Coastal Boundary Layer Influence on Pollutant Transport in New England

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ABSTRACT

Air pollution episodes in northern New England often are caused by transport of pollutants over water. Two such episodes in the summer of 2002 are examined (22–23 July and 11–14 August). In both cases, the pollutants that affected coastal New Hampshire and coastal southwest Maine were transported over coastal waters in stable layers at the surface. These layers were at least intermittently turbulent but retained their chemical constituents. The lack of deposition or deep vertical mixing on the overwater trajectories allowed pollutant concentrations to remain strong. The polluted plumes came directly from the Boston, Massachusetts, area. In the 22–23 July case, the trajectories were relatively straight and dominated by synoptic-scale effects, transporting pollution to the Maine coast. On 11–14 August, sea breezes brought polluted air from the coastal waters inland into New Hampshire.

1. Introduction

Most of New England's population lives near the coast, and most of its industry is found there. It is hardly surprising, then, that air pollution in New England is also linked to the coast. Mountainous terrain and distant sources play a role, but much of New England's air pollution comes from sources near its own or nearby coasts and is transported along the coast, either over land or across the near-shore waters. The overwater path has been fully appreciated only recently, and is the primary subject of this paper.

Locales in New England exceed national air quality standards for ozone on several days every year. Connecticut, Rhode Island, and Massachusetts record most of those exceedances, but even the downeast coast of Maine at Acadia National Park records several exceedances every year. The most important sources of pollutants and precursors in New England are in and near Boston, Massachusetts, but the Interstate 95 urban corridor from Washington, D.C., through New York City, New York, to southern Maine is also important. Figure 1 shows the intensity of pollutant emissions in the region on an average summer weekday.

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Air pollution in northern New England is, thus, primarily transported, rather than locally produced. If the air were stagnant, northern New England would be clean. High-pollution episodes in northern New England occur with light to moderate (but not stagnant) winds from the source regions. The paths taken by pollutants to reach coastal New Hampshire and Maine are not necessarily simple, however. Indeed, it seems that very high pollution in New Hampshire and Maine is unlikely to occur if the transport path is only over land.

Why is overwater transport important? Why is it different than transport over land? In northern New England, air transported from land encounters a cooler, smoother surface; convective mixing, therefore, decreases. A persistent pool of cold water exists offshore in the northern and eastern Gulf of Maine and the Bay of Fundy, with warmer water inshore. An early field study in the region is reported by Taylor (1917) who observed the cold water temperatures. Another factor is the lack of chemical deposition; ozone and most of its precursors are essentially not deposited to water surfaces, but they are rapidly deposited to leaf surfaces (Lenschow et al. 1982). Last, the reduction of convective mixing allows for differential advection, when polluted air at different heights is transported in different directions. We discuss two cases here; in both cases the primary impact is from polluted air transported at the

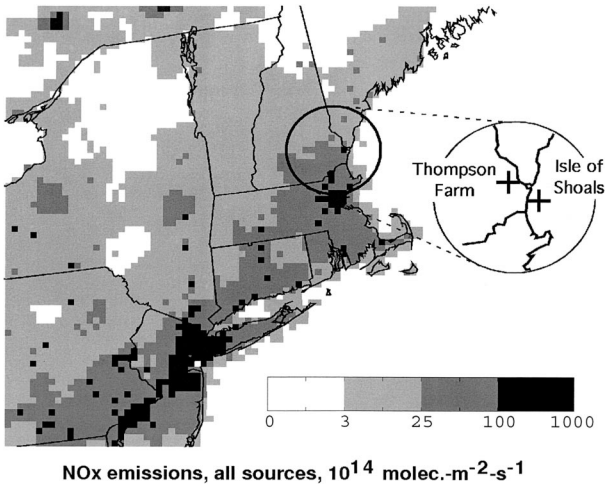

FIG. 1. Map of NO_x emissions for New England and vicinity with inset showing location of key sites.

surface, but in one case transport aloft also impacts a higher-elevation site.

The New England Air Quality Study (NEAQS; available online at http://www.al.noaa.gov/neaqs/) was conducted in July and August 2002. Many of the study sites and instruments, however, were active for a longer period. The core components of the study were four surface chemistry sites operated by the University of New Hampshire, six radar wind profilers and a Doppler lidar of the National Oceanic and Atmospheric Administration (NOAA), and the NOAA Research Vessel *Ronald H. Brown.* The ship carried a suite of atmospheric chemistry instrumentation, a lidar measuring vertical profiles of ozone and aerosol, and a radar wind profiler. Scientific staff on board launched radiosondes. Figure 1 shows the locations of the primary study sites.

One-hour-averaged ozone exceeded 100 ppb at one or more regulatory monitors in New England on 12 days in July and 10 days in August 2002. These days fall into episodes as follows: 1–4, 8–9, 13–15, 17–19, and 22–23 July and 2–4 and 10–18 August. One-hour ozone exceeding 125 ppb at one or more monitors occurred on 1, 2, 9, 18, 22, and 23 July and on 4 and 11–14 August. Figure 2 shows the ozone mixing ratio measured by the ''AIRMAP'' sites at Thompson Farm (Durham, New Hampshire, approximately 20 km inland) and the Isles of Shoals (approximately 8 km offshore) for July and August. Many of the larger ozone values were recorded in southern New England. Only 5 days approached or exceeded 100 ppb at Thompson Farm. Ozone mixing ratios were consistently greater at the Isles of Shoals than at Thompson Farm. The synoptic meteorological cycle (5–8 days) and the diurnal cycle are clearly visible.

Ozone episodes occurred during periods of moderate synoptic forcing. The most prominent feature of all episodes was a low pressure system over northern Ontario and/or Quebec, Canada, producing the requisite south-

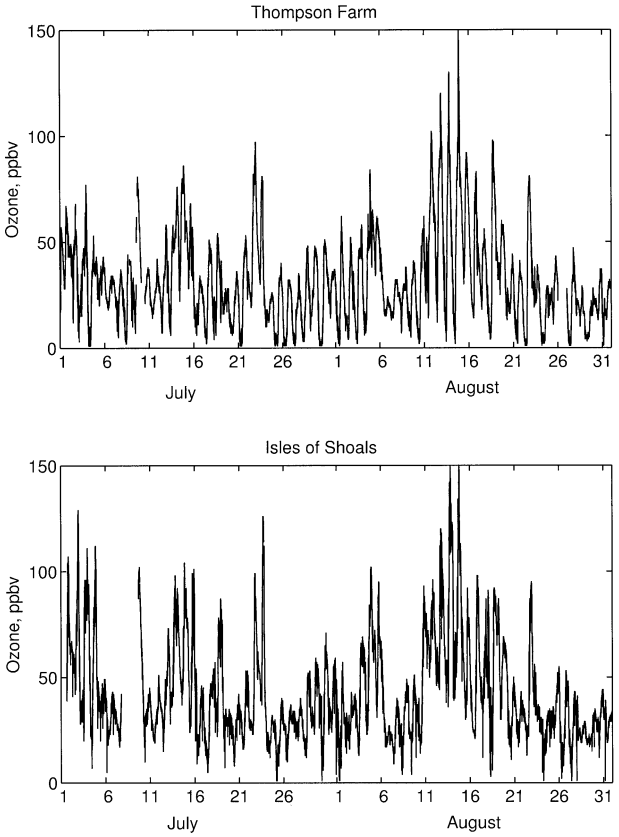

FIG. 2. Ozone mixing ratio measured at Thompson Farm and the Isles of Shoals (Appledore Island) during Jul and Aug 2002.

westerly flow. Episodes were terminated by the passage of cold fronts associated with those lows. This is consistent with the findings of Merrill and Moody (1996) for the 1993 season. Seaman and Michelson (2000) analyzed a 1995 episode in detail, with emphasis on areas somewhat farther south than the NEAQS area.

