

Article

An Investigation of Two Highest Ozone Episodes During the Last Decade in New England

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Abstract: This study examined the role of meteorological processes in two of the highest ozone (O₃) episodes within the last decade at monitoring sites in southern New Hampshire (NH), USA. The highest O₃ levels occurred on 14 August 2002 at Thompson Farm (TF) and 22 July 2004 at Castle Springs (CS). Ozone mixing ratios in the 2002 episode showed continual high values (>100 ppbv) at the beginning of the episode, and reached 151 ppbv on 14 August. The 2004 episode consisted of one day of high O₃ (>100 ppbv) on 22 July at CS with the peak level of 111 ppbv. Our analysis suggested that the August 2002 high O₃ event at TF occurred under stagnant synoptic high-pressure conditions that prevailed over the entire eastern USA for an unusually extended time period. The clear skies and stable meteorological conditions resulted in accumulation of pollutants in the boundary layer. At the same time, the mesoscale low-level-jet (LLJ) played an important role in transporting air masses from the polluted Mid-Atlantic areas to the Northeast. Local land-sea-breeze circulations also added to the impact of this event. Our examination showed that the unprecedented high levels of O₃ on 22 July 2004 at CS was driven by two mechanisms, stratospheric intrusion and the Appalachian lee trough (APLT), which was not found during other O₃ episodes at the site in the decade long data record. This study demonstrated that unusually high O₃ levels at New England rural sites were driven by multi-scale processes, and

the regional/local scale processes controlled the magnitude and timing of the local pollution episodes.

Keywords: O₃ episode; meteorological impact; synoptic; mesoscale processes

1. Introduction

Ground-level ozone (O₃) formation is dependent on a series of complex chemical reactions with nitrogen oxides (NO_x) and volatile organic compounds (VOCs), and is also sensitive to meteorological conditions, such as solar radiation, temperature, and wind speed/direction. Tropospheric O₃ is one of the six criteria pollutants, defined by Environmental Protection Agency (EPA), and it is considered harmful to plants and human health. To strengthen the protection of public health and welfare, EPA has proposed the latest 8-hour primary O₃ standard and lowered it from 80 ppbv down to 75 ppbv.

Many urban and rural areas throughout the United States are plagued by high levels of O₃. Numerous studies have reported widespread O₃ episodes during the summertime across the eastern United States [1-4]. Previous studies of elevated O₃ episodes have shown that the spatial and temporal patterns of O₃ in the eastern United States encompass various scales of air circulation and topographic influence [5-7]. On the synoptic scale, studies have demonstrated that a common characteristic of many elevated O₃ events in the eastern USA was the presence of a slow moving high-pressure system [1,3,8-16]. The weather under high-pressure systems is typically clear skies, high temperatures, and the stable meteorological conditions, which promotes O₃ formation and also tends to trap pollutants within the boundary layer [1,10,17-20]. In the Northeastern rural areas where less anthropogenic sources are present, an important source of pollutions is the southwesterly flow associated with the anticyclone flow of a high-pressure system bringing pollutants from upwind industrial areas to this region [15].

Another synoptic mechanism potentially affecting surface O₃ mixing ratios is stratospheric subsidence. Some studies have found that several high O₃ events during summertime were dramatically influenced by injection of O₃-rich stratospheric air. Occurrence of stratospheric subsidence is most frequent in spring. However, studies have found that it also occurs in summer, which dramatically enhances tropospheric O₃ levels [21-27]. The identification of stratospheric injections relies directly on vertical sounding measurements and aircraft observations, or indirectly on model simulations. The work most relevant to this mechanism is the Intercontinental Transport Experiment (INTEX) Ozonesonde Network Study in 2004 (IONS-04) [27,28], which launched nearly 300 O₃ soundings during July and August at eleven sites in North America. Several studies have analyzed the IONS-04 data for the tropospheric budget of O₃, and also examined interaction between impacts of the stratospheric intrusion on tropospheric composition in areas of Canada, Europe, and the USA [27-35]. In the eastern USA, the contribution of stratospheric subsidence to the tropospheric O₃ budget in summer 2004 was around $23 \pm 3\%$ [27,28].

In general, synoptic systems dominate the distributions of O₃ patterns and affect the intensity and duration of O₃ episodes over the eastern USA; regional or local smaller meteorological scales control the magnitude and timing of pollution episodes individually [6,13]. One of the significant regional scale

transport mechanisms influencing New England air quality is the nocturnal low-level-jet (LLJ). Many studies have focused on the correlation between the LLJ and air quality in the eastern United States [6,15,36-42]. For instance, Banta *et al.*, [38] demonstrated that the high O₃ events in rural areas were often caused by the nighttime transport and subsequent daytime downward mixing processes. Seaman *et al.* [6] presented a case study of 14–15 July 1995 being contributed by LLJ transport under weak synoptic forcing. Mao and Talbot [15] speculated that the LLJ transported O₃ and O₃ precursor-rich air masses overnight from the northeast urban corridor to New England in the residual layer, which were brought down to the surface during the day.

Another well-known regional scale transport mechanism is the Appalachian lee trough (APLT). In an earlier study, Pagnotti [43] found that the APLT was associated with around 70% of the high O₃ cases in the Mid-Atlantic States. Model simulations by Seaman [6] suggested that APLT favored accumulation of pollutants on the eastern side of the trough, causing elevated O₃ along the Northeastern urban corridor, and associated upward flows lifted pollutants from surface. Gaza [44] and Kleinman *et al.* [45] also demonstrated the role of APLT in occurrence of high O₃ days in the northeastern urban corridor using a combination of measurement data and backward trajectories. The model results from Mao and Talbot [15] demonstrated that APLT facilitated effectively the transport of O₃-rich air masses to inland locations in southern New Hampshire.

Along the coastal regions of Massachusetts (MA), New Hampshire (NH), and Maine (ME) a local scale of land-sea circulations commonly develops and impacts air quality during summertime [6,15,44,46-51]. Coastal New England can receive pollutants which are transported from upwind polluted areas around 50–500 km distances away and cause O₃ to exceed national air quality standards [51]. During the past few decades, studies have demonstrated many elevated O₃ episodes resulting from sea breeze circulations. For example, Gaza [44] concluded that the sea breeze was an important factor in the 4 July 1995 tropospheric O₃ pollution event in southern New England and New York State. Another high O₃ case associated with the sea breeze was identified on 12–16 July 1995 in the northeastern United States by Zhang *et al.* [13]. Mao and Talbot [15] found that surface O₃ mixing ratios greater than 90 ppbv at Rye, NH during 1 May to 3 November 2001 were coincided with occurrence of the sea breeze.

New Hampshire is located in the downwind region of these transport pathways that usually pass through metropolitan/industrial areas in the eastern USA. As a result, the air quality in rural areas of NH is not only dependent on its local pollutant production, but also is highly affected by the different transport pathways from other source regions. The AIRMAP program has nearly a decade of chemical compound records and meteorological data at several sites [52] for studying air quality and climate change in the New England region. The focus of this study was to examine the two highest O₃ episodes over the past decade captured in the AIRMAP measurements. Thompson Farm (TF) had a continual 4-days of high O₃ (>100 ppbv) from 11 to 14 August 2002 with 1-hour-daily maximum O₃ values increasing from 100 ppbv on 11 August to 151 ppbv on 14 August. In 2004, Castle Springs (CS) had one day of high O₃ (>100 ppbv) on 22 July. Although these events resulted in the highest O₃ mixing ratios at these sites during the last decade, the dominant meteorological conditions were quite different. The meteorological conditions during these two event periods were reported by NOAA National Climate Data as climate extremes with an unusually hot and dry period in 2002, and a cool and wet time

locations of Figure 1. Detailed information on the IONS-04 mission can be found in the studies of Cooper *et al.* [29], Thompson *et al.* [27,28], and Tarasick *et al.* [32].

Volatile organic compounds (VOCs) in this study were collected at TF from July to mid-August every 40 min. Details about the VOCs sampling and analysis are described in Sive *et al.* [56], Zhou *et al.* [57,58], and White *et al.* [59]. Daily meteorological patterns were identified using $1^\circ \times 1^\circ$ meteorology data from National Centers for Environmental Prediction (NCEP) Global Final Analysis (FNL) [60]. Analysis of potential vorticity was performed to identify stratospheric intrusions using the NCEP reanalysis $2.5^\circ \times 2.5^\circ$ dataset from NCAR/NCEP [61]. Skew-T and vertical potential temperature (θ , K) observational data were provided by National Weather Service sites archived at Plymouth State Weather Center [62]. Wind directions and speeds were obtained from NOAA radar wind observations together with local environment observatories at AIRMAP and surface data from National Weather Service sites archived at Plymouth State Weather Center.

3. Results and Discussion

3.1. 2002 Case

During this episode O_3 mixing ratios at TF reached 151 ppbv on 14 August, 2002, which was the highest level observed during the ten years of AIRMAP observations from 2001 to 2010 (Table 1). In fact, the EPA AIRNOW records and NH State data show that O_3 had not peaked this high in New England since the late 1980s. Other AIRMAP measurement sites in the Northeast also exhibited coincidentally high O_3 , such as at CS (88 ppbv) and on Appledore Island in the Gulf of Maine (154 ppbv). In the following sections we focus on the meteorological conditions and the air mass transport mechanisms which shaped this extremely high O_3 episode at TF.

