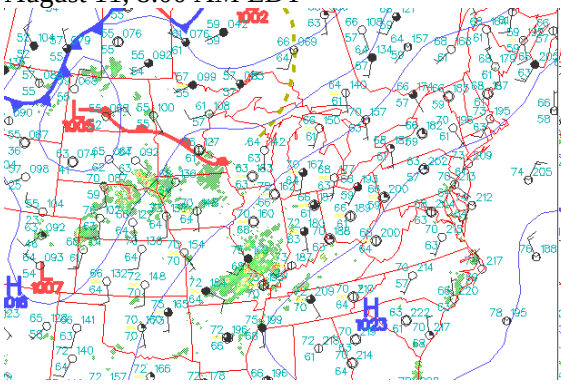
August 9, 8:00AM EDT



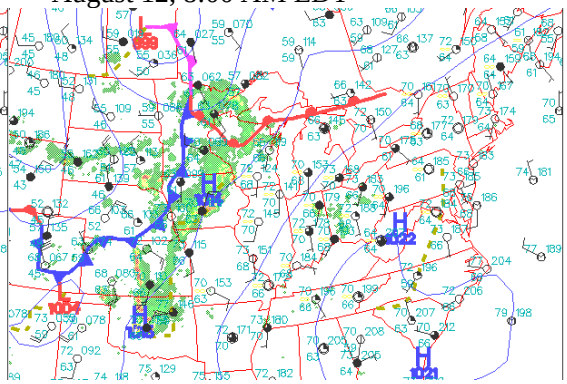
August 10, 8:00 AM EDT



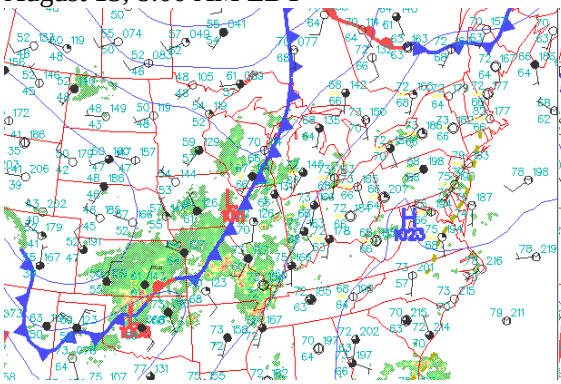
August 11, 8:00 AM EDT



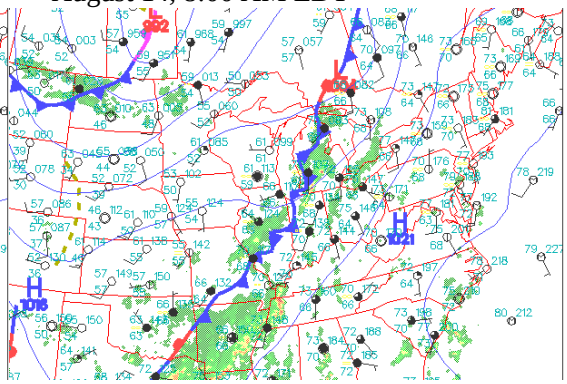
August 12, 8:00 AM EDT



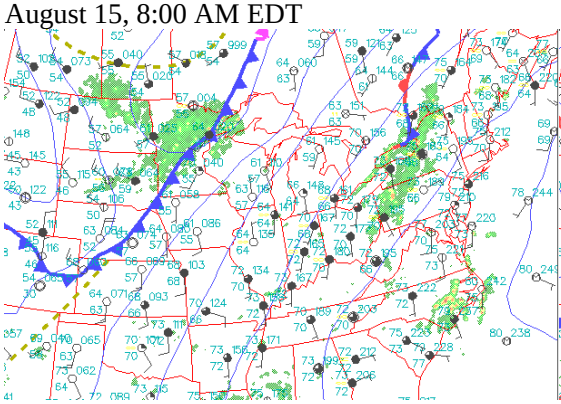
August 13, 8:00 AM EDT



August 14, 8:00 AM EDT



August 15, 8:00 AM EDT



August 16, 8:00 AM EDT

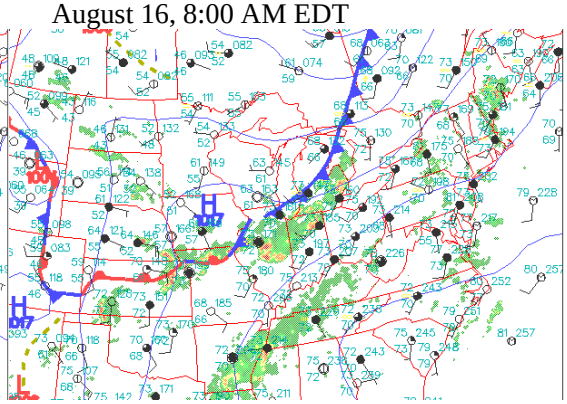
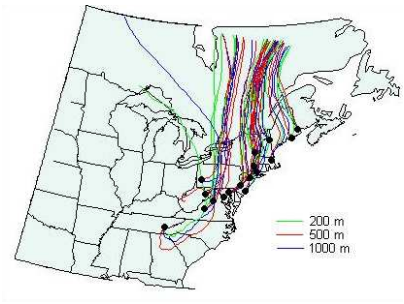


Figure 4-4. HYSPLIT 72-hour back trajectories for August 9-16, 2002

Aug 9, 2002 8 am EDT



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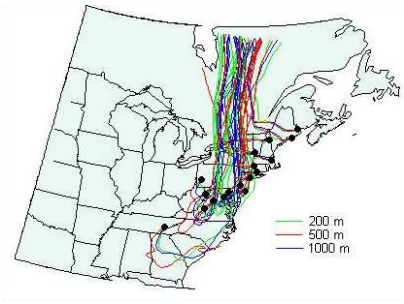

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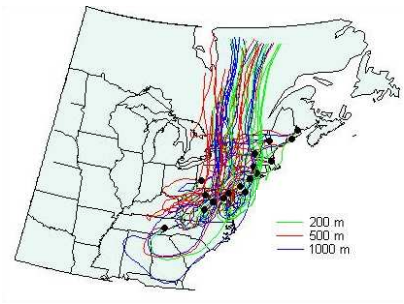
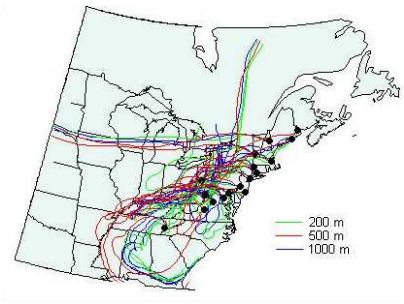
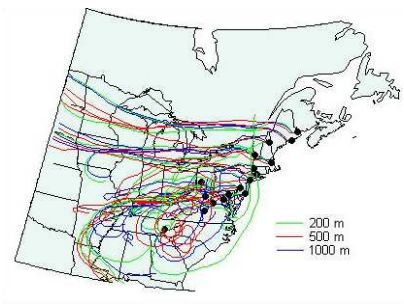


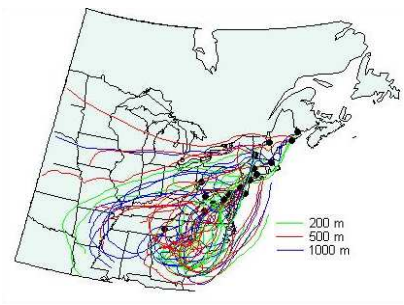
Aug 12, 2002 8 am EDT



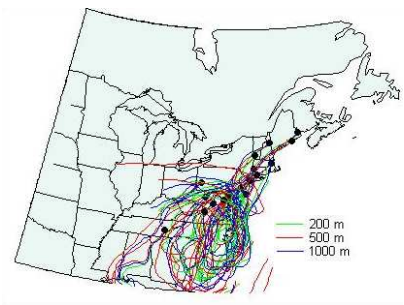
Aug 13, 2002 8 am EDT



Aug 14, 2002 8 am EDT



Aug 15, 2002 8 am EDT



Aug 16, 2002 8 am EDT

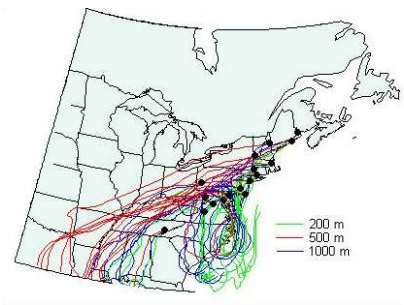
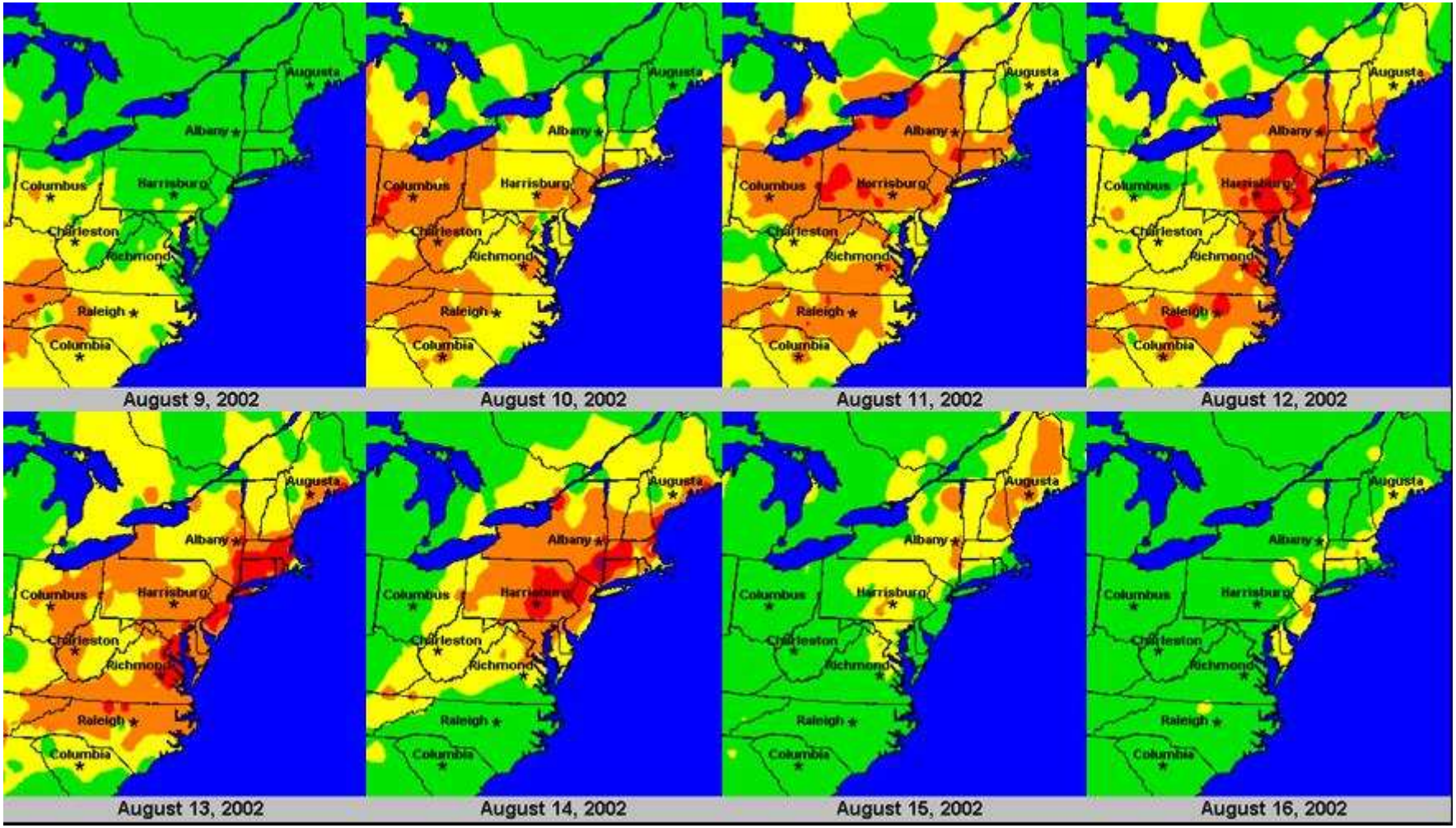


Figure 4-5. Spatially interpolated maps of maximum 8-hour surface ozone concentrations August 9 – 16, 2002



4.3. Clean Air Act provisions

As is evident from the myriad source regions and transport pathways affecting the OTR, the regional ozone nonattainment problem presents a significant challenge to air quality planners. To improve air quality, emission reductions of the appropriate pollutants must occur at the appropriate levels (i.e., stringency of controls) and over the appropriate geographic extent. States have primary responsibility for achieving the goals of the Clean Air Act, as they are responsible for developing State Implementation Plans and implementing and enforcing emission reduction programs to meet the health-protective National Ambient Air Quality Standards (NAAQS).