A previous field campaign in this area, the North Atlantic Regional Experiment 1993 Intensive (NARE 1993), explored many of these issues and produced extensive documentation. The focus of NARE 1993 was on transport to the remote North Atlantic and Europe, but considerable work was done in coastal New England and the Gulf of Maine as well. Angevine et al. (1996) give an overview of the mesoscale meteorological situation and the state of understanding of the coastal boundary layer that was then current. Ray et al. (1996) showed measurements from coastal Maine and described the circumstances leading to high-ozone episodes there. Strong layering of the atmosphere caused by the cold water offshore was a theme of many of the papers, including those analyzing aircraft observations (Buhr et al. 1996; Daum et al. 1996; Kleinman et al. 1996) and modeling studies (Fast and Berkowitz 1996).

Considerable work has been done since NARE 1993 on coastal, stable (nocturnal), and transitional boundary layers. For example, the Risø Air Sea Experiment (RAS-

EX) data have been used to test and adapt the theoretical framework for stable boundary layers (Mahrt et al. 1998b, 2001). Long-term measurements in the Kattegat between Denmark and Sweden were analyzed by Sempreviva and Gryning (2000). Spatially resolved measurements of turbulence in flow of warm air over cold water at the coast of North Carolina were reported by Vickers et al. (2001). Aircraft measurements in the Baltic were reported by Källstrand et al. (2000) showing the behavior of the internal boundary layer in two cases—one with a stronger geostrophic wind and no sea breeze, and one with a sea breeze and weaker large-scale winds. Smedman et al. (1997) combined theory, modeling, and measurements to show a pattern of internal boundary layer development consisting of a stable layer at short overwater transport times followed by development of a near-neutral layer at longer transport times. A thorough discussion of offshore flow in general can be found in Mahrt et al. (2001). Žagar et al. (2003) give useful scaling arguments for fluxes near shore. The recent and ongoing Baltic Sea Experiment (BALTEX; online at http://w3.gkss.de/baltex/) is being carried out in that area, which has a similar situation to the east coast of the United States. Studies in the U.S. Great Lakes region are also relevant (Dye et al. 1995; Fast and Heilman 2003).

Nocturnal boundary layers over land have features that are relevant to the coastal problem. Mahrt et al. (1998a) draw a distinction between weakly stable and very stable boundary layers and discuss the difficulties inherent in measurements and interpretation in very stable cases. These issues are explored further by Mahrt and Vickers (2002) using data from the Cooperative Atmosphere-Surface Exchange Study—1999 (CASES-99) experiment, which also yielded many other interesting results (Coulter and Doran 2002; Poulos et al. 2002). The afternoon transition from a convective to a stable boundary layer over land, which is analogous to the coastal transition, has been described by Grimsdell and Angevine (2002). Key findings are that the afternoon transition is a gradual reduction in the intensity and vertical extent of turbulence, not a sudden collapse, and that the transition begins as early as several hours before sunset. Despite this impressive body of work, many challenges yet remain in describing and understanding the immensely complex coastal boundary layer.

2. Case studies

a. 22–23 July

The largest ozone mixing ratios during this episode were found inland in Connecticut and Massachusetts, but large values also occurred along the coasts of Massachusetts and Maine. This pattern is also reflected in the research measurements from AIRMAP (Figs. 2 and 3). The episode began with a wind shift to southwesterly about 1800 UTC 21 July and continued with southwesterly flow until it was terminated abruptly by the passage of a cold front with thunderstorms in the afternoon of 23 July. Operational ozone monitors along the southwest coast of Maine and inland provide a picture of the episode (Fig. 4). On 22 July, the monitor at Kittery, Maine (on the coast at the border with New Hampshire), had a peak of 100 ppb; monitors farther northeast at Kennebunkport and Cape Elizabeth had peaks of only 80 and 65 ppb, respectively, with smaller peaks farther northeast along the Maine coastline. Inland Maine monitors at Hollis (~25 km west of Cape Elizabeth) and Gardiner (just south of Augusta) recorded peaks of 95 and 80 ppb, respectively, suggesting that the plume moved inland with southerly surface winds (not shown) instead of along the coast of Maine. On 23 July, many monitors along the Maine coast as far northeast as Acadia National Park observed ozone peaks of between 88 and 120 ppb, with a peak of 65 ppb at the Roosevelt Campobello International Park monitor further downeast. Inland sites in Maine on 23 July recorded peaks between 70 and 79 ppb. Peak ozone mixing ratio on the Scotia Prince Ferry (between Portland, Maine, and Yarmouth, Nova Scotia, Canada) was only 72 ppb during this episode. Again, the surface winds indicate that the same plume traveling along the coast impacted all these sites.

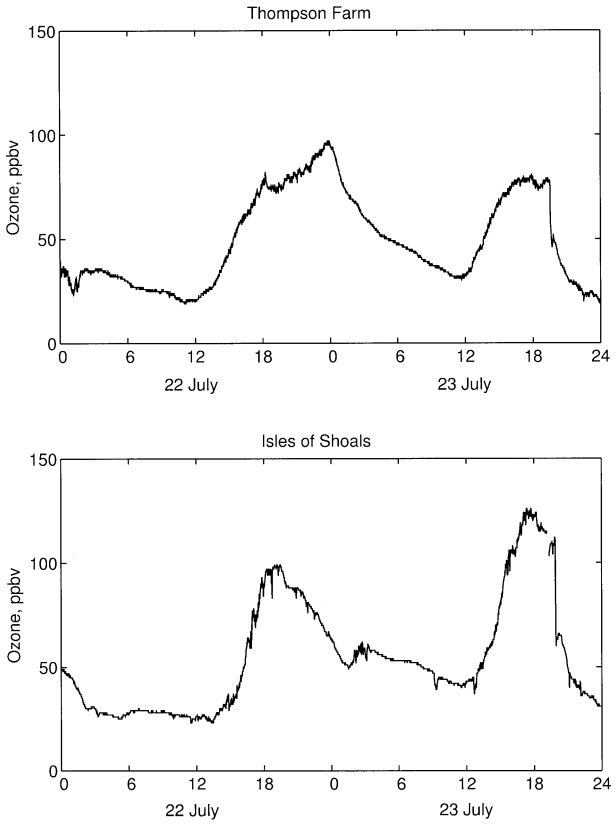

FIG. 3. Ozone mixing ratio measured at Thompson Farm and the Isles of Shoals on 22 and 23 Jul 2002 (time in UTC).