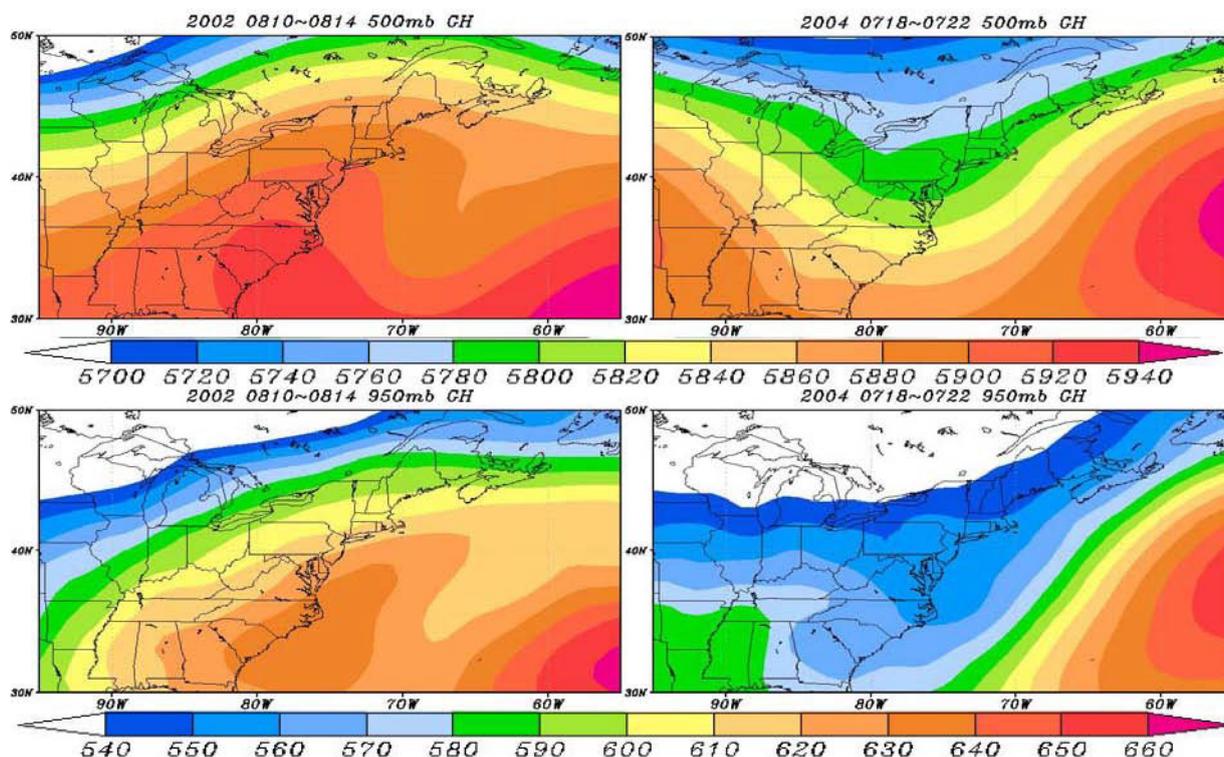
Table 1. The highest O_3 mixing ratio in summers from 2001 to 2010 at TF and CS. Table included the date and time in UTC of seasonal maxima.

Ozone Max During June~September		
Year	TF	CS
2001	100 (6/20, 0-3UTC)	82 (8/20, 8UTC)
2002	151 (8/14, 21UTC)	88 (8/15, 3UTC)
2003	137 (6/27, 18UTC)	90 (7/31, 00UTC)
2004	117 (7/22, 18UTC)	111 (7/22 22UTC)
2005	108 (9/14, 20UTC)	92 (6/10 4UTC)
2006	98 (7/17, 21UTC)	81 (7/26, 2UTC)
2007	113 (8/30, 19UTC)	84 (9/8, 2UTC)
2008	106 (7/18, 20UTC)	-
2009	84 (5/22, 17UTC)	-
2010	86 (7/28, 23UTC)	-

3.1.1. Synoptic Analysis

The meteorological conditions during the August 2002 episode were illustrated using the NCEP $1^\circ \times 1^\circ$ meteorological reanalysis data [60]. Meteorology in August 2002 was characterized by dry weather and high temperatures. The NOAA Climate Prediction Center reported that the weekly temperature average for 11–17 August 2002 in the Northeast was +5 to +8.3 °C higher than the 30 years average for the same week over 1961–1990. A steady high (H)-pressure system was positioned over the eastern USA coastline, as demonstrated by the 5-day (10–15 August) average geopotential height at 500 hPa and 950 hPa (Figure 2). At 950 hPa on 10 August an H-pressure centered over the eastern coastline and its range covered most of the coastal northeast urban corridor extending from southwest to northeast. From 11–13 August, the H-pressure system moved slightly southward along an eastward approaching trough with its influence remaining over the eastern coastline. Circulation of the inland H-pressure inter-connected on 13 August with the oceanic Bermuda H-pressure system which was located offshore of South and North Carolina (SC and NC). Meanwhile, the center of the inland H-pressure extended a ridge from West Virginia (WV), to Pennsylvania (PA), New York (NY), Vermont (VT), and NH. At this time, the northeast urban corridor was on the east side of the ridge, and this area experienced subsidence flow. On 14 August the center of the inland H-pressure moved eastward off the continent and merged with the oceanic Bermuda H-pressure system. Strong southwesterly flow dominated the eastern coastline, which was caused by the large pressure gradient formed by the eastward approaching trough and the westward extending oceanic H-pressure system.

Figure 2. NCEP $1^\circ \times 1^\circ$ geopotential height in 5-days average in 2002 (left) and 2004 (right) at 500 hPa (top) and 950 hPa (bottom). Shaded interval is 10 hPa.



During 10–14 August, a stable and deep boundary layer was shown in the regional potential temperature (θ) profiles from sounding data collected at KALB (Albany County, NY, (42.75°N, -73.80°W)), KOKX (Upton, NY, (40.87°N, -72.87°W)), and KCHH (Chatham, MA, (41.7°N, -69.9°W)), KIAD (VA, (38.95°N, -77.45°W)), and KGYX (Gray, ME, (43.90°N, -70.25°W)) (listed in the blue locations in Figure 1), obtained from National Weather Service sites. The θ -profile is provided for one or two days before the episode date depending on the availability of sounding data. The boundary layer height at KIAD (Figure 3) was around 2.5–3 km at 12 UTC. There is a stable nocturnal boundary layer from the surface to near 500 m, and a mixed layer from 500 m to 2500–3000 m during 9–14 August. Similar sounding results showed at other locations as well. Moreover, satellite images showed clear sky over the east coast due to the subsidence flow of the H-pressure system [63]. Under conditions of clear skies, high temperatures, light winds, and subsidence flow on the synoptic scale, the region experienced a build-up of pollutants. The CS site, at 400 m elevation is above the nocturnal inversion [53], and provides information on the regional buildup of O_3 in the remnant boundary layer. Ozone mixing ratios averaged over the nighttime period of 0000–1100 UTC for non-episode days in August 2002 ranged from 20 to 40 ppbv at CS. However, during the episode (11–14 August) values were 60–80 ppbv (Figure 4), indicative of regional build-up of O_3 .

We also examined the EPA hourly O_3 observations across the eastern U.S. during 10–14 August, and these are shown in Figure 5. It is apparent that O_3 mixing ratios were rising every day at most measurement sites. Elevated O_3 (128–144 ppbv) first appeared around the Great Lakes, Ohio Valley, and in New Jersey (NJ). Then from 10–13 August the high O_3 area moved eastward. This seemed to be coincided with the motion of the H-pressure. On 14 August, the area of elevated O_3 extended northward reaching northern New England. It should be noted that higher O_3 values (>112 ppbv) persisted from 10 August through 14 August in the Mid-Atlantic States which were consistently under the influence of H-pressure. Such simultaneous occurrence of H-pressure system and O_3 episodes were also documented for this region in earlier studies [15,16].

Figure 3. Potential temperature (Theda: θ) vertical profile at KIAD from 9–16 August 2002. X-axis is θ values; Y-axis is height in meter (m).

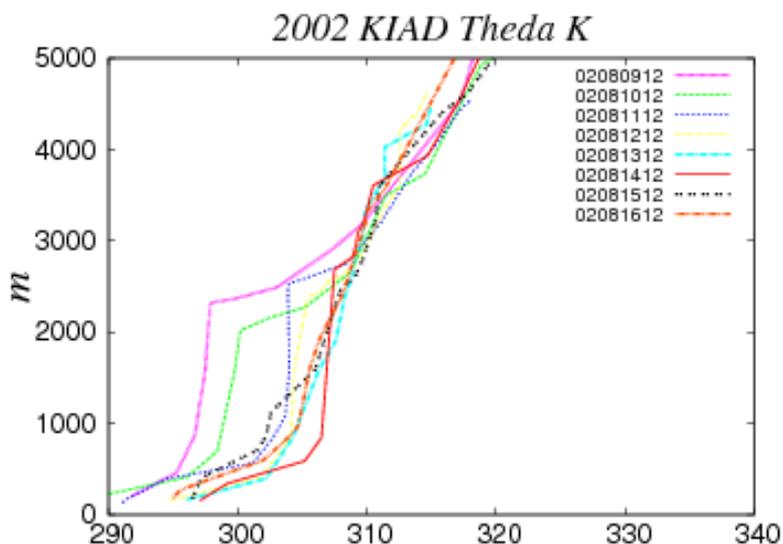


Figure 4. Ozone averages during nighttime (00-11UTC) during 1–31 August 2002 at CS. The dotted line is the monthly-nighttime-average of August.

