Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) 2004: 2. Tropospheric ozone budgets and variability over northeastern North America

Anne M. Thompson,[1] Jesse B. Stone,[1] Jacquelyn C. Witte,[2,3] Sonya K. Miller,[1] Samuel J. Oltmans,[4] Tom L. Kucsera,[3,5] Kelly L. Ross,[1] Kenneth E. Pickering,[3] John T. Merrill,[6] Gerry Forbes,[7] David W. Tarasick,[8] Everette Joseph,[9] F. J. Schmidlin,[10] W. Wallace McMillan,[11] Juying Warner,[12] Eric J. Hintsa,[13,14] and James E. Johnson[15,16]

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[1] Daily ozone soundings taken from the R/V *Ronald H. Brown* from 7 July through 11 August 2004 as part of the Intercontinental Chemical Transport Experiment (INTEX) Ozonesonde Network Study (IONS) are used to investigate the vertical structure of ozone over the Gulf of Maine and to characterize variability in sources of tropospheric ozone: stratosphere, regional convection and lightning, advection, and local boundary layer pollution. These soundings were part of a network of twelve IONS (http://croc.gsfc.nasa.gov/intex/ions.html) stations that launched ozonesonde-radiosonde packages over the United States and maritime Canada during the INTEX/International Consortium for Atmospheric Research on Transport and Transformation (ICARTT)/New England Air Quality Study (NEAQS) project from 1 July to 15 August 2004. Four of the IONS stations were in mid-Atlantic and northeast United States; four were in southeastern Canada. Although the INTEX/ICARTT goal was to examine pollution influences under stable high-pressure systems, northeastern North America (NENA) during IONS was dominated by weak frontal systems that mixed aged pollution and stratospheric ozone with ozone from more recent pollution and lightning. These sources are quantified to give tropospheric ozone budgets for individual soundings that are consistent with tracers and meteorological analyses. On average, for NENA stations in July-August 2004, tropospheric ozone was composed of the following: 10–15% each local boundary layer and regional sources (the latter including that due to lightning-derived NO) and 20–25% stratospheric ozone, with the balance (~50%) a mixture of recently advected ozone and aged air of indeterminate origin.

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1. Introduction

[2] Intercontinental transport is a topic of considerable interest because relatively modest amounts of imported pollution can modify the background burden of key constituents related to air quality: O_3, CO and aerosols. An expanding body of observations has delineated processes

[1]Meteorology Department, Pennsylvania State University, University Park, Pennsylvania, USA.
[2]Science Systems and Applications, Inc., Lanham, Maryland, USA.
[3]NASA Goddard Space Flight Center, Greenbelt, Maryland, USA.
[4]Earth Systems Research Laboratory, NOAA, Boulder, Colorado, USA.
[5]Also at Joint Center for Environmental Technology, University of Maryland Baltimore County, Baltimore, Maryland, USA.
[6]Graduate School of Oceanography, University of Rhode Island, Narragansett, Rhode Island, USA.
[7]Environment Canada Sable Island, Dartmouth, Nova Scotia, Canada.
[8]Meteorological Services Centre, Environment Canada, Downsview, Ontario, Canada.
[9]Department of Physics and Astronomy, Howard University, Washington, D. C., USA.
[10]Wallops Flight Facility, NASA Goddard Space Flight Center, Wallops Island, Virginia, USA.
[11]Department of Physics, University of Maryland Baltimore County, Baltimore, Maryland, USA.
[12]Joint Center for Environmental Technology, University of Maryland Baltimore County, Baltimore, Maryland, USA.
[13]Woods Hole Oceanographic Institution, Woods Hole, Massachusetts, USA.
[14]Now at Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts, USA.
[15]Joint Institute for Study of Atmospheres and Oceans, University of Washington, Seattle, Washington, USA.
[16]Also at Pacific Marine Environmental Laboratory, NOAA, Seattle, Washington, USA.

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and key transport patterns to and from North America [*Singh et al.*, 1994; *Parrish et al.*, 1998; *Forster et al.*, 2001; *Honrath et al.*, 2004; *Price et al.*, 2004; *Cooper et al.*, 2005]. A large complement of experiments came together in July-August 2004 to study North American import and export more closely. Designated as ICARTT (http://www.esrl.noaa.gov/csd/ICARTT/) [*Fehsenfeld et al.*, 2006] and INTEX-NA (Intercontinental Chemical Transport Experiment [*Singh et al.*, 2006]), approaches included multiple ground sites, aircraft, satellites and associated models.

[3] *Thompson et al.* [2007a] describe a network of ozonesonde stations called IONS (INTEX Ozonesonde Network Study) that was set up during INTEX/ICARTT to accomplish the following: (1) coordinated daily launches with same-day transmission of data to guide Lagrangian aircraft sampling, (2) geographical coverage with a cross-sectional view from the south central United States through maritime Canada (eight fixed sites and the R/V *Ronald H. Brown* ship operating in the Gulf of Maine) to characterize eastern North American ozone outflow, (3) a west Coast site allowed study of trans-Pacific flows, and (4) spatial and temporal density optimization for comparison of sondes with satellite ozone data incorporated into a regional assimilation model [*Pierce et al.*, 2007].

[4] Nearly three hundred ozone profiles from 12 stations (Figure 1) were collected during IONS. The first findings of IONS are summarized by *Thompson et al.* [2007a] as follows:

[5] 1. Typical summertime stagnation-pollution was relatively rare in June-July-August (JJA) 2004 over the northeastern United States and maritime Canada. Precipitation and cloudiness were ~25% greater than normal, conditions that tend to suppress pollution formation and persistence. Ozone over the mid-Atlantic and northeastern United States, as well as maritime Canada, appeared to be below normal. This was confirmed with statistical analysis of the IONS sondes, related tracers and a longer-term ozonesonde record at Wallops Island (37.9°N, 75.5°W). Surface O_3 mixing ratios over U.S. northeastern urban areas fell in the lowest 35% of values recorded during JJA in 1996–2004.

[6] 2. A persistent low-pressure system centered around the Great Lakes led to frequent exchanges between stratosphere and troposphere, leading to a contribution of stratospheric O_3 to the tropospheric O_3 budget, ~20–30% depending on the site [*Thompson et al.*, 2007a, Table 3].

[7] 3. Viewing IONS sondes across NENA (northeastern North America) shows that O_3 over the R/V *Ronald H. Brown* was in a transition zone in terms of pollution and stratospheric influence. Sable Island to the east (Figure 1) was more frequently influenced by subtropical marine air. Narragansett, to the west, was more polluted in the lower troposphere than over the Gulf of Maine.

[8] In the present paper, soundings and surface O_3 from the R/V *Ronald H. Brown,* along with meteorological analyses, trajectories and satellite observations are used to refine the interpretation of tropospheric O_3 variability and sources over New England and maritime Canada as presented by *Thompson et al.* [2007a]. Our investigation gives an overview of O_3 characteristics over the ship from 7 July to 11 August 2004, with context provided by Narragansett, Sable Island and other IONS sites. Useful perspectives are given by (1) O_3 mixing ratio variability, altitude versus time, at individual sites, and (2) an Eulerian cross section of O_3 distributions across a series of stations on a given day (section 3.1). Our investigation also provides budgets for O_3 contributions from the boundary layer (BL), stratosphere (ST), regional convection and lightning (RCL), and advection (AD) (section 3.2). Finally, our investigation examines representative episodes over the course of the R/V *Ronald H. Brown* cruise, using shipboard data, sondes and a buoy record of O_3 in the Gulf of Maine (section 3.3).

2. Measurements and Methods

2.1. IONS-04 and Related Observations

2.1.1. Ozone Data From Sondes, Ship, and Buoy

[9] Figure 1 shows locations of all IONS sites in 2004. Coordinates are given by *Thompson et al.* [2007a]. Soundings were made with ECC (electrochemical concentration cell) sondes and standard radiosondes (Vaisala or Sippican) at all locations. The system used at Pellston, Narragansett, Beltsville and the R/V *Ronald H. Brown* is described by *Thompson et al.* [2000] and *Johnson et al.* [2002]. The accuracy of the sonde is assured by calibration with colocated surface O_3 sensors prior to launch at most sites. Recent tests of the ECC sonde put accuracy in the troposphere and lower stratosphere at 5–10% [*Smit et al.*, 2007] and precision, defined here as the reproducibility of a single sounding is 5–7% [*Thompson et al.*, 2007b; *Smit et al.*, 2007]. Given the time response and ascent rate of the ECC sonde, the effective vertical resolution is ~100 m. At five IONS sites (Trinidad Head, Houston, Pellston, Narragansett, the R/V *Ronald H. Brown*) soundings were made daily between noon and 1400 local time to capture overpasses of the Aqua and Aura satellites. The remaining stations launched midday 1–3 times per week, except for Sable Island, where the ozonesonde accompanied the standard 2300 UTC radio sounding.

[10] Observations from the R/V *Ronald H. Brown* data were taken over two legs operated out of Portsmouth, NH. The first sailing was 5–23 July 2004; the second leg was from 26 July to 12 August 2004. The full suite of instrumentation on the R/V *Ronald H. Brown* appears in *Fehsenfeld et al.* [2006, Table A3]. Ozone mixing ratios were measured at 18 m above sea level using two different instruments operated by NOAA/Pacific Marine Environmental Laboratory (PMEL): a Dasibi model 1008AH and a TEI model 49 that had been calibrated to a standard O_3 instrument at NOAA Earth Systems Research Laboratory's Global Monitoring Division. The shipboard concentrations reported here are the average of the two PMEL instruments. This mean PMEL O_3 concentration agreed to within 1–2% to a third TEI O_3 instrument operated by NOAA Earth Systems Research Laboratory's Chemical Sciences Division).

[11] Also used are O_3 data measured by a 2B Technologies uv sensor-based instrument deployed on a buoy operated by Woods Hole Oceanographic Institution [*Hintsa et al.*, 2004, 2005] at 42.8°N, 70.3°W from 27 June to 6 September 2004. When buoy O_3 was compared to nearby

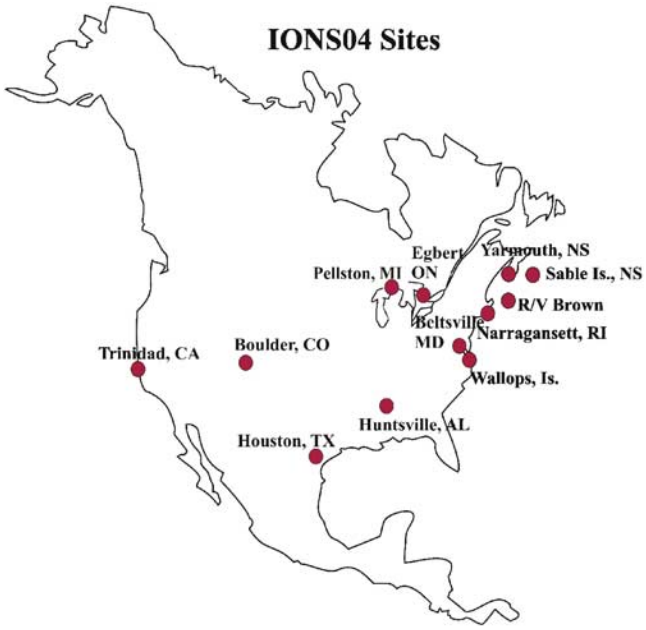

Figure 1. Map of IONS sites in July and August 2004, with the R/V *Ronald H. Brown* operating in the Gulf of Maine. Selected cruise segments appear in Figure 11.

O_3 from the R/V *Ronald H. Brown* on a couple occasions, agreement was within 2 ppbv.

2.1.2. Meteorological Analyses, Satellite Data, and Trajectories

[12] In addition to the O_3, temperature and relative humidity (P-T-U) profiles returned from the sondes, forward and backward air parcel trajectories were run from standard pressure levels at each site. The kinematic version of the *Schoeberl and Newman* [1995] trajectory model was used with meteorological fields generated from the GEOS-4 version of the GSFC Assimilation Model [*Bloom et al.*, 2005]. Images of the profiles and trajectories are viewable at: http://croc.gsfc.nasa.gov/intex/ions.html. Ancillary meteorological information, satellite imagery and trajectory-based domain-filling products used in the analyses can be viewed at http://croc.gsfc.nasa.gov/intex. Among the variables used most frequently from GEOS-4 are the RD (reverse-domain-filled) Ertel's potential vorticity (pv; 1 potential vorticity unit = 10^{-6} K kg^{-1} m^2 s^{-1}), tropopause height and cloud-detrainment mass flux. Useful potential source information for lightning and advected pollution is taken from back trajectories that incorporate weighted input from lightning flashes (EL, for exposure to lightning flashes recorded by the National Lightning Detection Network) and absorbing aerosols (ED, dust exposure), with the latter measured by TOMS, the Total Ozone Monitoring Spectrometer, on the Earth Probe satellite. Convective exposure maps, based on satellite-derived cloud-top temperatures, are found at http://bocachica.arc.nasa.gov/INTEX-NA. Maps of tropospheric CO from the Atmospheric InfraRed Sounder (AIRS) on board the Aqua satellite are used to evaluate the impacts from large fires in Alaska and Canada during IONS [*McMillan et al.*, 2005; W. W. McMillan et al., AIRS views of transport from 10–23 July 2004 Alaskan/Canadian fires: Correlation of AIRS CO and MODIS AOD and validation of AIRS CO and O3 retrievals from INTEX-NA aircraft in situ measurements, unpublished manuscript, 2007, hereinafter referred to as McMillan et al., unpublished manuscript, 2007].