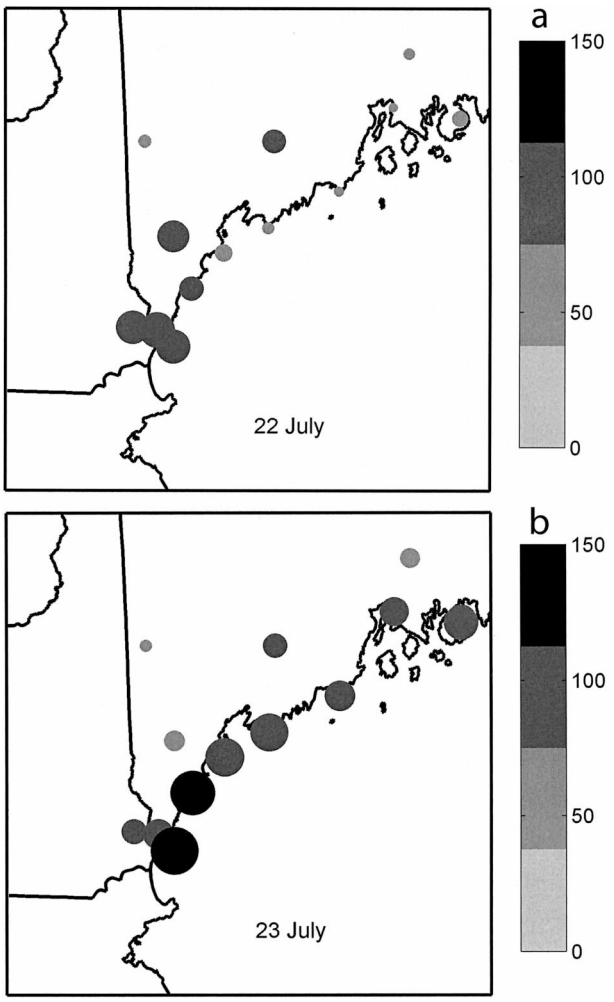

FIG. 4. Maximum 1-h-average ozone mixing ratios (ppbv) measured at operational monitors in Maine and at the Thompson Farm and the Isles of Shoals sites on (a) 22 and (b) 23 Jul.

Trajectories computed from the wind profiler network data (Figs. 5 and 6) indicate that polluted air reaching the Atlantic near Cape Ann came along the urban corridor over major source areas, including New York City and Boston in the 24 h previous to 1800 UTC 23 July. These trajectories were calculated from a regional network of eight fixed, land-based wind profilers and a mobile wind profiler deployed on the R/V *Ronald H. Brown.* The land-based wind profilers were located at Appledore Island, New Hampshire (ADI); Concord, New Hampshire (CCD); Orange, Massachusetts (ORE); Pease International Tradeport, New Hampshire (PEA); Pinnacles State Park, New York (PSP; location not shown in Figs. 5 and 6); Plymouth, Massachusetts (PYM); Rutgers University, New Jersey (RUT); and Schenectady, New York (SCH). The hourly wind profiler data were first averaged in the vertical between 300 and 400 m above mean sea level (MSL), and from these a weighted average of wind speed and direction was

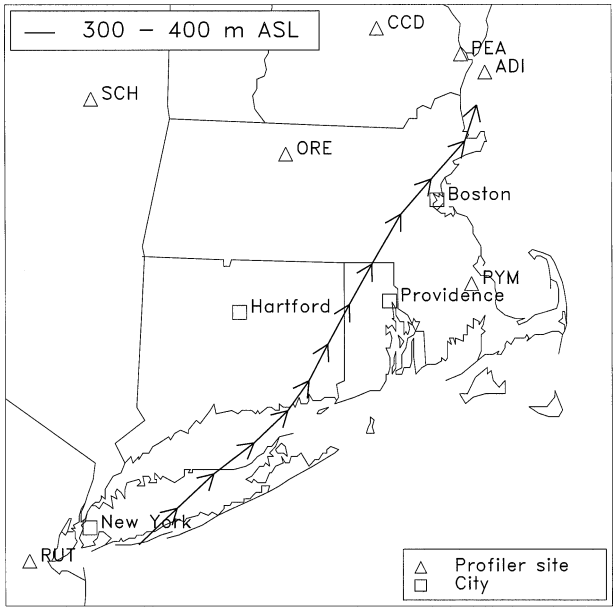

FIG. 5. Twelve-hour back trajectory at 300–400 m MSL ending at the position of the *Ronald H. Brown* at 2100 UTC 22 Jul. See text for details of method.

computed at the trajectory locations. The data from each individual profiler were weighted according to the inverse squared distance between the trajectory and the profiler location. These trajectories are generally consistent with trajectories computed from the operational Eta Model (not shown). Surface winds were slightly more southerly than winds aloft throughout the episode. The alignment of sources along the trajectories contributes to the large ozone mixing ratios and is part of the

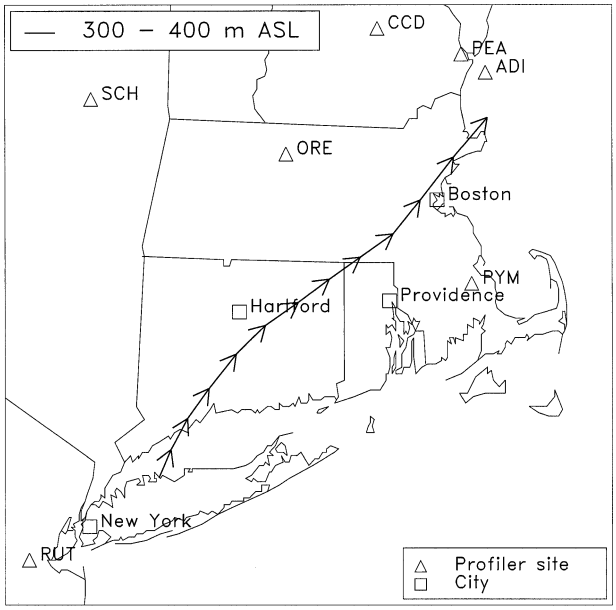

FIG. 6. As in Fig. 5 at 1800 UTC 23 Jul.

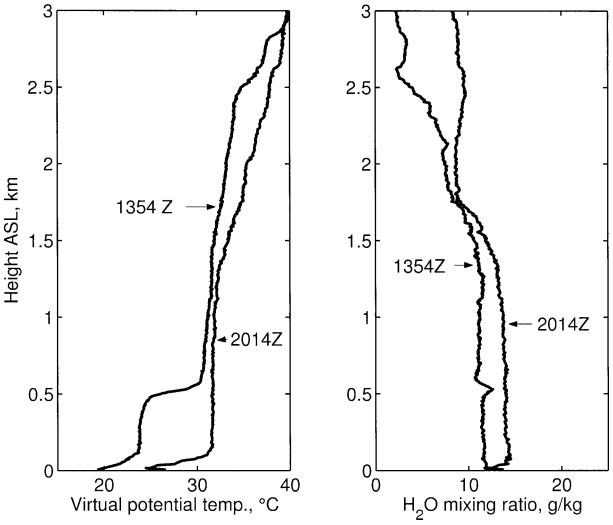

FIG. 7. (left) Potential temperature and (right) water vapor mixing ratio from radiosoundings launched from the ship at 1354 and 2014 UTC 22 Jul.

reason that they are localized along the coast north of Boston rather than inland. Trajectories arriving off Cape Ann at times from 0300 UTC 22 July through the episode have similar directions. Transit times from Boston to the Isles of Shoals are approximately 2–3 h throughout the episode, and transit times from the New York City area are approximately 12 h. Comparison of time series of 10-m-AGL air temperatures at Beverly, Massachusetts, and the Isles of Shoals show a phase lag that supports 2–3-h transit times from land to the isles. Both modeled trajectories and those computed from measurements must be used with some caution. For example, modeled trajectories to the Isles of Shoals and to the coast of Maine where the strongest ozone was measured show the effects of the cold front too early on 23 July. The measurement-based trajectories are uncertain outside the region of the dense wind profiler network and because they do not take into account vertical motion.