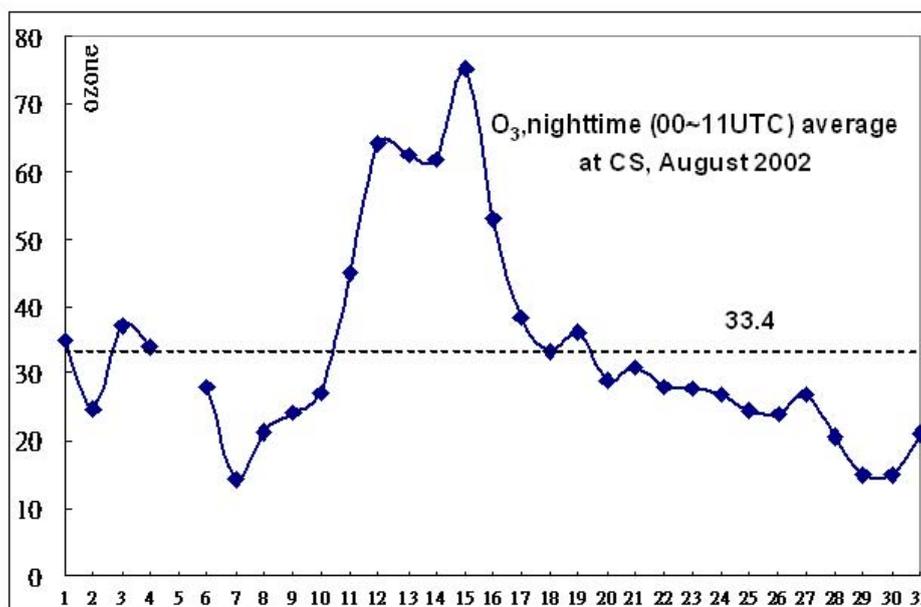
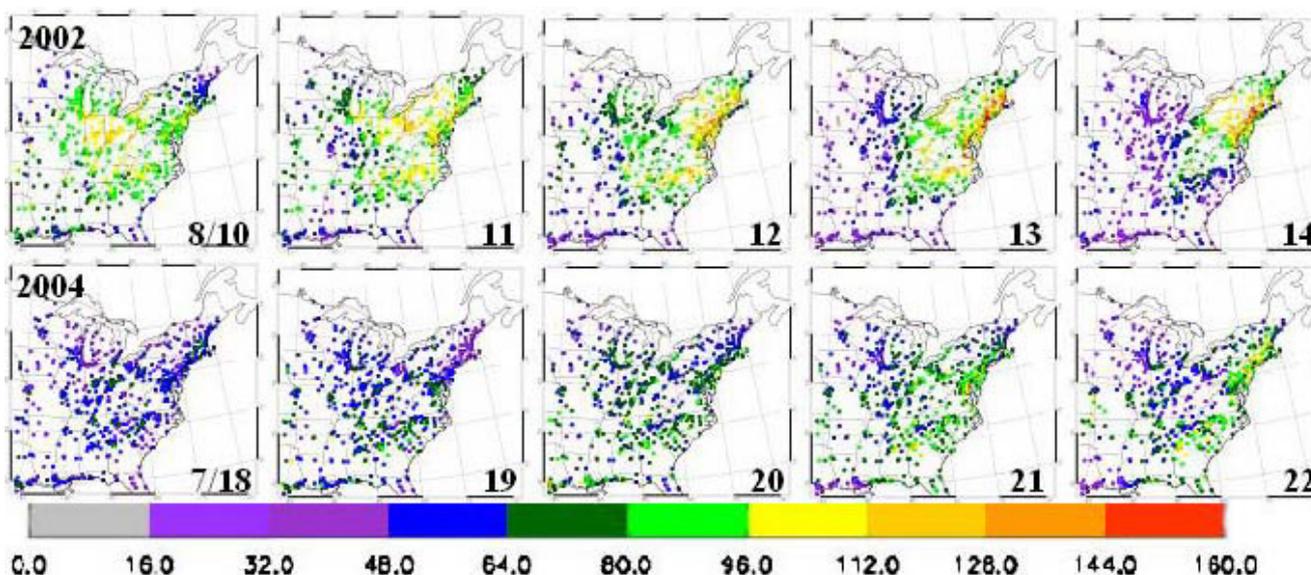
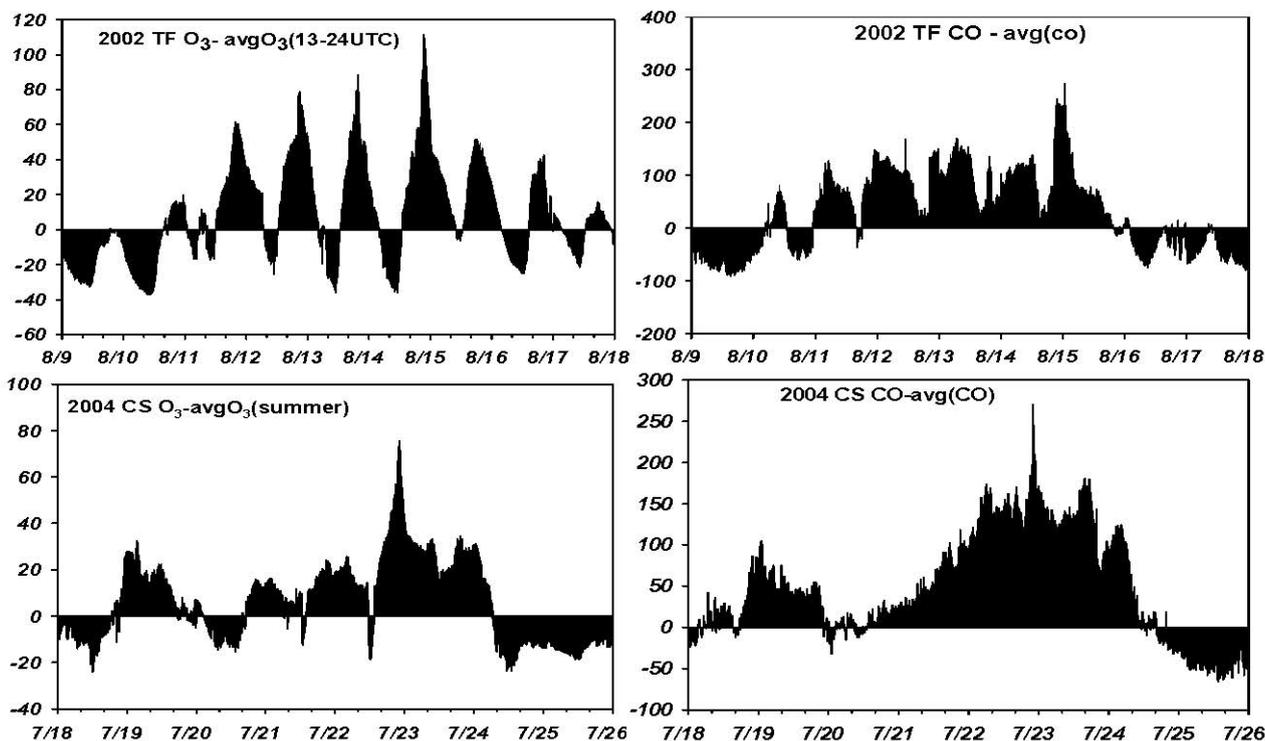


Figure 5. AIRNOW hourly ozone data on 10–14 August 2002 (top), and 18–22 July 2004 (below). Values range is from 0 to 160-ppbv. The color interval is 16-ppbv.



Changes in mixing ratios of other pollutants, such as CO and NO_y, for the episode are presented in Figure 6 (We only present the pattern of O₃ and CO here. NO_y had similar patterns) as deviation from their summer average values (denoted as ΔCO and ΔO₃, *i.e.*, anomalies). The values of ΔCO showed positive anomalies from 11–15 August and the CO levels were more than 100 ppbv higher than the summertime average. On 14 August ΔCO was elevated up to ~250 ppbv. Similar positive anomalies during the period of the event were also observed for O₃ and NO_y (not shown). The trends of these two pollutants during the episode again strongly suggest that the week-long calm weather and reduced ventilation caused a built up of pollution in the boundary layer.

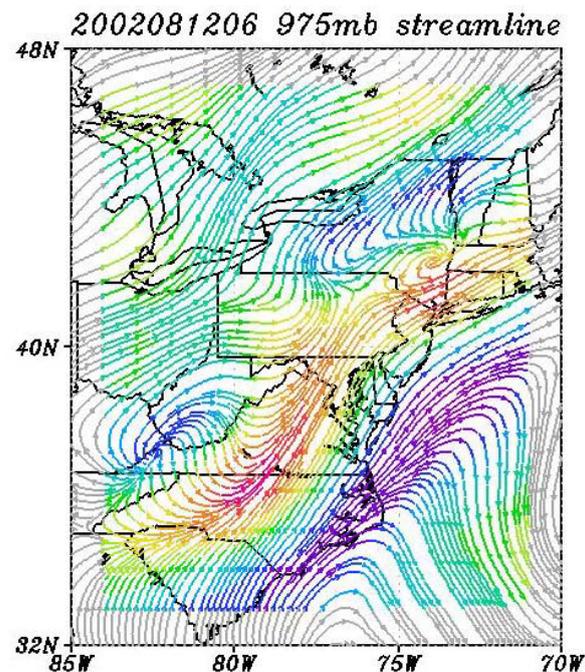
Figure 6. Top: The ΔO_3 (left) and ΔCO (right) (Δ = actual observed data minus its summer average) at TF during 9–19 August 2002; Below: The ΔO_3 (left) and ΔCO (right) at CS on 18–26 July 2004.



3.1.2. Mesoscale Influence

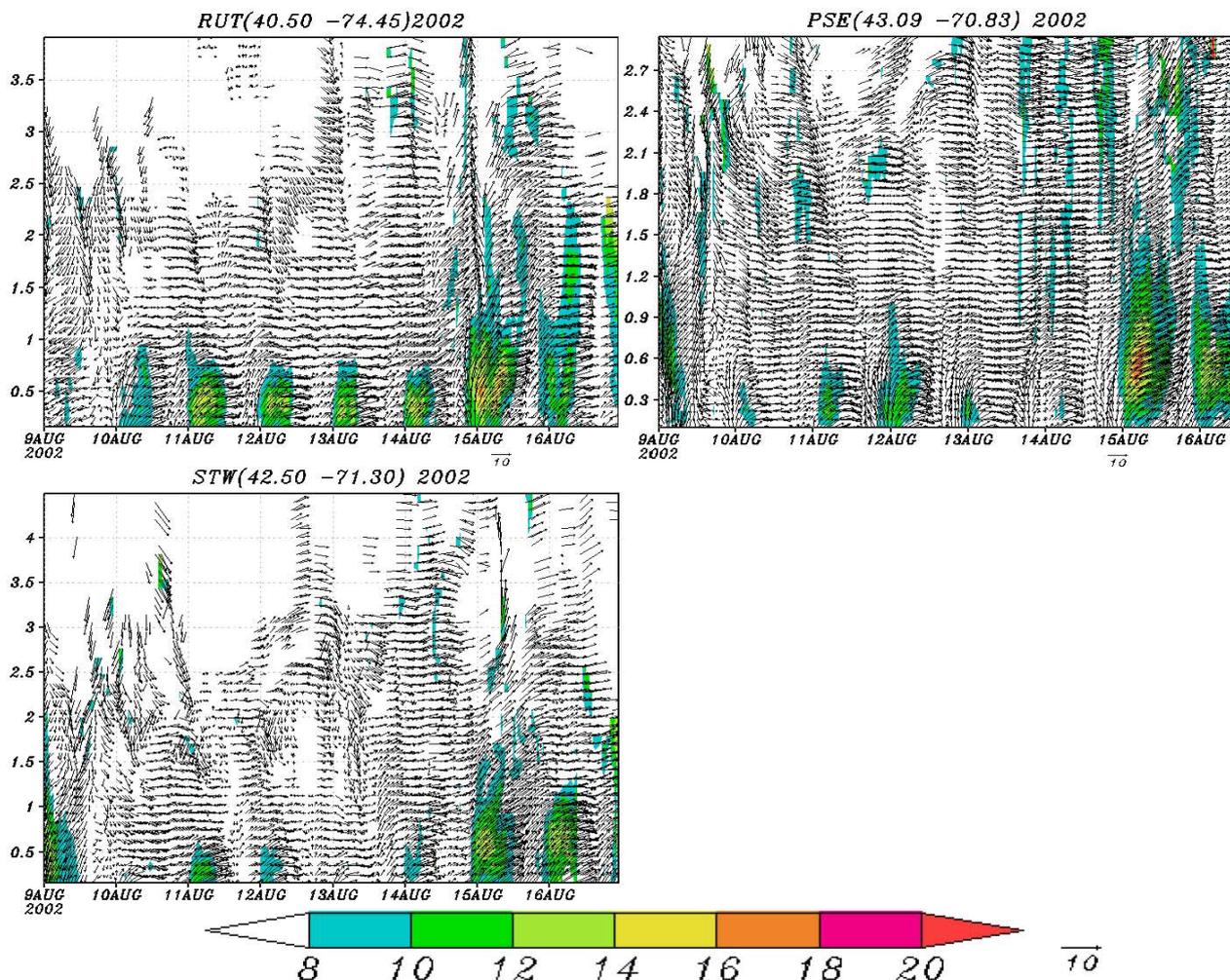
On meso-scale the low-level jet (LLJ), sea breeze circulation, and APLT were also identified in previous studies as efficient transport mechanisms to elevate O_3 along the Northeastern coastline [13-15,40,44,45]. The structure of the LLJ is shown on a geographic map (Figure 7) with cross-sections of vertical wind profiles from the NOAA radar wind observations (Figure 8) located at New Brunswick, NJ (40.5°N, -74.45°W) (RUT), Pease, NH (43.09°N, -70.83°W) (PSE), and Stow, MA (42.5°N, -71.3°W) (STW). The geographic map with streamlines from NCEP 1° × 1° meteorology data demonstrates southwesterly flow along the eastern coastline toward NH at night during the event (here we presented the streamline map on August 12 in Figure 7). Meanwhile, the vertical wind profiles from radar observations (Figure 8) showed strong southwesterly winds on August 10 of 10~14 m/s and on 11–13 August up to ~14 m/s at late night and early morning hours at altitudes below 500 m. These meteorological patterns are characteristic of the nocturnal LLJ [15,40].