2.2. Analysis for Ozone Budgets

[13] The method used to evaluate contributions to the IONS tropospheric O_3 budget, described by *Thompson et al.* [2007a], is based on the assumption that O_3 in each sounding originates from four processes: boundary layer photochemistry (BL); stratosphere to troposphere transfer (ST); interaction of regional pollution with convection plus photochemical reactions from lightning-generated NO (RCL); and advection, including recent transport and aged ozone (AD).

[14] The individual terms for each sounding are computed in the following steps.

[15] 1. The BL term is integrated O_3 from the surface to 1 km, a typical altitude for the top of the mixed layer (see discussions of boundary layer structure over the R/V *Ronald H. Brown* by *Angevine et al.* [2006]).

[16] 2. The ST and RCL amounts are based on the *Thompson et al.* [2007a] modification of classifying waves in stable O_3 layers that was introduced by *Teitelbaum et al.* [1994, 1996] and *Pierce and Grant* [1998]. Each stable lamina of O_3 that is associated with a Rossby wave (RW) is a potential source of stratospheric influence. To confirm that a layer is of stratospheric origin, humidity from the radiosonde is used to scale RW to ST, with back trajectory origins, measured in terms of coincidence of intersection with pv > 2 pvu, as a secondary weighting factor [*Stone*, 2006; *Thompson et al.*, 2007a]. This is performed for each lamina in each sounding. For the three sites described in detail in the present paper, an average of 68% of RW layers are assigned to ST. Stratospherically influenced O_3 layer amounts are added together to give the total ST amount. Column integrated quantities (Dobson Units; 1 DU = 2.69×10^{16}cm^{-2}) are reported. The RCL identification is based on the Gravity Wave, GW, classification of laminae [*Teitelbaum et al.*, 1994, 1996; *Thompson et al.*, 2007a]. Convectively generated GW are assumed to represent conditions in which O_3 or O_3 precursors are injected into the free troposphere, possibly with lightning leading to NO release and photochemical O_3 formation [*Dickerson et al.*, 1987; *Pickering et al.*, 1991, 1996; *Thompson et al.*, 1996]. The GW layers are assigned to RCL when lightning exposure (EL), cloud-detrainment mass flux, and elevated RH indicate that convection has occurred. The GW and RCL designations based on tracers coincide in more than 90% of the profiles analyzed. Over the entire IONS data set, however, the RCL identification may be underestimated because lamina-labeling requires stable layers [*Teitelbaum et al.*, 1996; *Pierce and Grant*, 1998; *Thompson et al.*, 2007a]; soundings that reflect active convection are not amenable to the technique. All but 5% of the Sable Island soundings are analyzed by the lamina technique. The effect of the missing information for other sites is estimated as follows. Section 3.3 shows that fresh advection can add 10 DU to the tropospheric O_3 column in a day. The highest RCL column amount in several individual profiles is 20 DU.

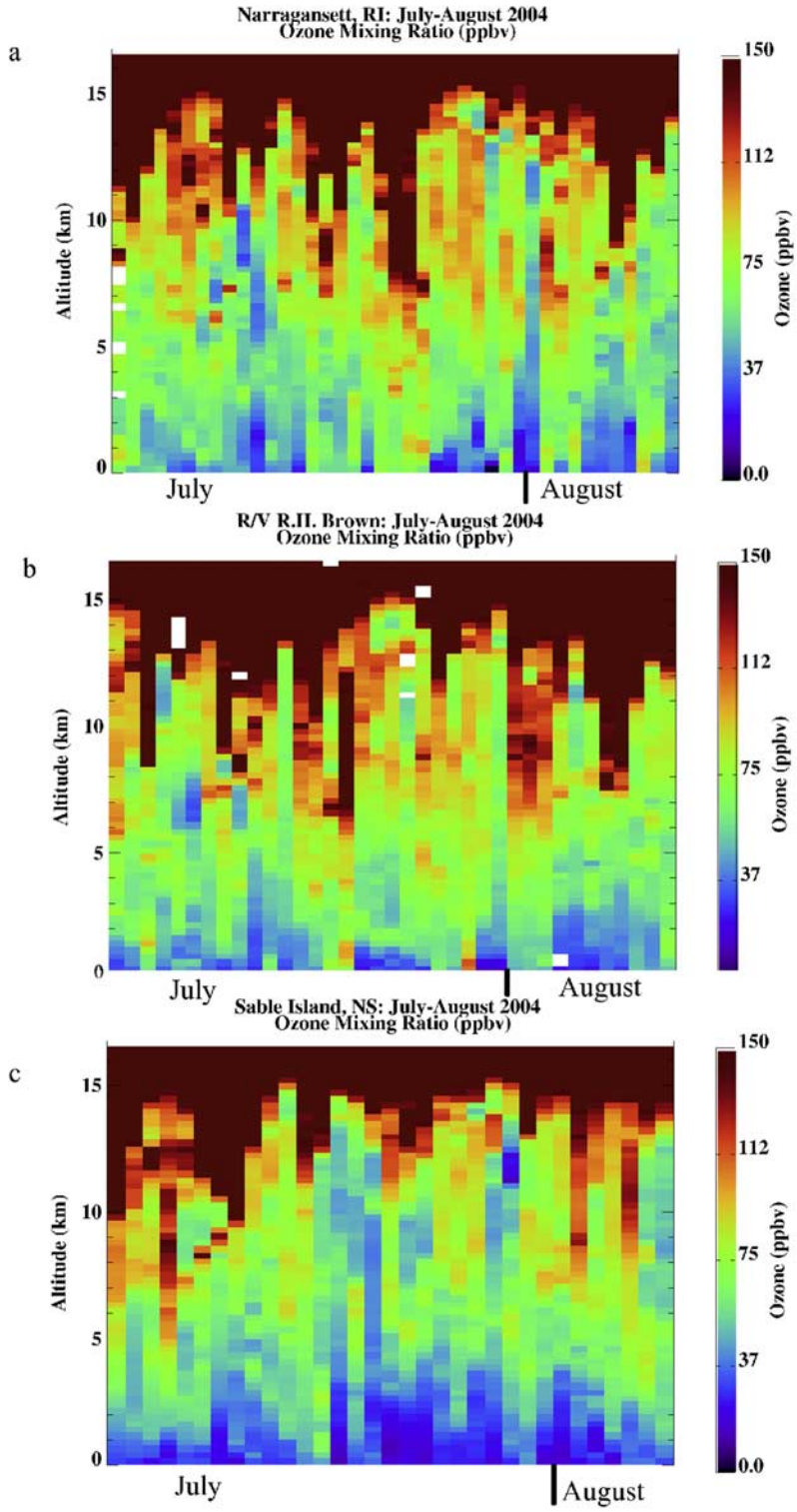

Figure 2. Cross section of O_3 mixing ratios. IONS soundings at (a) Narragansett (41.5°N, 71.4°W), (b) R/V *Ronald H. Brown*, and (c) Sable Island (43.9°N, 60.0°W).

Undercounting convection by 10–20 DU for the 20% of profiles not analyzed for the R/V *Ronald H. Brown* and Narragansett suggests that 2–4 DU or 5–10% of a profile with pollution (~50 DU), on average, might be underestimated for RCL.

[17] 3. After ST, RCL and BL O_3 amounts are determined, their sum is subtracted from total tropospheric O_3 to give a residual amount, designated AD, that includes recently advected O_3 and O_3 of indeterminate origins. The latter is estimated at 10–15 DU for any given sounding, on

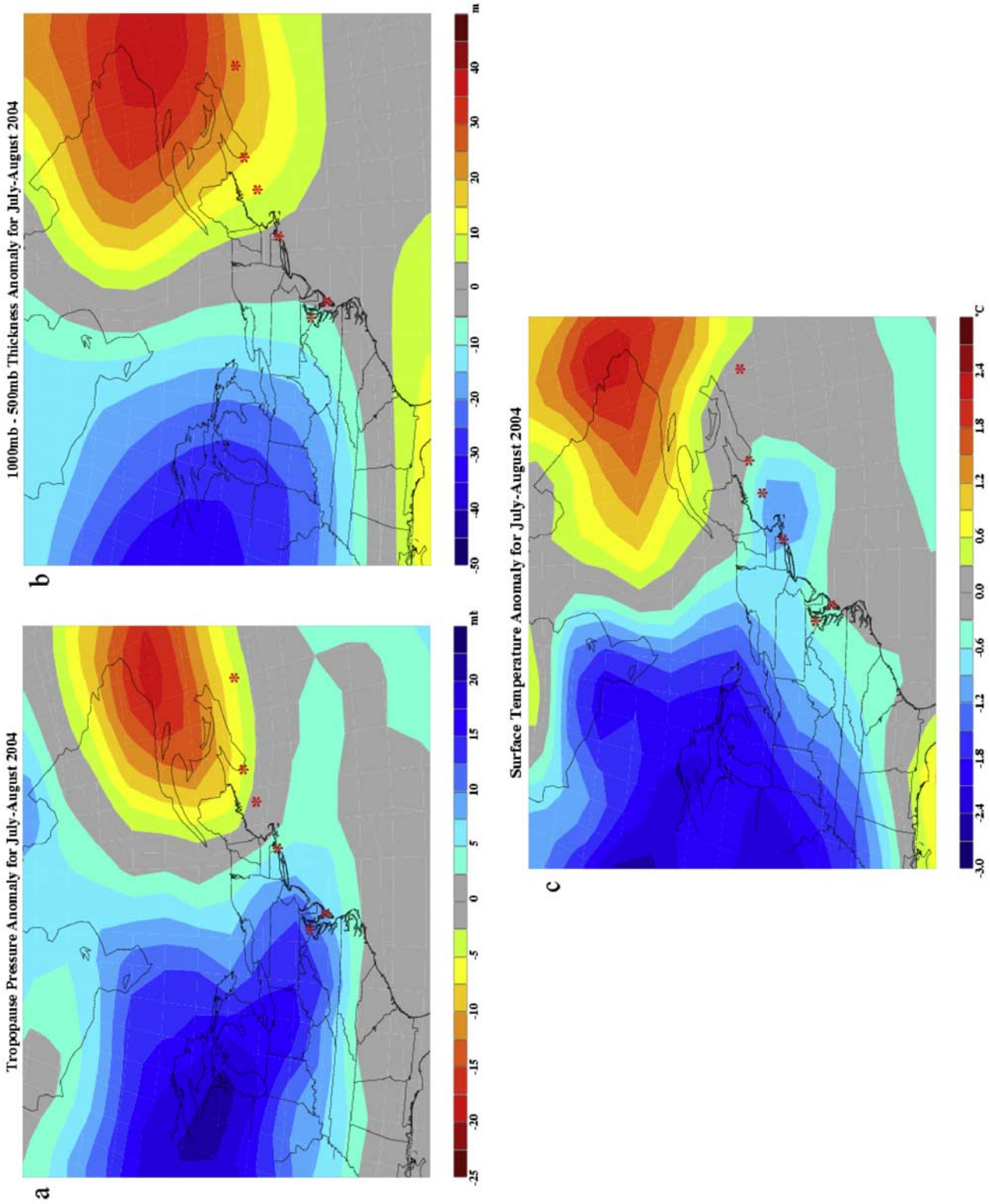
a
Tropopause Pressure Anomaly for July-August 2004
-25 -20 -15 -10 -5 0 5 10 15 20 mb
b
1000mb - 500mb Thickness Anomaly for July-August 2004
-50 -40 -30 -20 -10 0 10 20 30 40 m
c
Surface Temperature Anomaly for July-August 2004
-3.0 -2.4 -1.8 -1.2 -0.6 0.0 0.6 1.2 1.8 2.4 °C

Figure 3

Table 1. Tropospheric Ozone Budget Summaries for IONS-04 Soundings (1 July to 13 August 2004) at NENA Locations[a]

IONS-04	Belts	WFF	Narr	*RHB*	Yarm	Sable
O_3 (to 15 km), DU	58.2	49.6	57.2	60.6	52.8	47.5
O_3 (to OTP),[b] DU	47.1	47.5	44.7	45.6	45.4	40.7
OTP, km	12.1	13.8	12.2	12.5	12.6	14.8
ST O_3,[c] DU	12.6	13.0	10.5	8.3	13.6	8.9
ST O_3, fraction	0.27	0.27	0.27	0.19	0.30	0.22
RCL O_3,[c] DU	4.7	7.0	3.6	3.7	4.4	5.6
RCL O_3, fraction	0.10	0.15	0.08	0.08	0.10	0.14
AD O_3, fraction	0.53	0.49	0.55	0.65	0.52	0.58
BL (surface to 1.0 km), DU	5.0	4.4	4.4	3.6	3.8	2.6
BL (1.0 km fraction to OTP)	0.11	0.09	0.10	0.08	0.08	0.06
Surface O_3 (0–100 m),[d] ppbv	66	47	46	30	35	25

[a]WFF, Wallops.
[b]OTP, ozonopause, which is close to mean thermal tropopause [*Thompson et al.*, 2007a].
[c]Detailed descriptions of derivation of ST and RCL O_3 given by *Thompson et al.* [2007a].
[d]Refer to *Thompson et al.* [2007a] for discussion of surface O_3 and for statistics of major U.S. cities between Washington and Boston in 2004.

the basis of a minimum AD value over the NENA sites (illustrated in section 3.2, Figure 8) and can be designated as "background O_3," consisting of aged, unresolvable contributions from ST, RCL and advection.

[18] The ST and RCL O_3 components represent processes on the timescale of a week or so. During INTEX-A, an O_3 sensor with guided balloon technology followed O_3 laminae with > 100 ppbv mixing ratios from New England to the North Atlantic for a week or more [*Mao et al.*, 2006]. Certain trajectory-based techniques allow budget estimates based on longer-term histories. Using a FLEXPART approach [*Stohl et al.*, 1998; *Stohl*, 2001], *Cooper et al.* [2006] conclude that NO from lightning was responsible for most upper tropospheric O_3 measured in IONS soundings over eastern North America. Relevant source apportionments of summertime European O_3 soundings and aircraft profiles are given by *Collette and Ancellet* [2005].