When Congress passed the Clean Air Act Amendments of 1990, it recognized that air pollution transcends political boundaries and that tools for addressing transport must be made available to state and federal governments. Accordingly, several Clean Air Act provisions deal with transported pollution, including: (1) prohibiting the USEPA from approving State Implementation Plans that interfere with another state's ability to attain or maintain a NAAQS; (2) requiring the USEPA to work with states to prevent emissions that contribute to air pollution in a foreign country; (3) allowing states to form ozone transport regions; (4) requiring states in ozone transport regions to adopt a prescribed set of controls in order to achieve a minimum level of regional emission reductions; and (5) allowing states to petition the USEPA for timely relief from stationary source emissions that interfere with attainment or maintenance of a NAAQS, and requiring the USEPA to act on such petitions within a very short, prescribed timeframe. Taken together, these provisions provide a framework for air quality planning. Its inherent principles are:

- Timely action is critical in order to protect public health;
- States must act locally to address air pollution;
- While acting locally, states must also consider their impacts downwind in addition to in-state impacts when developing state implementation plans (SIPs), and ameliorate such impacts through SIPs;
- Regional actions have been and can continue to be effective;
- To be effective on a regional level, states working together must work off of a level playing field;
- National rules are important and will play a more critical role as more states and nonattainment areas become involved in air quality planning.

What the science tells us of the nature of the ozone problem in the OTR supports this framework. The smaller scale weather patterns that affect pollution accumulation and transport underscore the importance of local (in-state) controls for NO_X and VOC emissions. Larger synoptic scale weather patterns, and pollution patterns associated with them, support the need for NO_X controls across the eastern United States. Studies and characterizations of interstate ozone transport also support the need for local and regional controls on NO_X and VOC sources. The key issues is to identify and control the source of these emissions, whether they are transported in the free troposphere, nocturnal low level jet, or land, sea, bay, mountain, and valley breezes.

The mix of emission controls is also important for states to consider. While long-range transport of ozone is primarily due to NO_X, VOCs are important because they

contribute to ozone formation by influencing how efficiently ozone is produced by NO_X, particularly within urban centers. While reductions in anthropogenic VOCs will typically have less of an impact on the long-range transport of ozone, they can be effective in reducing ozone in those urban areas where ozone production may be limited by the availability of VOCs.

Another dynamic in play is the level of the NAAQS in relation to the ozone levels of the elevated reservoir. As NAAQS are set at lower levels, transported ozone will likely play a more prominent role in the ozone problem that states will need to address through their SIPs. Therefore, a combination of localized VOC reductions in urban centers with additional NO_X reductions (from both mobile and point sources) across a larger region will help to reduce ozone and precursors in nonattainment areas as well as their downwind transport across the entire region (NESCAUM, 1997).

4.4. Past regional efforts

While states are somewhat limited in their ability to directly affect emissions reductions beyond their own geo-political boundaries, over the past 15-20 years, the Northeast states have acted regionally with tremendous success. Such efforts have included:

- In 1989, regional low volatility gasoline (i.e., Reid Vapor Pressure pf 9.0 pounds per square inch (psi)) was introduced into the NESCAUM region, resulting in significant VOC reductions;
- In 1994, the California Low Emission Vehicle (LEV) program commenced in the Northeast Corridor as regulations were adopted by Maine, Massachusetts, New York, and Vermont. To date, four additional states have joined the program, which continues to yield reductions in NO_X, VOC, CO, and air toxics.
- In 1994, the states of the Ozone Transport Commission agreed to promulgate regional NO_X RACT controls and a NO_X cap-and-trade program. The adopted regional RACT deadline was 1995. By 1999, the NO_X Budget Program was implemented over the 12-state region from Maine to Washington, DC. In 2002, the USEPA reported that the NO_X Budget sources "emitted at a level approximately 12 percent below 2001 allocations" (USEPA, 2002). Progress continues with a more stringent cap taking effect in 2003.
- In 1997, eight OTR states petitioned the USEPA under section 126 of the Clean Air Act, requesting NO_X emissions reductions on certain stationary sources in the Eastern U.S. In 1999, four more OTR members filed section 126 petitions. The USEPA granted four of the initial eight state petitions in 2000.[f]
- Beginning in 2001, the states of the Ozone Transport Commission agreed to support a suite of model rules for inclusion in SIPs as appropriate to address ozone nonattainment. Model rules to address the 1-hour ozone NAAQS included

[f] The initial eight section 126 OTR states were Connecticut, Maine, Massachusetts, New Hampshire, New York, Pennsylvania, Rhode Island, and Vermont. The additional four OTR members filing section 126 petitions were Delaware, the District of Columbia, Maryland, and New Jersey. The four granted petitions were from Connecticut, Massachusetts, New York, and Pennsylvania.

controls for: (1) architectural and industrial maintenance coatings; (2) portable fuel containers; (3) consumer products; (4) solvent cleaning; (5) mobile equipment repair and refinishing; and (5) additional NO_X controls for industrial boilers, cement kilns, stationary reciprocating engines, and stationary combustion engines. Several of the model rules have been or are being updated to reflect more current technology and emission reductions needed to address the 8-hour ozone standard.

These regional efforts led the way for similar broader regional and national programs. For mobile sources, the USEPA promulgated its federal Reformulated Gasoline Program in 1995 and the National LEV program in 1998. For stationary sources, the USEPA in 1998 promulgated the NOx SIP Call. This program expanded the OTR NO_X Budget Program, involving 20 states, and put in place a regional NO_X cap in 2003. The NO_X SIP Call also served as a response to the aforementioned Section 126 petitions under the Clean Air Act, which were filed by states in 1997.

In addition to the OTR, other regions in the eastern United States affected by regional transport have experienced benefits from the geographically expanded ozone control initiatives. For example, ozone levels in western Michigan are dominated by transport from major urban areas in the Lake Michigan area (e.g., Chicago, Gary, and Milwaukee) as well as from other source areas in the eastern United States. Regional control programs were found to be an effective method to address ozone transport and achieve the ozone NAAQS in this region (USEPA, 2009).

In 2005, the USEPA took a further step to address the regional ozone problem by issuing the Clean Air Interstate Rule (CAIR), which required additional NO_X reductions in 25 eastern states and the District of Columbia. The CAIR rule establishes an annual NO_X cap of 1.3 million tons in the eastern United States to be achieved by 2015. The cap, however, was virtually achieved by 2009, as NO_X emissions from sources covered by state adoption of CAIR were only 1.305 million tons (USEPA, 2010).

The USEPA projected that CAIR would achieve NO_X reductions of 2 million tons in 2015, a 61% decrease from 2003 levels. This would be a significant step forward in improving air quality, but the time allowed to achieve these reductions is later than the deadline many eastern states are facing to meet the current 8-hour ozone NAAQS. CAIR, therefore, only partially provides the OTR with a regional measure to help achieve the Clean Air Act's goal of attaining the ozone air quality health standard within the Act's mandatory deadlines.

A number of parties challenged CAIR in court, and it was initially vacated in its entirety on July 11, 2008 by the U.S. Court of Appeals for the District of Columbia Circuit (*North Carolina v. EPA*, 531 F.3d 896 (D.C. Cir.)). The D.C. Circuit subsequently modified its opinion by remanding CAIR back to EPA without vacatur so that CAIR remains in effect while the USEPA develops a replacement rule consistent with the court's opinion.

4.5. New ozone standards

While the OTR states have made demonstrable progress towards meeting the 1997 ozone NAAQS through a combination of local and regional VOC and NO_X control measures, evolving understanding of ozone's impacts on human health has led to the need for a newer, more protective primary ozone NAAQS. In 2006, the Clean Air Scientific Advisory Committee (CASAC) – an independent review committee of expert scientists – made a unanimous recommendation to the USEPA to revise the primary ozone standard within a range of 0.060 – 0.070 ppm (CASAC, 2006). After initially promulgating a revised primary ozone NAAQS of 0.075 ppm 8-hour average in 2008, the USEPA reconsidered its initial action and re-proposed in 2010 a more stringent primary ozone NAAQS within the CASAC-recommended range (75 Fed. Reg. 2938-3052 (January 19, 2010)). The USEPA has also proposed secondary ozone NAAQS of a different form from the primary ozone NAAQS to better protect welfare values, such as forests and agricultural crops. The proposed secondary standard would be a weighted sum of daytime hourly ozone concentrations over a three month warm weather growing season. Regardless of the final revised ozone NAAQS levels, achieving new more protective standards will require maintaining the momentum of states' efforts to reduce VOC and NO_X emissions. New strategies will be needed that may be a combination of deeper emission reductions from existing sources and new measures on previously uncontrolled sources or types of activities.

Even with the lower NO_X emissions of 2009, many monitoring locations in the OTR recorded ozone levels that would put them in violation of a more stringent revised ozone primary NAAQS in the USEPA-proposed range of 0.060 – 0.070 ppm, 8-hour average. This indicates that the CAIR NO_X cap, originally established to assist states in meeting an ozone NAAQS of 0.08 ppm, will not be sufficient to address the more stringent ozone NAAQS expected from the USEPA. Analysis by the OTC indicates that it is feasible to cost effectively further reduce power plant NO_X emissions in the eastern United States down to an annual cap of 900,000 tons by 2015.

A more stringent 8-hour ozone NAAQS also has implications for what may be considered an ozone "episode." As mentioned in Section 1 and described in Appendix B, the classic OTR-wide high ozone episode is typically a summertime multi-day event associated with the slow passage of a high pressure system from the west to east that also brings sunny skies and high temperatures. There are also four other types of patterns associated with elevated ozone in the OTR. With a more stringent 8-hour ozone NAAQS, about 60 percent of days during an extended six month ozone season could experience an ozone exceedance somewhere in the OTR, based on recent history. For example, between 2003 and 2009, the average annual number of days during which at least one ozone monitor in the OTR exceeded 64 ppb was 112 out of ~180. This number of ozone exceedance days may implicate other weather patterns in addition to those identified in Appendix B. Futhermore, a conceptual model of ozone formation and transport for a multi-day ozone "episode" is not readily applicable to a secondary ozone NAAQS that is a summation of weighted hourly ozone concentrations over a three month period. It is known that the relative effectiveness of VOC versus NO_X control measures at reducing ozone can vary in location and time, thus a greater diversity of control measures will be needed to address a broader range of conditions leading to ozone formation.

4.6. Future climate change and ozone

Warmer temperatures can enhance ozone formation, thus leading to the potential for a "climate penalty" of increasing ozone with a warming climate. A statistical analysis by Bloomer *et al*. (2009) that looked at 21 years of ozone and temperature observations across the rural eastern U.S. discerned a relationship between rising temperatures and increasing ozone. A change in the relationship around 2002 also indicated reducing NO_X precursor emissions has the potential to mitigate the climate penalty. According to the Bloomer *et al*. analysis, ozone increased by an average of ~3.2 ppbv/°C prior to 2002. After 2002, a smaller climate penalty factor of ~2.2 ppbv/°C was observed as ozone levels fell by ~10 percent. The decrease in ozone was attributed to a 43 percent reduction in power plant NO_X emissions.

This historical analysis is directionally consistent with air quality modeling studies summarized below that projected deteriorating air quality in a warmer future climate when anthropogenic precursor emissions were held constant. When emissions reductions were incorporated into the modeling, air quality improvements were predicted, although perhaps not as great as would have otherwise occurred in the absence of future warmer temperatures.

A review article by Weaver *et al*. (2009) summarizes and compares the results of several modeling experiments of U.S. regional ozone impacts from climate change projected to *circa* 2050. All the modeling experiments held anthropogenic emissions of ozone precursors constant at present-day levels, but allowed biogenic VOC emissions to respond to simulated climate changes. This was done to investigate solely the influence of climate change on ground-level ozone independent of changes in anthropogenic precursor emissions in the future. The studies compared present and future ozone levels corresponding to the maximum daily eight-hour average. On a national scale, the different modeling experiments were in general agreement in projecting an increase in ozone of roughly 2-8 ppb for a large part of the country due to future climate change alone. Some regions, however, were predicted to experience little change in ozone concentrations, or even decreases. While these general features were common across the various modeling experiments, the regional patterns of increasing or decreasing future ozone varied across the models. These differences were attributed to differences in the modeling systems, model configuration, and experimental design choices among the different research groups. Specific examples included differences in cloud cover affecting surface sunlight levels, average maximum daily temperature, number of rainy days, and boundary layer depth.

Of special note were modeling results predicting a greater increase in the future 95^{th} percentile maximum daily eight-hour averages as compared to the mean maximum (Nolte *et al*., 2008). Because the current primary ozone air quality standard is based on the 4^{th} maximum annual eight-hour average, this accentuated projected increase at the higher end of the daily maximum eight-hour averages has implications for air quality planners in developing control strategies to achieve as well as maintain attainment ozone air quality standards (Weaver *et al*., 2009).

When the modeling included future decreases in ozone precursor emissions consistent with the reductions expected from EPA's Clean Air Interstate Rule (to be re-

proposed by EPA following a court-ordered remand), the projected 95th percentile eight-hour ozone levels no longer increased in much of the United States. Projected ozone levels decreased by at least 5 ppb nearly everywhere across the U.S., with large portions of the eastern and central U.S. predicted to see decreases of up to 15 ppb (Nolte *et al*., 2008). A modeling study by Liao *et al.* (2007) also projected ozone decreases in 2050 for 4th maximum 8-hour ozone averages, with ozone becoming even more sensitive to NO_X in the future as NO_X reductions continue to occur while anthropogenic VOC reductions are offset by increases in biogenic VOCs in a warmer climate.