Near-surface air temperatures were warmer over land than at the Isles of Shoals throughout the episode at all times of day and night. Temperatures at Beverly or Portsmouth, New Hampshire, were up to 9°C warmer than at the Isles of Shoals. Soundings from the R/V *Ronald H. Brown* in the area between Cape Ann and the Isles of Shoals at several times on 22 and 23 July showed a statically stable layer near the surface (Fig. 7), a few tens of meters deep. An advected continental mixed layer, up to about 500 m deep at 1354 UTC and 1300 m deep at 2014 UTC, lies above the stable marine layer. In the earlier sounding, a near-neutral residual layer with similar water vapor content extends to approximately 1.5 km, and a statically stable layer with decreasing water vapor lies atop that. In the later (2014 UTC) sounding, the transition between the advected mixed layer and the stable layer above 1.3 km is smooth, without a pronounced temperature inversion, and the transition is more easily seen in the water vapor profile. Profiles of the bulk Richardson number (not shown) indicate that the shallow surface-based layer is dynamically unstable (very small or slightly negative bulk Richardson number). The turbulence produced by shear-driven instability allows for some of the mixing within the layer that cools it; if no turbulence were present at all, the layer cooled by contact with the water would only be on the order of 1 m deep. During the night, the land–sea temperature difference is small, requiring relatively little shear to produce dynamic instability; during the day, the land–sea temperature difference increases, the static stability increases, but turbulent kinetic energy advected from the land is available to enhance mixing. We cannot distinguish the effects of locally produced shear-driven turbulence and advected turbulence with the data we have available. The basic structure shown in these soundings was quite typical; all soundings in offshore flow had surface-based statically stable layers.

The wind profiler data from Portsmouth and the Isles of Shoals have interesting similarities and differences (Fig. 8). The wind speed at Portsmouth below 1 km was near 10 m s^{-1}, and the wind speed at the Isles of Shoals was consistently 3–4 m s^{-1} faster than at Portsmouth during the day. This is not a stagnant air mass. Wind directions are very consistent between the two sites except below 300 m, where there is some directional shear over the Isles of Shoals. The wind direction profiles at Portsmouth at midday (1830 UTC) are nearly constant with height up to about 1.2 km, indicating a deep mixed layer, which does not exist at the Isles of Shoals. The mixing depths diagnosed from the Portsmouth profiler reflectivity are about 1.5 km at midday on both 22 and 23 July, and similar depths are measured inland at Concord, New Hampshire. A low-level jet was observed at night until 1200 UTC at both sites on 22 July and at the Isles of Shoals only on 23 July.

Ozone at Thompson Farm (Fig. 3) peaked at approximately 100 ppb at 0000 UTC 23 July, several hours later than, but with almost the same concentration as, that at the Isles of Shoals. Surface wind observations and trajectories indicate that the air at Thompson Farm came over land from the New York City area.

The ship, patrolling north of Cape Ann on 22 July, observed a broad peak of ozone at the surface after 1500 UTC, with maximum values near 90 ppbv. The ozone-laden air mass was warm (20°–30°C) and hazy. The pattern on 23 July was similar but the ozone peak was near 120 ppbv. The shipboard ozone lidar provides interesting information about the vertical structure of the polluted layer. Between 1600 UTC 22 July and 1000 UTC 23 July the lidar saw a layer of ozone mixing ratios greater than 100 ppb at 300–500 m MSL (Fig. 9), with slightly smaller values until 1400 UTC. The varying position of the moving ship complicates interpretation somewhat (Fig. 10). Large ozone concentrations reap-

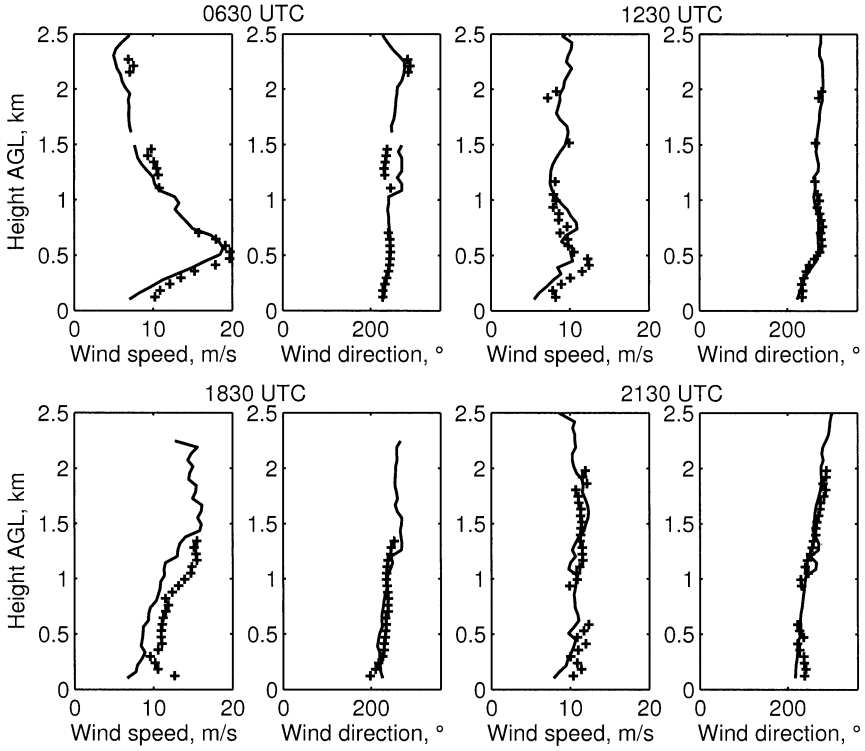

FIG. 8. Wind speed and direction measured by wind profilers at Portsmouth (solid) and the Isles of Shoals (plus signs) on 22 Jul.

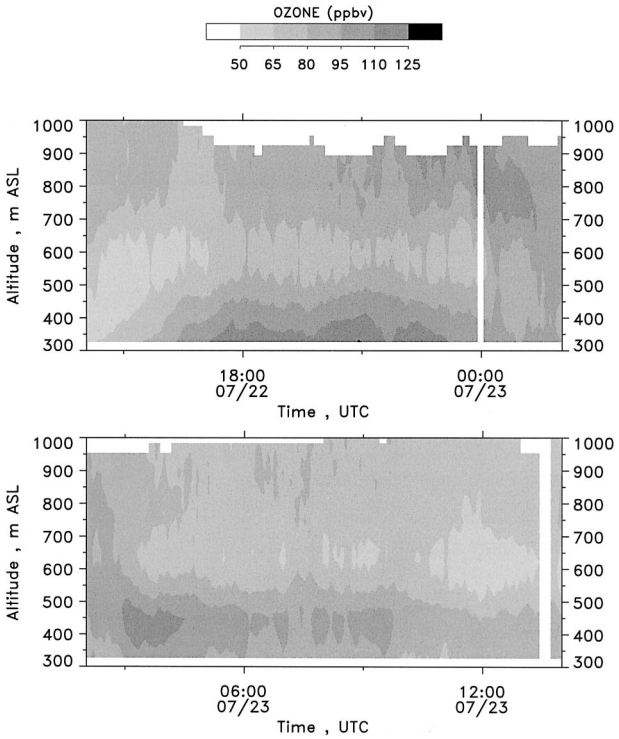

FIG. 9. Ozone concentration observed above the ship on 22 and 23 Jul.