3. Results and Discussion

3.1. Overall NENA Variability

[19] During IONS-04, the R/V *Ronald H. Brown*, Narragansett, and Sable Island were within 1000 km of one another. However, Figure 2, showing the day-to-day variability at each site, displays differences in O_3 distributions among the stations. Variations in structure and mean altitude of the tropopause, at the orange-brown transition, is one area of contrast. The troposphere above 1–2 km, where green-yellow patterns vary greatly, is also distinct over each station.

[20] The tropopause during July-August 2004 was lower than normal south and west of the R/V *Ronald H. Brown* (Figure 3a), which is consistent with the persistent low-pressure system centered over the Great Lakes during much of this period. On average, the lower troposphere was warmer (Figure 3b) over the Gulf of Maine and eastern Canada. However, the warm anomaly did not extend to the surface because of an anomalous onshore component to the surface winds (Figure 3c). A summary of synoptic analysis for NENA during IONS appears at http://croc.gsfc.nasa.gov/intex/ions.html.

[21] The boundary layer, nearly always very stable during the cruise [*Angevine et al.*, 2006], ranged from 0.5 to 1 km in most cases, and also differed in O_3 over the three sites. The light green-yellow shades (~75–80 ppbv O_3) near the surface over Narragansett (Figure 2a) signify more pollution. There is almost no surface pollution at Sable Island and few occurrences over the R/V *Ronald H. Brown* (Figures 2b and 2c). A daily inventory of air parcel origins over the R/V *Ronald H. Brown*, Narragansett and Sable Island, based on back trajectories initialized for each IONS O_3 profile, shows differences in transport patterns. Trajectory images are at the IONS website: http://croc.gsfc.nasa.gov/intex/ions.html.

[22] In the following sections, variations in O_3 among the R/V *Ronald H. Brown*, Narragansett and Sable Island are explored further. A summary view is provided with probability distributions of O_3 mixing ratio in the lower, middle and upper troposphere (section 3.1.1). Aspects of UT/LS (upper troposphere–lower stratosphere) O_3 are summarized with tracers and meteorological analyses (section 3.1.2). Ozone budgets are presented in section 3.2. Representative periods during the R/V *Ronald H. Brown* cruise are described with Lagrangian views of O_3 and interpretation of sources based on trajectories, tracers and satellite observations (section 3.3).

3.1.1. Upper, Middle, and Lower Tropospheric Ozone Summary

[23] For convenience, upper (UT), middle (MT) and lower tropospheric (LT) designations refer to 10–15 km, 5–10 km and 0–5 km, respectively. The mean tropopause for the NENA IONS sites is between 12 and 13 km (Table 1), except at Wallops and Sable where it is 14 km. Figure 4 presents probability distribution functions for O_3 in the three layers over the R/V *Ronald H. Brown*, Narragansett and Sable Island. Narragansett (Figure 4a) has the sharpest LT distribution with the peak at > 60 ppbv O_3; this is also where the R/V *Ronald H. Brown* LT O_3 peaks (Figure 4b). The mean mixing ratio at Narragansett near the surface is 10–20 ppbv greater than for the other two sites (Figure 5) and the O_3 mixing ratio peak, just above the BL, signifies accumulation of pollution at that location (Figure 5a). Sable Island LT O_3 is nearly all below 60 ppbv (Figure 4c).

[24] Sable Island is distinct in the middle and upper troposphere as well. The MT O_3 distribution (Figure 4c) peaks approximately 10 ppbv lower than at the other two stations, 70 ppbv instead of 80 ppbv. The high O_3 tail,

Figure 3. July–August 2004 anomaly fields derived from NCEP/NCAR reanalysis data. (a) Tropopause pressure in mbar, (b) 1000–500 mbar thickness in m, and (c) surface temperature in K (shaded). West to east, the stars are locations of Beltsville, Wallops, Narragansett, the R/V *Ronald H. Brown*, Yarmouth, and Sable Island.

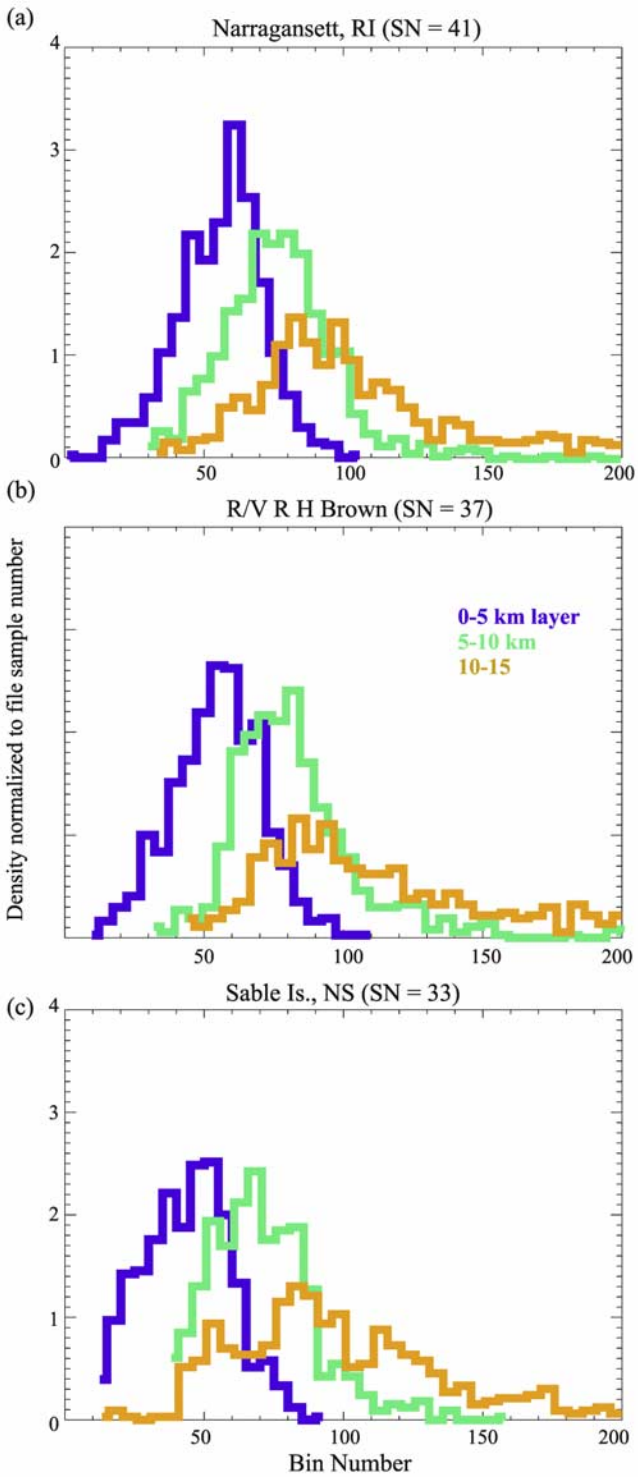

Figure 4. Density distribution of O_3 mixing ratios in 0–5 km, 5–10 km, and 10–15 km layers for IONS soundings over (a) Narragansett, Rhode Island; (b) R/V *Ronald H. Brown*; and (c) Sable Island. These are based on 25-m averages; sample number is at top of panel.

>100 ppbv, is less significant at Sable than at the R/V *Ronald H. Brown* and Narragansett. The UT distribution at Sable (Figure 4c) has three peaks, centered at 55 ppbv, 75 ppbv and 115 ppbv, instead of a single broad distribution with a maximum centered at 90 ppbv as for the R/V *Ronald H. Brown* and Narragansett (Figures 4a and 4b). This is reflected in the shapes and deviation of the mean profiles for the three sites (Figure 5). The lower 1-sigma limit of UT O_3 mixing ratio over Sable is ~20 ppbv, half the value for Narragansett and the R/V *Ronald H. Brown*. The greater frequency of UT low-O_3 values for Sable (Figure 4c) suggests convective transport of unpolluted BL O_3 above 10 km (compare INTEX-A aircraft observations from which *Bertram et al.* [2007] infer ages of convectively influenced air).

3.1.2. Upper Troposphere/Lower Stratosphere (UT/LS)

[25] In Figure 2, the brown color signifies stratospheric O_3. Incursions of stratospheric O_3 below the mean tropopause, ~13 km, are prominent over Narragansett and the R/V *Ronald H. Brown* during the first three weeks of July (Figures 2a and 2b). Similar incursions, with 200 ppbv O_3 below 10 km, resumed after 5 August (section 3.3.3). That these episodes correspond to stratospheric influence is indicated by comparing mean O_3 in the UT with pv at 150 hPa, the approximate midpoint in the 10–15 km layer (Figure 6). In general, the highest O_3 mixing ratios occurred when pv was greater than 3 pvu.

[26] Another indicator of stratospheric influence is dry air in the UT. When the O_3 profiles for 10–15 km are separated by a criterion of mean UT O_3 mixing ratio being greater than or less than 250 ppbv (Figure 7), those with the higher mixing ratio are significantly drier in the UT. In addition, in the LT and MT, the shapes and range of RH (shown shaded, to ±1-sigma, in Figure 7) of the two O_3 categories are distinct. In the 5–12 km range for all three sites, the profiles with the higher UT O_3 mixing ratios display RH roughly half as high (20–25%) as RH from the soundings with higher O_3 mixing ratio (compare Figures 7c to 7d and Figures 7g to 7h). These profiles point to stratospheric influence in the MT as well as in the UT, even though the mean O_3 profile between 5 km and 12 km is nearly the same for both classes. The shapes of the mean O_3 profile in the UT (10–15 km), for the O_3 > 250 ppbv category, differ among sites with Sable showing a double tropopause, it appears (compare Figure 7j with Figures 7b and 7f).

3.2. Ozone Budgets in NENA Profiles

[27] In Figure 8 the column tropospheric O_3 totals for the R/V *Ronald H. Brown*, Narragansett and Sable Island are illustrated with individual O_3 budget terms. Averages of the individual terms over the IONS period appear in Table 1, with summaries for Yarmouth, Wallops Island and Beltsville.

3.2.1. BL Ozone

[28] The average BL O_3 amount for Narragansett is greater, 4.4 DU, than at the R/V *Ronald H. Brown* and Sable Island, 3.6 DU and 2.6 DU, respectively (Table 1). At Sable, where the sondes are launched midevening, the O_3 may be near the daily maximum, whereas the pollution impact implicit in BL O_3 at Narragansett is a midday snapshot, probably 5–6 hours prior to the daily surface O_3 maximum. Figure 9, based on hourly averages of continuous O_3 data from the R/V *Ronald H. Brown*, shows maximum O_3 shortly before midnight UTC, 2000 hours Local for most of the cruise. The ship traveled throughout the Gulf of Maine (see track in the work by *Fehsenfeld et al.*

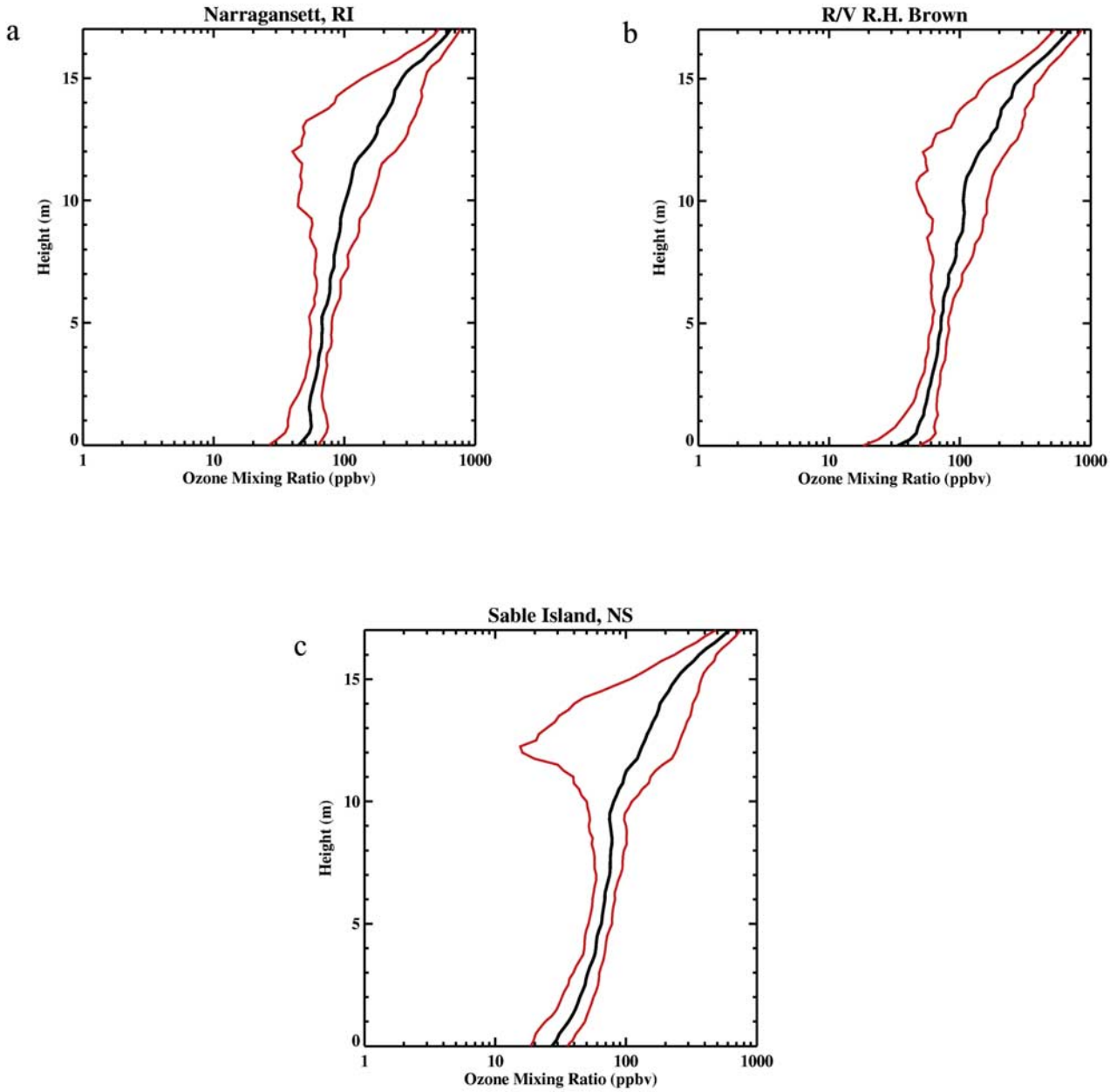

Figure 5. Mean vertical O_3, from surface to 17 km; mixing ratio with 1-sigma standard deviation, in red, for IONS soundings over (a) Narragansett, Rhode Island; (b) R/V *Ronald H. Brown*; and (c) Sable Island.