This suggests that continuing efforts to reduce ozone precursor emissions will have success in mitigating and improving ozone air quality despite countervailing pressure from future climate change. Maintaining precursor emissions at current levels, however, could result in higher future ozone levels under projected climate change through *circa* 2050. Even with continued emission reductions, it has been pointed out that the "climate penalty" will require stronger emission controls to meet a given air quality standard (Bloomer *et al*., 2009; Jacob & Winner, 2009; Racherla & Adams, 2009).

4.7. Summary: Building upon success

A conceptual understanding of ozone as a regional problem in the OTR and throughout the eastern U.S. is now well established. With this evolution in understanding, regional approaches to the ozone problem are now underway, starting with the 1990 Clean Air Act Amendments that created the Ozone Transport Region. This initial regional approach, however, did not include large source regions outside of the OTR containing many large coal-fired power plants and other pollution sources contributing to the long-range transport of ozone into the OTR.

In 1998, the USEPA took another step in addressing the regional problem by finalizing the NO_X SIP Call, which covered emissions of NO_X, the main precursor of regional ozone, in additional parts of the East. Even with these reductions, air quality modeling has projected continuing significant contributions from upwind sources in out-of-state regions. As a result, the USEPA promulgated a further round of regional NO_X reductions in the East with the adoption of CAIR in 2005. With the modeling foundation for CAIR, the USEPA has presented a compelling technical case on the need for additional regional NO_X reductions in the eastern U.S. to reduce ozone levels and protect public health. While states in the Northeast disagree with the extent of NO_X reductions and the timeline for those reductions to occur, the program is an excellent next step toward reducing ozone in the OTR.

There is a tendency to characterize the nonattainment problems persisting after implementation of CAIR and other federal programs as "residual," but care must be taken in assessing these continuing nonattainment problems. A "residual" ozone problem is better characterized as a persistent nonattainment problem that still requires broad regional responses coupled with local controls. As this conceptual description points out, one of the great lessons and successes seen in the history of air quality policy was the shift from urban-only air pollution control strategies to broader regional approaches in the East at the end of the 1990s (e.g., NO_X SIP Call). The danger exists, however, that the perception of a "residual" ozone problem as being only a local issue will ignore the

lessons learned from effective regional approaches and possibly stall much needed national rules.

The current suite of local and regional controls have a proven track record of success, and have helped to significantly lower NO_X, VOC, and ozone levels across the eastern U.S. As described earlier in this report, monitored NO_X emissions and ambient concentrations have dropped between 1997 and 2008, and the frequency and magnitude of ozone exceedances have declined within the OTR. In the near future, however, there will very likely be a new health-based primary ozone NAAQS within the range of 0.060 – 0.070 ppm averaged over eight hours. The conceptual model of regional ozone formation and transport, along with a track record of demonstrated progress in achieving cleaner air within this conceptual model framework, provides direction to air quality planners for meeting the new challenge of a more health-protective ozone NAAQS. To maintain the current momentum for improving air quality so that the OTR states can meet current and future attainment deadlines, the direction is for deeper and broader regional NO_X reductions coupled with appropriate local NO_X controls and regional and local VOC controls.

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Appendix A: USEPA Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze

From "Guidance on the Use of Models and Other Analyses for Demonstrating Attainment of Air Quality Goals for Ozone, $PM_{2.5}$, and Regional Haze," U.S. Environmental Protection Agency, EPA-454/B-07-002, Section 11, April 2007.

Appendix A: EPA Guidance Document Excerpt

11.0 How Do I Get Started? - A "Conceptual Description"

A State/Tribe should start developing information to support a modeled attainment demonstration by assembling and reviewing available air quality, emissions and meteorological data. Baseline design values should be calculated at each monitoring site, as described in Section 3. For PM applications, speciated data should be reviewed to get a sense of what component(s) might be contributing most significantly to nonattainment or light extinction. If past modeling has been performed, the emission scenarios examined and air quality predictions may also be useful. Readily available information should be used by a State/Tribe to develop an initial conceptual description of the nonattainment or reasonable haze problem in the area which is the focus of a modeled demonstration. A conceptual description is instrumental for identifying potential stakeholders and for developing a modeling/analysis protocol. It may also influence a State's choice of air quality model, modeling domain, grid cell size, priorities for quality assuring and refining emissions estimates, and the choice of initial diagnostic tests to identify potentially effective control strategies. In general, a conceptual description is useful for helping a State/Tribe identify priorities and allocate resources in performing a modeled demonstration.

In this Section, we identify key parts of a conceptual description. We then present examples of analyses which could be used to describe each of these parts. We note that initial analyses may be complemented later by additional efforts performed by those implementing the protocol.

11.1 What Is A "Conceptual Description"?

A "conceptual description" is a qualitative way of characterizing the nature of an area's nonattainment or regional haze problem. It is best described by identifying key components of a description. Examples are listed below. There are 3 different examples. One each for ozone, annual $PM_{2.5}$, and regional haze. The examples are not necessarily comprehensive. There could be other features of an area's problem which are important in particular cases. For purposes of illustration later in the discussion, we have answered each of the questions posed below. Our responses appear in parentheses.

11.1.1 8-Hour Ozone NAAQS

1. Is the nonattainment problem primarily a local one, or are regional factors important?

(Surface measurements suggest transport of ozone close to 84 ppb is likely. There are some other nonattainment areas not too far distant.)

2. Are ozone and/or precursor concentrations aloft also high?

(There are no such measurements.)

3. Do violations of the NAAQS occur at several monitoring sites throughout the nonattainment area, or are they confined to one or a small number of sites in proximity to one another?

(Violations occur at a limited number of sites, located throughout the area.)

4. Do observed 8-hour daily maximum ozone concentrations exceed 84 ppb frequently or just on a few occasions?

(This varies among the monitors from 4 times up to 12 times per year.)

5. When 8-hour daily maxima in excess of 84 ppb occur, is there an accompanying characteristic spatial pattern, or is there a variety of spatial patterns?

(A variety of patterns is seen.)

6. Do monitored violations occur at locations subject to mesoscale wind patterns (e.g., at a coastline) which may differ from the general wind flow?

(No.)

7. Have there been any recent major changes in emissions of VOC or NO_X in or near the nonattainment area? If so, what changes have occurred?

(Yes, several local measures [include a list] believed to result in major reductions in VOC [quantify in tons per summer day] have been implemented in the last five years. Additionally, the area has seen large regional NO_X reductions from the NO_X SIP call.)

8. Are there discernible trends in design values or other air quality indicators which have accompanied a change in emissions?

(Yes, design values have decreased by about 10% at four sites over the past [x] years. Smaller or no reductions are seen at three other sites.)

9. Is there any apparent spatial pattern to the trends in design values?

(No.)

10. Have ambient precursor concentrations or measured VOC species profiles changed?

(There are no measurements.)

11. What past modeling has been performed and what do the results suggest?

(A regional modeling analysis has been performed. Two emission scenarios were modeled: current emissions and a substantial reduction in NO_X emissions throughout the regional domain. Reduced NO_X emissions led to substantial predicted reductions in 8-hour daily maximum ozone in most locations, but changes near the most populated area in the nonattainment area in question were small or nonexistent.)

12. Are there any distinctive meteorological measurements at the surface or aloft which appear to coincide with occasions with 8-hour daily maxima greater than 84 ppb?

(Other than routine soundings taken twice per day, there are no measurements aloft. There is no obvious correspondence with meteorological measurements other than daily maximum temperatures are always > 85 F on these days.)

Using responses to the preceding questions in this example, it is possible to construct an initial conceptual description of the nonattainment area's ozone problem. First, responses to questions 1 and 11 suggest there is a significant regional component to the area's nonattainment problem. Second, responses to questions 3, 4, 7, 8, and 11 indicate there is an important local component to the area's nonattainment problem. The responses to questions 4, 5 and 12 indicate that high ozone concentrations may be observed under several sets of meteorological conditions. The responses to questions 7, 8, and 11 suggest that ozone in and near the nonattainment area may be responsive to both VOC and NO_X controls and that the extent of this response may vary spatially. The response to question 6 suggests that it may be appropriate to develop a strategy using a model with 12 km grid cells.

The preceding conceptual description implies that the State/Tribe containing the nonattainment area in this example will need to involve stakeholders from other, nearby States/Tribes to develop and implement a modeling/analysis protocol. It also suggests that a nested regional modeling analysis will be needed to address the problem. Further, it may be necessary to model at least several distinctive types of episodes and additional analyses will be needed to select episodes. Finally, sensitivity (i.e., diagnostic) tests, or other modeling probing tools, will be needed to assess the effects of reducing VOC and NO_X emissions separately and at the same time.

11.1.2 Annual $PM_{2.5}$ NAAQS

1. Is the nonattainment problem primarily a local one, or are regional factors important?

(Surface measurements suggest that only design values in or immediately downwind of the city violate the NAAQS. However, other nearby design values come close to the concentration specified in the NAAQS.)

2. What is the relative importance of measured primary and secondary components of $PM_{2.5}$ measured at sites violating the NAAQS?

(Secondary components (i.e., SO_4, NO_3, OC) constitute about 80% of the measured mass of $PM_{2.5}$. There are higher concentrations of primary $PM_{2.5}$ in the core urban area compared to the suburbs and more rural areas.)

3. What are the most prevalent components of measured $PM_{2.5}$?

(The most important components in ranked order are mass associated with SO_4, OC and inorganic primary particulate matter (IP).)

4. Does the measured mix of PM components appear to roughly agree with mix of emission categories surrounding the monitoring sites?

(No. Relative importance of measured crustal material (IP) appears less than what might be inferred from the inventory.)

5. Do there appear to be any areas with large gradients of primary $PM_{2.5}$ in monitored or unmonitored areas?

(Cannot really tell for sources of crustal material until we resolve the preceding inventory/monitoring discrepancy. There are no other obvious major sources of primary particulate matter.)

6. Is there any indication of what precursor might be limiting formation of secondary particulate matter?

(No indicator species analyses have been performed. Past analyses performed for ozone-related SIP revisions suggest that ozone in this area may be limited by availability of VOC.)

7. Do monitored violations occur at locations subject to mesoscale wind patterns (e.g., at a coastline) which may differ from the general wind flow?

(No.)

8. Have there been any recent major changes in emissions of PM or its precursors in or near the nonattainment area? What?

(Yes, measures believed to result in major reductions in VOC and NO_X have been implemented in the last 5 years. Reductions in power plant NO_X have resulted from the NO_X SIP call and SO_2 emissions reductions have resulted from the national program to reduce acid deposition.)

9. Are there discernible trends in design values or other air quality indicators which have accompanied a change in emissions?

(The trend appears to be downward, but the most recent air quality data has been higher. Overall, the period of record is insufficiently long to tell.)

10. Is there any apparent spatial pattern to the trends in design values?

(No.)

11. What past modeling has been performed and what do the results suggest?

(A regional modeling analysis has been performed for ozone and $PM_{2.5}$. Two emission scenarios were modeled: current emissions and a substantial reduction in NO_X and SO_2 emissions throughout a regional domain. Reduced NO_X emissions led to substantial predicted reductions in 8-hour daily maximum ozone in most locations. Modeled SO_2 reductions from the CAIR rule had a strong impact on sulfate concentrations.)

12. Are there any distinctive meteorological measurements at the surface or aloft which appear to coincide with occasions with $PM_{2.5}$ concentrations in excess of 15.0 $\mu g/m^3$?

(Other than routine soundings taken twice per day, there are no measurements aloft. There is no obvious correspondence with meteorological measurements other than daily maximum temperatures are often > 85 F on days with the highest $PM_{2.5}$ observations.)

13. Do periods with high measured particulate matter or components of particulate matter appear to track each other or any other measured pollutant?

(There appears to be some correspondence between measured high concentrations of SO_4 and ozone.)

Using responses to the preceding questions in this example, it is possible to construct an initial conceptual description of the nonattainment area's ozone problem. First, responses to questions 1, 2 and 3 suggest there is a significant regional component to the area's nonattainment problem. Second, responses to questions 1 and 3 indicate there is a local component to the problem. The responses to questions 11, 12 and 13 suggest that there may be a link between reducing ozone and reducing particulate matter. Thus, it may be appropriate to assess effects of previously committed to strategies to reduce ozone and national PM control measures before simulating additional control measures. The responses to questions 4 and 5 suggest that it is premature to determine whether a "local area analysis" will be needed. The response to question 7 suggests that it may not be necessary to model with very small grid cells, at least for the secondary components of $PM_{2.5}$.

The preceding conceptual description implies that the State containing the nonattainment area in this example will need to involve stakeholders from other, nearby States to develop and implement a modeling/analysis protocol. It also suggests that a nested regional modeling analysis will be needed to address the problem.

11.1.3 Example Regional Haze Application

1. What components of particulate matter appear to have high concentrations on days with poor visibility?

(Mass associated with SO_4 and coarse particulate matter (CM) seem to have the highest concentrations on most such days.)

2. What are typical values for the humidity adjustment factor during the times of year when most of the days with poor visibility occur?

(Typical values appear to be about “4.0”.)

3. Does visibility appear to track well among nearby Class I areas?

(Yes, but not always.)

4. Does poor visibility seem to occur under any specific meteorological conditions?

(This information is not readily available.)

5. Does poor visibility seem to coincide with high observed concentrations of any particular other pollutant?

(There seems to be some correspondence with high regional ozone concentrations.)