Figure 7. The NCEP $1^\circ \times 1^\circ$ data of streamline at 975 mb on 12 August 2002 at 06UTC.



In addition, to illustrate the influence of the LLJ transport to air quality, we examined variability in pollutants such as O_3 , CO, $PM_{2.5}$ and NO_y . Mao and Talbot [15] pointed out that the average diurnal amplitude for CO is around 20 ppbv in rural New England. Moreover, the North East Oxidant and Particle Study (NEOPS) pointed out that the transport of air masses by the LLJ was usually associated with simultaneously high O_3 and $PM_{2.5}$ [64]. In this event, the averaged $PM_{2.5}$ level at TF was $\sim 32 \mu\text{g}/\text{m}^3$ compared to its summer average of $\sim 18 \mu\text{g}/\text{m}^3$, while the average O_3 mixing ratio was 76 ppbv compared to its 2002 summer daytime average of ~ 40 ppbv. The daily amplitudes of CO, $PM_{2.5}$, and NO_y mixing ratios were examined from 1600 UTC (local noon time) to 0400 UTC (local midnight) during 10–14 August. It was found that CO mixing ratios rose from 146 at noontime on 10 August to 310 ppbv at 0400 UTC on 11 August, $PM_{2.5}$ from 13 to $19 \mu\text{g}/\text{m}^3$, and NO_y from 3.3 to 5.9 ppbv. These pollutants greatly exceeded their summer average values at midnight. During the episode there was consistently a stable nocturnal boundary layer, as identified in the 1200 UTC profiles at KIAD in Figure 3. The LLJ (Figures 7 and 8) was above the nocturnal boundary layer and the wind speed was sufficiently fast to transport polluted air overnight along the urban corridor toward the northeast [6,15,44]. Then, when the nocturnal boundary layer was eroded by thermal and turbulent mixing after sunrise, remnant boundary layer air was mixed down to the surface [53], elevating the mixing ratios of O_3 , CO, NO_y , and $PM_{2.5}$. Due to poor ventilation in the region resulting from the stagnant H-pressure system, the effects of the LLJ nighttime transport and morning mixing down were accumulative during the episode leading to the increasing trends in nighttime mixing ratios and daily amplitudes of pollutants.

Figure 8. The radar wind observations at RUT, PSE, and STW. X-axis is observational date from 9–16 August. Y-axis is height in kilometers. Wind vector was drawn when data was available. Shaded areas are wind speeds higher than 8 m/s in 2 m/s interval [65].



The other effective transport component of mesoscale systems in the Northeast is the sea breeze circulation. Previous studies demonstrated that the sea breeze is efficient in carrying polluted air from over the Gulf of Maine to the eastern coast of New England [6,15,51,66,67]. The cold water of Gulf of Maine creates a stable and less turbulent surface, which confines air pollutants within this layer and allows pollutants to be transported directly from Boston to northern New England [51]. Mao and Talbot [15] found that the sea breeze could transport air pollutants that originated from Mid-Atlantic States to coastal areas of southern NH and ME.

We examined the characteristics of the sea breeze circulation by analyzing horizontal meteorological patterns and local wind observations. Maps of surface wind at 18Z every day during 11–14 August clearly showed a southeasterly wind direction (on-shore winds) in the afternoon along the Northeast coast during the episode, and here we only show the map for 14 August (Figure 9) when the daily maximum O₃ mixing ratio reached the highest of the episode. Local winds observed at the Pease Air Base in NH and Boston, MA (in Figure 10 top panel) showed an off-shore wind (southwesterly) in the morning and then on-shore wind (southeasterly) in the afternoon with wind speed of ~5 m/s. Observations at TF showed similar wind patterns (Figure 10 bottom panel). EPA AIRNOW O₃

observations showed that O_3 values were unusually high (>112 ppbv) along the Boston coastal areas from 11–14 August. The off-shore winds could carry O_3 -rich air masses from Boston, MA to the Gulf of Maine. Figure 11 presented the relation between O_3 mixing ratios and wind directions at TF from 10–14 August. Wind direction sectors of 0° – 90° (Sector I), 90° – 180° (Sector II), 180° – 270° (Sector III), and 270° – 360° (Sector IV) were defined as northerly-to-easterly winds (I), easterly-to-southerly winds (II), southerly-to-westerly winds (III), and westerly-to-northerly winds (IV). It showed that the majority of wind directions came from southwesterly and southeasterly (II and III) during the episode. Furthermore, when $O_3 > 120$ ppbv all were from southeasterly (II). In addition, observations at Appledore Island (Figure 12) showed that in the afternoon/evening (1800–2300 UTC), the average winds were southeasterly, and corresponded to both elevated O_3 and CO values. It is apparent that the sea breeze occurred along the New England coastal region during the episode and it was a very important transport mechanism for the occurrence of the August episode.

Figure 9. Surface winds (m/s) at 18Z on 14 August 2002 from Plymouth State Weather Center.

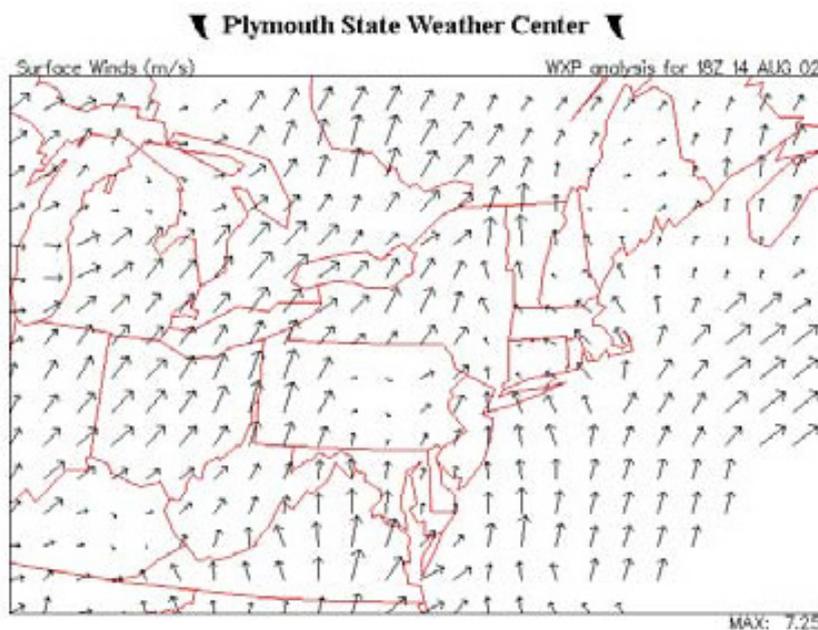


Figure 10. (Top) Local hourly wind observations from 10–14 August at KPSM and KBOS. X-axis is observational date from August 10 to 14; (Below) Hourly measurements at TF on 8/9~8/19 2002. Ozone is in green (use left axis), CO is in red (use right axis); and wind vector is in black (top line of the figure).