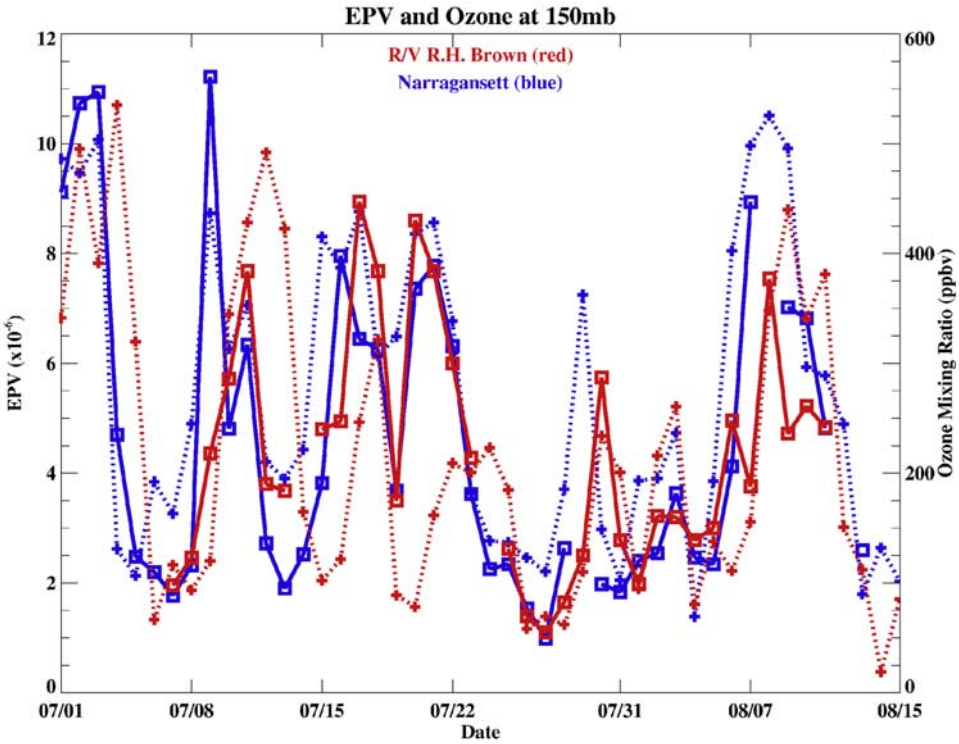

Figure 6. Daily pv (interpolated at 150 hPa, ~13 km, from GEOS-4 analyses) from 1 July to 15 August 2004 (pluses) with O_3 mixing ratio averaged between 10 and 15 km (squares) at Narragansett, Sable Island, and the R/V *Ronald H. Brown*.

[2006]). For comparison to the R/V *Ronald H. Brown* observations, Figure 9 displays hourly averages of O_3 measured on the buoy moored 35 km southeast of Portsmouth, NH. The buoy, closer to land on average than the ship, cycles between onshore flow and offshore flow, the latter with more pollution. *Hintsa et al.* [2005] noted that for offshore flow, the daily O_3 cycle at the buoy peaks sharply at 64 ± 4 ppbv O_3 at 2100 UTC. For onshore conditions there is a less distinct pattern, with a 53 ± 2 ppbv O_3 peak at 1900 UTC.

3.2.2. Free Tropospheric Ozone: ST, RCL, and AD

[29] *Thompson et al.* [2007a] describe stratospheric influence on the tropospheric O_3 budget for 21 July 2004 in detail, partly to examine ambiguities in ST O_3 amounts arising from different designations of the tropopause or UT/LS region. By any criterion, the ST O_3 fraction over the NENA sites is prominent throughout IONS, ~25% on average, on the basis of the amounts in Table 1. Others have noted stratospheric influences in northern midlatitude summer [*Moody et al.*, 1995; *Cooper et al.*, 2005]. In the work by *Thompson et al.* [2007a], evaluation of the Wallops ozonesonde time series shows that a 25% value for ST O_3 for NENA may be typical. This figure also agrees with the ST fraction of tropospheric O_3 deduced from tracer and trajectory calculations for more than 25000 profiles taken over Europe from 1970 through 2003 [*Collette and Ancellet*, 2005]. Examples of ST O_3 variability in IONS are given in section 3.3.

[30] In Figure 10a the 25 July 2004 Narragansett O_3 profile is shown. On this day more than 15% of the tropospheric column is assigned to RCL (Figure 8a). Up to 500 hPa, the O_3 mixing ratio and RH are both high (Figure 10a); the latter denotes convective mixing to this level. From 500 hPa to 300 hPa O_3 increases, RH falls to ~50%, also consistent with convective redistribution of O_3 or its precursors. Convection and lightning are both plausible. Figure 10b shows evidence for lightning in the trajectory-mapped lightning exposure product at 300 hPa. The GEOS-4 computed cloud mass flux and cloud-detrainment mass flux (not shown) are appreciable at 300 and 500 hPa in the New England coastal region. Convective influence persisted to 26 July at Narragansett and was also detected in the R/V *Ronald H. Brown* sounding on that day.

[31] The residual (AD) category is typically 50% or more of the total column for the tropospheric O_3 budgets (Figure 8). Noting that aged O_3 may be 10–15 DU, or ~1/2 to 2/3 of the AD O_3 column (on average 22 DU, Table 1), we can infer that freshly advected O_3 is approximately 10 DU for the NENA sites.

3.3. Representative Episodes During IONS

[32] Key aspects of tropospheric O_3 measured over the R/V *Ronald H. Brown* during IONS were (1) relatively few pollution episodes at the surface, consistent with surface wind and temperature anomalies, and (2) complex free tropospheric O_3 structure, with interaction of ST, RCL O_3 and advection impacts varying almost daily. The remainder of this section examines several periods with reference to sondes, surface O_3 and tracers, back trajectories and meteorological conditions: 7–12 July, mixed stratospheric-

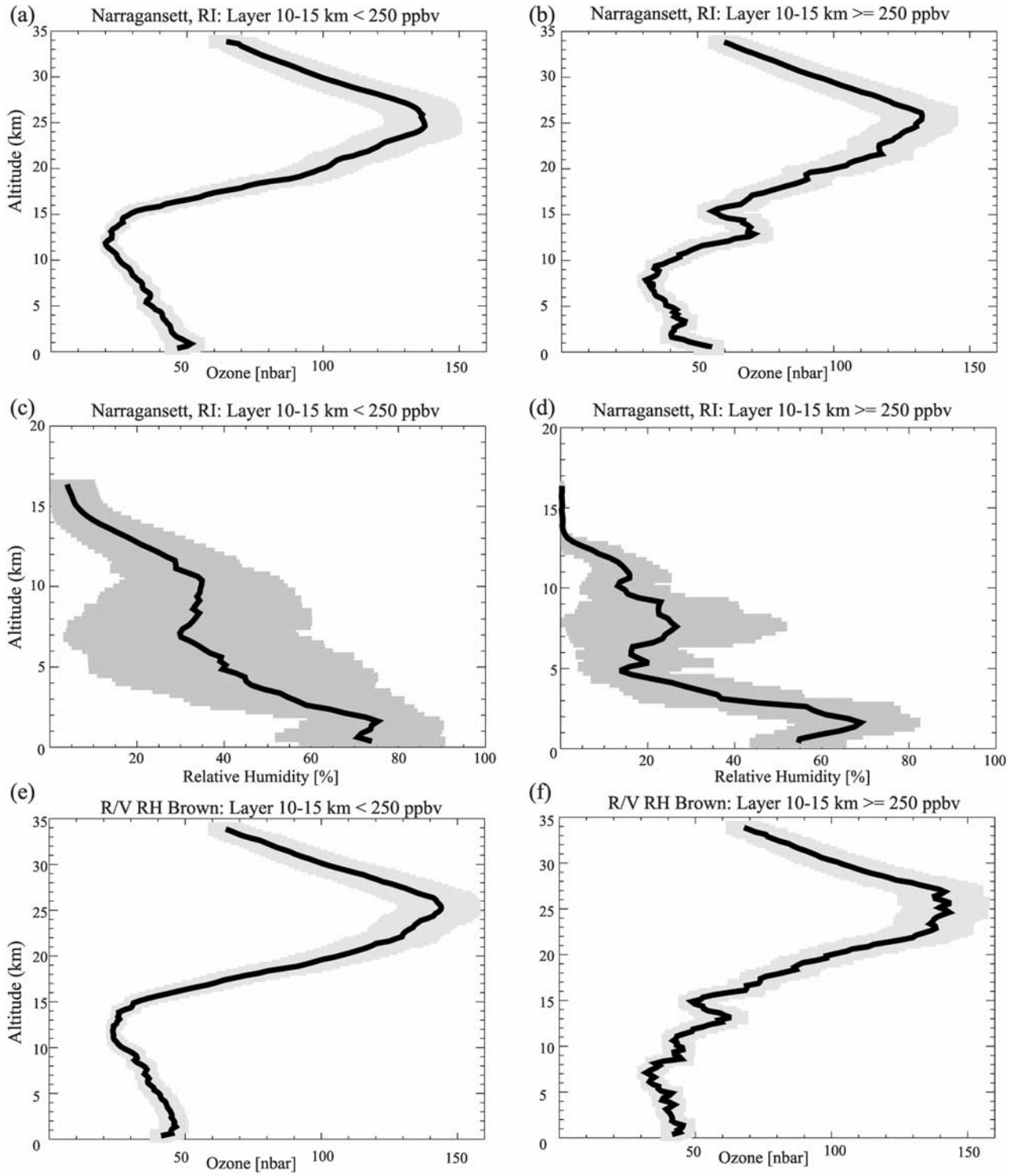

Figure 7. (a–f) Mean O_3 mixing ratio, with shaded range to ±1-sigma, apportioned by mean mixing ratio in the 10–15 km segment, as given in top legend. Profiles, based on ozone- and radio-soundings, are for Narragansett and the R/V *Ronald H. Brown*, with O_3 mean <250 ppbv (Figures 7a and 7e) and with O_3 mean mixing ratio >250 ppbv (Figure 7b and 7f). Also shown are corresponding figures showing relative humidity for Narragansett (Figures 7c and 7d). (g–l) Mean O_3 mixing ratio and corresponding relative humidity, with shaded range to ±1-sigma, apportioned by mean O_3 mixing ratio in the 10–15 km segment. Profiles are taken from ozone- and radio-soundings, for the R/V *Ronald H. Brown* (Figures 7g and 7h) and Sable Island (Figures 7i–7l).

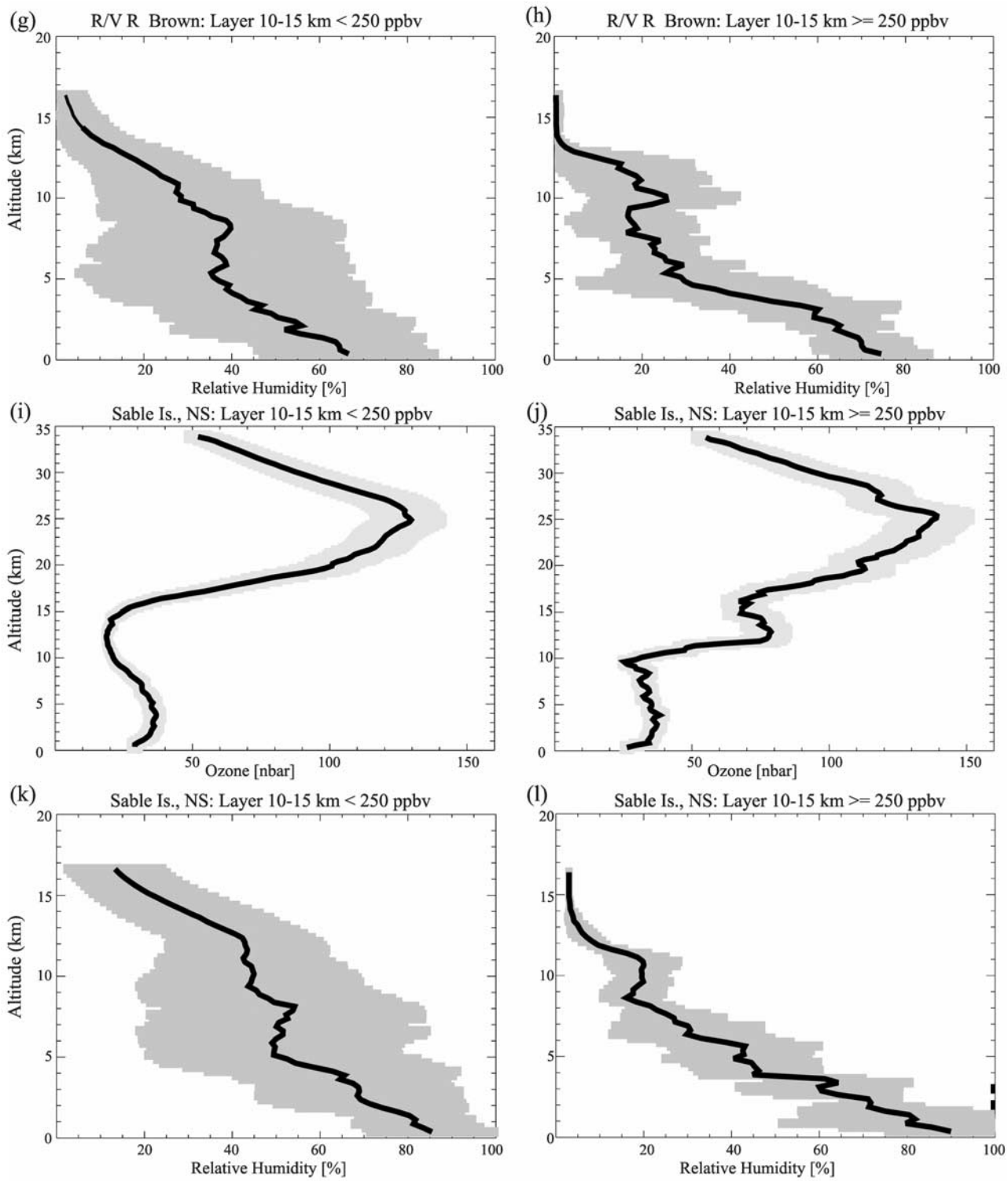

Figure 7. (continued)

