Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) 2004: 1. Summertime upper troposphere/lower stratosphere ozone over northeastern North America

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Received 25 April 2006; revised 15 November 2006; accepted 10 January 2007; published 15 May 2007.

[1] Coordinated ozonesonde launches from the Intercontinental Transport Experiment (INTEX) Ozonesonde Network Study (IONS) (http://croc.gsfc.nasa.gov/intex/ions.html) in July-August 2004 provided nearly 300 O_3 profiles from eleven North American sites and the R/V *Ronald H. Brown* in the Gulf of Maine. With the IONS period dominated by low-pressure conditions over northeastern North America (NENA), the free troposphere in that region was frequently enriched by stratospheric O_3. Stratospheric O_3 contributions to the NENA tropospheric O_3 budget are computed through analyses of O_3 laminae (Pierce and Grant, 1998; Teitelbaum et al., 1996), tracers (potential vorticity, water vapor), and trajectories. The lasting influence of stratospheric incursions into the troposphere is demonstrated, and the computed stratospheric contribution to tropospheric column O_3 over the R/V *Ronald H. Brown* and six sites in Michigan, Virginia, Maryland, Rhode Island, and Nova Scotia, 23% ± 3%, is similar to summertime budgets derived from European O_3 profiles (Collette and Ancellet, 2005). Analysis of potential vorticity, Wallops ozonesondes (37.9°N, 75.5°W), and Measurements of Ozone by Airbus In-service Aircraft (MOZAIC) O_3 profiles for NENA airports in June-July-August 1996–2004 shows that the stratospheric fraction in 2004 may be typical. Boundary layer O_3 at Wallops and northeast U.S. sites during IONS also resembled O_3 climatology (June-July-August 1996–2003). However, statistical classification of Wallops O_3 profiles shows the frequency of profiles with background, nonpolluted boundary layer O_3 was greater than normal during IONS.

Citation: Thompson, A. M., et al. (2007), Intercontinental Chemical Transport Experiment Ozonesonde Network Study (IONS) 2004: 1. Summertime upper troposphere/lower stratosphere ozone over northeastern North America, *J. Geophys. Res.*, *112*, D12S12, doi:10.1029/2006JD007441.

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0148-0227/07/2006JD007441

1. Introduction

[2] An ozonesonde network provides consistently placed and well-resolved (100 m in vertical) observations of tropospheric and stratosphere O_3 that satellite and aircraft platforms do not achieve. Relative ease of operations allows for strategic design and sampling to address the mechanistic requirements of a field campaign. When combined with meteorological fields and P-T-U (pressure-temperature-humidity) radiosonde data, portions of the O_3 budget can be deduced without complex models. Illustrations are given by *Teitelbaum et al.* [1994, 1996] where O_3 and water vapor laminae were used to infer gravity and Rossby wave influences in soundings. Similarly, *Merrill et al.* [1996] used O_3 soundings to identify a wave-breaking mechanism for introducing stratospheric O_3 into the middle and upper troposphere in northern hemisphere summer. The work of Collette and colleagues [*Collette et al.*, 2005; *Collette and Ancellet*, 2005], based on thousands of European O_3 profiles from soundings and aircraft, show how persistent are stratospheric influences on O_3 in the free troposphere.

[3] The importance of stratospheric O_3 on the midlatitude free troposphere has also been demonstrated with aircraft data [*Parrish et al.*, 2000; *Esler et al.*, 2003] and by trajectories related to surface O_3 [*Moody et al.*, 1995; *Merrill et al.*, 1996]. These observations show more stratospheric flux during springtime (as expected from models [e.g., *Meloen et al.*, 2003; *Olsen et al.*, 2004]) but the frequency of stratospheric encounters at other seasons is remarkable. Aircraft O_3 profile classifications [*Browell et al.*, 1996a] displayed stratospheric influences in 40–50% (frequency, not mass content) of midlatitude O_3 profiles above 300 hPa during late summer and early fall. In northeastern U.S. and North Atlantic flights designed to sample aircraft and surface-influenced pollution in early fall 1997 [*Singh et al.*, 1999; *Thompson et al.*, 1999], stratospheric O_3 was encountered on 40% of flights. *Cho et al.* [1999, 2001] detailed gravity wave influences on the UT/LS and showed that pollution transport can be modified by stratospheric capping.

[4] During the ICARTT (International Consortium on Atmospheric Research on Transport and Transformation [*Fehsenfeld et al.*, 2006]) and INTEX-NA (Intercontinental Chemical Transport Experiment–North America [*Singh et al.*, 2006]) experiments in July-August 2004, coordinated ozonesonde launches were made through IONS (INTEX Ozonesonde Network Study). In this study, we apply the methods of *Teitelbaum et al.* [1994] and *Pierce and Grant* [1998] to IONS O_3 profiles to address the following: (1) How much tropospheric O_3 in northeastern North America (NENA) originated in the stratosphere? (2) Given that July-August 2004 meteorology over NENA was dominated by persistent low-pressure conditions that favored injection of stratospheric O_3 to the troposphere, was stratospheric influence unusually high compared to other years? (3) How did 2004 BL (boundary layer) O_3 in NENA compare to climatology? Experimental and analytical methods are described with a case study in section 2, followed by two sections that answer the questions.

2. Observations and Analysis

2.1. IONS-04: Experiment and Ancillary Data

[5] The strategic objectives defining IONS were threefold. (1) With coordinated launches at participating stations (Table 1), measurements were timed for satellite overpasses; sonde data were transmitted within 8 hours of launch to North American and European research aircraft field sites to guide Lagrangian sampling. (2) Coverage of IONS from south central United States through maritime Canada, spanning eight of the stations and a ship in the Gulf of Maine, was designed for a cross-sectional view over a major pollution pathway. This is illustrated by the ozone mixing ratio "curtain" for 21 July 2004 (Figure 1). (3) Soundings over coastal California, Colorado, Michigan and Ontario allowed study of intercontinental and cross-continental flows.

[6] Over 290 soundings were taken between 1 July and 15 August 2004, with ECC (electrochemical concentration cell) ozonesondes at all locations. Water vapor was derived from P-T-U profiles measured by radiosondes that flew with the ozonesondes. The ozonesonde-radiosonde system used at Trinidad Head, Houston, Pellston, Huntsville, Narragansett and the R/V *Ronald H. Brown* [*Thompson et al.*, 2000] recorded daily profiles at 1.2-s frequency, generally during overpasses of the Aqua and Aura satellites. The latter was launched on 15 July 2004 with four O_3 instruments (http://aura.gsfc.nasa.gov). Other IONS sites launched sondes 1–3 times/week. Images of all IONS O_3 profiles are at http://croc.gsfc.nasa.gov/intex/ions.html.

[7] For comparisons with O_3 climatology over NENA, MOZAIC profiles (Measurements of Ozone and Water Vapour by Airbus In-service Aircraft; http://mozaic.aero.obs-mip.fr/web) and surface O_3 from an EPA network, http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm, are used. Satellite data, trajectory-enhanced meteorological fields and imagery as well as standard analyses (e.g., potential vorticity, potential temperature; all determined with GEOS-4, http://croc.gsfc.nasa.gov/intex) are used to interpret O_3 and P-T-U profiles. All trajectories are run with a kinematic version of the Goddard trajectory model [*Schoeberl and Newman*, 1995] using GEOS-4 analyses.

2.2. Evaluation of Boundary Layer and Stratospherically Influenced Ozone

[8] Tropospheric O_3 within each sounding is assumed to originate from four processes: boundary layer photochemistry (BL), stratospheric intrusion (ST), interaction of regional pollution with convection plus photochemical reactions from lightning-generated NO (RCL), and advection, including recent transport and aged ozone (AD).