peared at the same heights in the morning of 23 July (not shown), although the lidar was undergoing maintenance during much of that time so that data are available only for a few short time periods. Figure 11 compares the ozone measured at the surface on the ship with that measured by the lidar at 300–400 m MSL. During the day on 22 July ozone at the surface was somewhat less than aloft. During the period of large surface ozone values on 23 July, there was remarkable agreement between the lidar and surface mixing ratios when the lidar data were available. However, the surface ozone mixing ratios were much lower during the night while the values aloft remained strong. Trajectories (not shown) suggest that the nighttime ozone at 300–500 m MSL, observed farther offshore, came from the New York City vicinity rather than directly from Boston. The layer of ozone at approximately 800 m AGL near 0000 UTC 23 July also appears to have come from New York City.

Ozone mixing ratios on the ship (at the surface) and at the Isles of Shoals on 22 and 23 July during the day were similar. The isles and the ship were not exactly aligned along the profiler-measured trajectories, but the surface winds were slightly more southerly, which would place the sites in better alignment. The plume observed by the ship was sufficiently broad that we can be confident that the observations were in basically the same air mass.

In summary, during this episode the boundary layer

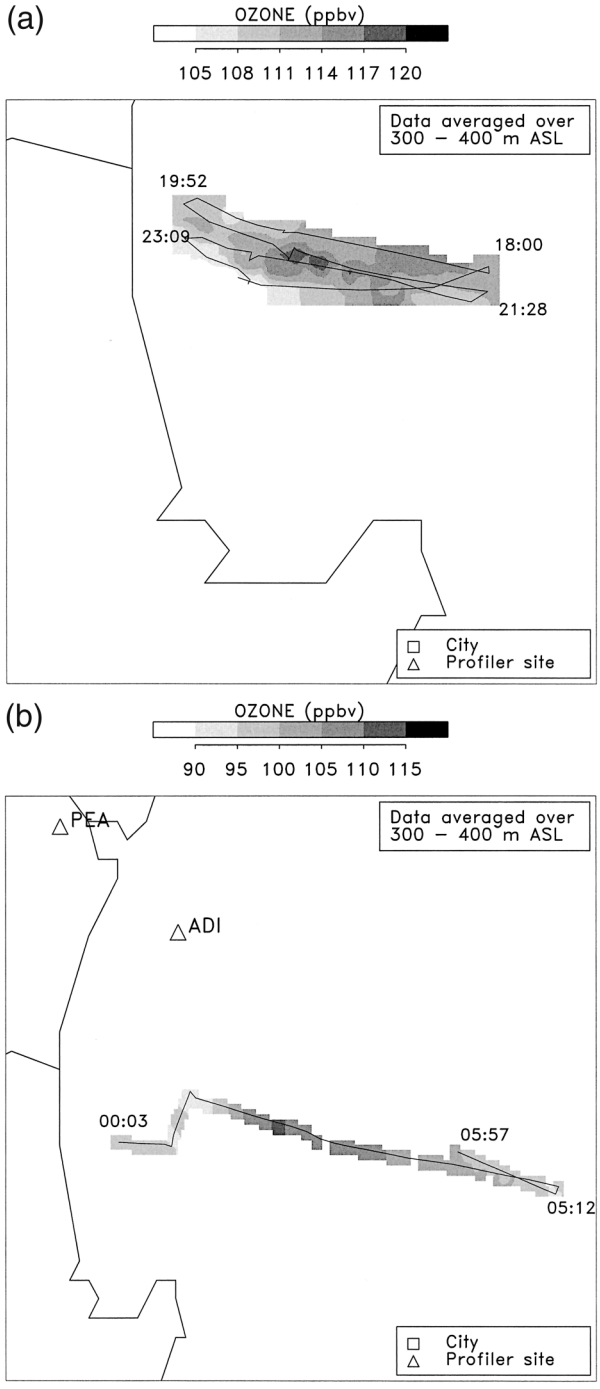

FIG. 10. Ship tracks on portions of (a) 22 and (b) 23 Jul with lidar-observed ozone concentration overplotted.

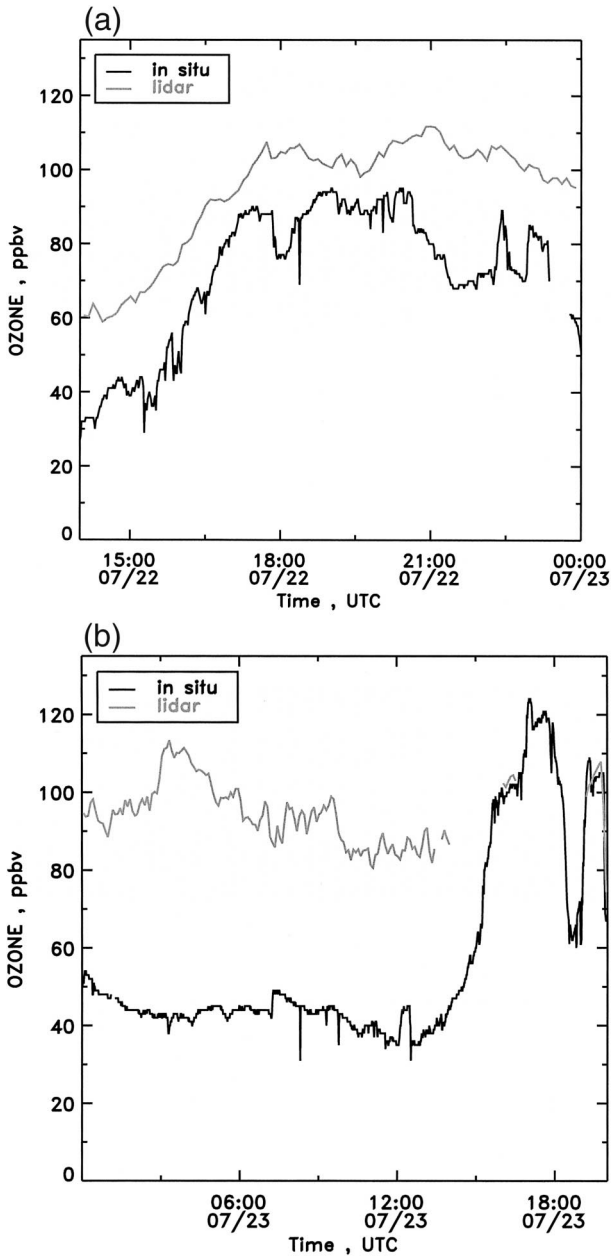

FIG. 11. Surface (in situ) and lidar-observed (at 300–400 m MSL) ozone concentrations on (a) 22 and (b) 23 Jul.

offshore was statically stable, with polluted air from land-based sources (primarily Boston) remaining near the surface. The lowest 100–200 m of the surface-based layer was cooled by contact with the cooler sea surface but not strongly modified chemically. The horizontal extent of the pollution plume as observed both at the surface and aloft was at least 10–20 km. The lowest polluted layer, that is, the layer that impacted the surface-based sensors, was 400–600-m deep and, at least at the ship location, contiguous with the layer observed by the shipborne lidar during the days. The transport was primarily large scale without major contributions from local or mesoscale effects such as land–sea breezes.