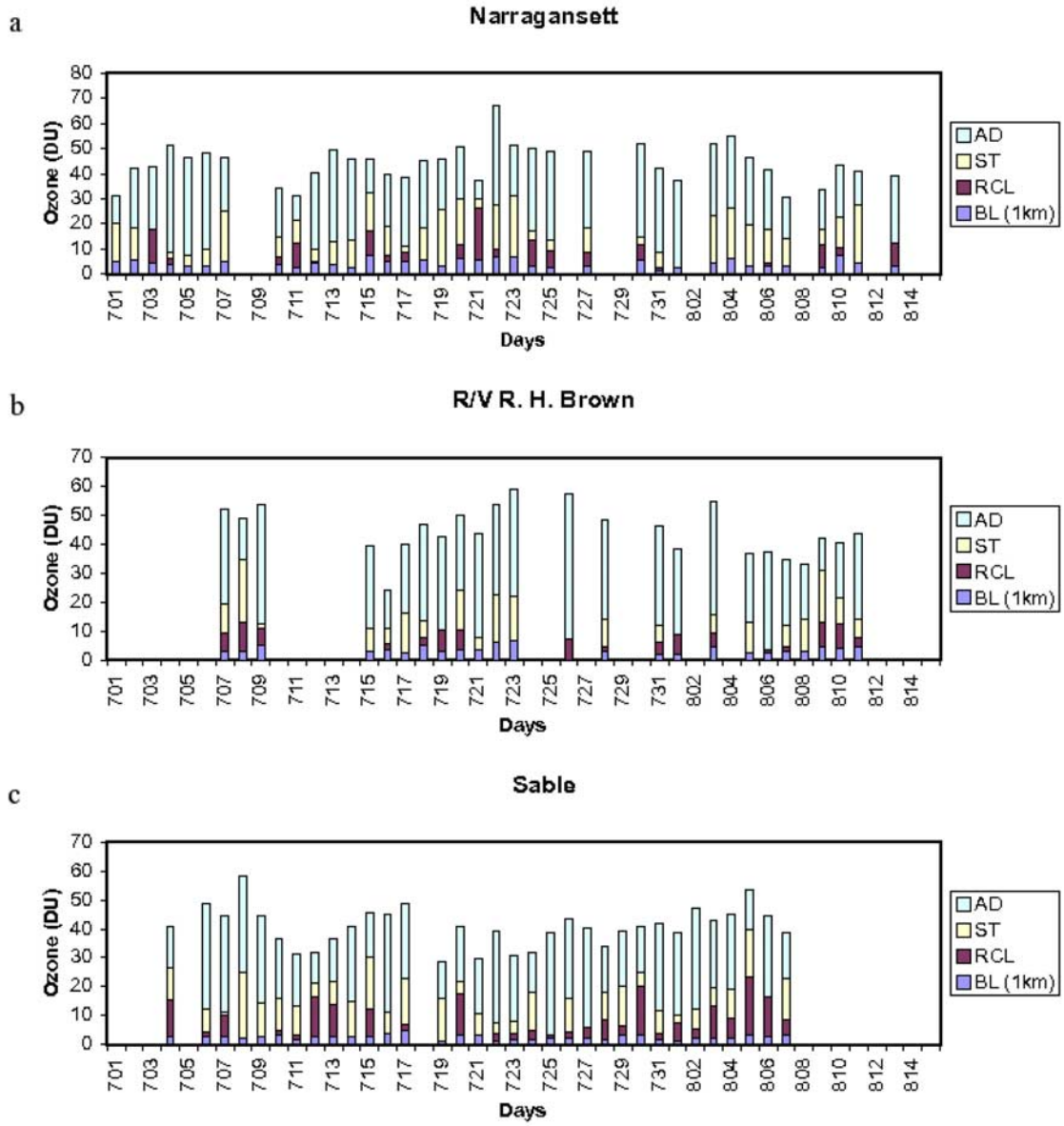

Figure 8. Ozone budgets in DU for (a) Narragansett, (b) R/V *Ronald H. Brown*, and (c) Sable Island. BL ozone is based on integrating from the surface to 1 km. ST and RCL O_3 are determined from lamina-labeling [after *Pierce and Grant*, 1998; *Teitelbaum et al.*, 1996] as modified by *Thompson et al.* [2007a]. Individual laminae are added to give the composite ST and RCL O_3 terms. After summing BL, ST, and RCL O_3, the residual tropospheric O_3 is assigned to advection (AD), a term that includes freshly transported and aged air of indeterminate origins.

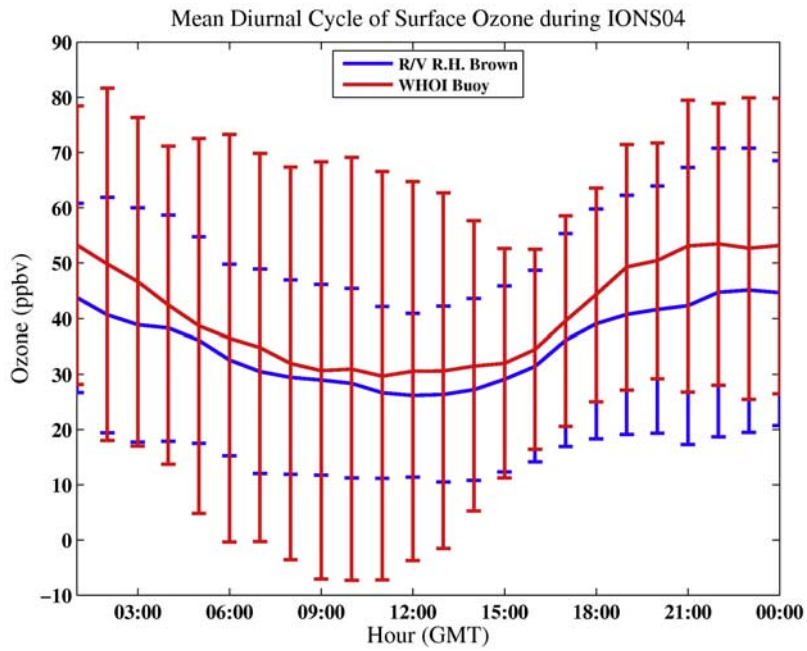

Figure 9. Diurnally averaged O_3 mixing ratio measured over the Gulf of Maine based on continuous sampling on the R/V *Ronald H. Brown* and an O_3 sensor operated on a buoy at 42.8°N, 70.3°W from 27 June to 6 September 2004, 35 km southeast of Portsmouth, New Hampshire. Hourly averages, with 1-sigma standard deviations, are illustrated. Higher values and greater variability from the buoy instrument occur because it is closer to coastal pollution than the ship and is more influenced by a diurnal onshore-offshore wind cycle.

pollution effects, followed by convection; 15–23 July, stratospheric influence with RCL and advected pollution, the latter most pronounced on 22 July (see E. V. Browell et al., Large-scale air mass characteristics observed over North America and western Atlantic Ocean, submitted to *Journal of Geophysical Research,* 2006, hereinafter referred to as Browell et al., submitted manuscript, 2006); and 31 July to 2 August and 5–9 August, mixed sources, followed by stronger stratospheric influence in low-pressure systems.

3.3.1. Measurements on 7–12 July 2004: Mixed Stratospheric-Pollution Effects

[33] Figure 11a displays the cruise track and surface O_3 measured on the R/V *Ronald H. Brown* during this period. Few episodes of O_3 mixing ratio exceed 50 ppbv. On 8 July 2004 the O_3 profile from the shipboard launch shows relatively high stratospheric influence (Figure 12a) above 200 hPa where the RH profile is very dry. Stratospheric influence may also be present in some of the drier layers below 400 hPa. The ST O_3 fraction of tropospheric O_3 on 8 July is 45% (Figure 8b). Similar appearance and fractions hold for Narragansett on 7 July 2004 (Figure 12b). By 9 July 2004, the O_3 soundings from the ship, Narragansett and Yarmouth all display a low tropopause (Figure 13a) due to a filament of stratospheric air. The latter is prominent in the meteorological analyses, e.g., in the RD mapped pv (340 K) at http://croc.gsfc.nasa.gov/intex for 9 July 2004 (Figure 13b). Back trajectories initialized from the R/V *Ronald H. Brown* sounding O_3 peaks at upper levels (200, 300 hPa) have origins over south central Canada (Figure 13b) where pv at 340 K was 2–5 pvu 1–2 days earlier. In contrast, the 200 hPa back trajectories over Sable Island originate over the central United States (Figure 13b). The starting pressure levels for the trajectories initialized above Narragansett and the R/V *Ronald H. Brown* are above the tropopause. Near Sable Island the pv map for 9 July shows residual stratospheric influence, consistent with the budget in Figure 8c.

[34] A consequence of the meteorological transition from 7 to 9 July 2004 is that the Narragansett soundings are not sufficiently stable for laminar analysis [*Teitelbaum et al.*, 1994; *Pierce and Grant*, 1998; *Thompson et al.*, 2007a]. A similar pattern holds for the R/V *Ronald H. Brown* soundings from 10 to 12 July. Convective influence (RCL = 33% for 11 July) is prominent in the Narragansett soundings on 11 and 12 July 2004 (see Figure 2a). Water vapor (not shown) increases from 6 km to the tropopause, signifying detrainment in the UT. The lightning exposure maps for 11 and 12 July 2004 are similar to those for 25 July (Figure 10b). Ozone profiles during 11–12 July also displayed influences of Alaskan and Canadian fires [*Pfister et al.*, 2005, 2007; McMillan et al., unpublished manuscript, 2007].

3.3.2. Measurements on 15–23 July 2004: Mixed Stratospheric and Pollution Influences

[35] The R/V *Ronald H. Brown* cruise track over this period is depicted in Figure 11b. The earlier and final segments were near the New England coastline; 18–20 July sampling was in the eastern part of the Gulf of Maine. Surface O_3 readings > 70 ppbv occurred on 22 and 23 July 2004 (compare Figure 2b). Throughout the period 15–18 July 2004, there was considerable fine structure in O_3