[9] The first two terms in the tropospheric O_3 budget, BL and ST, are foci for the present study. These are computed as follows:

[10] 1. The first term is boundary layer (BL) O_3. Except for a few low-level inversions (200–300 m), the P-T-U profiles show that the BL over IONS stations was 0.5–1.5 km. The lower limit held for the shipboard soundings [*Angevine et al.*, 2006]. Computed BL O_3 is based on integration from the surface to 0.5 or 1 km, with column amounts given in DU (Dobson Units; 1 DU = 2.69×10^{16} cm^{-2}). For comparison to O_3 measured at surface monitoring locations, IONS "surface O_3" is determined by averaging O_3 measured by the sonde from the surface to 100 m. Including more than the lowest 2–3 values allows for the sonde response time and correction for interferences with surface ECC O_3 readings, although no definite interferences

Table 1. IONS Stations in 2004: R/V *Ronald H. Brown* Operating From Portsmouth, New Hampshire

Station	Location
Beltsville, Maryland, USA	39.04°N, 76.52°W
Boulder, Colorado, USA	40.30°N, 105.20°W
Egbert, Ontario, Canada	44.23°N, 79.78°W
Houston, Texas, USA	29.72°N, 95.40°W
Huntsville, Alabama, USA	35.28°N, 86.58°W
Narragansett, Rhode Island, USA	41.49°N, 71.42°W
Pellston, Michigan, USA	45.57°N, 84.68°W
Sable Island, Nova Scotia, Canada	43.93°N, 60.01°W
Trinidad Head, California, USA	40.80°N, 124.15°W
Wallops Island, Virginia, USA	37.85°N, 75.50°W
Yarmouth, Nova Scotia, Canada	43.87°N, 66.12°W

were detected. A similar calculation gives "surface O_3" from the MOZAIC profiles.

[11] 2. The second term is ST O_3 amount. Determination of the stratospheric contribution to tropospheric O_3 is based on modification of the technique for classifying waves in stable O_3 layers that was introduced by *Teitelbaum et al.* [1994, 1996] and *Pierce and Grant* [1998]. Designated here as the PT (for Pierce and Teitelbaum) or lamina-labeling method, the use of laminae is based on the observation by *Holton* [1987] and others [*Dobson*, 1973; *Reid and Vaughan*, 1991], that most soundings show a relatively constant O_3 profile within the troposphere or stratosphere except where regional perturbations lead to stable layers that deviate significantly from the mean. *Newell et al.* [1999] made similar observations about aircraft O_3 profiles.

[12] In Figure 1, a cross section of O_3 mixing ratios from IONS soundings, all the profiles illustrated display a relatively constant mixing ratio between the BL and the tropopause, whether the latter is determined by conventional thermal or ozone-based definitions. This is evident in individual O_3 profiles depicted in Figure 2a. In Figure 1, Huntsville and Beltsville display a distinct tropopause; the region below the sharp red-to-brown transition can be considered as the upper troposphere (UT). The UT has a more transitional character in a 2-km thick region shaded red-orange-yellow for the other stations. For Sable Island, the mean tropospheric mixing ratio is 40–45 ppbv, corresponding to light blue-light green in Figure 1. For all profiles except Sable Island, the midtropospheric mean O_3 is ~75 ppbv, green-to-yellow shading, and the UT mean is ~100 ppbv O_3, orange in Figure 1.

[13] In the PT method, stable laminae in O_3 and potential temperature are identified within an ozonesonde-radiosonde pair. Each stable lamina of tropospheric O_3 that is designated as a Rossby wave (RW) is associated with a filament from wave-breaking, i.e., stratospheric influence, provided there is supporting evidence from trajectories and/or tracers, H_2O or pv (compare the GW studies of *Teitelbaum et al.* [1994]). To apply the laminar method to the tropospheric O_3 budget, the tropopause is first determined. Both a TTP (thermal tropopause) and OTP, an ozone-defined tropopause, are used. The latter is determined with the method of Browell and coworkers [*Browell et al.*, 1996a, 1996b; *Fenn et al.*, 1999] where the OTP is the first 100 ppbv O_3 mixing ratio point below a specified rate of stratospheric ozone decline. In Figure 1 this generally corresponds to the first occurrence of 100 ppbv O_3 below the brown color. The pv-tropopause has been compared to the OTP and TTP from the soundings. Our GEOS-4 fields, at 1 × 1-degree resolution, do not resolve the differences among the northeastern and North Atlantic sites studied here. For 21 July 2004, the

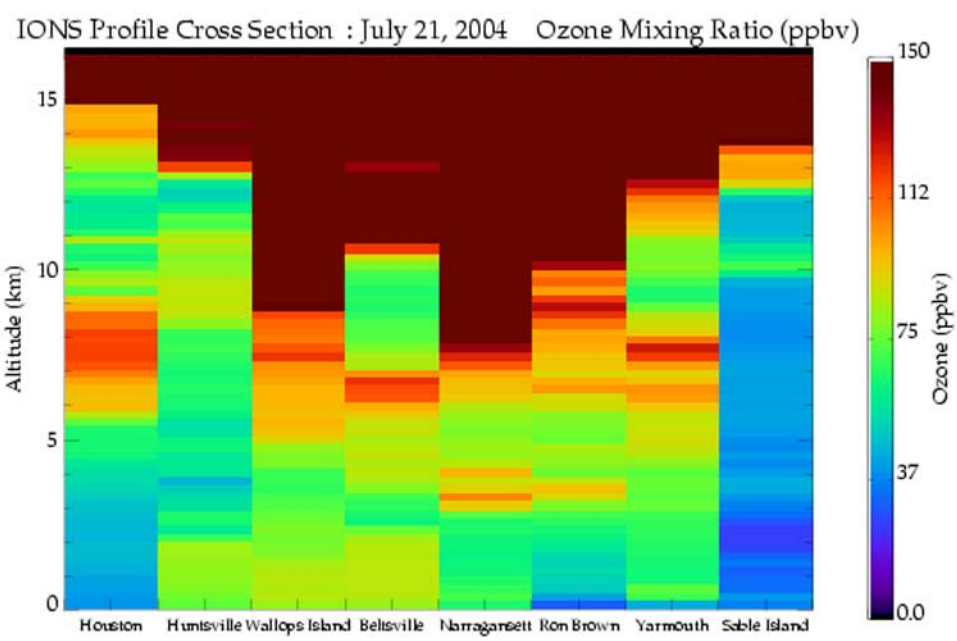

Figure 1. Cross section of O_3 mixing ratios below 17 km on 21 July 2004 from IONS soundings with sites arranged by increasing latitude. The corresponding longitudes order sites from south central to northeast United States–maritime Canada. The four New England–maritime Canada stations (Narragansett, R/V *Ronald H. Brown*, Yarmouth, and Sable Island) display a progression of tropopause height (the red-brown transition in most cases) that increases with increasing latitude and eastward increasing longitude (coordinates in Table 1).