b. 11–14 August

This prolonged episode produced large ozone mixing ratios for several days and was only the most intense

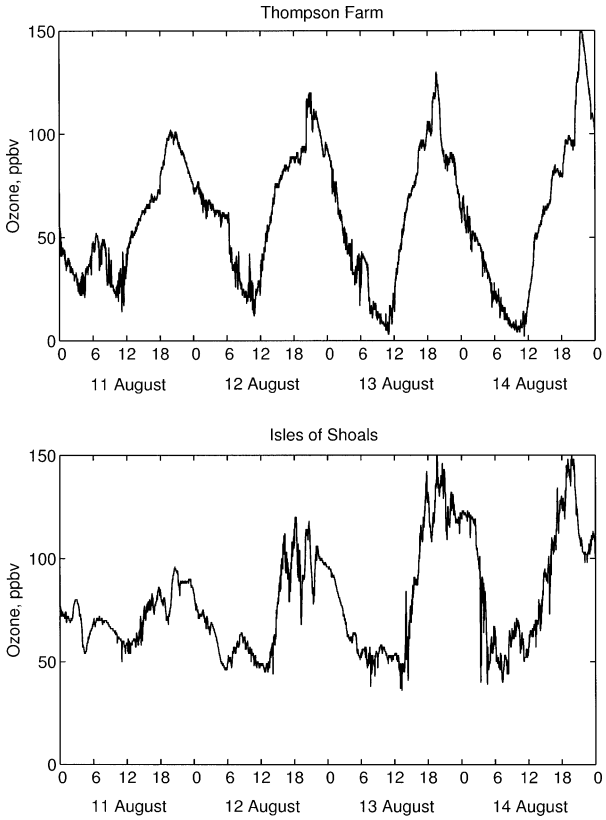

FIG. 12. Ozone mixing ratio measured at (top) Thompson Farm and (bottom) the Isles of Shoals on 11–14 Aug 2002.

part of a longer episode that extended from 10 to 19 August. Again, the largest ozone mixing ratios were found inland in Connecticut and Massachusetts and along the Massachusetts, New Hampshire, and Maine coasts, but inland New Hampshire, Maine, and Vermont were also impacted. This difference from the 22–23 July episode is also evident in the AIRMAP data (Figs. 2 and 12) where the Thompson Farm ozone mixing ratios (inland) were comparable to those at the Isles of Shoals. The episode began as the winds shifted to southwesterly early on 10 August. Winds at and near the surface remained generally southerly to southwesterly until 19 August, although northwesterly flow was observed on some nights at some sites. Observed winds from the Portsmouth and Isles of Shoals profilers are shown in Fig. 13. Winds during the most polluted period (12–14 August) were lighter than during the 22–23 July episode but never fell below 2 m s^{-1} in the profiler observations (above 100 m AGL). Diurnal vector-averaged wind magnitudes were about 2 m s^{-1} at 150 m AGL on 14 August, the day of lightest overall winds, indicating that the air mass near and offshore was never stagnant but always had some net motion. Transport times from Boston are rather uncertain but could be a substantial fraction of a day, and transport times from other source areas are likely more than a day.

Surface observations and the timing of the ozone peaks at Thompson Farm indicate that a sea breeze carried the ozone onshore. The sea-breeze onset is clear in the meteorological measurements, especially wind direction, at Thompson Farm on 11, 12, and 14 August but is not as clear on 13 August. Surface winds inland were very light at night on 12 and 13 August.

Near-surface air temperatures were warmer over land than at the Isles of Shoals throughout the episode at all times except for short periods in the early morning. Temperatures at Beverly or Portsmouth were as much as 12°C warmer than at the Isles of Shoals (Fig. 14).

The profiler observations show a sea-breeze layer (southeasterly wind direction) of varying depth during the days. The layer is only about 300 m deep at 1530 UTC when it is first observed at Portsmouth on 13 August and deepens to about 400 m by 1930 UTC. It is deeper on 14 August, approximately 600 m between 1500 and 1800 UTC, and then becomes shallower and less well-defined later in the day (Fig. 15).

At Acadia National Park, approximately 250 km northeast of the Isles of Shoals, the timing and vertical structure of ozone peaks were quite different. Figure 16 shows ozone measured at two sites—in Acadia National Park at Cadillac Mountain (466 m elevation) and McFarland Hill (122 m), as well as at Schoodic Point (30 m, ~11 km east of Cadillac Mountain), and in Kittery (on the Maine–New Hampshire border at the coast). The ozone peaks at Cadillac Mountain and McFarland Hill indicate medium-range transport rather than local production, because they occurred at night. In contrast, the peaks at Kittery occurred at midday, indicating that the production time scale was at least as important as the time scale of transport to this site, relatively near the source. The time lag between the peaks at Kittery and at Acadia National Park is consistent with transport at 8–10 m s^{-1}, roughly consistent with the wind profiler measurements, although there is some uncertainty that arises from the possibility of ongoing ozone production during the transport. The most interesting point to be taken from Fig. 16, however, is the difference between the ozone at the elevated sites and at sea level. Cadillac Mountain, the highest-elevation site, saw greater peak ozone than McFarland Hill, which is somewhat lower. Schoodic Point received much less ozone than either of the two more elevated sites. This pattern holds true in general over several years of measurements at sites near Acadia National Park, indicating that elevated transport of pollution to that area is common.

This episode can be summarized as follows. The winds were relatively weak but not stagnant near the coast or offshore. The most polluted air masses were transported offshore (as in the 22–23 July episode) but were carried onshore in New Hampshire by the sea breeze in a layer 400–600 m deep. Polluted air near the surface was transported in a layer that remained in contact with the surface despite passing over cooler water. The surface layer was at least intermittently turbulent,