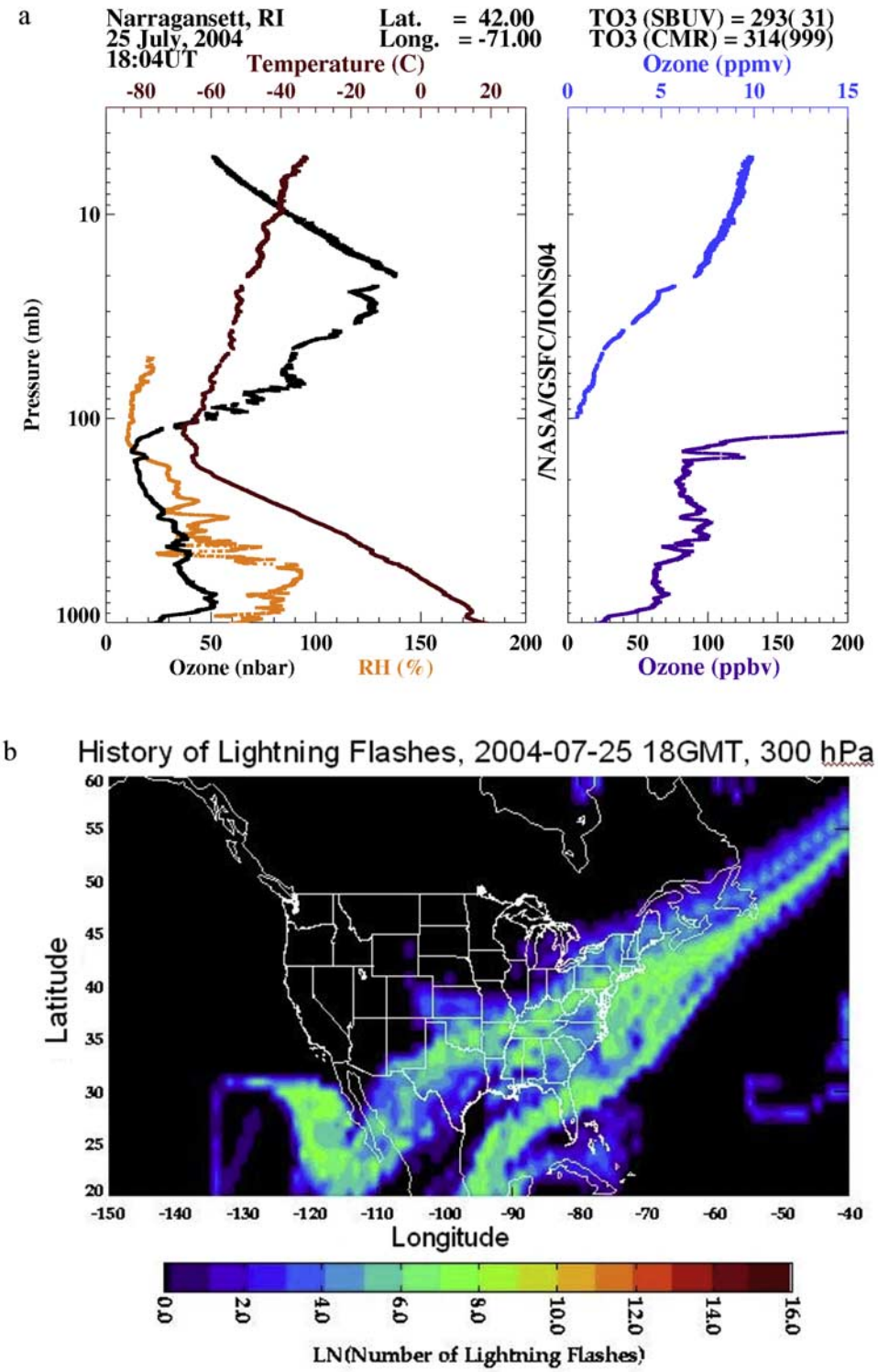

Figure 10. (a) Narragansett 25 July 2004 profiles, showing evidence for convective and lightning influence in (b) EL product from http://croc.gsfc.nasa.gov/intex determined from coincidence between the previous 5 days' National Lightning Detection Network flashes and trajectories initialized at 300 hPa on 25 July 2004.

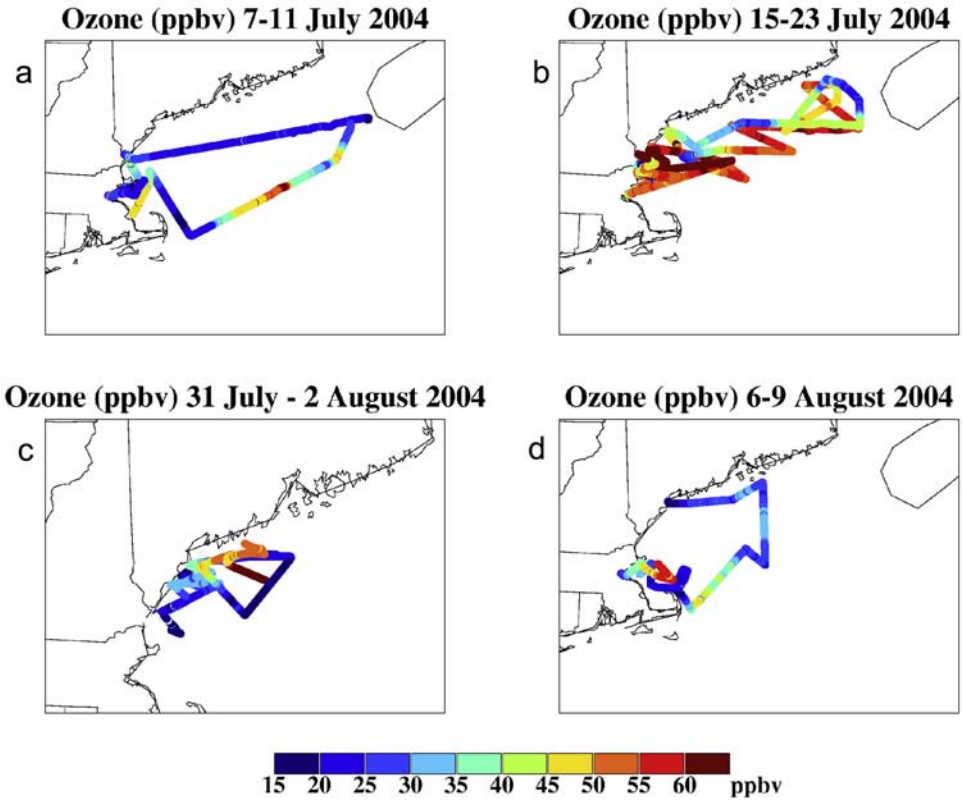

Figure 11. Segments of R/V *Ronald H. Brown* cruise with O_3 mixing ratio. Three O_3 instruments on the ship operated continuously and recorded values within 2% of one another. Data shown are the average from two PMEL instruments, a Dasibi, and a TEI Model 49. See http://saga.pmel.noaa.gov/Field/NEAQS-ITCT and *Fehsenfeld et al.* [2006] for complete list of cruise measurements. (a) 7–11 July 2004, (b) 15–23 July 2004, (c) 31 July to 2 August 2004, and (d) 6–9 August 2004.

from the Narragansett and R/V *Ronald H. Brown* O_3 profiles (Figures 2a and 2b). The penetration of orange-red colors below the tropopause, the red-brown transition point, suggests stratospheric influence, but AD and RCL also play a role, as the budgets in Figure 8b indicate. Mixed ST, RCL and advected pollution influences contribute to the layering, with the fractional amounts varying day by day (Figure 8b). On 15 July, the tropopause was relatively low (Figures 2a and 2b). On 16 and 17 July, the tropopause retreated to a higher level but residual high, dry layers remained. The fine structure in O_3 and RH profiles over the R/V *Ronald H. Brown* appears in Figure 14a. The layer from 5 km to the tropopause on 17 July is identified as ST O_3 over the R/V *Ronald H. Brown*: 35% of tropospheric O_3 that day. A relatively high ST O_3 is also determined from the 17 and 18 July Narragansett profiles. On 18 July 2004, a different fine structure is observed over the R/V *Ronald H. Brown* (Figure 14b). Ozone peaks are present as on 17 July 2004, but they are lower in concentration, ~100 ppbv on 18 July compared to 150 ppbv for 17 July. There is more moisture between 8 and 10 km on 18 July. The ST O_3 fraction falls and the RCL amount increases. T. D. Fairlie et al. (Impact of multiscale dynamical processes and mixing on the chemical composition of the upper troposphere and lower stratosphere during INTEX-A, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Fairlie et al., submitted manuscript, 2006) use a model and INTEX-A DC-8 data to describe a mixture of influences in the NENA region on 18 July 2004.

[36] Ozone over the R/V *Ronald H. Brown* on 19 July 2004 is very different from the prior 2 days. Figure 14c shows a well-mixed layer of O_3, averaging 65 ppbv above 4 km, with RH >80% up to 6 km. The fraction of RCL O_3 increases. From 17 and 18 July, where the 4-day origins are from the northern United States and southern Canada, back trajectories origins for the ship soundings (not shown; see http://croc.gsfc.nasa.gov/intex/ions.html) shifted from west and NW to southwesterly at midtropospheric level and from the Atlantic at 25°N for surface and near-surface O_3. This changes again on 20 July, as Figures 2b and 14d show. The tropopause descends from 13 km to 10 km. Elevated O_3 with dry layers persist to 5 km (Figure 14d). Stratospheric influence is high for 20 July over the R/V *Ronald H. Brown*; RCL O_3 is also relatively high for that day. The origins of the air parcels shift back to higher latitudes.

[37] There is further transition on 21–23 July 2004. On 21 July 2004, for example, the tropopause is as low as 7 km over NENA sites, except at Sable Island. These profiles, where there is also considerable ST O_3 over Narragansett and the R/V *Ronald H. Brown* are analyzed in detail by *Thompson et al.* [2007a, see Table 2]. Two factors contribute: (1) a filament of elevated pv over the Atlantic coast and

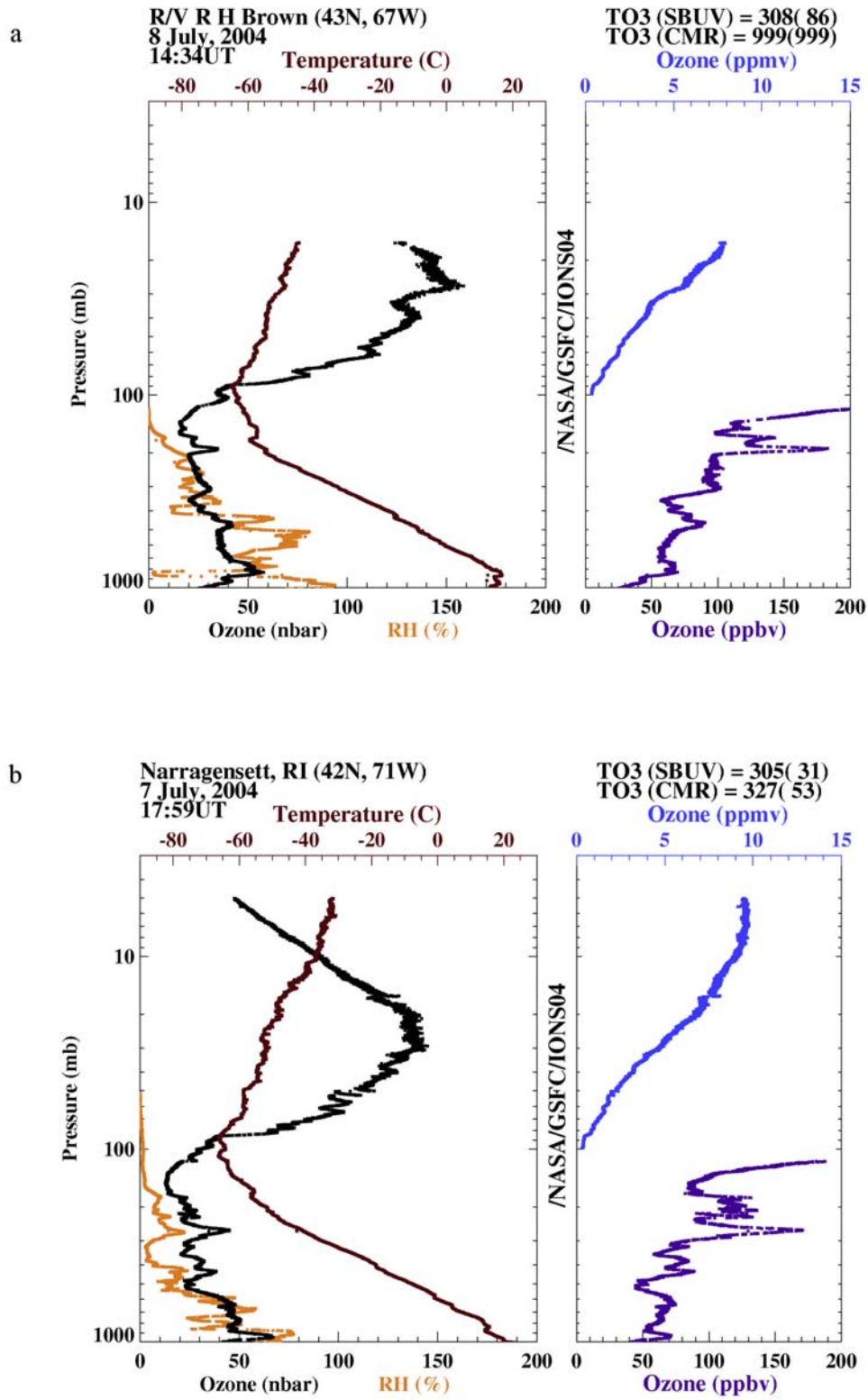

Figure 12. Profiles from (a) the R/V *Ronald H. Brown* on 8 July 2004 and (b) Narragansett on 7 July 2004.