6. What components of particulate matter appear to have relatively high concentrations on days with good visibility?

(Coarse particulate matter and OC.)

7. What are typical values for the humidity adjustment factor during times of year when most of the days with good visibility occur?

(About “2.3”.)

8. Does good visibility appear to occur under any specific meteorological conditions?

(Don’t know.)

Answers to the preceding questions suggest that strategies to reduce sulfate concentrations and, perhaps, regional ozone concentrations might be effective in reducing light extinction on days when visibility is currently poor. The responses suggest that a strategy which focuses on this alone should first be tried for the days with good visibility as well. Even though sulfate concentrations appear low on such days, the fact that sulfates scatter light efficiently (see Equation (6.1)) and relative humidity is still high enough to enhance this effect is worth considering. Responses suggest that further meteorological analyses would be worthwhile prior to selecting strategies to simulate with a resource intensive regional model.

It should be clear from the preceding examples that the initial conceptual description of an area's nonattainment problem draws on readily available information and need not be detailed. It is intended to help launch development and implementation of a modeling/analysis protocol in a productive direction. It will likely be supplemented by subsequent, more extensive modeling and ambient analyses performed by or for those implementing the modeling/analysis protocol discussed in Section 12.0.

Questions like those posed in Section 11.1 can be addressed using a variety of analyses ranging in complexity from an inspection of air quality data to sophisticated mathematical analyses. We anticipate the simpler analyses will often be used to develop the initial conceptual description. These will be followed by more complex approaches or by approaches requiring more extensive data bases as the need later becomes apparent. These analyses are intended to channel resources available to support modeled attainment demonstrations onto the most productive paths possible. They will also provide other pieces of information which can be used to reinforce conclusions reached with an air quality model, or cause a reassessment of assumptions made previously in applying the model. As noted in Section 7, corroboratory analyses should be used to help assess whether a simulated control strategy is sufficient to meet the NAAQS.

Appendix B: Ozone pattern classifications in the OTR

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The following five types of ozone patterns in the OTR are taken from: Stoeckenius, T. and Kemball-Cook, S. "Determination of representativeness of 2002 ozone season for ozone transport region SIP modeling." Final Report prepared for the Ozone Transport Commission, 2005. Figure B-1 shows the 850 mb height and wind fields and Figure B-2 shows the surface temperatures and 10 meter wind fields for the five patterns (reproduced from Figures 3-2 and 3-5 of Stoeckenius & Kemball-Cook, 2005).

"Type A" – High ozone throughout the OTR. This pattern is characterized by strong high pressure over the southeastern states extending from the surface to 500 mb with high temperatures extending into New England and southwest surface winds throughout the OTR. The 850 mb temperatures and heights, and surface temperatures are above average at all locations except Washington DC; winds are southwest to west throughout the OTR except more variable at LaGuardia and magnitudes of resultant wind vectors are higher than average (indicative of a fairly steady, well defined flow regime), east-west surface pressure gradients are near neutral but southwest-northeast gradients along the I-95 corridor and in the west (Pittsburgh to Buffalo) are positive, which is consistent with the southwest flow. The stable air mass and high temperatures promote ozone formation throughout the OTR under these conditions.

"Type B" – High ozone confined to the extreme southeastern OTR. This pattern is characterized by an upper-level trough offshore of the OTR and a surface high centered over Kentucky. This results in cooler air advection over nearly all of the OTR with northwest flow aloft and a more westerly flow at the surface. The 850 mb heights are lower than average (especially in New England) and surface winds are more frequently from the northwest along the I-95 corridor than under Type A. Temperatures at 850 mb along the I-95 corridor are only slightly cooler than under Type A but inland temperatures, especially in the north, are much cooler (e.g., at Buffalo); similarly, surface temperatures along the I-95 corridor are about the same as under Type A but temperatures are cooler in Buffalo and Albany. Type B events have the strongest positive west-east surface pressure gradients of any category, consistent with the northwest winds but gradients from Washington to New York and Boston are positive. The cooler air over the western OTR and westerly to northwesterly flow result in the higher ozone levels being confined to just the extreme southern portion of the OTR under this pattern.

"Type C" – High ozone along the I-95 corridor and northern New England. This pattern is characterized by an extension of the semi-permanent Bermuda high into the southeastern U.S. and an area of high surface and 850 mb temperatures extending from Maryland to Maine; the 500 mb pattern is nearly zonal (east-west flow) while flow at the surface is generally from the southwest. The 850 mb heights are intermediate between Type A and Type B but 850 mb temperatures are very high along the I-95 corridor and slightly cooler further inland. Winds are more consistently south - southwest at all sites than under other episode types and almost no northwest-north-northeast winds are seen at LaGuardia in contrast to other types. Resultant wind vector magnitudes are much higher

than average, consistent with the steady southwest flow. Southwest – northeast pressure gradients along the I-95 corridor and from Pittsburgh to Buffalo are positive, consistent with the southwest flow. Average east-west pressure gradients are near zero. These conditions result in above average ozone levels all along the I-95 corridor with advection north into coastal and interior New England. Ozone levels are slightly below average in the extreme southeastern and western OTR.

"Type D" – High ozone in the western OTR. This pattern is characterized by an area of mean upper level divergence with associated cut-off low at 850 mb off the Outer Banks of North Carolina. A relatively vigorous mean low pressure center can be seen at the surface. An east-west temperature gradient across the OTR is evident at 850 mb. Surface temperatures along the I-95 corridor and in Albany are below average but surface temperature is above average at Buffalo. The 850 mb heights are the highest of any episode type due to a strong ridge over New England. Surface winds are mostly east-northeast along the I-95 corridor from DC to New York but more variable further north. In contrast to episode types A, B, or C, the southwest-northeast pressure gradients along the I-95 corridor are negative, consistent with the northeast surface winds. West-east pressure gradients are flat. These conditions result in below average ozone in the eastern OTR due to the on-shore flow in the north and cyclonic conditions in the south but above average ozone levels in the western OTR due to stable, warm conditions with light winds.

"Type E" – Generally low ozone throughout the OTR. This category includes days with moderately low to lowest average ozone readings of all OTR exceedance days used in the characterization scheme. The Bermuda high is shifted east relative to the other types and flow over the southeastern U.S. is only weakly anti-cyclonic with a nearly zonal flow pattern at the 850 and 500 mb levels over the OTR. Temperatures at the surface and aloft are the coolest of any episode type. While winds aloft are nearly westerly, surface winds are generally south-southeast over most of the OTR. The southwest-northeast pressure gradients are negative along the I-95 corridor and east-west gradients are positive, consistent with the southeast flow. These conditions result in below average ozone throughout the OTR due to the relatively low temperatures and southeasterly onshore flow at coastal locations.

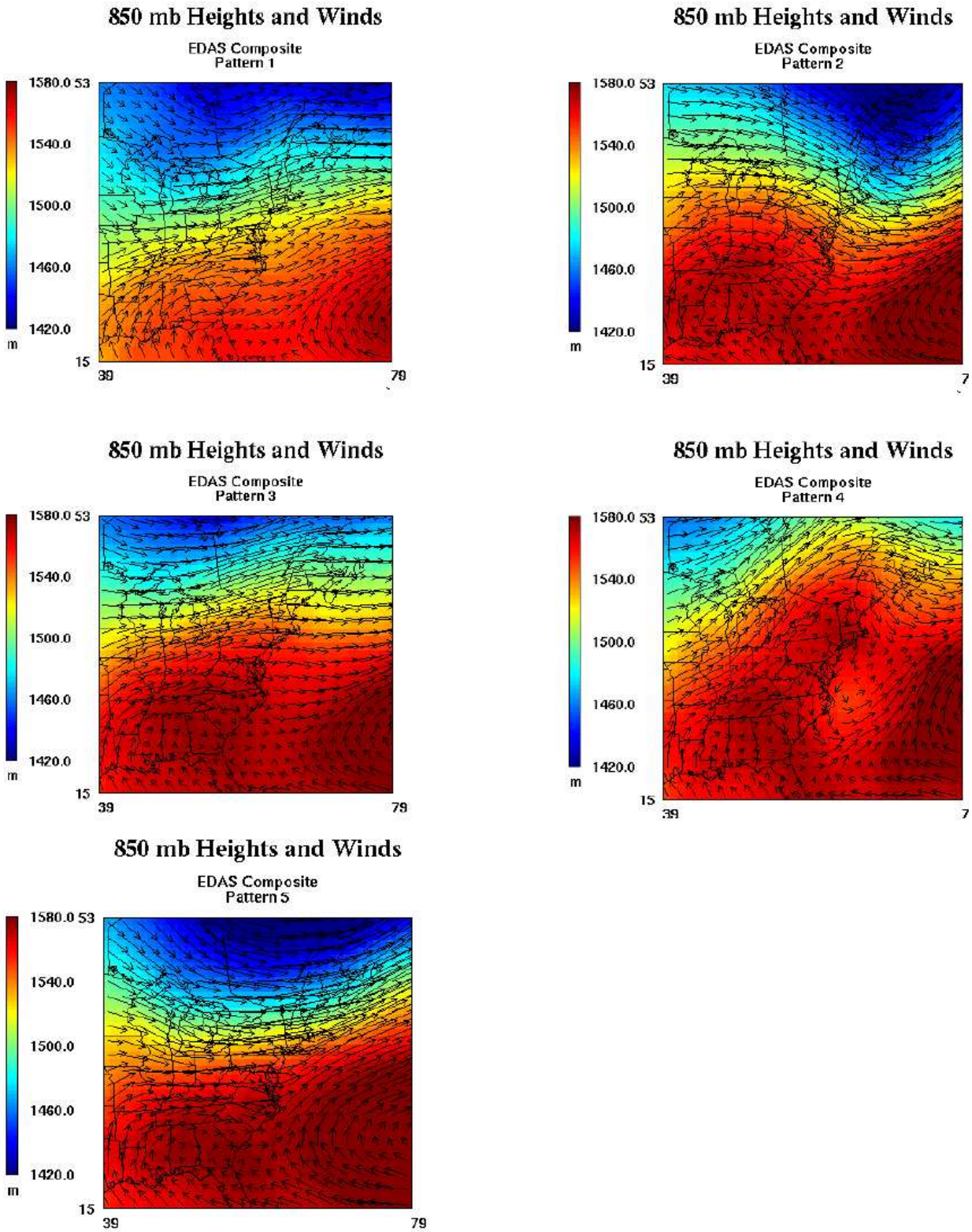

Figure B-1. Average 850 mb height and wind fields for each episode (pattern) type identified by Stoeckenius and Kemball-Cook (pattern numbers refer to the episode types listed in text): Pattern 1 = Episode Type E, 2 = B, 3 = A, 4 = D, 5 = C) (Figure 3-2 of Stoeckenius & Kemball-Cook (2005)).

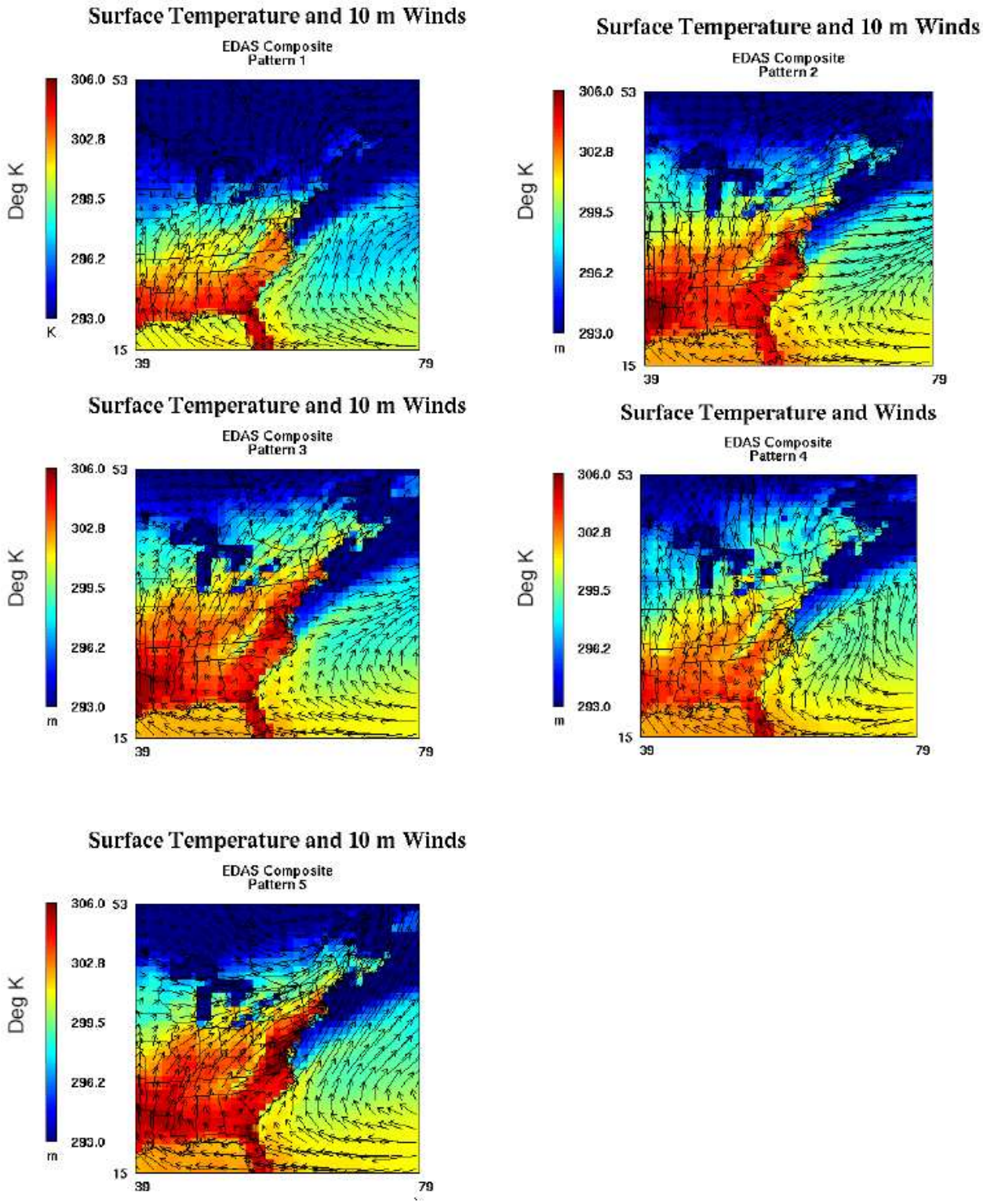

Figure B-2. Average surface temperature and 10 m wind fields for each episode (pattern) type identified by Stoeckenius and Kemball-Cook (pattern numbers refer to the episode types listed in text): Pattern 1 = Episode Type E, 2 = B, 3 = A, 4 = D, 5 = C) (Figure 3-5 of Stoeckenius & Kemball-Cook (2005)).