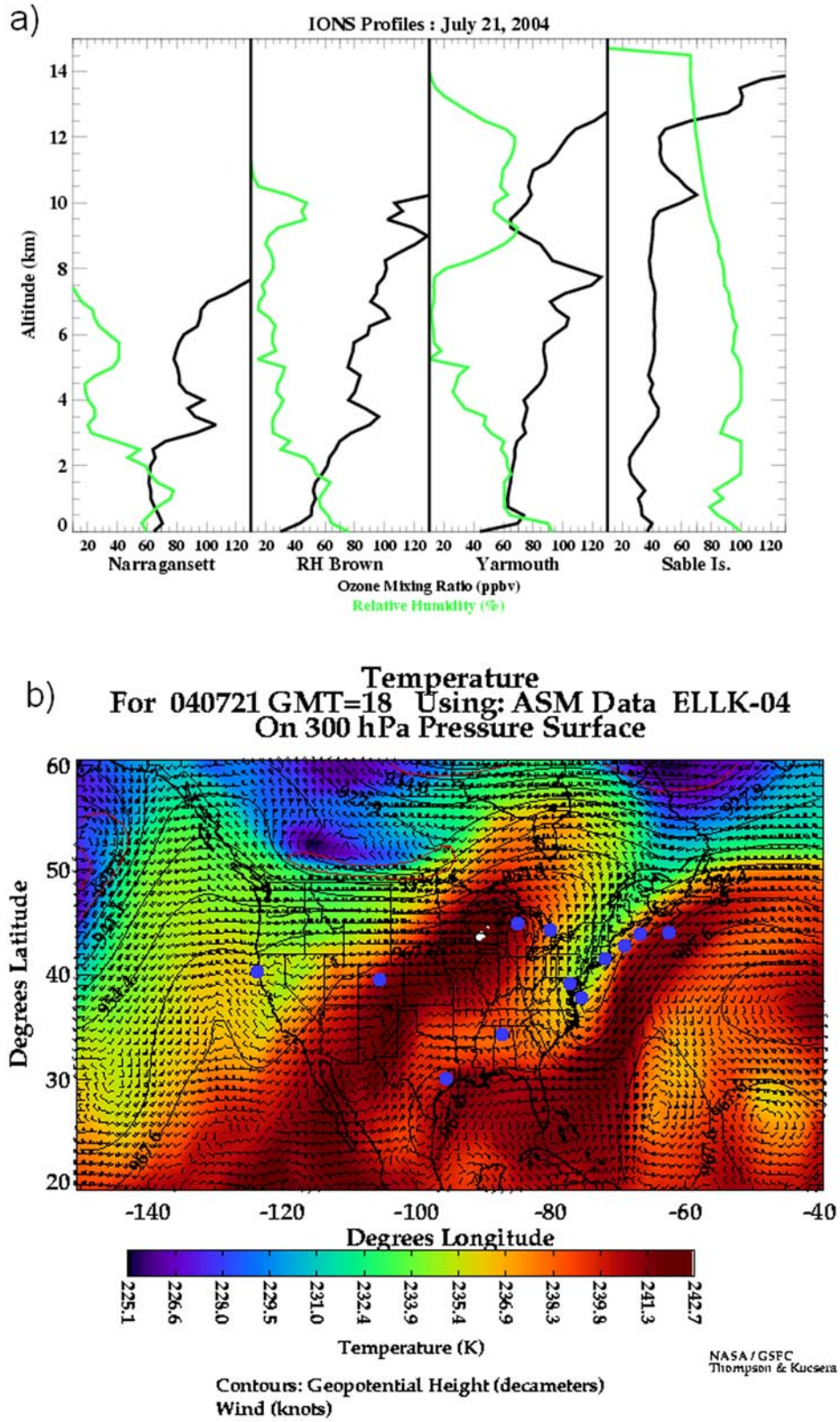

Figure 2. (a) Progression of O_3 and relative humidity (RH) profile mixing ratios for four of the stations illustrated in Figure 1, for 21 July 2004. (b) The 300 hPa surface winds and temperature, with blue dots denoting the IONS sites (Table 1), showing cooler temperature along a northeast coastal tongue. This is associated with higher epv (not shown) in a pattern of increasing tropopause height from west to east.

Table 2. Ozone on 21 July 2004 at IONS Locations, With Stratospherically Influenced Tropospheric O_3 (ST) and BL O_3[a]

21 Jul 2004	Pells	Belts	WFF	Narra	*RHB*	Yarm	Sable Island
O_3 (to OTP), DU	42	67.3	69.9	37.3	43.6	52.3	29.7
OTP, km	13.9	13.8	12.4	7.0	9.5	12.7	14.1
RW-OTP (minimum),[b] DU	5.8	6.6	14.2	0.0	0.0	14.6	2.7
RW-OTP (maximum),[b] DU	15.7	32.2	22.1	6.7	12.6	21.2	12.8
RW-OTP (minimum fraction)	0.14	0.10	0.20	0.0	0.0	0.28	0.09
RW-OTP (maximum fraction)	0.37	0.48	0.32	0.18	0.29	0.41	0.43
O_3 (to TTP), DU	42.0	49.4	77.2	85.0	60.0	52.3	28.2
TTP, km	13.4	10.9	14.2	13.9	12.7	12.8	13.9
RW-TTP (fraction)	0.23	0.15	0.30	0.33	0.16	0.34	0.16
RW to ST scale factor	...	0.5	0.65	1.0	0.65	0.77	0.0
BL (surface to 0.5 km), DU	1.5	3.4	2.5	2.7	1.6	2.5	1.6
BL (0.5 km fraction to OTP)	0.04	0.05	0.05	0.07	0.04	0.05	0.03
BL (surface to 1.0 km), DU	4.4	5.0	4.4	4.4	3.6	3.8	2.6
BL (1.0 km fraction to OTP)	0.10	0.11	0.09	0.10	0.08	0.08	0.06

[a]Pells, Pellston; Belts, Beltsville; WFF, Wallops Island; Narra, Narragansett; *RHB*, R/V *Ronald H. Brown*; Yarm, Yarmouth.
[b]Minimum, maximum determinations defined in section 2.2 and Figure 3.

example followed in this section (Table 2) Beltsville, Wallops and Narragansett are assigned the same tropopause whereas analysis of the soundings (Figures 1 and 2a) implies a 2.9- to 6.8-km difference, depending on whether OTP or TTP is used.

[14] Following *Pierce and Grant* [1998], laminae are analyzed to 25 or 30 km in the following procedure:

[15] Step 1 is to locate laminae in O_3 through normalization to a running mean and in potential temperature, θ, from the radiosonde data. For the 21 July 2004 example, Sable Island has pronounced laminae at 13 km, 10.5 km and 4 km. These appear as solid lines and positive deviations from the 40–45 ppbv O_3 mean in Figure 3a. The θ deviations are dotted lines. Negative deviations define O_3 laminae at 13.5 km, 12.2 km, 9 km and 2.5 km. Positive O_3 deviations for the R/V *Ronald H. Brown* and Narragansett (Figures 3b and 3c) are significant at 9 km, where Figure 1 displays more red than orange, and at 3–4 km, which is more yellow-orange than the 75 ppbv O_3 mean.

[16] Step 2 is to compare correlations between O_3 and θ gradients to identify gravity waves (GW) and RW. The correlations (dashed lines in Figure 3), with positive values >0.7 define GW. A range of RW limits, with RW denoted by a "low-correlation" criterion, is investigated. Standard PT practice sets the GW limit for correlation within ±0.3. We also considered a RW classification defined with the correlation cutoff raised to ±0.4 and lowered to ±0.2, to bracket, respectively, maximum and minimum RW values.

[17] Step 3 is to compute integrated ozone in a layer, in Dobson Units.

[18] Step 4 is to add all such layers within a profile to obtain total RW, absolute and fraction, within the O_3 profile from surface to OTP or TTP.