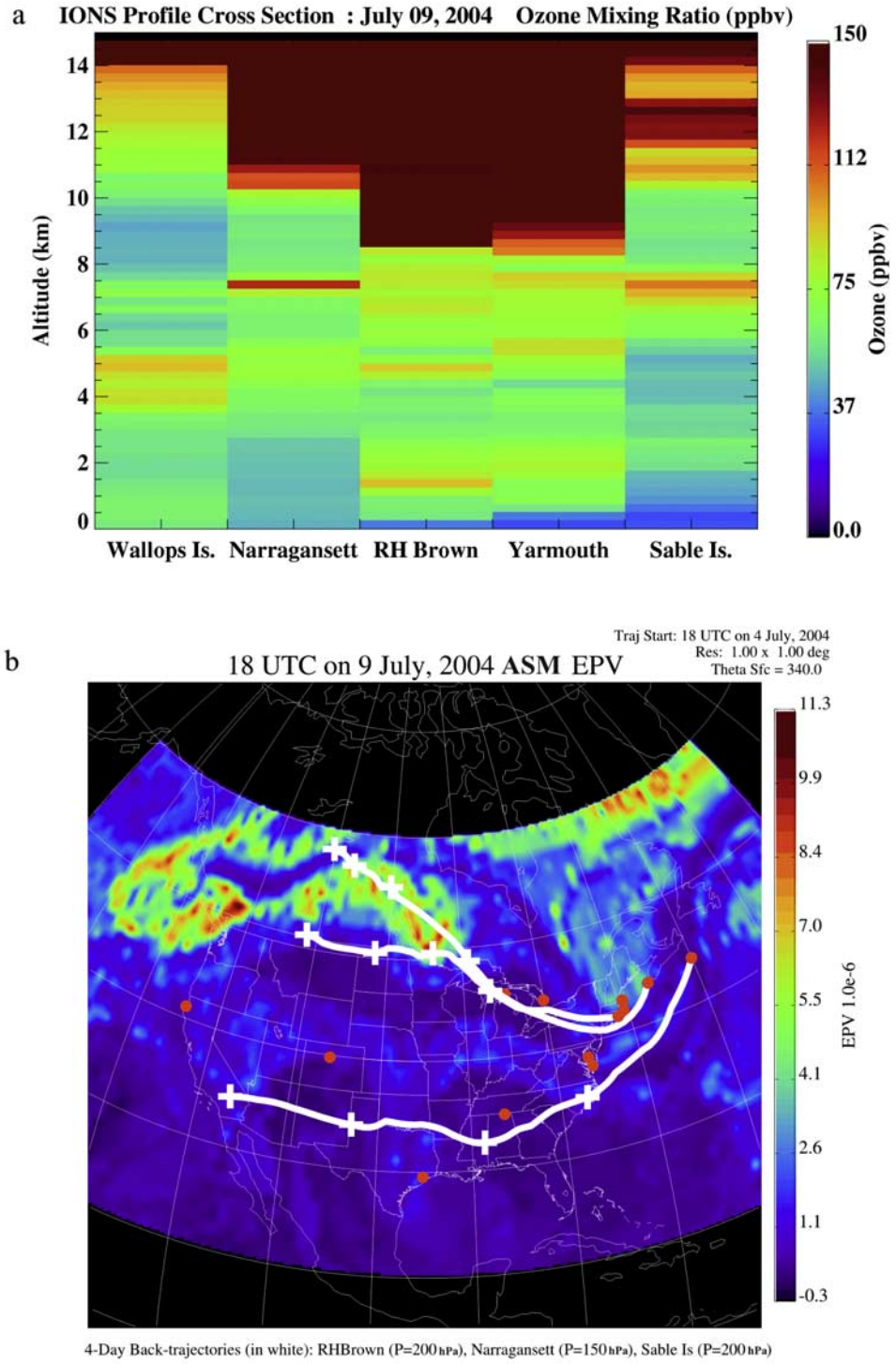

Figure 13. (a) Eulerian view of O_3 mixing ratio on 9 July 2004, showing the low tropopause and layers of mixed ST O_3 with RCL (Figure 8b for R/V *Ronald H. Brown*). (b) Four-day air parcel history initialized 4 July from Narragansett, R/V *Ronald H. Brown*, and Sable Island at tropopause heights 200 hPa, 150 hPa, and 200 hPa, respectively, plotted over trajectory-mapped epv. Crosses mark 24 hour periods. Starting pressure levels for trajectories initialized over Narragansett and the R/V *Ronald H. Brown* are above tropopause. For Sable Island the pv map shows some stratospheric influence in the region; compare Figure 8c budget.

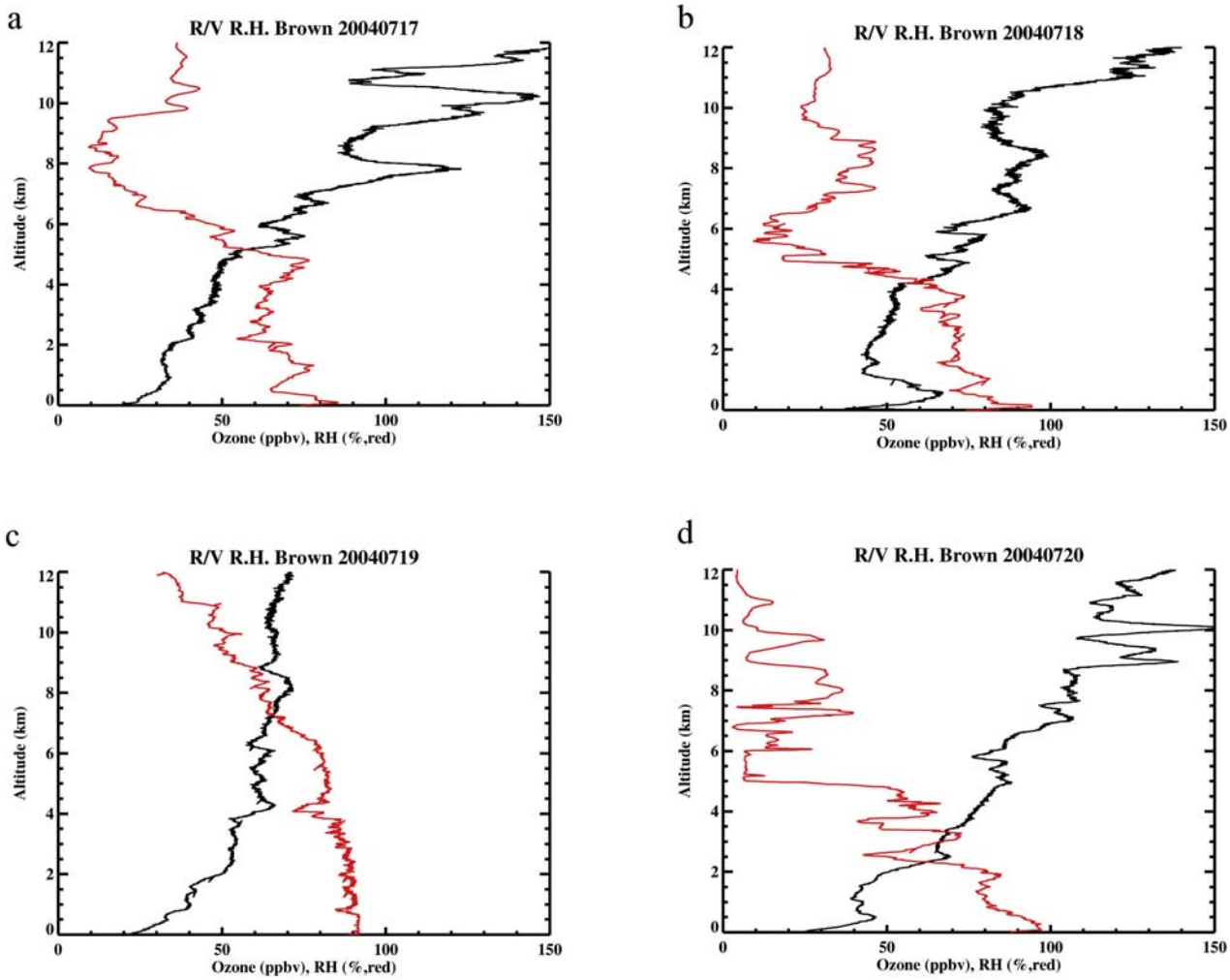

Figure 14. O_3 mixing ratio and RH in R/V *Ronald H. Brown* soundings for (a) 17 July 2004, (b) 18 July 2004, (c) 19 July 2004, and (d) 20 July 2004.

(2) origins of midtropospheric air parcels arriving at those stations are over southern Canada, where pv is also elevated. Trajectories and the local pv environment over Sable suggest subtropical Atlantic air [*Thompson et al.*, 2007a, Figure 4].

[38] On 22 July 2004, the tropospheric O_3 column over all three stations, the R/V *Ronald H. Brown,* Narragansett and Sable Island, increased 10 DU from the previous day (Figure 8). An Eulerian cross section of O_3 appears in Figure 15a. The fractions of AD O_3 increase from 21 to 22 July, although ST is partially responsible for the R/V *Ronald H. Brown* increase (Figure 8). Advected O_3 and tracers of biomass fires were detected in DC-8 sampling over New England (Browell et al., submitted manuscript, 2006) and at the Azores PICO site [*Val Martín et al.*, 2006]. Air parcels arriving at Narragansett and the R/V *Ronald H. Brown* from the midwestern United States and southern Canada were probably exposed to Canadian and Alaskan fires (Figure 15b). A major outbreak occurred from 11 to 14 July 2004 [*Pfister et al.*, 2005, 2007] with the ensuing CO plume reaching the Gulf of Mexico by 19 July [*Morris et al.*, 2006] and Europe by 23 July 2004 (McMillan et al., unpublished manuscript, 2007).

3.3.3. Measurements on 31 July to 2 August and 5–9 August 2004: Mixed Ozone Sources

[39] Another pollution episode occurred in the beginning of August. Figure 11c shows that July ended with low surface O_3, except for one segment in the second half of 31 July, where O_3 was > 60 ppbv and CO (not shown) was > 150 ppbv, instead of a typical cruise value of 100–120 ppbv. Surface O_3 over the R/V *Ronald H. Brown* at launch on 31 July and 1 August was ~25 ppbv but mixing ratios of 80 ppbv aloft extended to a higher-than-average tropopause (Figure 2b). A cold front passed through the Gulf of Maine on 1 August 2004. By 2 August 2004, when there were two launches from the ship, the free troposphere showed more evidence of high-O_3 layers, as it did over all the NENA sites except Wallops (Figure 16a). The sounding pictured in Figure 16a is the earlier one for 2 August, timed with the other profiles shown except Sable.

[40] The total tropospheric O_3 column for Yarmouth, the R/V *Ronald H. Brown*, Narragansett and Sable Island on

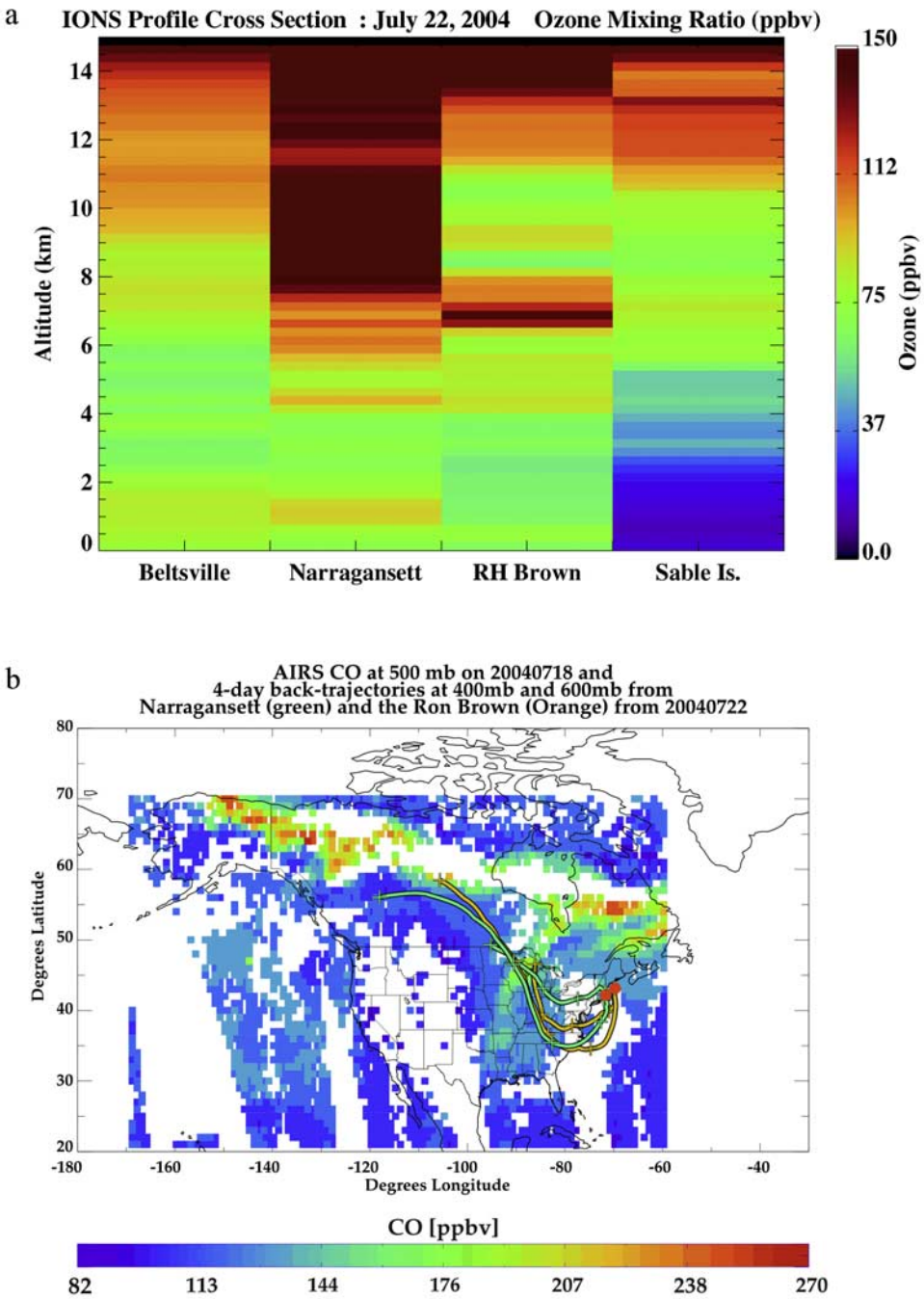

Figure 15. (a) Eulerian curtain of O_3 mixing ratio on 22 July 2004, with ST, RCL, and relatively fresh pollution (see DC-8 O_3 (Browell et al., submitted manuscript, 2006)). (b) Pollution origins indicated by 4-day back trajectories over AIRS CO map of 18 July 2004. Trajectories are initialized from elevated O_3 layers over Narragansett and the R/V *Ronald H. Brown*. The CO is at 500 hPa, near the peak of AIRS vertical sensitivity; the CO values are accurate to within 15% [*McMillan et al.*, 2005, also unpublished manuscript, 2007].