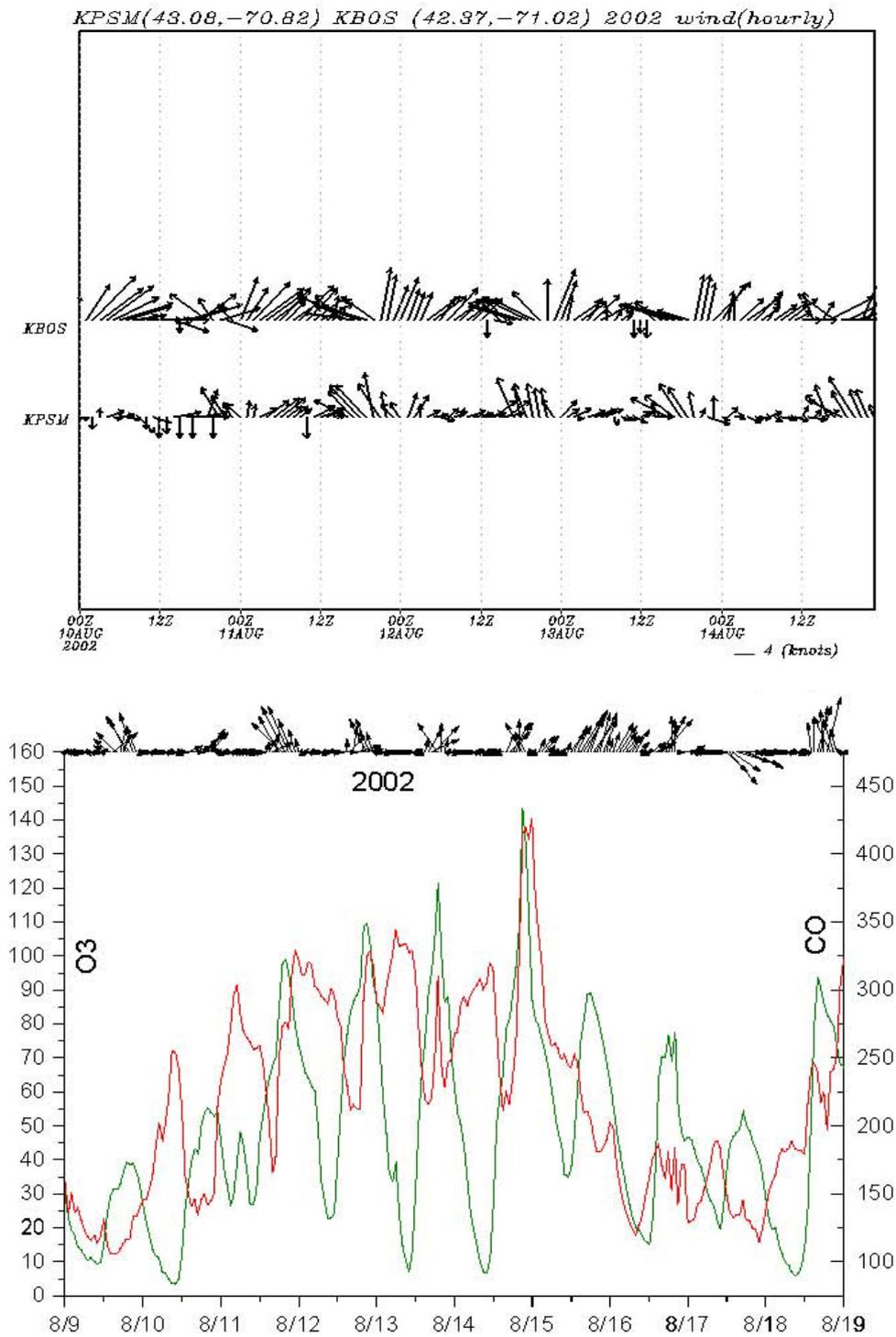


Figure 11. The observations of O₃ and wind directions at TF from 10–14 August. Radial scales present O₃ mixing ratios from 0 to 160 ppbv, in 10 ppbv interval. Angular degrees in clockwise present wind directions. In degrees of 0°–90° are northerly-to-easterly winds (I); 90°–180° are easterly-to-southerly winds (II); 180°–270° are southerly-to-westerly winds (III); and 270°–360° are westerly-to-northerly winds (IV).

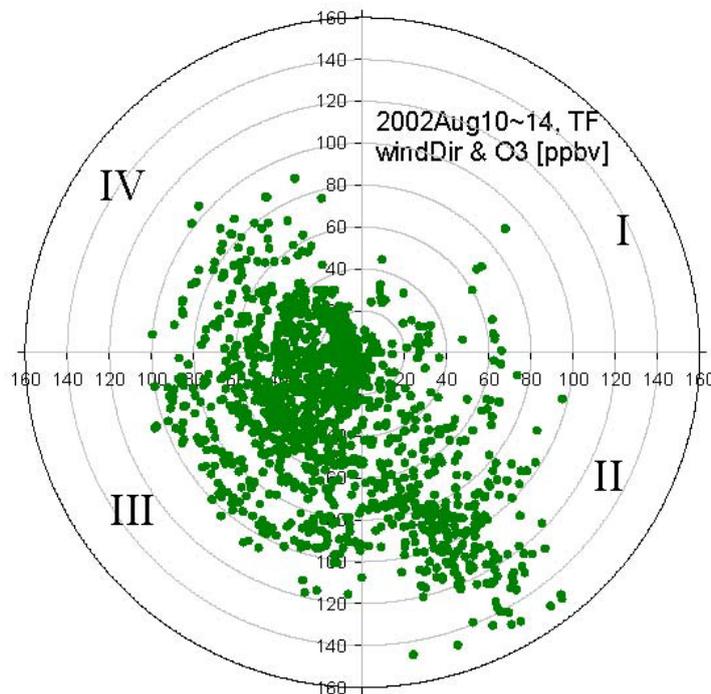
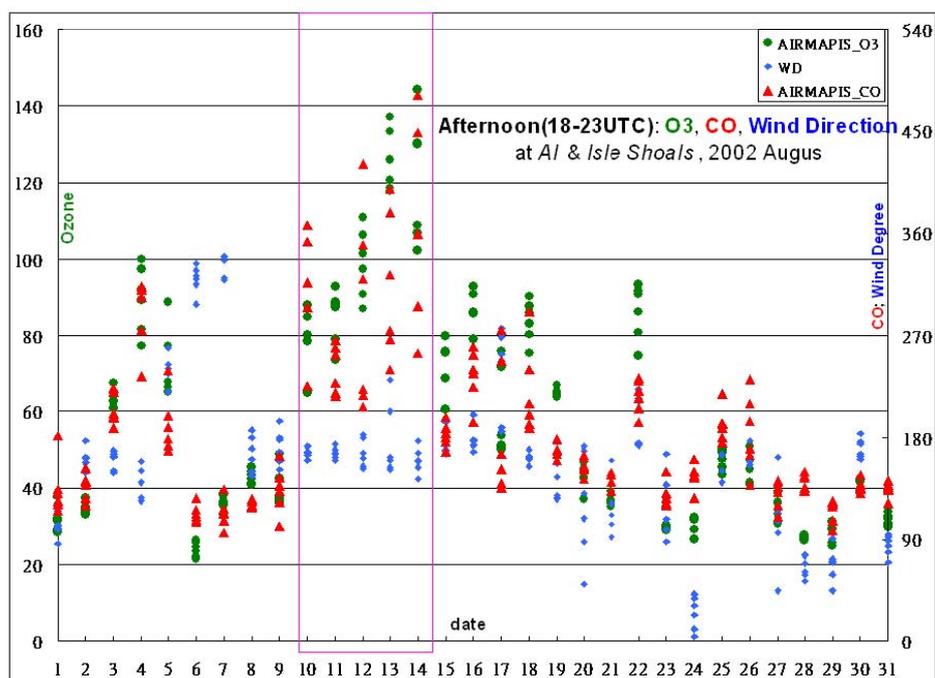


Figure 12. The observations of O₃ (in green dots, left-axis), CO (in red dots, right-axis), and wind directions (in blue dots, right-axis) during afternoon time (18–23 UTC) from 1–31 August 2002 at Appledore Island.



3.2. 2004 Case

The meteorological conditions in the Northeast during summer 2004 were dominated by wet weather with mild temperatures and frequent passage of frontal systems. NOAA National Climate Data Center records showed that the precipitation anomaly in the Northeast during the week of 18–24 July was 140% to ~200% higher than from the same week averaged over 1971–2000. Temperatures for the week averaged 21–27 °C. Analysis of the 5-day average (18–22 July) geopotential height revealed an overall deep trough covering the middle and eastern USA at 500 hPa and 950 hPa (Figure 2). Unlike summer 2002, summer 2004 lacked consistent presence of H-pressure systems. However, under these conditions, CS surprisingly observed the seasonal maximum mixing ratio of 111 ppbv on 22 July, more than 20 ppbv higher than the other years ranking the highest O₃ episode in our 8 years of record. It was also the third highest event at TF (Table 1). The event was short, only lasting one day.

3.2.1. Influence of Stratospheric Subsidence

The average meteorological patterns for July 2004 showed a deep trough covering the middle and eastern USA (Figure 2). Dynamically, the deep trough induces a descending air motion on its trailing side. In the upper troposphere it usually causes tropopause folding with stratospheric air mixing downward into the troposphere. The physical characteristics of stratospheric air are its dry low water vapor nature with high potential vorticity (PV). Values of PV in the troposphere are usually less than 1.5 PV (1 PV = 10⁻⁶ m² s⁻¹ K kg⁻¹). We used NCEP 2.5° × 2.5° data [61] to examine the PV patterns during the time period of interest. On 20 July the highest PV areas were found over Hudson Bay (>2.6 PV), the Great Lakes (2.2–2.4 PV), and western New York State (2–2.4 PV). The next day high PV areas extended from eastern Canada along the coastline of ME down to VA (Figure 13, top). The highest PV values (>2.6) were present over the DE coastal region. As the areas of enhanced PV moved eastward, values along the coastline of ME to NJ and to southern CT remained greater than 1.6 PV. The highest PV values ranged over 2.2–2.4 on 22 July, and were located just south of Rhode Island. This pattern of PV indicates that southern Canada and the northeastern USA coastline could potentially be affected by stratospheric subsidence and its associated higher O₃ mixing ratios. The NCEP 1° × 1° data showed a lower tropopause height from the Great Lakes to the east coast (Figure 14 top), which was consistent with the low-pressure system located in the Great Lakes (Figure 2), and had the positive anomaly (which was calculated by subtracting the climatology data of 1979–1995) of tropopause pressure (lower tropopause height) during 19–22 July (Figure 14, bottom panel).

Furthermore, total column O₃ (in Dobson Units, 1 DU = 2.69 × 10¹⁶ cm⁻²) from the NCEP 1° × 1° data showed that the O₃ value was around 325 DU along the east coast from VA to NH during the event (Figure 13, bottom panel). The TEMIS (Tropospheric Emission Monitor Service) image presented that the monthly mean total O₃ value based on multi-sensor-reanalysis data for July 2004 was around 325 DU along the U.S. east coast. Higher total column O₃ values (>325 DU represented in blue and red colors) were present over Hudson Bay and the Great Lakes area on 20 July, coincided with the higher PV areas aforementioned, and followed the same evolutionary patterns as PV during 20–22 July.

During this time period total column O₃ values had ~15 DU higher than the monthly mean value for July in the areas of interest.

We examined 2004 vertical O₃ profiles using sounding data from the IONS network [27,28,32,35]. Wallops exhibited O₃ values exceeding 340 ppbv on 21 July in the upper troposphere at 10–12 km, while O₃ was higher than 70 ppbv at altitudes below 6 km (Figure 15). Compared to other days in July and early August, 21 July had the highest O₃ mixing ratios in the altitude bins of 0-6 km (>70 ppbv). More specifically, in the mid-troposphere of 4–6 km, O₃ was up to 100 ppbv, which was 30 ppbv larger than other days. The Narragansett and Ron Brown locations also observed similar vertical patterns as found over Wallops. The O₃ values were usually <70 ppbv at altitudes between 0–6 km, but they increased on 21 and 22 July by 10–30 ppbv. Below 1 km, O₃ values were up to 90 ppbv at Wallops, 100 ppbv at Narragansett, and 100 ppbv at the Ron Brown.

Figure 13. (Top) The NCEP 2.5° × 2.5° Potential Vorticity (PV) at 1200 UTC at 300 mb on 20–22 July 2004. The shaded interval is 0.2 PV. (Bottom) NCEP 1° × 1° Total column ozone (Dobson) on 20–22 July 2004. The interval is 5 Dobson Units.