[19] Two sources of uncertainty affect the RW determination in steps 1–4. First, not all O_3 profiles display sufficiently stable layers to give meaningful results with lamina-labeling. This eliminates profiles affected by active mixing, presumably from both surface and the stratosphere. Depending on the site, 10–20% of the NENA IONS profiles are excluded by this criterion. Second, O_3 and P-T-U data between 750 and 200 hPa during IONS were such that the $O_3-\theta$ gradient correlations often shifted near the edges of the RW range, e.g., the correlation curve for R/V *Ronald H. Brown* near 9 km in Figure 3b. These uncertainties are estimated as equivalent to a typical O_3 column depth in 0.5 km (±2–3 DU).

[20] Table 2 summarizes the BL and RW amounts and fractions for six NENA stations on 21 July, along with the same day's soundings over Pellston, Michigan. Three RW values are given, two based on the minimum and maximum RW criteria for the O_3-defined tropopause (OTP) and one derived from the standard RW criterion, 0.3 absolute for the low-correlation limit [*Teitelbaum et al.*, 1994; *Pierce and Grant*, 1998], with a thermally defined tropopause (TTP).

[21] For 21 July 2004, the TTP for the NENA sites is 11–14 km. For all but two stations (R/V *Ronald H. Brown* and Narragansett), the OTP and TTP difference is less than 3 km. The greatest divergence between the OTP and TTP, nearly 7 km at Narragansett, leads to a large difference between tropospheric O_3 column and RW fractions computed with the two criteria. The tropospheric O_3 is 37 DU integrated to the OTP (at 7.0 km) whereas total O_3 is 85 DU to 13.9 km. This is equivalent to an RW minimum fraction of 0 (maximum of 0.18), based on the OTP. The conventional RW criterion, correlation $\leq|0.3|$ with the TTP, gives a fractional value for RW O_3 = 0.33 at Narragansett.

[22] The final step in lamina-labeling as applied to IONS data is the use of tracers to assign the amount of O_3 identified as RW to ST O_3 within the profile.

[23] In step 5 each layer labeled as RW is examined with respect to its corresponding RH or degree of undersaturation; back trajectory origins and potential vorticity [*Stone*, 2006] to assign a scale factor. The scale factor for the 21 July laminae appears in Table 2. Stratospherically influenced O_3 layer amounts are added together to give the total ST O_3 amount.

[24] A systematic analysis of layers in European summer O_3 profiles was presented by *Collette et al.* [2005], who assigned ST amounts affecting the free tropospheric segment of soundings through a 21-factor principal component analysis. They found no differences when the number of factors was reduced to six and the humidity-O_3 relationship turned out to be the most robust indicator of ST impact. Thus our method maps well to the more statistically comprehensive analysis of *Collette et al.* [2005].

[25] In the 21 July 2004 IONS examples, RW is equivalent to ST at Narragansett because the RW-labeled laminae correspond to relatively dry segments of the profile. For

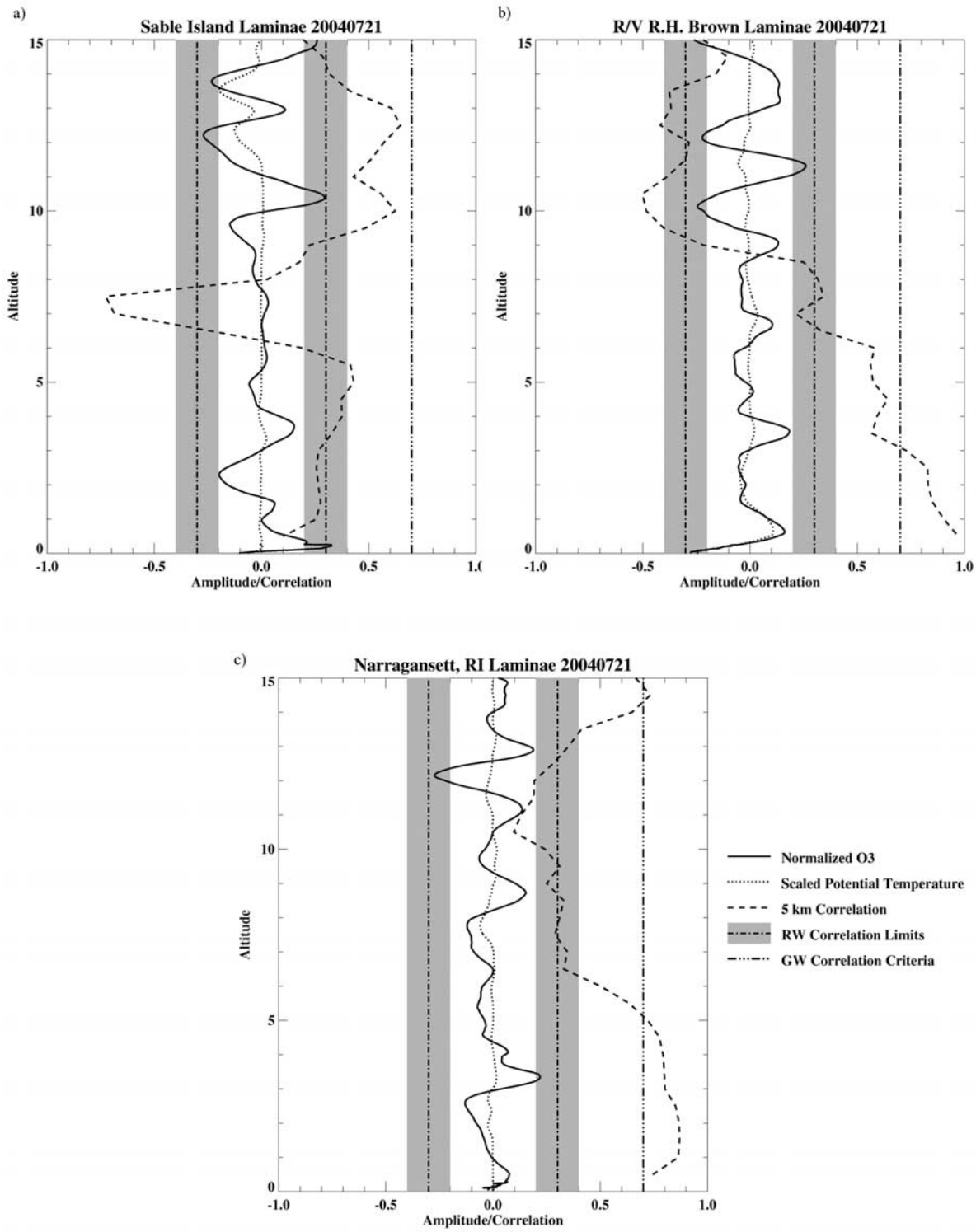

Figure 3. Elements of PT analysis for three ozone profiles shown in Figures 1 and 2a: (a) Sable Island, (b) R/V *Ronald H. Brown*, and (c) Narragansett. Ozone laminae (normalized relative to a running mean within the profile) are solid lines. Dotted lines are normalized potential temperature (θ) laminae from P-T-U measurements of the radiosonde flown with the ozonesonde. Dashed lines signify correlations between O_3 and θ gradients. Low correlations, within the shaded ranges, are attributed to Rossby waves (RW). The corresponding O_3 layer amounts for a "minimum" RW O_3 in Table 2 are based on dashed lines falling between -0.2 and 0.2; upper limits for RW O_3 (Table 2 "maximum") are based on -0.4 to 0.4. The PT method assigns gravity wave (GW) character to high correlations (>0.7) between O_3 and θ gradients.