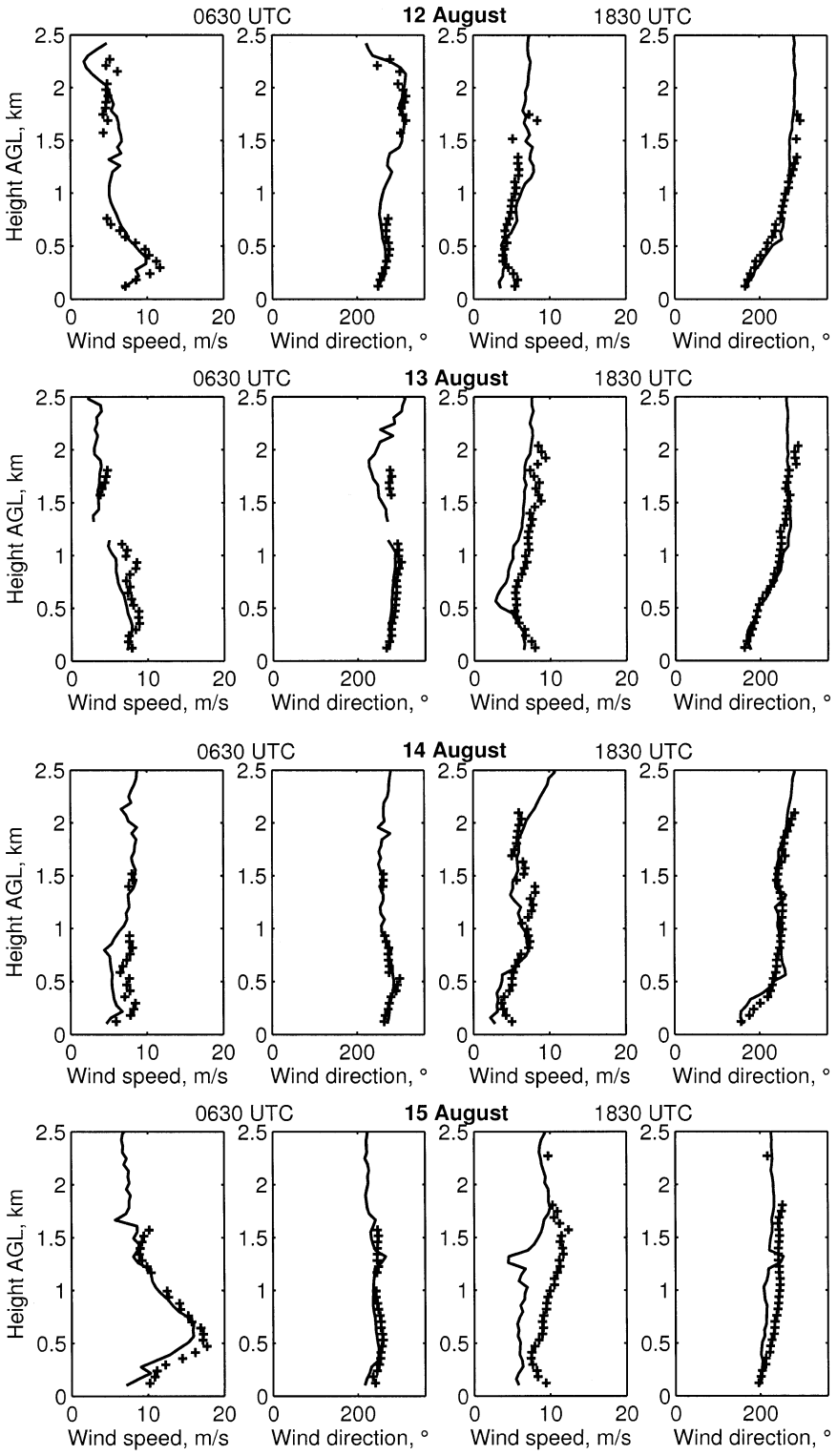

FIG. 13. Wind speed and direction measured by wind profilers at Portsmouth (solid) and the Isles of Shoals (plus signs) at 0630 and 1830 UTC 12–15 Aug showing the sea-breeze pattern.

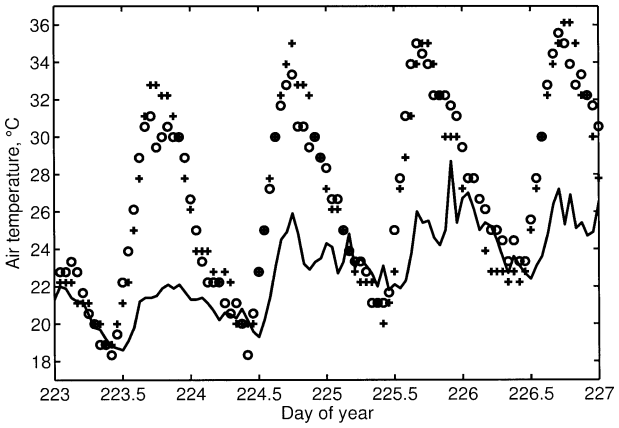

FIG. 14. Surface air temperatures at Beverly (plus signs), Portsmouth (circles), and the Isles of Shoals (line) on 11–14 Aug.

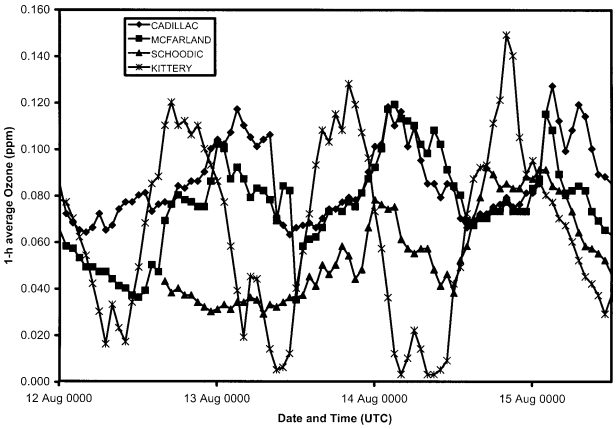

FIG. 16. Ozone concentration observed by operational monitors at three sites in and near Acadia National Park (Cadillac Mountain, McFarland Hill, and Schoodic Point) and at Kittery (on the New Hampshire border at the coast) on 12–15 Aug 2002.

with advection and shear production likely both contributing. A higher layer reached Cadillac Mountain in Acadia National Park, at 466 m MSL, carrying ozone concentrations similar to those that reached sea-level monitors farther south, but the surface layer near Acadia National Park had less ozone.

3. Discussion

The most polluted air in coastal New Hampshire and Maine in 2002 came over the coastal waters from the Boston area. Pollutants from Boston were augmented by longer-range transport from the more distant east coast urban corridor, and smaller (but still significant) pollutant concentrations were transported over land. Pollution episodes in coastal areas were associated with moderate, definitely nonstagnant winds. During the 11–14 August episode, winds in source areas were light, and so were winds in some inland areas with moderately

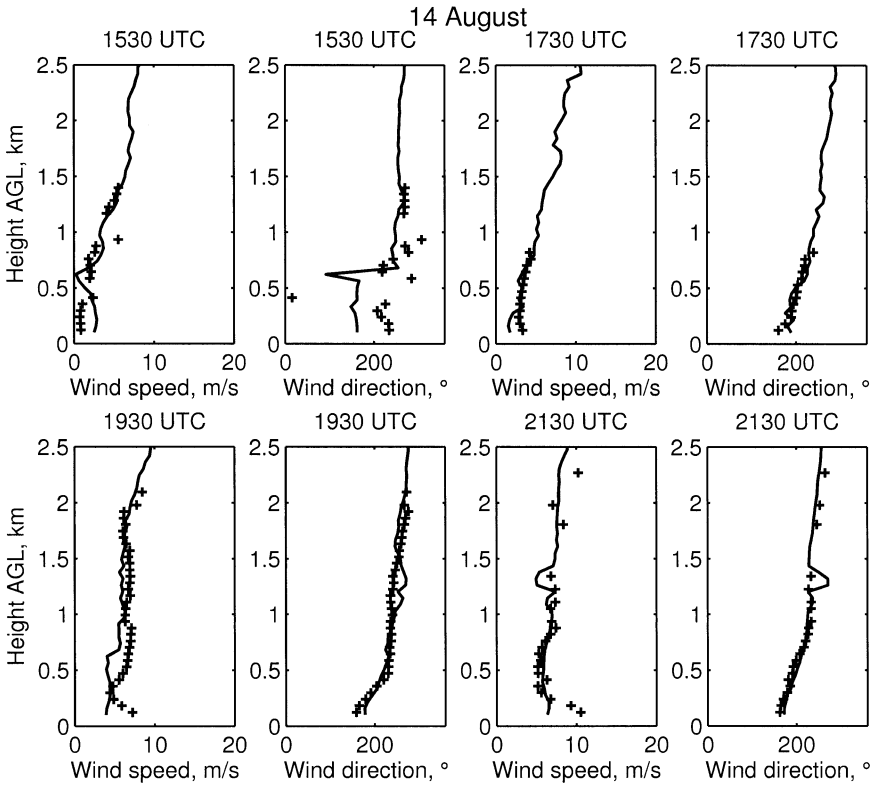

FIG. 15. Wind speed and direction measured by wind profilers at Portsmouth (solid) and the Isles of Shoals (plus signs) on 14 Aug showing the sea-breeze development.