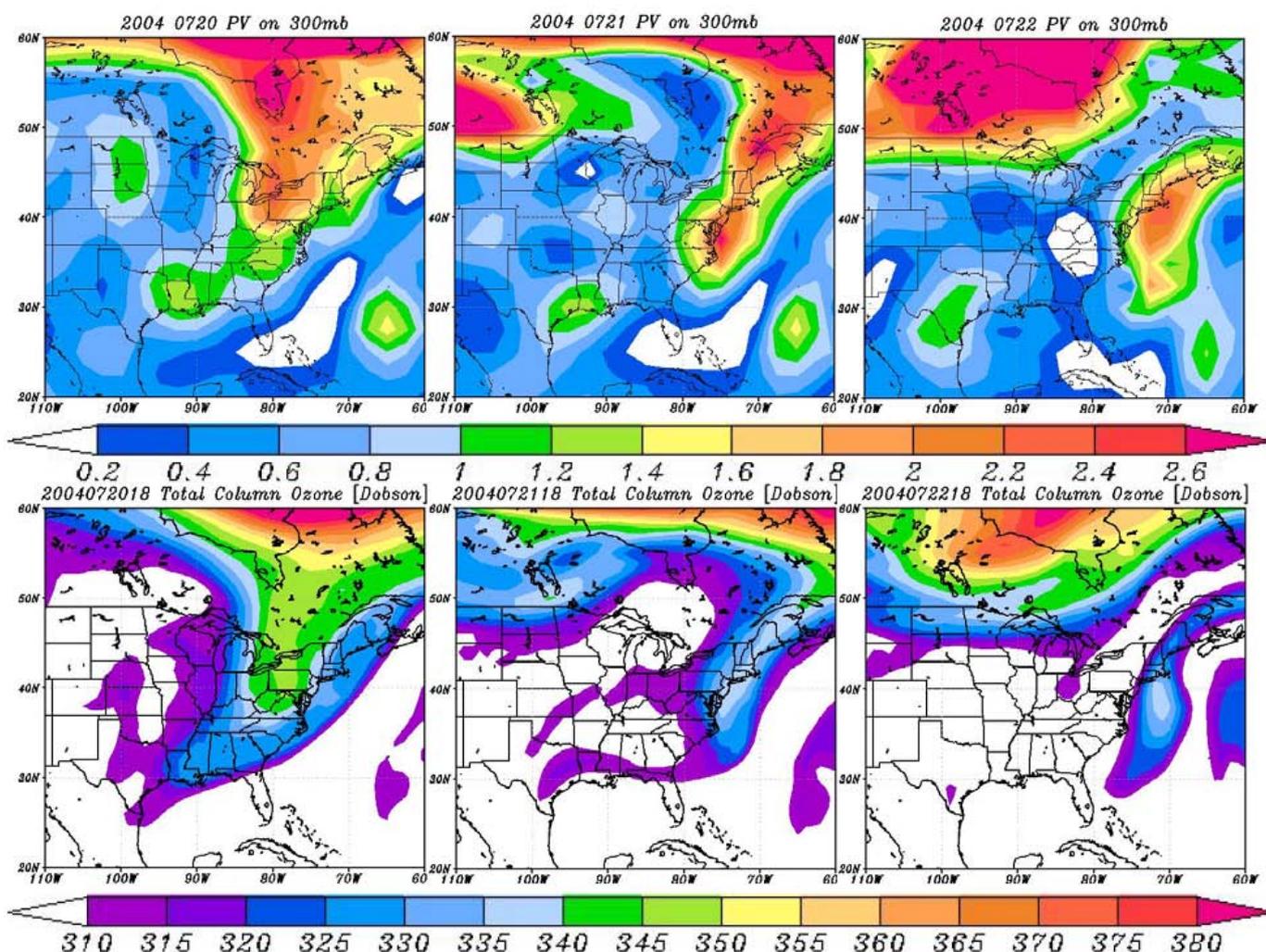


Figure 14. (Top) The NCEP $1^\circ \times 1^\circ$ tropopause pressure on 21 July 2004. (Bottom) The 4-day anomaly (which was calculated by subtracting the climatology data of 1979–1995) of tropopause pressure from NCEP daily analysis data. The Northeast showed the positive anomaly of tropopause pressure (mean lower tropopause height) during 19–22 July [68].

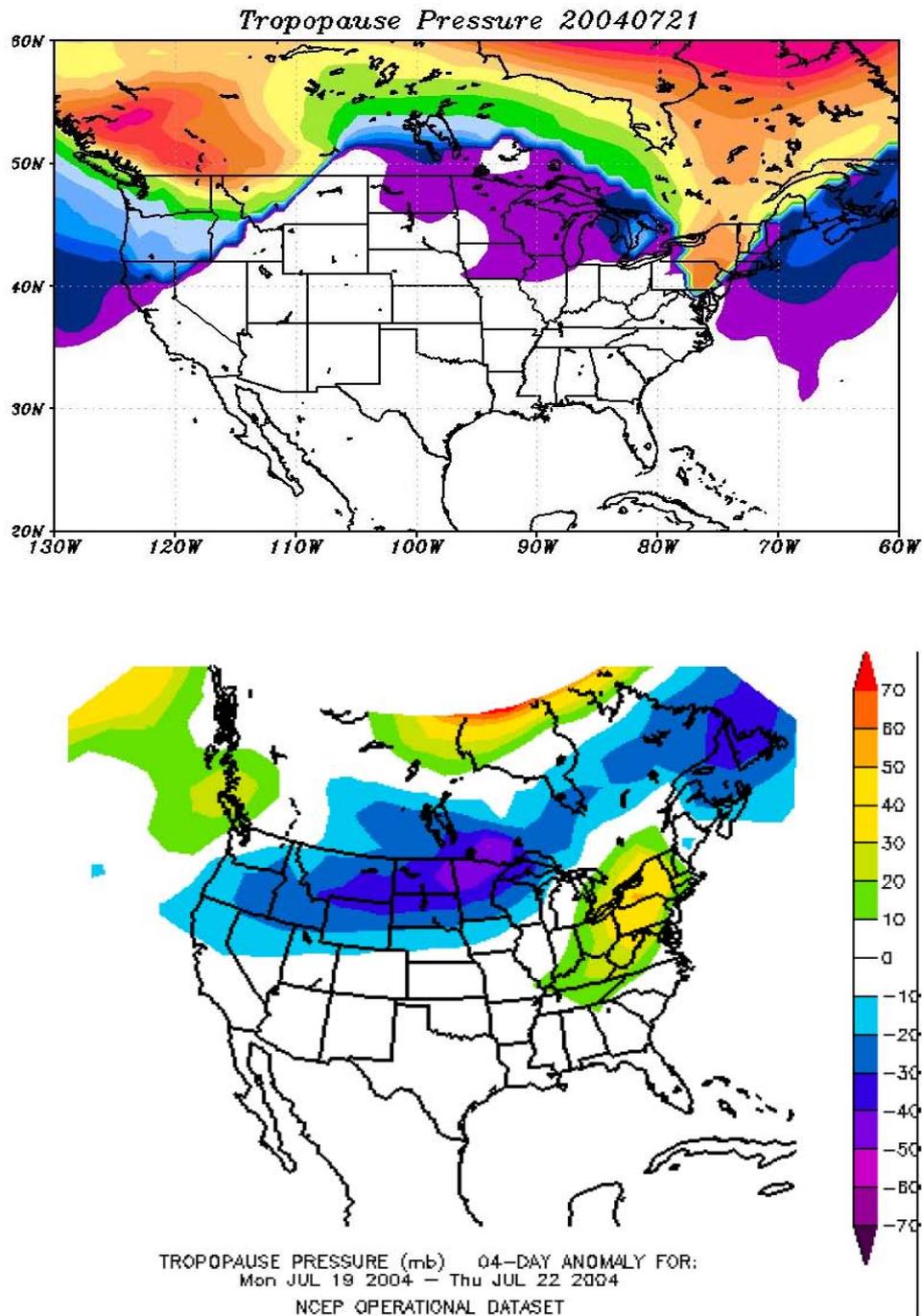
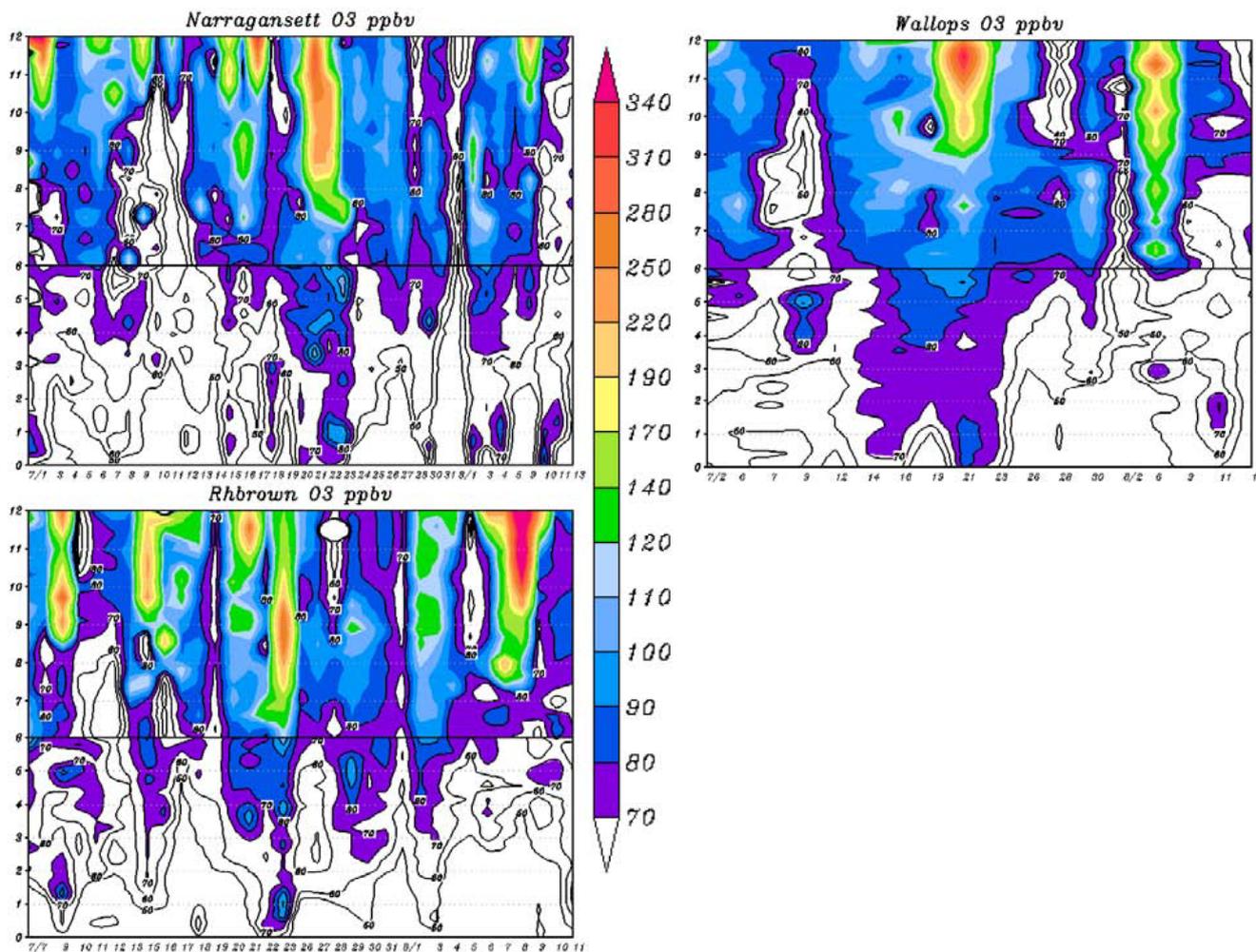


Figure 15. IONs-04 Ozone vertical profiles from surface to 12 km during their launching days (July-August) at Narragansett, Wallops, and RH Brown. Ozone values >70 ppbv are shaded. X-axis: launching dates. Y-axis: altitude in km.