Appendix C: The sea breeze and flow over the ocean in-depth

Appendix C: The sea breeze and flow over the ocean in-depth

Figure C-1 displays a general description of ozone transport in coastal New England. This figure shows 90th percentile ozone concentration wind direction plots at four sites along the coast. For the first site, Lynn, MA, high ozone days are affected mainly by winds from the southwest bringing ozone up the coast to the site. At the second site, Newbury, MA, winds arrive to the site from two directions, up the coast, in a similar pattern seen at Lynn, but also from the ocean. The high ozone days therefore can result from ozone and its precursors coming from inland or from the ocean in the sea breeze. At the two northern sites in Maine, Cape Elizabeth and Acadia National Park, winds on high ozone days come mostly off the ocean. This is mainly due to the orientation of the Maine coastline, as summertime winds generally come from the southwest, therefore traveling over the ocean before arriving to these sites.

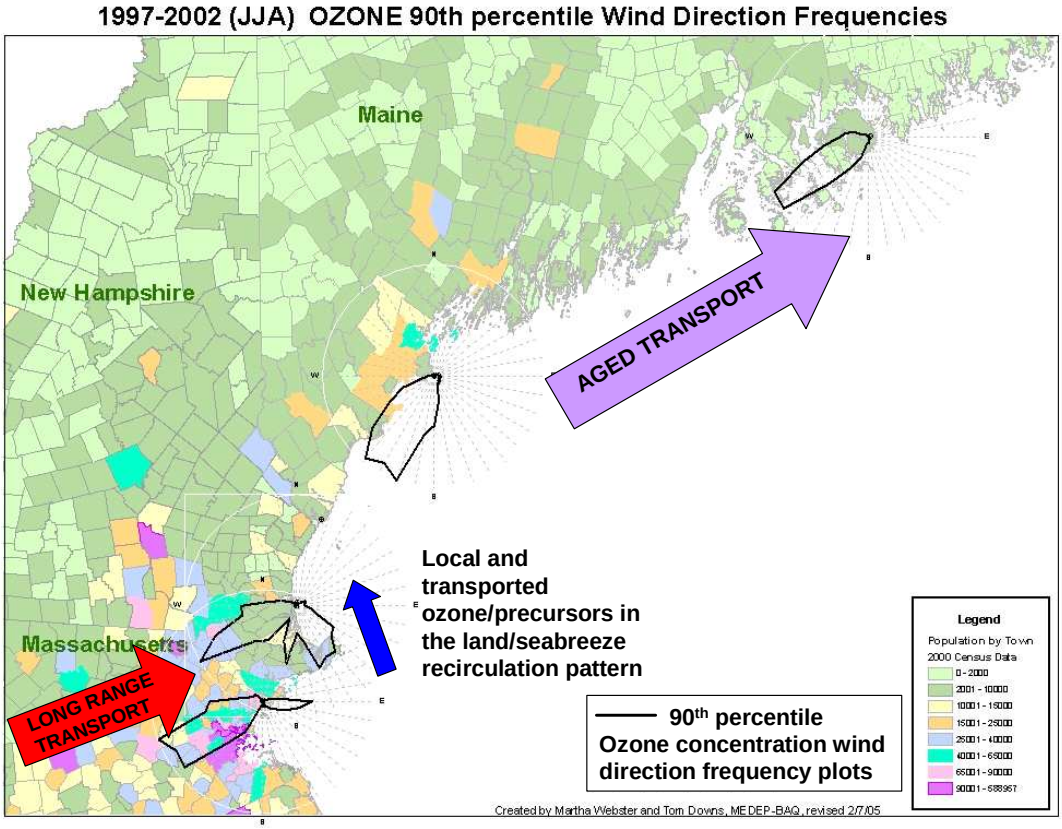

Figure C-1. 90th percentile ozone concentration wind direction frequency plots at four coastal sites in northern New England (figure provided by Tom Downs, Maine Department of Environmental Protection).

Figure C-2 displays wind directions at Newbury, MA on June 29, 1997 where hourly ozone concentrations ranged from 88 ppb to 107 ppb during the afternoon hours and a sea breeze can be identified. The forward trajectory starting in Boston at 6 a.m. shows winds pushing air from the Boston metro area out into the harbor throughout the day. The hourly ozone wind rose at Newbury, MA shows the afternoon wind shift that

occurred on this day where vector direction indicates wind direction and magnitude indicates ozone concentrations. Morning winds came from a west/northwesterly direction when hourly ozone concentrations at the site ranged from 47 to 68 ppb. At 1 p.m., the wind shifted direction, now coming off the ocean from the southeast, accompanied by a 20 ppb increase in hourly ozone. Hourly ozone levels then continued to increase in the early afternoon, peaking at 107 ppb at 3 p.m. This increase in ozone levels accompanying a shift in winds pushing air masses from the ocean to a coastal site illustrates how the sea breeze can contribute to poor air quality along the coast. The poor air quality could be a result of polluted air from Boston being pushed back to the site in the sea breeze. Sea breezes, however, are not always associated with worsening air quality as the afternoon sea breeze doesn't always bring in polluted air.

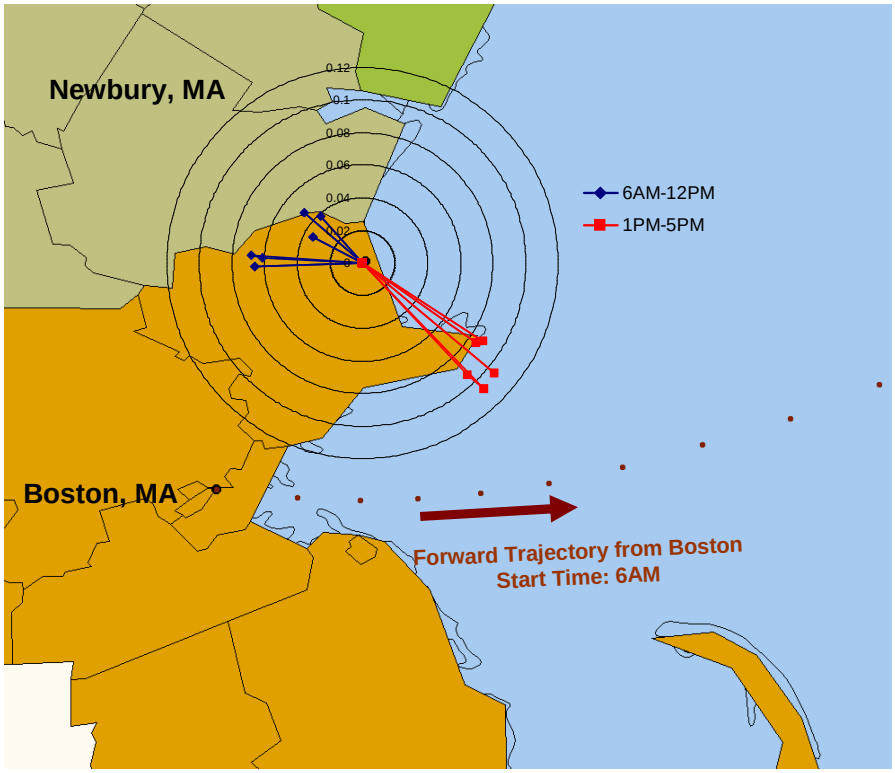

Figure C-2. Example of a sea breeze effect occurring in Newbury, MA on June 29, 1997 (figure data provided by Tom Downs, Maine Department of Environmental Protection).

At sites further north in Maine, the sea breeze effect is less dramatic due to the orientation of the Maine coastline. Figure C-3 shows a similar ozone wind rose plot for Cape Elizabeth, ME on the same day illustrated in Figure C-2. With the exception of the winds at 6 a.m. that came from the northwest, the winds arrived to the site from the southwest direction. There are some slight shifts in wind direction, particularly a shift after 5 p.m. that began to bring winds from the inland side of the coast, but it is difficult to determine whether these shifts are due to a sea breeze effect or if the evening shift is due to the weakened sea breeze. Winds are generally moving up the coast, over water, and winds in the same direction of the sea breeze can bring poor air quality. On this day, ozone concentrations ranged between 89 and 102 ppb between 3 p.m. and 7 p.m.

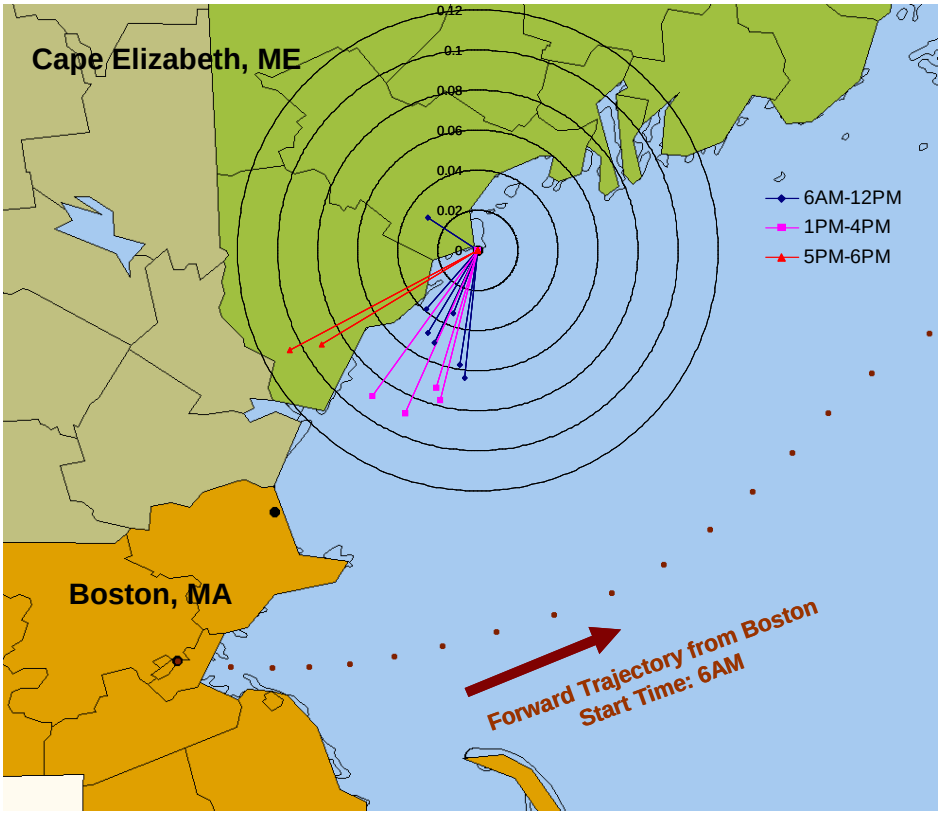

Figure C-3. Wind directions and ozone concentrations at Cape Elizabeth, ME on June 29, 1997 (figure data provided by Tom Downs, Maine Department of Environmental Protection).

Transport over the ocean is commonly observed downwind of the New York City metropolitan area during the summer months due its proximity to the Atlantic Ocean and the Long Island Sound. The four pollution rose plots presented in Figure C-4 represent the frequency of wind direction on the highest 10 percentile ozone concentration days from April 1 to October 31 during the years 1997 to 2005. The winds on the highest ozone days point at the New York City metropolitan area at all locations along the Connecticut shoreline. Going along the Connecticut shoreline to the east (towards Groton), the predominant wind frequency direction shifts increasingly to the west, tracking the upwind location of the New York City metropolitan area.