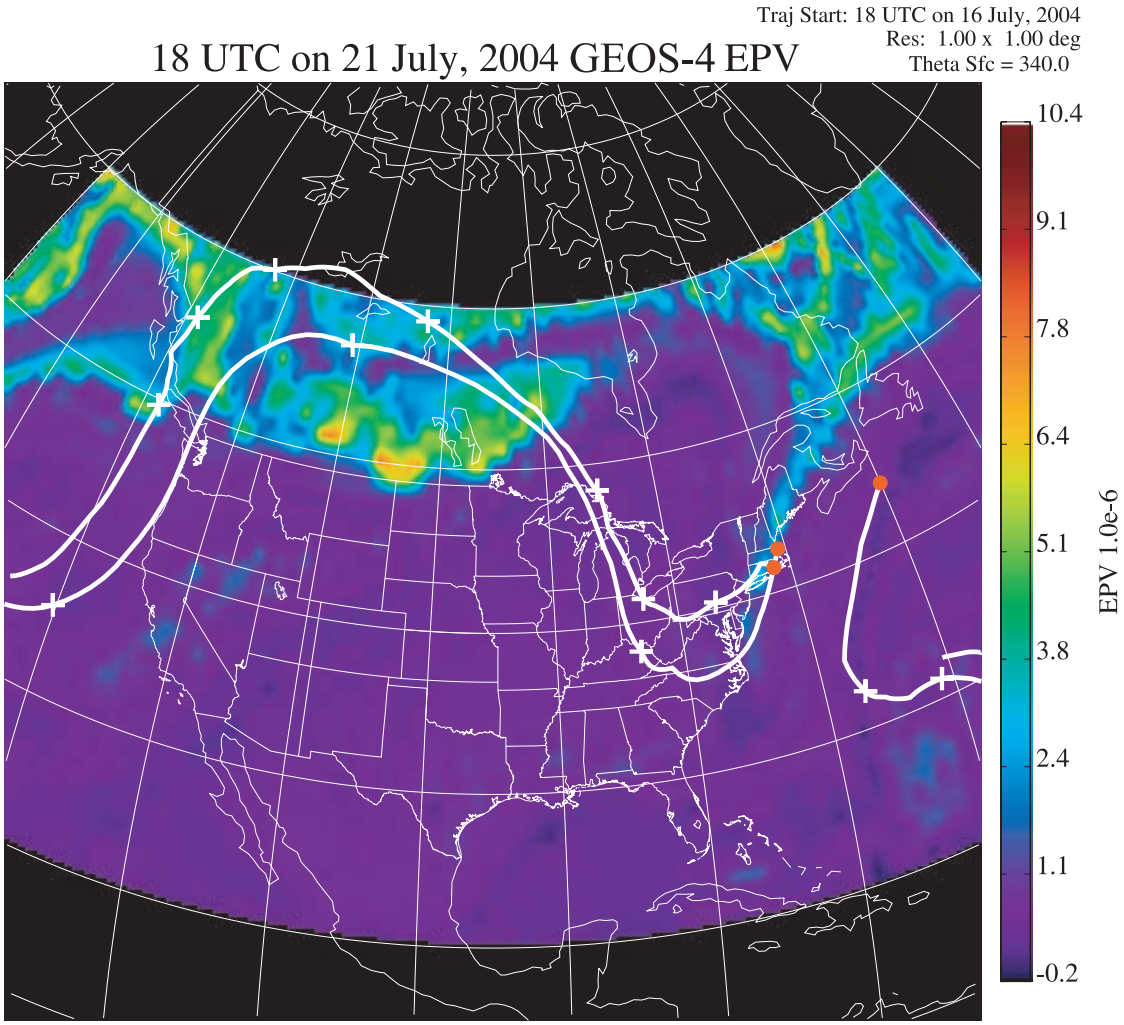

Figure 4. IONS sites (red dots) with origins of 8-day back trajectories (in white) from sites Narragansett, R/V *Ronald H. Brown*, and Sable Island, superimposed on trajectory-mapped epv for 21 July 2004. Levels chosen for the trajectory initialization correspond to RW laminae (Figure 3). Pv and meteorological fields used in trajectories, initialized at 1° × 1° resolution, are from the Goddard Assimilation Model (GEOS-4 version [*Bloom et al.*, 2005]). Trajectory mapping (as depicted in the map) displays a 5-day impact of pv in air parcels arriving at each grid point in contrast to an instantaneous pv value at the location.

Sable Island the entire troposphere is near-saturation and the scale factor is 0. These interpretations are reinforced by pv (in the analyses here, Ertel's pv, computed from GEOS-4) and the origins of back trajectories (Figure 4). Back trajectories initiated from the midtroposphere over the R/V *Ronald H. Brown* and Narragansett show O_3 in those air parcels originating over Canada at 60°N (Figure 4) where the trajectory-mapped pv corresponds to stratospheric values. The levels for the O_3 soundings at which the air parcel trajectories were initialized correspond to laminae associated with RW. The trajectory-mapped pv in Figure 4 displays a filament with elevated pv (>3 pvu at ~10 km in Figure 4) over the R/V *Ronald H. Brown* and Narragansett, extending south to the mid-Atlantic region. The temperature on 21 July 2004 (Figure 2b) shows the narrow filament to be cooler than the surrounding regions. To the east of the filament is a transition toward subtropical pv levels (denoted purple in Figure 4) that includes Yarmouth (red spot in western Nova Scotia; cf. Table 1) and Sable Island (easternmost red spot in Figure 4). As the transition suggests, Yarmouth's 10-km O_3 mixing ratio (~80 ppbv; Figure 1) is less than the 10-km O_3 mixing ratio at Narragansett and the R/V *Ronald H. Brown*, both >100 ppbv. Lamina-labeling is sufficiently sensitive to pick up RW character in the Yarmouth sounding (Table 2); relatively low RH (Figure 2a) confirms stratospheric influence below 12 km.

3. Results: The 2004 IONS ST and BL Ozone

[26] Section 3.1 summarizes statistics for BL and ST O_3 over all the NENA IONS soundings. These are discussed