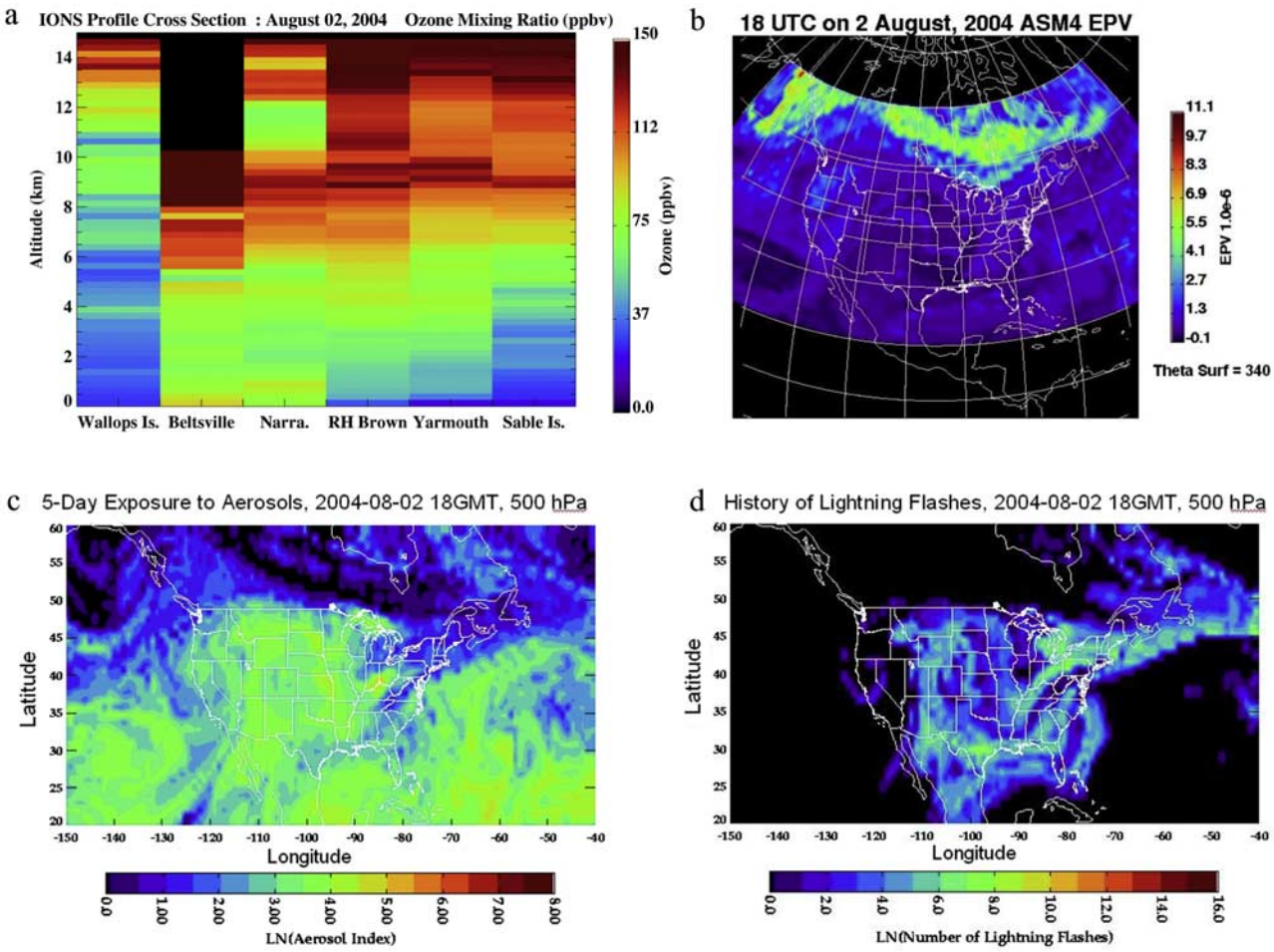

Figure 16. (a) Eulerian O_3 curtain for 2 August 2004. Influences from (b) stratosphere in pv, 340 K surface; (c) pollution in trajectory-mapped aerosol; and (d) lightning.

2 August 2004 is 7–12 DU above their respective IONS average (Table 1). Back trajectories at midtropospheric levels from all these stations show continental origins on 1 and 2 August, over the east coast in the former case, shifting over midcontinent on 2 August, where aerosol pollution is significant (Figure 16c). The frontal passage meant that only two O_3 budgets could be evaluated on 2 August. At Sable Island most of the additional O_3 is attributed to fresh advection, but ST and RCL O_3 account for 20% of the tropospheric O_3 total. Similar fractions hold for the R/V *Ronald H. Brown* on 3 August 2004 (Figure 2b). This is consistent with the mixture of sources suggested by the analyses in Figures 16b–16d and by tracers from the INTEX DC-8 flight on 2 August (Browell et al., submitted manuscript, 2006; Fairlie et al., submitted manuscript, 2006). There is evidence for stratospheric influence in the pv analysis (Figure 16b), pollution (using absorbing aerosols as a tracer, Figure 16c) and lightning and convection along the coast (Figure 16d). The 2 August budget for Yarmouth (not illustrated) assigns all the additional O_3 to ST.

[41] After 2 August 2004, low pressure associated with Hurricane Alex and systems afterward affected the NENA region (Synoptic Summary at http://croc.gsfc.nasa.gov/intex/ions.html). Surface O_3 measured on the R/V *Ronald H. Brown* over the Gulf of Maine (Figure 11d) was low as was O_3 over most of the NENA sites (6 August 2004, Figure 17a). On 6 August 2004, the total O_3 column amounts for Narragansett, 42 DU, and the R/V *Ronald H. Brown*, 37 DU, were 10 DU and 18 DU lower, respectively, than O_3 on 3 August (Figures 2a and 2b; 3 August structure resembles Figure 16a). NENA shows elevated pv (through the RD mapping in Figure 17b) and low pollution and convective-lightning influences (Figures 17c and 17d). This implies that reduced O_3 column on 6 August is a consequence of fewer photochemical sources and a lower-than-average tropopause.

4. Summary

[42] The variability of tropospheric O_3 over New England and the North Atlantic during the ICARTT/INTEX-NA experiments in July-August 2004 has been characterized with soundings launched as part of IONS. Comparisons among soundings from the R/V *Ronald H. Brown* in the Gulf of Maine and IONS profiles over Narragansett, Rhode Island, and Sable Island illustrate the complexity of O_3 vertical structure and temporal variability over a 1000 km wide region in the transition between summertime midlatitude and high-latitude air. Regional features are consistent

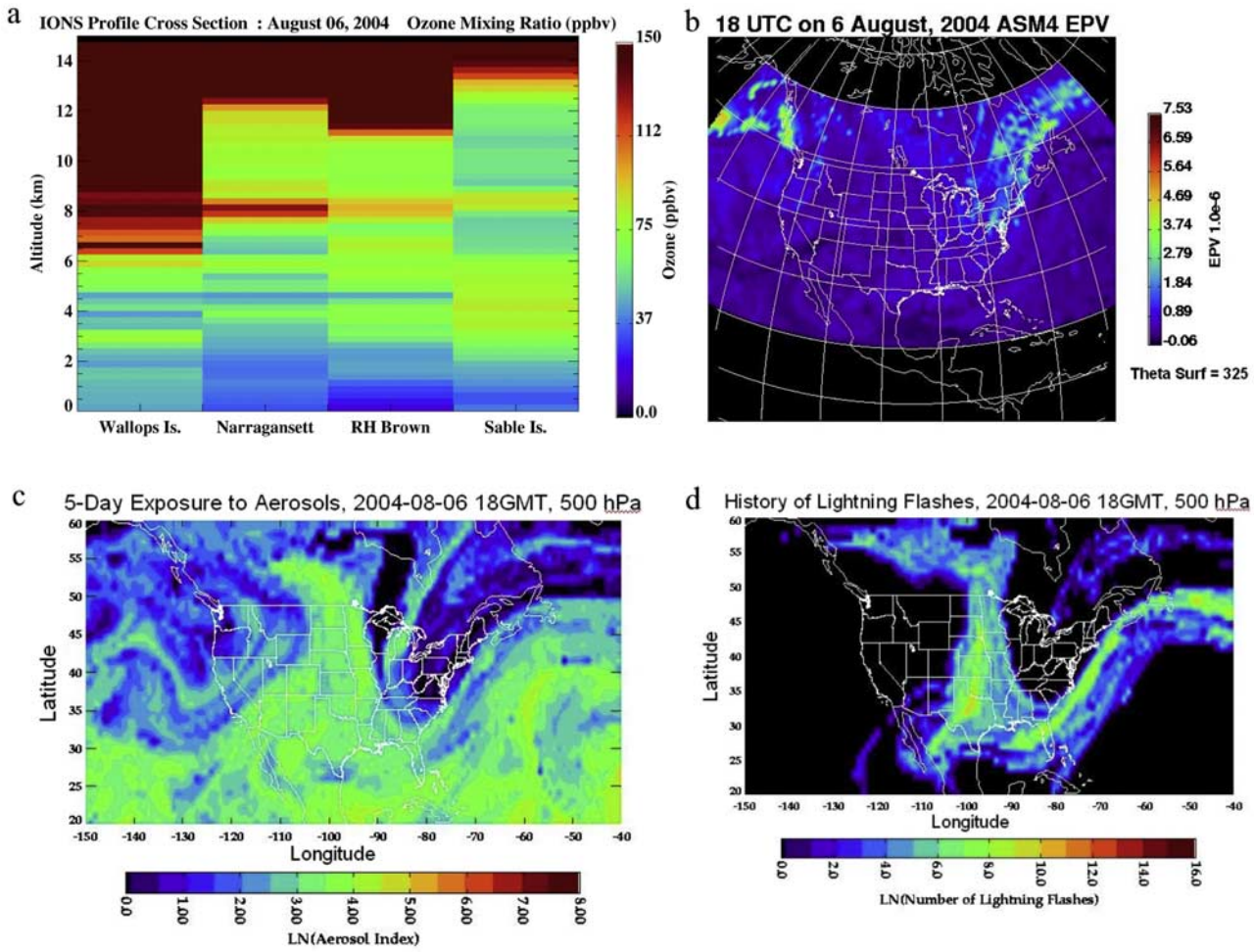

Figure 17. (a) Eulerian O_3 curtain for 6 August 2004. Influences from (b) stratospheric in pv, (c) pollution in trajectory-mapped aerosol, and (d) lightning.

with recurrent low-pressure systems and associated anomalies in tropopause height, temperature and surface winds in July-August 2004.

[43] Representative episodes of pollution, convective and stratospheric O_3 influence, as observed in ozone profiles, are interpreted in terms of meteorological variability and tracers for pollution sources. These analyses are consistent with tropospheric O_3 budgets for individual soundings determined from lamina identification [*Thompson et al.*, 2007a]. A composite tropospheric O_3 budget from northeastern North American IONS sampling is 10–15% each BL O_3 and O_3 from regional pollution and lightning, with 20–25% from the stratosphere. The remaining ~50% of tropospheric ozone is a mixture of freshly advected pollution and aged background air of indeterminate origins.

[44] **Acknowledgments.** We are grateful to chief scientist T. S. Bates, D. Hamilton (NOAA/PMEL), and the crew of the R/V *Ronald H. Brown* for superb support during the NEAQS-2004 cruise. Thanks to J. F. Liesch (University of Maryland) who made ozone soundings on the second leg. The lightning flash data were provided by Vaisala (National Lightning Detection Network) via NASA Marshall Space Flight Center (H. Christian). NASA's Tropospheric Chemistry Program, NOAA's ESRL Global Monitoring Division and Chemical Sciences Division, and Environment Canada were the primary IONS sponsors. Analysis by A.M.T. and J.B.S., whose M.S. Thesis is based on this work, was supported by NASA's Tropospheric Chemistry and Aura Validation Programs and by Penn State's Meteorology Department.

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G. Forbes, Environment Canada Sable Island, 45 Alderney Drive, Dartmouth, NS, Canada B2Y 2N6. (gerry.forbes@ec.gc.ca)

E. J. Hintsa, Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA 02138, USA. (ehintsa@nsf.gov)

J. E. Johnson, Pacific Marine Environmental Laboratory, NOAA, 7600 Sand Point Way NE, Seattle, WA 98195, USA. (james.e.johnson@noaa.gov)

E. Joseph, Department of Physics and Astronomy, Howard University, 2355 6th Street NW, Washington, DC 20059, USA. (ejoseph@howard.edu)

T. L. Kucsera and J. Warner, Joint Center for Environmental Technology, University of Maryland Baltimore County, 1000 Hilltop Circle, Baltimore, MD 21250-0001, USA. (tlk@croc.gsfc.nasa.gov; juying@umbc.edu)

W. W. McMillan, Department of Physics, University of Maryland Baltimore County, 1000 Hilltop Circle, Baltimore, MD 21250-0001, USA. (mcmillan@umbc.edu)

J. T. Merrill, Graduate School of Oceanography, 312 CACS Building, University of Rhode Island, Narragansett, RI 02882, USA. (jmerrill@gso.uri.edu)

S. K. Miller, K. L. Ross, J. B. Stone, and A. M. Thompson, Meteorology Department, Pennsylvania State University, 503 Walker Building, University Park, PA 16802, USA. (anne@met.psu.edu)

S. J. Oltmans, Earth Systems Research Laboratory, NOAA, Boulder, CO 80305, USA. (samuel.j.oltmans@noaa.gov)

K. E. Pickering, NASA Goddard Space Flight Center, Code 613.3, Greenbelt, MD 20771, USA. (pickerin@gator1.gsfc.nasa.gov)

F. J. Schmidlin, Wallops Flight Facility, NASA Goddard Space Flight Center, Code 614.4, Wallops Island, VA 23337, USA. (fjs@osb1.wff.nasa.gov)

D. W. Tarasick, Meteorological Services Centre, Environment Canada, Downsview, ON, Canada M3H 5T4. (david.tarasick@ec.gc.ca)

J. C. Witte, Science Systems and Applications Inc., Lanham, MD 20706, USA. (witte@gavial.gsfc.nasa.gov)