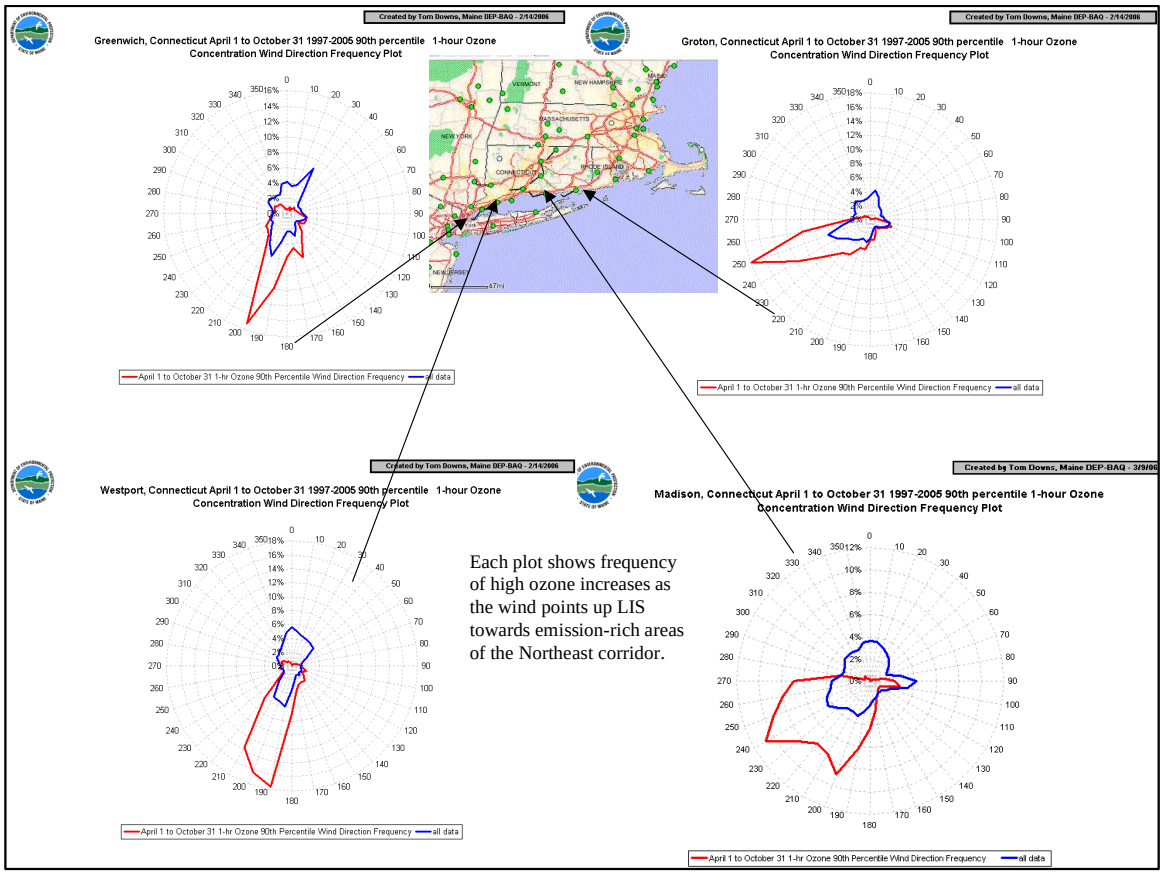

Figure C-4. Wind rose plots along Connecticut shoreline for the time period April 1 to October 31 during the years 1997 through 2005. The elongated red outlines pointing to the southwest to west are wind directions on the highest 10 percentile ozone concentration days at four Connecticut coastal locations. For comparison, the blue outlines are the wind rose plots for all days over the same period. The high ozone day wind rose plots indicate pollution flow over Long Island Sound that tracks the upwind location of the New York City metropolitan area.

Appendix D: Observed nocturnal low level jet across the OTR, July 2002

Appendix D: Observed nocturnal low level jet across the OTR, July 2002

An example of the nocturnal low level jet across the OTR can be seen on the nights of July 22 through July 24, 2002, as night time winds at altitudes between 450 m and 1500 m were observed at several coastal sites. Figure D-1 shows wind profiler data on the night of July 22-July 23, 2002 for five sites along the east coast: Fort Meade, MD (FME), Orange, MA (ORE), Stow, MA (STW), Appledore Island, ME (ADI), and Pease Air Force Base, NH (PSE). These wind "barb" plots show wind direction (direction of arrow indicating where wind is coming from), wind speed (wind barb color), time of day (UTC time, x-axis), and altitude (meters, y-axis). The location of the nocturnal low level jet appears within the circle in each wind barb plot of Figure D-1. The figure shows a weak nocturnal low level jet at the southernmost site, Fort Meade, with wind speeds of 15 to 25 knots between 300 m and 500 m in the early part of the night. Further north, the nocturnal low level jet is more pronounced with wind speeds between 500 m and 1500 m above ground reaching 40 knots. Figure D-1 shows on this day the nocturnal low level jet extending from Maryland up through southern Maine. In addition, the wind barb plots show the northeasterly direction of the nocturnal low level jet. Above this jet, we see slower winds coming from the west to all the sites.

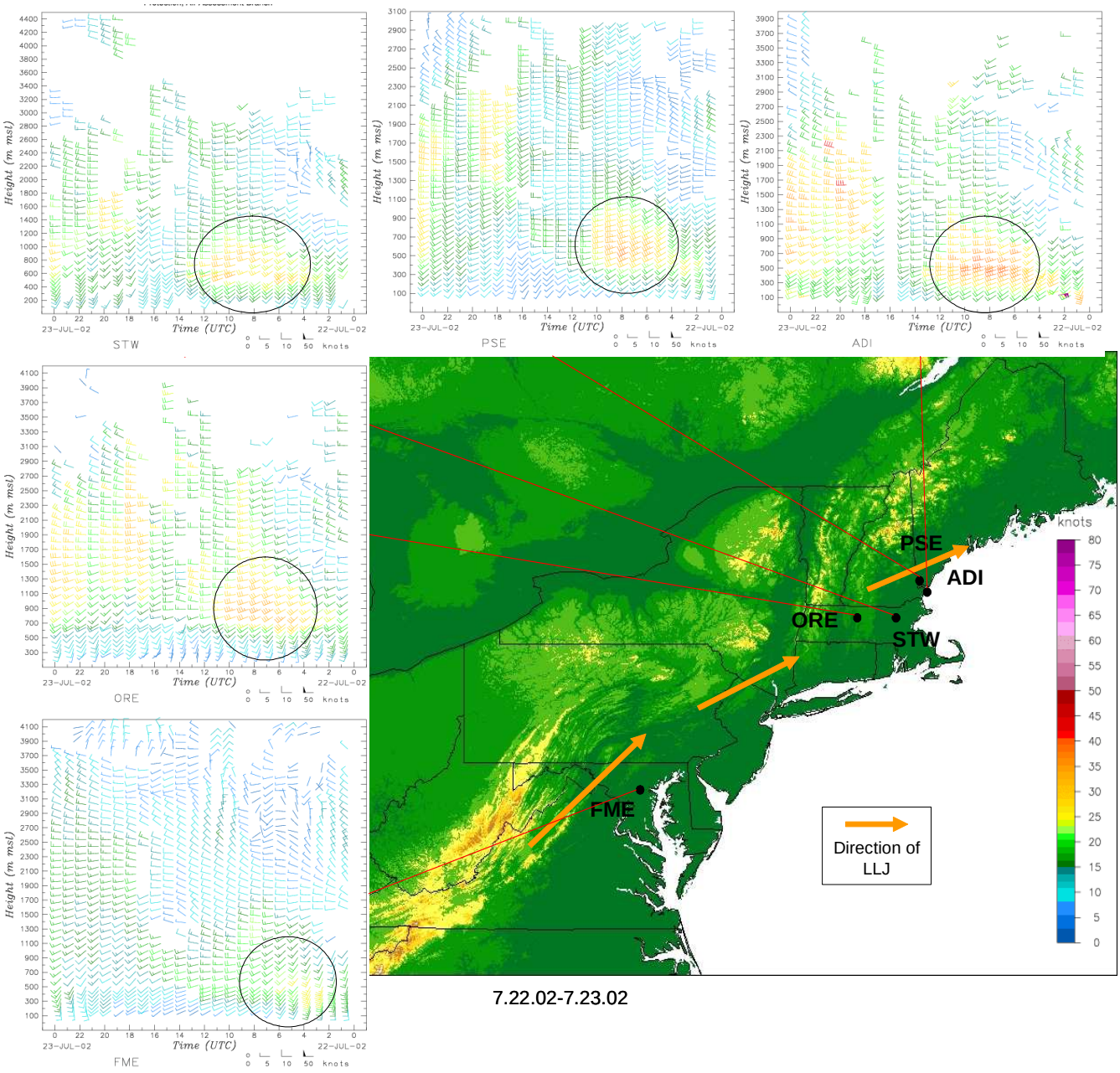

Figure D-1. Nocturnal low level jet on July 22 – 23, 2002. Note: Circles in the wind barb plots indicate the location of the nocturnal low level jet.

Figure D-1 shows that throughout the night, the nocturnal low level jet travels in a northeasterly direction along the east coast. The pollution implications of this nocturnal low level jet episode can be seen in Figure D-2. The Cadillac Mountain ozone monitor is located on the coast of Maine at an elevation of 466 m. At this elevated position, we can see how the nocturnal low level jet affects overnight and early morning ozone levels. Between midnight and 4 a.m. during the northeasterly nocturnal low level jet, hourly ozone concentrations at Cadillac Mountain are between 70 ppb and 80 ppb. Ozone levels

had begun to increase early in the evening on July 22 and continued to increase throughout the night and peak at 3 a.m. This increasing nighttime ozone at an elevated position corresponds to the nocturnal low level jet bringing air up the coast during the night. Conversely, at Cape Elizabeth, a ground level site relatively close to Cadillac Mountain, night time ozone levels are much lower than on top of Cadillac Mountain. This difference in ozone at upper and lower levels shows how the nocturnal inversion can isolate air masses above and below the inversion.

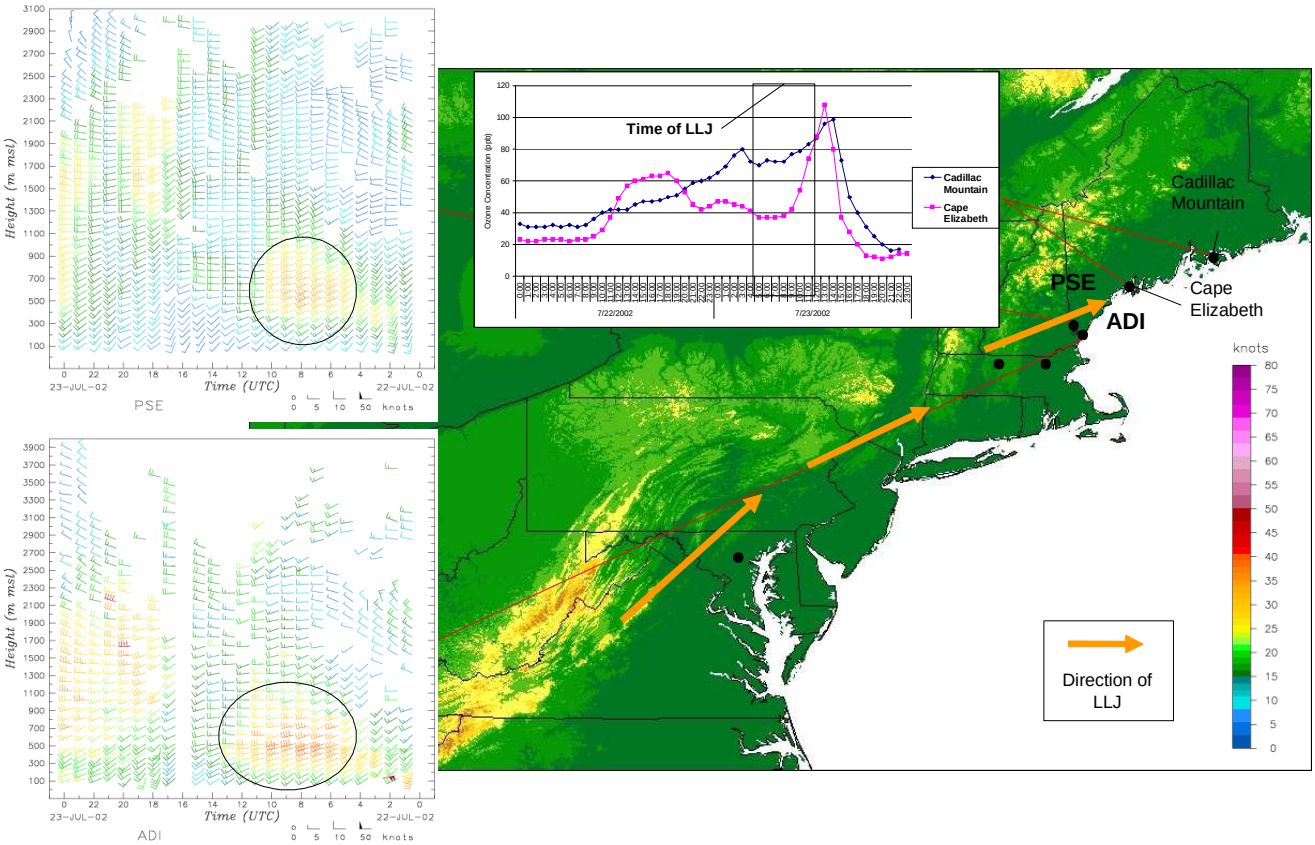

Figure D-2. Nocturnal low level jet with hourly ozone concentrations at Cadillac Mountain, ME and Cape Elizabeth, ME on July 22 – 23, 2002. Note: Circles in the wind barb plots indicate the location of the nocturnal low level jet.