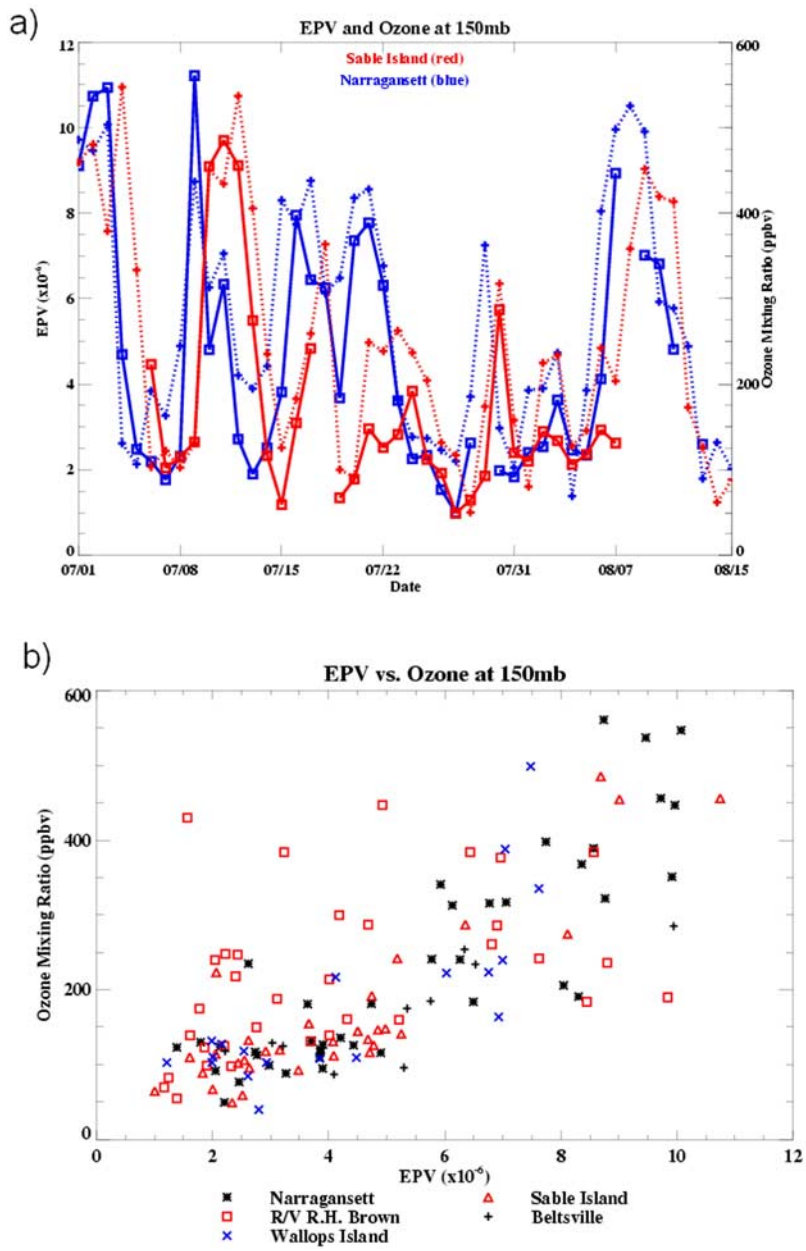

Figure 5. (a) Daily pv (interpolated at 150 hPa, ~13 km, from GEOS-4 analyses) from 1 July to 15 August 2004 (pluses) with O_3 mixing ratio at 150 hPa (squares) at Narragansett (blue) and Sable Island (red). The R/V *Ronald H. Brown* and Yarmouth pv and O_3 are similar to Narragansett. (b) Scatterplot of O_3 and pv (150 hPa) over five IONS sites for 1 July to 15 August 2004.

with reference to ancillary observations during the IONS period in section 3.2.

3.1. Composite ST and BL Ozone Statistics During IONS

[27] The Narragansett OTP-TTP discrepancy described in section 2.2 is not typical for IONS. Over the entire set of profiles analyzed by lamina-labeling, the mean OTP-TTP difference is 0.3 km. Thus, for statistical analysis of ST and BL fractions over IONS profiles as a whole, a single definition (OTP) is used. Averaged parameters for OTP, O_3 column amounts, ST and BL fractions appear in Table 3.

[28] Amounts and fractions of ST O_3 are similar among NENA sites. The mean ST fraction is 0.23 (±0.03). BL O_3 amounts for Trinidad Head (not shown) and Pellston, sites usually remote from pollution, are roughly half the amount at NENA locations. Sable Island is also low in BL O_3 because it usually had marine, southeasterly near-surface flows. The most urban site, Beltsville, has the highest BL O_3, ~20% greater than the other locations listed in Table 3.

3.2. Observations Related to IONS ST and BL Ozone

[29] Three additional analyses put 2004 ST and BL O_3 into perspective.

[30] 1. The first analysis involves ozone at 10–15 km. To further minimize ambiguity in the interpretation of ST O_3 due to tropopause definitions, the integrated 10–15 km O_3 column is computed (average value in Table 3). This can be

Table 3. Maximum and Minimum ST and BL O_3 Amounts and Fractions for IONS-04 Soundings (1 July to 13 August 2004)

IONS-04	Pells	Belts	WFF	Narr	*RHB*	Yarm	Sable	Trini
O_3 (to OTP), DU	39.9	47.1	47.5	44.7	45.6	45.4	40.7	40
OTP, km	11.7	12.1	13.8	12.2	12.5	12.6	14.8	12.6
RW-OTP (minimum),[a] DU	9.8	14.3	13.6	12.1	8.4	12.5	8.7	9.8
RW-OTP (maximum),[a] DU	17.4	21.8	25.2	20.4	19.3	20.9	17.7	20.3
RW-OTP (minimum fraction)	0.24	0.30	0.29	0.27	0.18	0.28	0.21	0.24
RW-OTP (maximum fraction)	0.44	0.46	0.53	0.46	0.42	0.46	0.43	0.51
RW to ST scale factor[a]	...	0.62	0.56	0.64	0.68	0.63	0.70	...
ST O_3,[b] DU	...	13.5	16.0	11.7	11.8	11.1	6.6	...
ST O_3 (fraction)	...	0.24	0.28	0.23	0.18	0.22	0.24	...
O_3 (to 15 km), DU	59.6	58.2	49.6	57.2	60.6	52.8	47.5	49.7
BL (surface to 1.0 km), DU	2.6	5.0	4.4	4.4	3.9	3.8	2.6	2.4
BL (1.0 km fraction to OTP)	0.07	0.11	0.09	0.10	0.09	0.08	0.06	0.06
Surface O_3 (0–100 m), ppbv	34	66	47	46	30	35	25	18

[a]Minimum, maximum determinations defined in section 2.2 and Figure 3.
[b]Average from scaling RW-OTP maximum and minimum to ST O_3.

considered a geometrically prescribed UT/LS O_3 amount. Figure 6a shows that the fraction of O_3 in the 10–15 km layer, relative to surface-to-15 km O_3, compares well with the below-tropopause RW fraction. RW O_3 fraction averages 0.35 among the six NENA sites; with scaling, the ST O_3 fraction is about 1/3 lower.

[31] 2. The second analysis uses MOZAIC LTO (landing and takeoff) profiles [*Thouret et al.*, 1998]. These are normally measured below 200 hPa (~11 km) and are useful for characterizing UT/LS O_3 when limited sonde data are available [*Cooper et al.*, 2005, 2006] (section 4). The 9–11 km O_3 segments from MOZAIC profiles during JJA 2004, mostly from LTO at New York and Washington, are compared with IONS segments between 9 and 11 km at NENA locations (Figure 6b). Strong similarity in the fractions suggests that characteristics of IONS sonde O_3 profiles, e.g., fractions of ST O_3, apply to the larger NENA region.

[32] 3. The third analysis employs potential vorticity as a tracer. The mean tropopause height (OTP in Table 3) for the six NENA sites corresponds to 150 hPa. A sequence of 150 hPa O_3 from two IONS sites shows typical tracking with pv (Figure 5a). Stratospheric O_3 is identified with $O_3 >$ 150 ppbv and pv > 3 pvu. When mean O_3 is ~100 ppbv and pv is less than 2 pvu, the PT method may show RW segments and ST O_3. The positive correlation between pv and O_3 at the sites in Figure 5b is typical of IONS data as are recurring episodes of stratospheric influence throughout July-early August 2004. Thus pv is used as a tracer for stratospheric influence when profile data are not available above 200 hPa (section 4). Figure 5a may suggest a tendency toward lower pv (and lower O_3 mixing ratio) as summer progresses. This is consistent with trajectory analysis of MOZAIC LTO profiles in which *Cooper et al.* [2005] conclude that stratosphere-to-troposphere injections occur throughout North American summer but with declining O_3 flux over time.