large pollutant concentrations, but even in this episode the largest concentrations were transported by winds with well-defined directions and nonvanishing daily vector average speeds. This contrasts with episodes in other parts of the United States, which occur in stagnant or recirculating conditions (Solomon et al. 2000; Hidy 2000). What appears to be a buildup of pollutants over a few days, similar to that caused by stagnation, is due either to the addition of pollutants transported from longer distances or to coincidences involving the transport directions and/or speeds.

In the episodes examined above, transport of polluted air to coastal New Hampshire and coastal southwest Maine occurred in a layer in contact with the surface. The lowest portion of this layer was turbulent despite being strongly statically stable. It was evidently turbulent because it cooled during the transport; without turbulence there would have been little or no cooling. The transport was definitely not isentropic, as assumed by Angevine et al. (1996) and Dye et al. (1995). The fact that the layer contained large pollutant concentrations justifies the assumption of Lagrangian transport; that is, the cooling could not have been due to temperature advection. The polluted layer was 400–600 m deep in both cases studied. The turbulently cooled portion of the layer was less than 100 m deep in the 22–23 July case where we have vertical profiles. Note that we are using the term ''stable'' in the sense in which is it used in the common phrase ''stable boundary layer,'' that is, a layer in which potential temperature increases with height (statically stable) but is not necessarily free of turbulence. Stable layers can have turbulence because of shear-driven instability, or turbulence may be advected into the layer either horizontally (from the land, e.g.) or vertically (from shear instability above the layer). See Vickers et al. (2001), Mahrt et al. (2001), and Van de Wiel et al. (2002) for current thinking on these boundary layer processes.

There were other layers, equally or more polluted, above the surface. They affected areas of the coast farther northeast, such as Acadia National Park, and contributed to long-range transport. Polluted layers at various heights were observed in NARE 1993 (Angevine et al. 1996; Buhr et al. 1996; Daum et al. 1996; Kleinman et al. 1996). Lofting of originally surface-based layers was not observed during NEAQS, except possibly by the routine observations at Acadia National Park, but the observation platforms were not in position to observe it far offshore, where the water was colder and lofting was more likely.

The particular area of the coast reached by the polluted air depends on details of the transport winds. The 22–23 July 2002 episode was dominated by large-scale transport, and, therefore, primarily impacted coastal New Hampshire and Maine. On the other hand, the 11–14 August 2002 episode had sea breezes that brought the polluted air to inland New Hampshire. A concise summary of the sea-breeze literature can be found in Källstrand et al. (2000). Savijärvi and Alestalo (1988) make a number of points relevant to the New England situation, including the fact that in their simulations, a background wind to the left of perpendicular to the coast, produces the strongest sea breezes.

Does the transport pattern depend on time of day? Observed ozone mixing ratios are strongly diurnal and strongly correlated with air temperature, but that might be true even if the ozone were being produced continuously from constant concentrations of fresh precursors. Emissions from urban areas depend on time of day, and so does static stability, both over land and over water. The urban boundary layer over Boston, for example, will be shallow at night while receiving relatively weak emissions and deeper during the day when emissions are stronger. The shallow nocturnal boundary layer is likely partitioned into fewer layers over the water, while the deeper daytime urban boundary layer may populate several layers, advected differentially, over the water. The data from the Isles of Shoals are consistent with essentially continuous large-scale transport that does not depend strongly on time of day, with a superimposed mesoscale sea-breeze circulation on some days. A plausible scenario is this: At night, the stability is weak because the land temperature is cool, comparable to the sea surface temperature. Therefore, wind shear is sufficient to keep the lower layer turbulent. During the day, the stability is stronger because the land is warmer, but advected turbulence from the land boundary layer is sufficient to keep the lowest layer turbulent, at least intermittently. It is difficult with the present dataset to determine precisely the transport time from the sources. In the 22–23 July case, we can estimate a 2–3-h transport time from Boston to Cape Ann and the Isles of Shoals. The shape of the ozone peaks at the isles and on the (moving) ship, which are strongly correlated with solar zenith angle, suggests that the surface ozone mixing ratio is dominated by production either during transport or over the sources rather than by diurnal changes of the transport itself. Transport times to distant monitors such as those at Acadia National Park (330 km from Boston) can vary from a few hours to a day, and the timing of the peaks there is dominated by transport effects. The monitors at Acadia National Park did not show strong concentrations on the first day of any of the episodes.

The behavior of the boundary layer at the downwind (receiving) coast also depends on the time of day and the relative temperatures of the incoming air and the land. In the 2002 episodes, because the most polluted air was transported at the surface, monitors at the coast in New Hampshire and southwest Maine measured large concentrations regardless of the stability inland. We do not need to invoke fumigation to explain the observations. In fact, it is likely that concentrations would decrease substantially as vertical mixing deepens if air was carried inland under convective conditions.

Is overwater transport more efficient than transport

over land? Because the overwater trajectory segments are always stable in these episodes, the pollutants in the surface layer are not diluted by deep vertical mixing. The surface layer is, however, turbulent, as evidenced by its cooling, and, therefore, pollutants could be lost to surface deposition. However, ozone and most of its precursors are deposited much more slowly to water surfaces than to vegetation, and so the polluted layers over water retain most of their ozone and precursors.

The model study of Smedman et al. (1997) shows a stable layer near shore, as we observe here, but also shows the reformation of a mixed layer farther downwind. That mixed layer is not observed in NEAQS or in NARE 1993. In NEAQS the fixed and shipborne observations were generally too close to shore during offshore flow to observe any reformed mixed layer, but the NARE 1993 observations were not so constrained. One possibility is that continually falling sea surface temperatures along trajectories renew the stable layer, equilibrium is never reached, and the mixed layer does not reform.

Some details of the coastal boundary layer situation are not perfectly constrained by the data available from NEAQS. Fine-resolution modeling studies would help to show the full four-dimensional structure of pollutant transport. It would be useful to have direct measurements of surface fluxes offshore. This would allow us to understand the intensity and possible intermittency of turbulence over the water. It would also be interesting to look for the reformed neutral layer at long overwater fetch, possibly with an aircraft platform.

This paper has emphasized periods with large ozone mixing ratios (generally over 100 ppbv), because those periods provide the clearest picture of specific sources and transport patterns. Weaker episodes may also be of interest to the policy and regulatory communities, and appear to occur under a broader range of conditions.

To summarize, the coastal boundary layer influences pollutant transport in northern New England by allowing for stable layers over water that carry pollutants, relatively undiluted and with minimal deposition, to distant (20–200 km) areas on other parts of the coast. The sea breeze modifies the large-scale flow to select the particular sites that receive polluted air. Elevated layers transport polluted air very long distances (200–2000 km).

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