The air mass affecting early morning ozone concentrations in Figure D-2 can be roughly tracked using wind speed and wind direction information from Cadillac Mountain, Pease, Appledore Island, and Orange. Assuming the nocturnal low level jet occurs for five hours that night (based on neighboring wind barb plots), the air mass arriving at Cadillac Mountain at 3 a.m. during peak ozone conditions was over central Massachusetts around 11 p.m. on July 22 when the nocturnal low level jet began to form. Tracking this farther back shows that the air mass affecting Cadillac Mountain was over western Connecticut around 6 p.m. on July 22. Looking at ozone levels in Cornwall, CT, we see that high ozone conditions existed in this region during the afternoon of July 22 with the average hourly ozone at 112 ppb between 4 p.m. and 7 p.m. Elevated ozone from this region first slowly traveled up the coast in the evening. When the nocturnal low level

jet formed, it quickly pushed ozone up the coast affecting ozone levels at Cadillac Mountain, an elevated site in the jet, in the early morning hours (~3 a.m.).

Figure D-3 shows wind profiler information for the next day, July 24, 2002. In this case we see a stronger nocturnal low level jet between midnight and 8am that originates further to the south. The Fort Meade and Rutgers (RUT) sites show the nocturnal low level jet in the early part of the evening with flow in the northeasterly direction. At higher altitudes slower winds from the west pass over the nocturnal low level jet. Further north, a strong nocturnal low level jet can be seen at Stow, Appledore Island, and Pease. It is difficult to determine if a nocturnal low level jet exists at Orange as high winds continue at the upper altitudes and data are missing for the highest altitudes. Figure D-3 demonstrates an example of the nocturnal low level jet passing along the east coast as far south as Maryland and as far north as southern Maine.

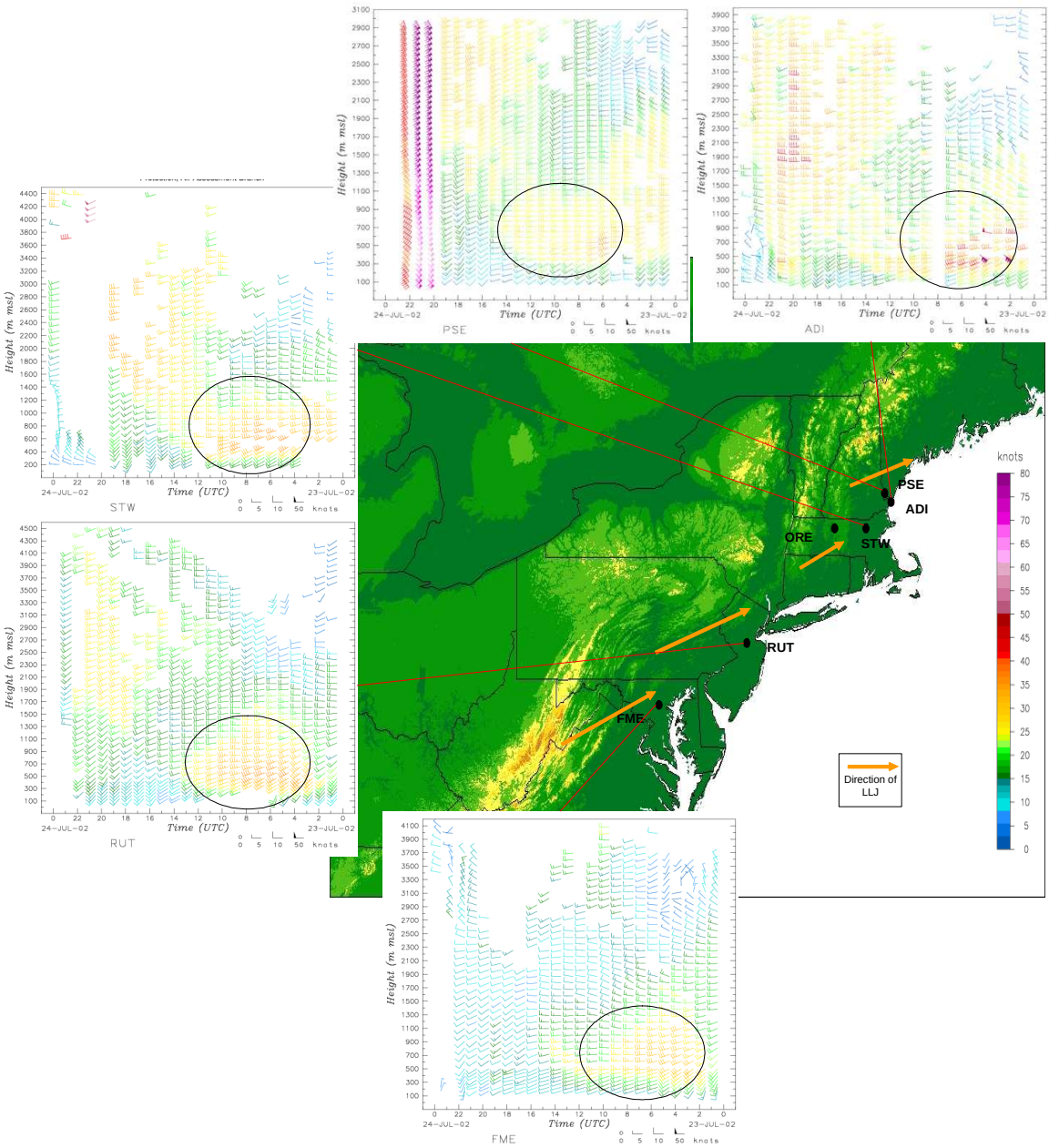

Figure D-3. Nocturnal low level jet on July 23 – 24, 2002. Note: Circles in the wind barb plots indicate the location of the nocturnal low level jet. Data are inconclusive for identifying a nocturnal low level jet at Orange, MA.

Figure D-3 shows that the nocturnal low level jet occurred on the night of July 23-24 as it did on the previous night. Figure D-4 shows ozone levels overnight on the July 23-24 at Cadillac Mountain and Cape Elizabeth. In this case, we see that low ozone is occurring at both sites during the early hours of July 24. Applying the same methods utilized earlier, wind speed and wind direction information from Cadillac Mountain indicate that the air arriving at Cadillac Mountain was also roughly over central Massachusetts at 10 p.m. on July 23 (same wind direction and wind speed as previous day). Wind profiler data show that winds moved this air mass from eastern New York and western Connecticut in the late afternoon. Average ozone levels between 4 p.m. and 7 p.m. were 53 ppb at Cornwall, CT. Therefore, much like on the previous day, air masses were tracked back to the western Connecticut area upwind. In this case, however, low levels of ozone existed in the air mass.

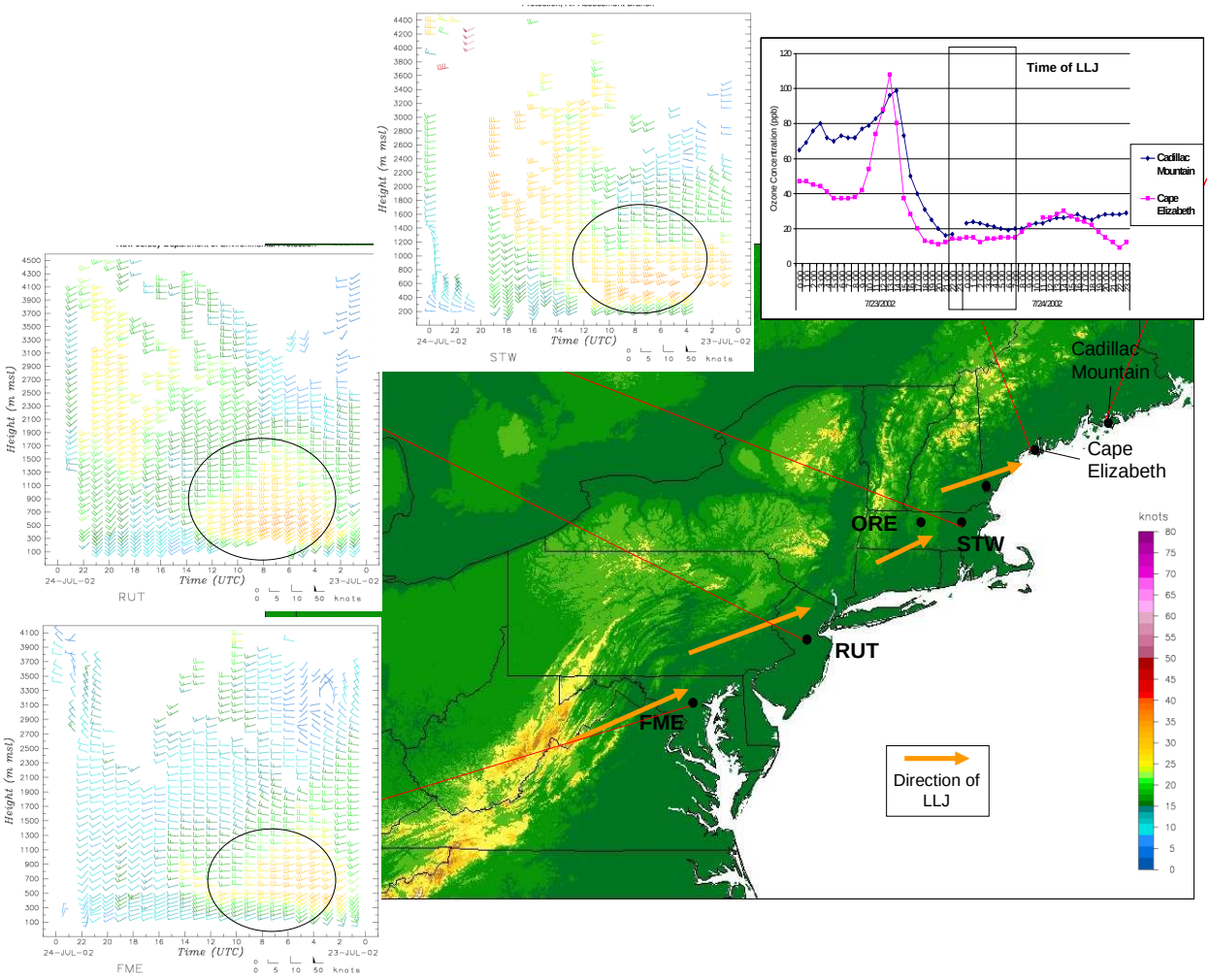

Figure D-4. Nocturnal low level jet with hourly ozone concentrations at Cadillac Mountain, ME and Cape Elizabeth, ME on July 23 – 24, 2002. Note: Circles in the wind barb plots indicate the location of the nocturnal low level jet. Data are inconclusive for identifying a nocturnal low level jet at Orange, MA.

Examining the wind profiler data from 4 p.m. to midnight on July 23 (Figure D-1 and Figure D-3), we see high winds at all altitudes developing throughout the region. Figure D-5 shows that these high winds are part of a weather front that passed through the region in the afternoon of July 23. This corresponds with the sharp drop in ozone levels at Cornwall, CT, Cadillac Mountain, ME, and Cape Elizabeth, ME (Figure D-6) as the front pushed ozone out of the region. This explains the low levels of ozone seen at Cadillac Mountain during the nocturnal low level jet in the early hours of July 24. This example demonstrates that not all nocturnal low level jets are associated with high ozone levels at elevated sites. A necessary condition for the transport of ozone in a nocturnal low level jet is the presence of upwind elevated ozone levels. The front that pushed through the region on the previous day resulted in “clean” air being transported in the nocturnal low level jet.