[33] Figure 7 presents O_3 surface mixing ratio for six NENA sites during IONS and for four MOZAIC airports (surface to 100 m means in JJA 2004). Because profile data represent 20 s of sampling, surface readings from O_3 monitoring sites (continuous readings) near each city are also presented. The latter are daytime averages from sites reporting to the EPA (http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm). The marine locations, R/V *Ronald H. Brown*, Sable Island and Yarmouth on the western Nova Scotia coast, are in the 25–35 ppbv range. This resembles O_3 in the lowest layer shown for 21 July 2004 in Figures 1 and 2a. On average for the IONS period, Wallops and Narragansett surface O_3, at 47 and 46 ppbv (Figure 7), respectively, are more affected by pollution than Yarmouth and the R/V *Ronald H. Brown*. The near-urban location, Beltsville, averaged 66 ppbv for the IONS period, greater than the nearby MOZAIC (Washington-Dulles) and EPA Baltimore-Washington data (JJA mean O_3 = 45 ppbv; Figure 7). On average, O_3 mixing ratios at IONS sites are lower than MOZAIC surface measurements. Besides the latter being more urban, aircraft profiling usually occurs a few hours after the sondes, at midafternoon to early evening, when surface O_3 is near its daily maximum.

4. The 2004 Ozone and Climatology

[34] To compare ozone from IONS with recent climatology (1996–2004), Wallops sondes and MOZAIC profiles are used to examine the free troposphere and UT/LS (section 4.1) and BL and surface ozone (section 4.2).

4.1. UT/LS Ozone and Potential Vorticity Over NENA (1996–2004)

[35] Wallops is the only NENA IONS station with a multiyear sounding record [*Newchurch et al.*, 2003]. Normal sampling frequency is weekly, usually midweek within a couple hours of local noon. ST and BL O_3, as in Table 3, are computed from Wallops soundings for JJA 1996–2004. Figure 8a, the time series of mean ST O_3, shows that 2004 is not unusual in the recent climatology. Nor is the fraction of UT/LS ozone in the 10–15 km layer, also displayed in Figure 8a. Wallops, at the southern end of the region designated as NENA in this study, may not be representative of the northeastern United States or maritimes. MOZAIC LTO O_3 profiles are used as a reference for the mid-upper troposphere (300–200 hPa, 9–11 km). A time series of 1996–2004 MOZAIC and Wallops 9–11 km O_3 column amounts (Figure 8b) shows remarkable consistency within each data set, though MOZAIC averages 2–3 DU greater in absolute O_3 amount and 30–40% fractionally. For 2004, the standard deviation of both data sets is small and the 9–11 km column and fractional amounts are identical, as implied by Figure 7b.

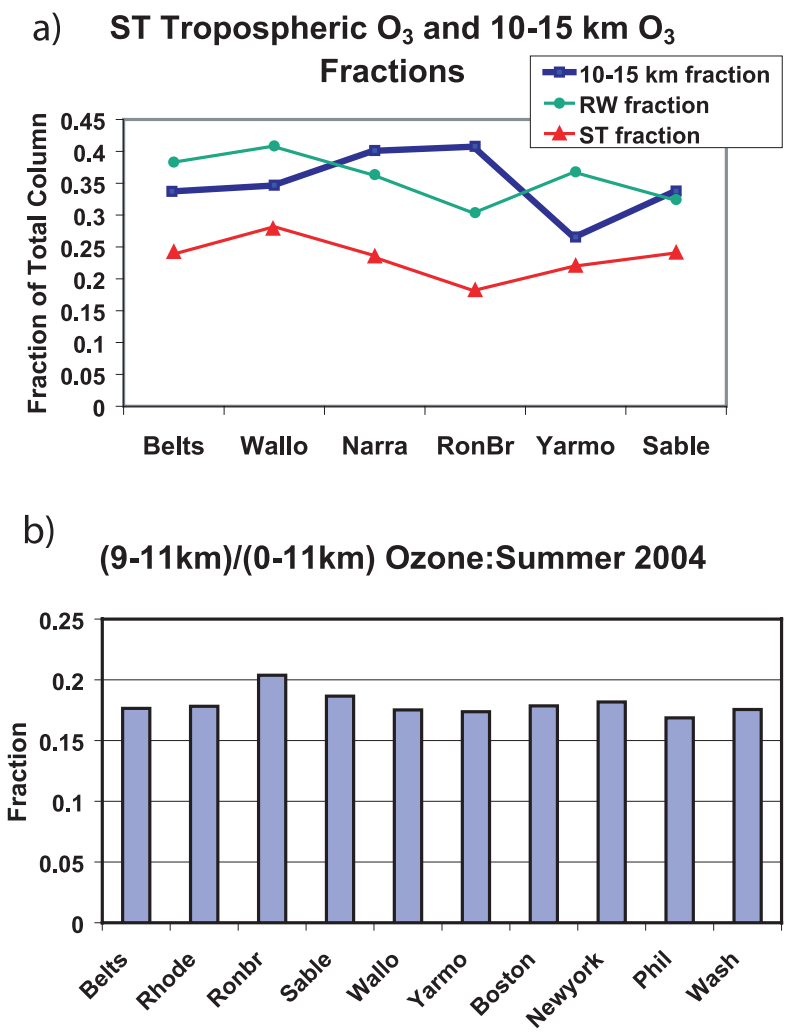

Figure 6. (a) Mean RW and ST, fraction of stratospherically influenced O_3 below tropopause (Table 3) at NENA IONS sites, and the mean fraction of O_3 within the layer 10–15 km compared to the total surface-to-15 km O_3 column. RW and ST O_3 amounts are based on averages of minimum and maximum RW-OTP (Table 3). (b) Fractions of O_3 within 9–11 km layer relative to O_3 column from surface to 11 km for IONS sites and MOZAIC LTO profiles at four NENA airports during JJA 2004.

[36] To compensate for limited pre-2004 O_3 data above 200 hPa over NENA, statistics for pv at 150 hPa [cf. *Thompson et al.*, 1999] are used to determine whether ST O_3 fractions during IONS might be unusual. Figure 9 displays probability distributions of daily epv for JJA in 1996–2004 over the locations of Pellston and the six NENA sites analyzed in Tables 2 and 3 and Figures 6 and 7. The 2004 distribution (red in Figure 9) is somewhat broader than other years, with more values >10 pvu. However, standard statistical tests do not show 2004 to be anomalous. Thus pv statistics at IONS sites, like the Wallops O_3 analysis, suggest that 2004 was a typical year for UT/LS O_3 over NENA.

4.2. BL Ozone at NENA IONS Sites in the 1996–2004 Context

[37] The synoptic conditions of NENA during IONS appeared to be anomalous when compared to climatology. Parameters like temperature, cloud cover, precipitation amount (not shown; see analyses at http://www.cdc.noaa.gov) describe a summer that was cooler and wetter than normal. Ozone at the surface, based on daily maximum O_3 values from continuous data over JJA 2004 at EPA sites around Washington, DC; Baltimore and Philadelphia, were in the lowest 25–30% of values recorded during the period 1984–2004 (not shown).

[38] Figure 10 shows the surface (lowest 100 meter averages) O_3 mixing ratio for Wallops and MOZAIC NENA locations, averaged over JJA from 1996 to 2004. EPA daytime mean O_3 averaged over the 5 locations of Figure 7 is also displayed. The individual city data, as means and medians based on data from http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm, appear in the Auxiliary Material.[1] The EPA O_3 resembles the Wallops

[1]Auxiliary materials are available at ftp://ftp.agu.org/apend/jd/2006jd007441.