Evidences from the patterns and the temporal variations in PV, lower tropopause heights, enhancements in total column O₃, and the elevated O₃ vertical distributions suggest that stratospheric influences enhanced tropospheric O₃ in Northeast coastal regions.

3.2.2. Influence of Mesoscale Systems

Besides stratospheric influence, regional meteorological systems could transport photochemically produced O₃ to CS. On 20 July sea level pressure showed that a high-pressure system formed over the Kentucky–West Virginia area. A ridge extended across PA and NY, with the east side connected to a shallow trough. The 950 hPa and 975 hPa geopotential heights (Figure 16) showed a short wave trough that was located along the coastline from VA to NH. This short wave trough was in fact the APLT. Previous studies had demonstrated that the APLT was connected with many high-pollution events in the Northeast [6,43-45]. In this event, surface O₃ patterns in the eastern U.S. from EPA AIRNOW showed that on 20 July higher O₃ values (~112 ppbv) appeared in VA and on Manhattan Island (Figure 5) when the APLT was present. Before 20 July O₃ mixing ratios in the Northeast were <80 ppbv. Values in NH,

however, were still low at this time ranging from about 50 to 65 ppbv. Over the next two days the area of elevated O₃ expanded northeasterly reaching NH. In southern New Hampshire mixing ratios were >100 ppbv, while at CS, situated in central NH, O₃ mixing ratios reached 111 ppbv on 22 July.

Figure 16. NCEP 1° × 1° geopotential height at 975 mb (right) and 950 mb (left) on 20 July 2004.

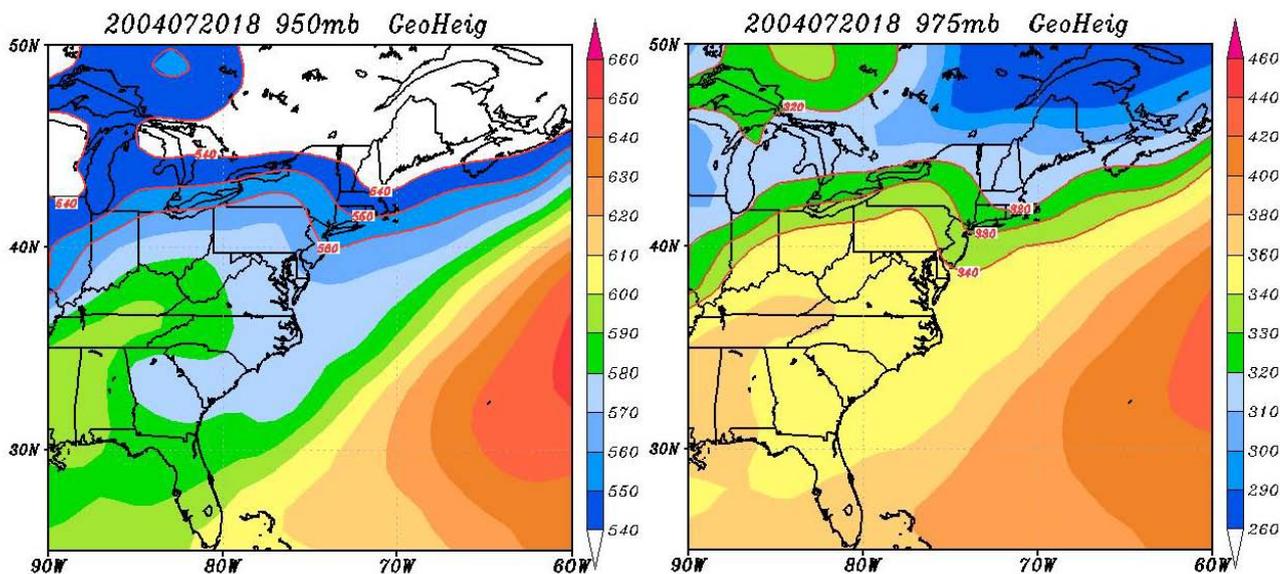
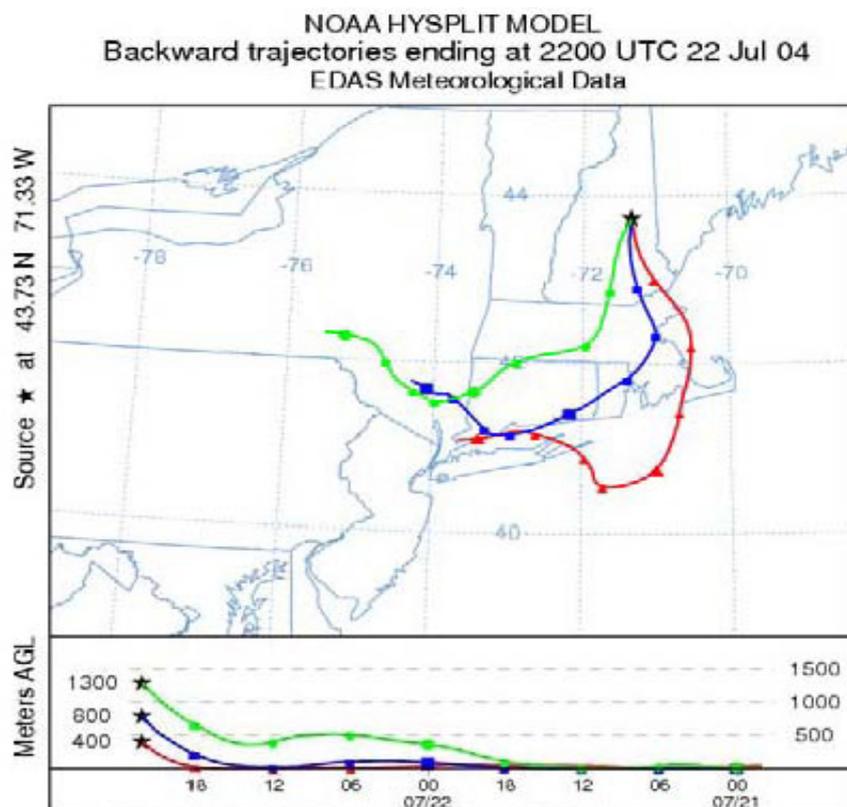


Figure 17. Forty-eight hours backward trajectories from NOAA HYSPLIT model starting at CS. The starting altitude 400 m is in red, 800 m in blue, and 1300 m in green [69].



Depending on the dynamical process, the APLT forms a convergent circulation at low altitude [6] bringing air from the surface to higher altitudes which was transported via the upper air stream (here it is a southwesterly wind) to downwind areas. This northeastward transport is demonstrated in the 48 h backward trajectories starting at CS in Figure 17. It showed that the air pathway originated around southeastern New York and Connecticut and circled over CT and MA in slow upward motion moving from 400 m to 1500 m altitude, indicating that the air masses spent ~2 days below 1500 m. Thus, once the southwesterly flow passed over these high O₃ regions, it could carry O₃-rich air to the downwind regions.

Polluted air from urban/industrial areas increased the mixing ratios of CO and NO_y at CS. For example, ΔCO (Δ=actual observed data minus its summer average) increased on 20 July and kept rising over the following days (Figure 6). It peaked on 22 July when ΔCO = ~250 ppbv. The ΔO₃ at CS (Figure 6) exhibited ~20 ppbv values on July 20 and 21. On 22 July, ΔO₃ increased to 80 ppbv. The ΔCO and ΔO₃ values showed a similar pattern over the 20–22 July timeframe with a correlation coefficient of ~0.65. Other pollutants like NO_y posted similarity with CO. The elevated CO and NO_y at CS support the hypothesis of the transport influence from the urban and industrial pollution. Furthermore, the examination of VOCs observations during 1 July–15 August at TF showed that most of the industrial/urban tracers were present at much higher values on 22 July than their average and median values (Table 2). For instance, C₂Cl₄ is the tracer of industrial/urban areas and its average value of 22 July (73 pptv) was three times more than its monthly average (22 pptv from 1 July to 15 August). The common combustion tracer ethyne also showed 677 pptv on 22 July compared to its monthly average of 289 pptv. Moreover, the vehicle emission tracers of acetone, benzene, and toluene at least doubled their values above their monthly average values (Table 2). From the enhanced chemical observations and the 48 h backward trajectories, we suggest that the APLT played an important role in transporting air from the urban/industrial areas northeasterly toward NH.

In addition, we compared the ratio of ΔO₃ to ΔCO in case 2002 at TF and case 2004 at CS. The average ratio of ΔO₃ to ΔCO in the timeframe 10–14 August 2002 was 0.24, while the average ratio during 20–22 July 2004 was 0.20. Our previous examinations (section 3.1) illustrated O₃ episode in 2002 was a typical case dominated by a high-pressure system and mesoscale transport of the LLJ and sea breeze in the east USA. In addition, Parrish *et al.* [21] compared the ratio of O₃ to CO at three measurement sites along the east coastal line of North America. Their result (in their Figure 2) suggested if air mass was transported from polluted/industrialized continental areas, the ratios of O₃ to CO were near 0.27 or 0.29. If the elevated O₃ attributed to stratospheric subsidence, this ration was around 0.22 in their study. In our study, the smaller ratio in 2004 indicated that the enhanced O₃ at CS not only might be contributed by the mesoscale transport of APLT (which brought polluted air from the urban/industrial area), but also be injected by the stratospheric subsidence with low CO. The relation between ΔO₃ and ΔCO was lower than in case 2002. This emphasized the important impact of the stratospheric intrusion on the highest O₃ event in 2004 at CS.

Table 2. VOC measurements at TF in 2004. The statistic values include the maximum on 7/22 (1st column), the averages on 7/22 (2nd column), the averages during 7/1~8/15 (3rd column), and the median values during 7/1~8/15(4th column).