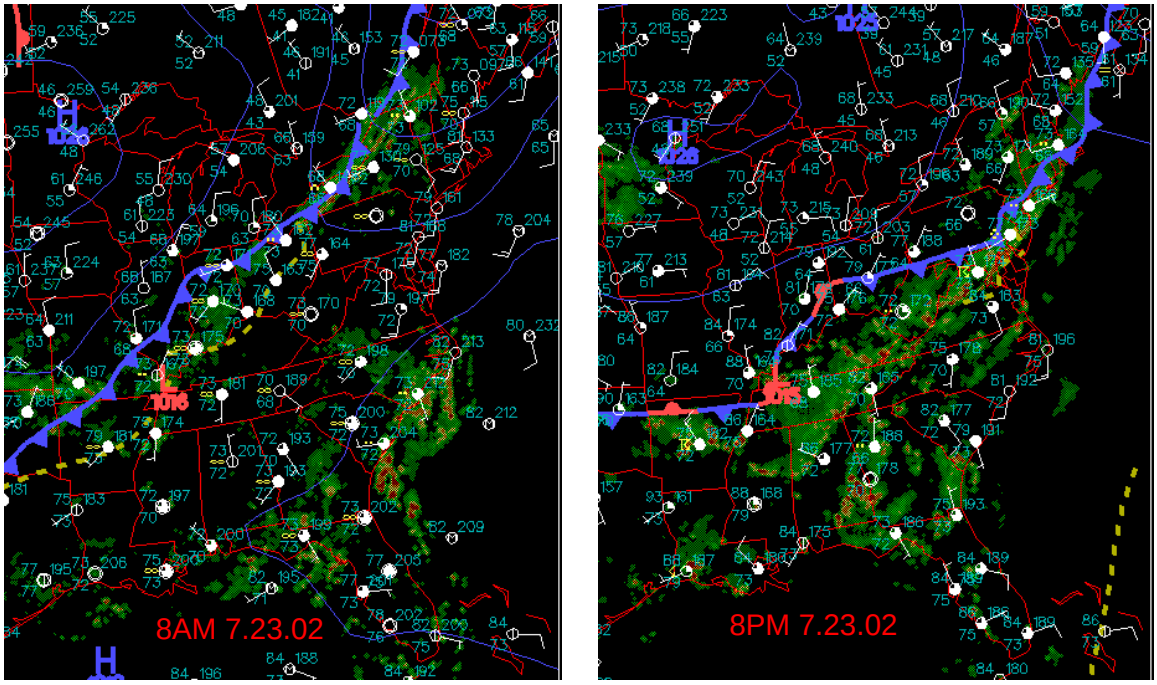

Figure D-5. Weather map displaying a front passing through the East on July 23, 2002.

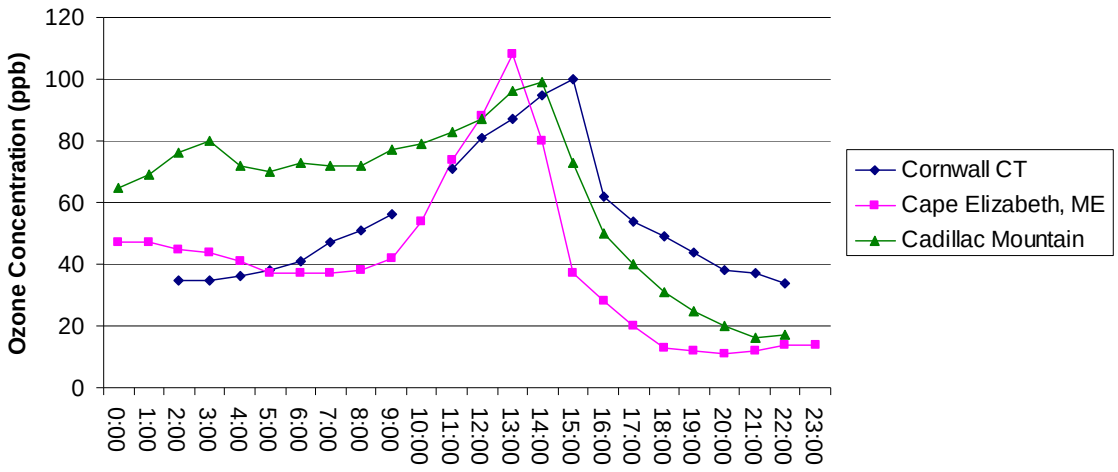

Figure D-6. Hourly ozone concentrations on July 23, 2002 at three sites.

Appendix E: Contributions to the ozone reservoir

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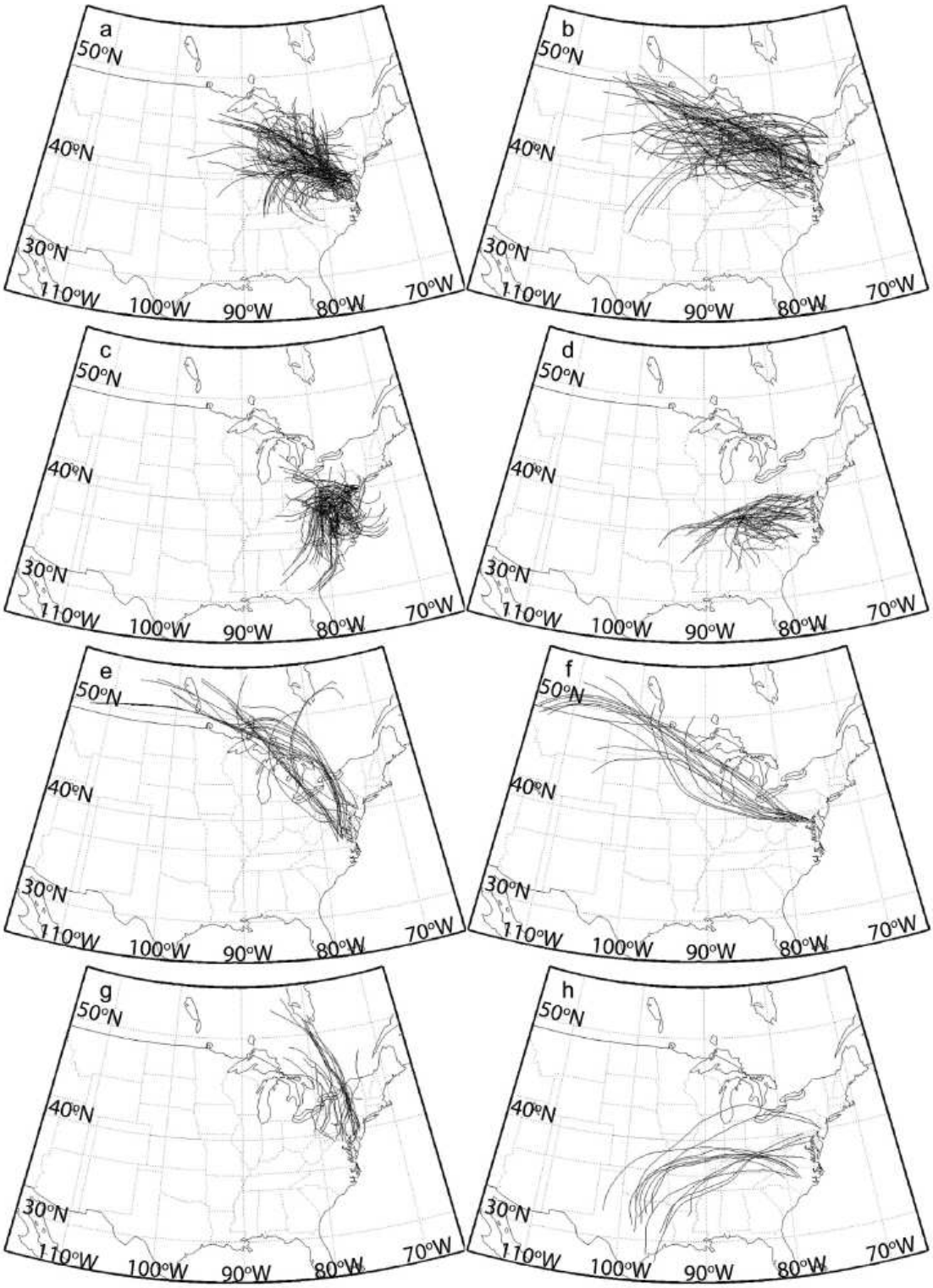
Contributions to the ozone reservoir can come from two sources. The first is from the residual local ozone and precursors in the atmosphere at sunset. The second is from transport of ozone and precursors from outside of the local region. To identify these outside sources, Taubman *et al*. (2006) have made an analysis of the complete set of aircraft flights undertaken by RAMMPP between 1992 and 2003 in the middle and southern portions of the Ozone Transport Region. Initially, the data were divided into morning and afternoon profiles to identify diurnal patterns. Little diurnal variation was observed in the carbon monoxide and sulfur dioxide profiles. The ozone values were greater in the afternoon than the morning, while ozone in the lower free troposphere (i.e., above the boundary level), where long-range transport is possible, was consistently ~55 ppb.

Transport patterns and source regions during summertime haze and ozone episodes were analyzed with a cluster analysis of back trajectory data. Eight clusters were identified, which were then divided into morning and afternoon profiles. Table G-1 lists the characteristics of each cluster, and Figure G-1 shows the back trajectories calculated for each profile divided by cluster at an altitude of 2000 meters. The median profile values were calculated and statistical differences were determined using a nonparametric procedure. When the greatest trajectory density lay over the northern Ohio River Valley, which has large NO_X and sulfur dioxide sources, the results were large ozone values, a large SO_2/CO ratio, large scattering particles, and high aerosol optical depth over the mid-Atlantic U.S. In contrast, relatively clean conditions over the mid-Atlantic occurred when the greatest trajectory density lay over the southern Ohio River Valley and nearly missed many large NO_X and SO_2 sources. The greatest afternoon ozone values occurred during periods of stagnation that were most conducive to photochemical production. The least pollution occurred when flow from the north-northwest was too fast for pollution to accumulate and when flow was from the north, where there are few urban or industrial sources.

Similar results were obtained in the northern portion of the Ozone Transport Region. Periods of ozone greater than 80 ppb measured at the summit of Mount Washington (elevation 1910 m) in northern New Hampshire over the summers of 1998 to 2003 coincided with westerly (71 percent) and southwesterly (29 percent) transport, based on backward trajectory analyses (Fischer *et al*., 2004). Low ozone events were more typically associated with transport trajectories from the northwest. Higher ozone transported from industrialized areas to the west/southwest and lower ozone from the northwest has also been found in back trajectory analyses by Yu *et al*. (2009).

Figure G-1: Maps of the 2 km, 48 hr HY-SPLIT back trajectory clusters for mid-Atlantic region



Note: Cluster groupings are a) cluster 1, b) cluster 2, c) cluster 3, d) cluster 4, e) cluster 5, f) cluster 6, g) cluster 7, and h) cluster 8. Figure from Taubman *et al.*, 2006.

In Taubman *et al*. (2006), ozone transport over several hundred kilometers into the mid-Atlantic U.S. was estimated by calculating the ratio of the residual layer ozone between 500 m and 2 km in the upwind morning profiles to the downwind afternoon boundary layer values between 100 m and 2 km. The greatest level of transported ozone (69-82 percent) occurred when the maximum trajectory density lay over the southern and northern Ohio River Valley (clusters 1, 2, 4, and 6); ~59 percent of the total profiles). The least amount of transported ozone (55-58 percent) was associated with fast southwesterly flow (cluster 8; ~3 percent of the total profiles), fast north-northwesterly flow or clean northerly flow from regions with relatively few urban or industrial pollution sources (clusters 5 and 7; ~6 percent of the total profiles), and stagnant conditions within the mid-Atlantic conducive to greater local ozone production (cluster 3; ~27 percent of the total profiles). The average amount of ozone transported into the Baltimore-Washington urban corridor is 64 percent of the total observed ozone in the afternoon boundary layer. If the background ozone is removed, then this value is lowered to 55 percent.

When trajectory density plots were overlaid on maps with the largest annual NO_X and SO_2 emitters, specific source regions were identified. The results indicate that the areas of maximum trajectory density together with wind speed are effective predictors of regional pollution and loadings. Additionally, due to the Lagrangian nature of the dataset, the regionally transported contribution to the total afternoon boundary layer column ozone content in each cluster could be quantified.

Table G-1. Cluster groups for air mass trajectories into mid-Atlantic Region

Cluster	Description	Upwind Region
1	Large ozone values, large SO_2/CO ratio, large highly scattering particles. Moderate northwesterly flow – aged point source air.	Northern Ohio River Valley
2	Small ozone values, large SO_2/CO ratio. Northwesterly flow at higher wind speeds than Cluster 1 – aged point source air.	Northern Ohio River Valley, extending into the Great Lakes region
3	Large ozone values, small SO_2/CO ratio. Stagnant conditions with light southerly flow.	Central mid-Atlantic region
4	Small ozone values, small SO_2/CO ratio. Moderate southwesterly flow, small pollution loading – fewer point sources.	Southern Ohio River Valley
5	Fairly fast north-northwesterly flow. Flow too fast for pollution to accumulate from source region.	Northern Great Lakes
6	Moderately large ozone values, SO_2/CO ratio very large, smaller less scattering particles. Northwesterly flow, but faster wind speeds than Clusters 1 and 2. Crosses several large SO_2 and NO_X sources.	Northern Ohio River Valley
7	Least pollution of any of the clusters. Flow is out of the north. Relatively cool, dry continental air.	Eastern Ontario, western Quebec
8	Small ozone values, small SO_2/CO ratio. Fast southwest flow. Very few trajectories.	Vicinity of Texas

Consistent results were seen in a trajectory cluster analysis by Hains *et al*. (2007). Groupings of trajectories identified six distinct cluster types associated with ozone in the mid-Atlantic region. Integrated NO_X emissions along the paths of each back trajectory cluster were positively correlated with ozone levels, and large ozone values in several cluster groups were associated with regions of high NO_X source density in the Northern Ohio River Valley.

A study by Davis *et al*. (2010) looked at air mass types coupled with back trajectories that provided results consistent with the studies described above, but which would not have been evident from looking at air mass types or back trajectories alone. For example, warm humid conditions are generally not conducive to ozone formation. Air masses of this type arriving at a site in Shenandoah Valley, Virginia, coming from the north and east have very low ozone concentrations. However, the same air mass type traveling moderate distances from the west at low speed over the Ohio Valley and Midwestern source regions has ozone levels 30 ppb higher than those traveling longer distances from the north or northeast.

References

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