	7/22(Max)	7/22(Avg)	Average (0701-0815)	Median(0701-0815)	Ratio [(7/22)/ July]
propane	4152	2058	840	570	2.5
<i>i</i> -butane	424	160	59	45	2.7
<i>n</i> -butane	503	269	116	97	2.3
ethyne	1101	677	289	241	2.3
cyclopentane	22	11	6.2	5.3	1.8
<i>i</i> -pentane	1330	601	247	187	2.4
<i>n</i> -pentane	440	188	83	62	2.3
methylcyclohexane	77	36	16	13	2.3
<i>n</i> -heptane	99	51	21	16	2.5
benzene	229	150	80	71	1.9
2,3,4-trimethylpentane	37	21	12	9.3	1.8
2-methylheptane	53	26	15	13	1.7
ethylbenzene	169	56	24	19	2.3
<i>m+p</i> -xylene	568	173	73	52	2.4
styrene	65	24	12	9.5	2.0
2-ethyltoluene	41	14	9.3	7.6	1.5
C2Cl4	166	73	22	16	3.4
MeONO2	8.0	5.0	2.7	2.5	1.9
EtONO2	8.9	6.6	2.7	2.5	2.4
1-PrONO2	4.5	3.4	1.1	0.95	3.1
2-PrONO2	30	24	9.1	8.3	2.7
2-BuONO2	25	17	5.5	4.7	3.1
3-PenONO2	10	6.6	1.9	1.6	3.4
2-PenONO2	16	8.7	2.5	1.9	3.6
	7/22(Max)	7/22(Avg)	Average (0711-0819)	Median(0711-0819)	Ratio [(7/22)/ July]
acetaldehyde	2.1	0.61	0.31	0.22	1.9
acetone	8.2	3.8	1.8	1.5	2.1
MEK	1.3	0.46	0.21	0.16	2.2
benzene	0.28	0.18	0.08	0.07	2.3
toluene	0.58	0.31	0.14	0.10	2.2
C8 aromatics	1.1	0.38	0.17	0.13	2.3
acetic acid	3.6	1.4	0.45	0.26	3.1

3.2.3. Comparison of the Four Highest O₃ Episodes

A comparison of the four highest O₃ events at CS, the PV values, the total column O₃, and the geopotential height at 975 hPa are presented in Figure 18. Values of PV (Figure 18 top) and the total column O₃ (Figure 18 middle panel) in the area around NH showed that 2004 had the highest values, which indicated CS during the 2004 episode was under the strongest stratospheric influence. Moreover, in the low-troposphere, the geopotential height (Figure 18, bottom panel) of these four events showed that the APLT only occurred in 2004. The event in 2004 was the only year having O₃ values >100 ppbv along with higher PV and total column O₃ (Table 3). These suggest that the highest O₃ episode at CS in 2004 was a unique case; its occurrence was a result of two mechanisms: stratospheric intrusion and mesoscale transport of the APLT.

Figure 18. (Top panel) The NCEP 2.5° × 2.5° Potential Vorticity (PV) at 300 mb for the four highest events. Images from left to right represent years from 2002 to 2005. The shaded interval is 0.2 PV. (Middle panel) NCEP 1° × 1° total column O₃ (in Dobson units) values are listed from 2002 (right) to 2005 (left). The interval is 5 Dobson Units. (Bottom panel) The NCEP 1° × 1° geopotential height at 975mb during the years of interest. A trough was present along the eastern coast in 2004.

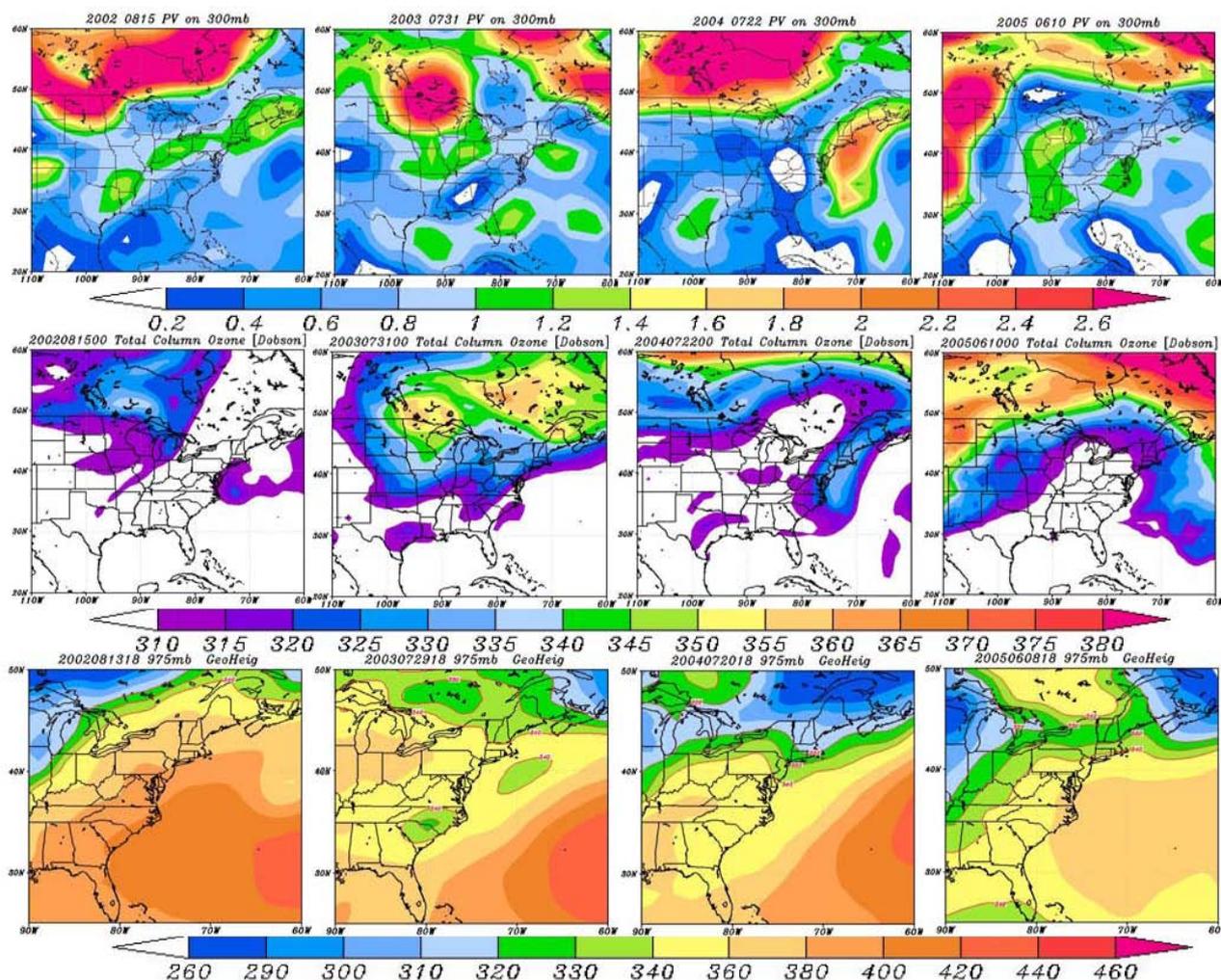


Table 3. The four highest O₃ episodes at CS: 2004 was the highest event with the only one year with O₃ > 100 ppbv; 2005 was the second highest event; 2003 was third and 2002 was fourth. Values of PV and total O₃ column were obtained from Figure 18 (top and middle panels) in the area over NH.

Year	O ₃ Max at CS	PV	Total O ₃ Column(Dobson)	APLT
2002	88 (8/15)	1-1.2 PV	<310	–
2003	90 (7/31)	<1 PV	325-335	–
2004	111 (7/22)	1.6-1.8 PV	330-335	+
2005	92 (6/10)	<1 PV	310-315	–

“–” means no APLT; “+” means having an APLT. They were identified by the geopotential height at 975 hPa in Figure 18 (bottom panel).

4. Conclusions

This paper presents an analysis of meteorological processes on the synoptic and mesoscale for 10–15 August 2002 at TF (151 ppbv) and 22 July 2004 at CS (111ppbv) with record-high O₃ mixing ratios. The 2002 episode was a typical high O₃ event with an H-pressure system stagnant over the Northeast for an unusually extended period. The 2004 episode occurred as a result of stratospheric influence and impact from the APLT. In general, synoptic systems controlled the overall transport patterns of the events, while mesoscale systems played a crucial role in determining the timing and the magnitude of the O₃ peaks.

The H-pressure synoptic system situated over the eastern USA dominated the majority of elevated-O₃ distributions in the 2002 case. The regional build-up of air pollutants was observed by nighttime measurements at CS, where O₃ increased from 27 ppbv on 10 August to 75 ppbv in the early morning on 15 August. The mesoscale LLJ characteristics were identified using horizontal maps and radar vertical wind observations in the Northeast corridor. The enhanced mixing ratio values of CO, PM_{2.5} and O₃ also illustrated accumulation of polluted air masses owing to the LLJ transport from upwind source regions. Finally, another mesoscale mechanism associated with this case was the transport facilitated by the sea breeze circulation. The repeated afternoon southeasterly wind direction along with the timing of O₃ peaks were observed at TF and AI during this period contributing to the occurrence of high O₃ values in this episode.

In 2004, a synoptic scale deep trough resulted in an intense stratospheric subsidence over the Northeastern USA. The total O₃ column increased by more than 50 DU over Hudson Bay in Canada, and by more than 20 DU in the upper troposphere over the U.S. east coast. The vertical O₃ distributions from IONS-04 soundings showed the high O₃ mixing ratio and dry air penetrated downward to the middle-to-lower troposphere. At 6–12 km altitude, O₃ increased from 120 ppbv to 340 ppbv. At the altitude of 0–6 km, O₃ increased by at least 10–30 ppbv during episode days. The average ration of ΔO_3 to ΔCO was lower than the value in episode 2002. Thus, combining meteorological patterns, sounding measurements, backward trajectories, and the lower relation between ΔO_3 to ΔCO , we suggest that stratospheric subsidence contributed directly to high O₃ levels in the Northeast.

Second mechanism important to this event was mesoscale transport related to the APLT. The meteorological features of the APLT were present in the lower troposphere on 20 July. Backward

trajectories initiated at the height of 400 m to 1500 m showed airflow caused by the lee-side-trough, which was upward motion and northeastward transport toward NH. Moreover, the elevated pollutants, like CO, NO_y, and some urban/industrial VOCs tracers at CS and TF supported the influence from urban and industrial areas.

This study illustrated the very different meteorological conditions and multi-scale dynamic systems resulting in two severe O₃ episodes at rural sites of southern NH. Meteorology played a crucial role in determining air quality in the region. Further the ability of air quality modeling systems to replicate these phenomena should be a focus of future studies.

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