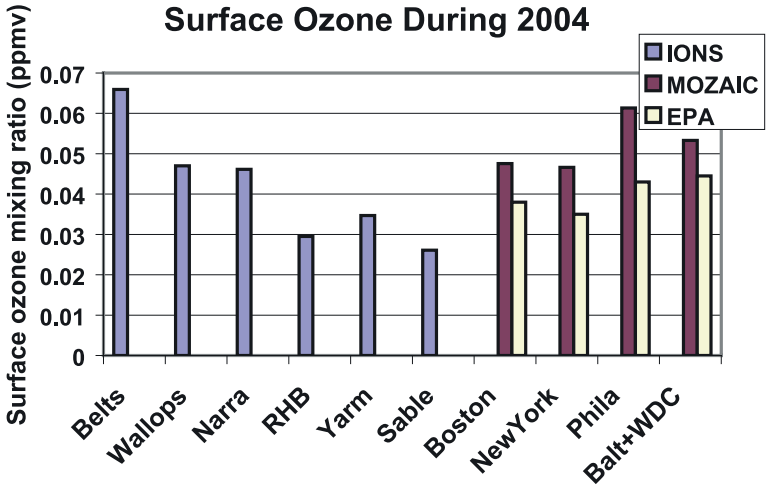

Figure 7. Near-surface O_3 mixing ratio (surface to 100 m) at six IONS sites, averaged during 1 July to 15 August 2004, and mean JJA 2004 surface O_3 from four MOZAIC airports (Boston, New York, Philadelphia, and Washington).

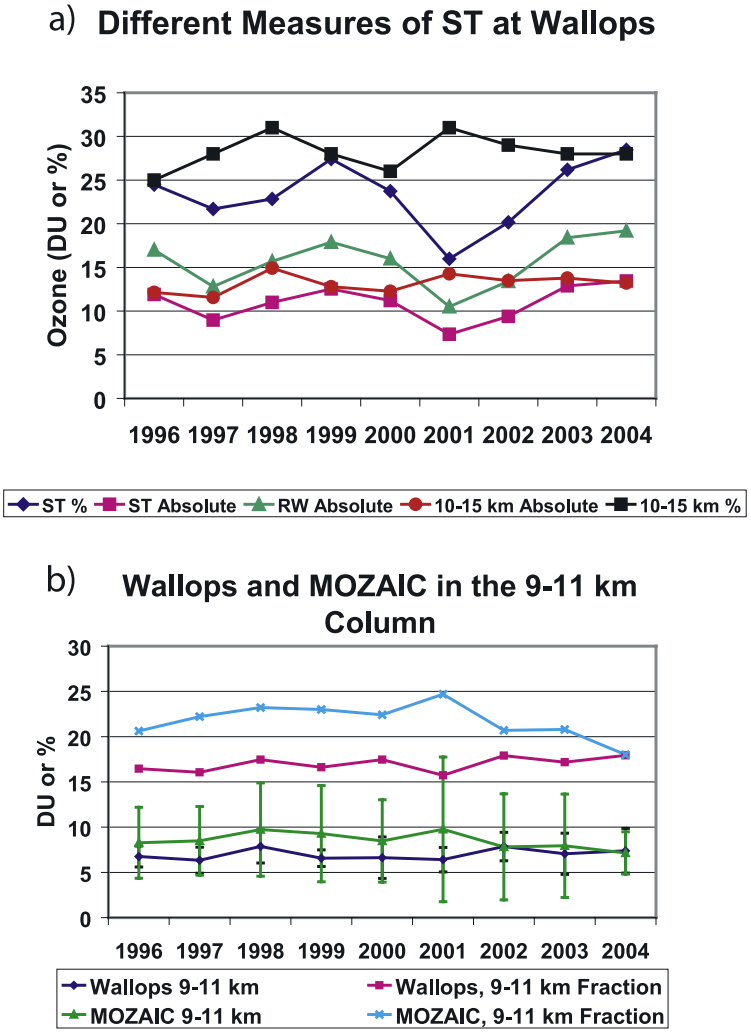

Figure 8. (a) Mean ST O_3 (DU and fraction) from JJA 1996–2004 at Wallops evaluated by the PT method with mean 10–15 km O_3 amounts from the same soundings. RW O_3 is also displayed. (b) The 9–11 km O_3 amount and fractions from JJA soundings at Wallops and for JJA MOZAIC profiles at New York, Washington, Boston, and Philadelphia.

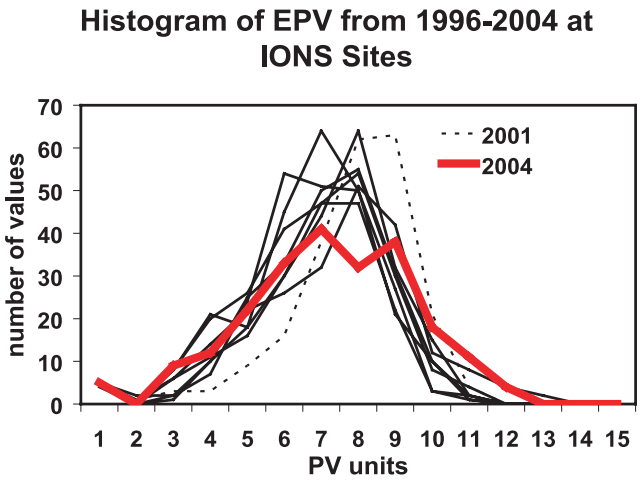

Figure 9. Histograms of pv for 1 July through 15 August for years 1996–2004 averaged over locations corresponding to Pellston, Beltsville, and Wallops (same grid point in the meteorological analysis); Narragansett-Yarmouth-R/V *Ronald H. Brown* (same value in the grid); and Sable Island. Statistical tests (e.g., Kruskal-Wallace) to determine whether any year(s) were distinct showed that 2004 (red) was not; only 2001 (dashed) was unique.

pattern, with the two means within 7 ppbv of one another, Wallops higher in all but 2 years. Wallops surface O_3 during 2004 does not stand out in the record, but mean surface O_3 is 15% lower than the 2001 and 2002 Wallops means. The O_3 column (to 2.0 km) at Wallops was 8.7 DU in 2004 compared to 10.5 ± 0.5 DU for 1996–2003. The MOZAIC surface O_3 data are not always consistent with Wallops and EPA records. It is not clear why MOZAIC O_3 is so low in 1998 and 2000.

[39] Means are not always the most effective way to evaluate O_3 profiles, as *Diab et al.* [2003, 2004] demonstrated with statistical classification of sonde and MOZAIC data over the Johannesburg (25°S, 28°E) area. Like the IONS sites, Johannesburg is a region of mixed pollution, marine and stratospheric influences. Prototype O_3 profiles for the middle and lower troposphere over Johannesburg, corresponding to distinct meteorological situations, emerged from agglomerative hierarchical clustering [*Diab et al.*, 2004] of the data. Following the latter study, we classified the O_3 mixing ratio measurements taken from Wallops sondes up to 200 hPa for JJA in 1996–2004 (148 total sondes in Figure 11). Three prototypes are distinguished in clustered distributions of the Wallops profiles. Category 1, resembling marine tropical air, is minimally polluted throughout the profile. A second category is relatively unpolluted in the lowest 5 km but displays higher O_3 mixing ratio in the middle troposphere. Category 3 is distinguished with high pollution frequency below 5 km. The BL O_3 means are 44 ppbv for category 1, 46 ppbv for category 2, and 56 ppbv for category 3. The yearly distribution for Wallops, displayed as fractions of each year's JJA profile count, appears in Figure 11. Wallops in 2004 was unique in having 20% of its profiles in category 1, compared to 5 years in 1996–2004 with 5–10% occurrence and 3 years with none. Only 30% of Wallops profiles (a minimum value, met only in one other year) were in category 3 (high-pollution) for 2004, compared to >40% of JJA profiles in five summers in the 1996–2003 period.

5. Discussion and Summary

5.1. Comparison With Other Ozone Studies

[40] The laminar- and trajectory-based analysis of ST O_3 is based on processes traced for a week or so. During INTEX-A, O_3 and other pollutants were followed by satellite [*Martin et al.*, 2006; *Pfister et al.*, 2005, 2006;

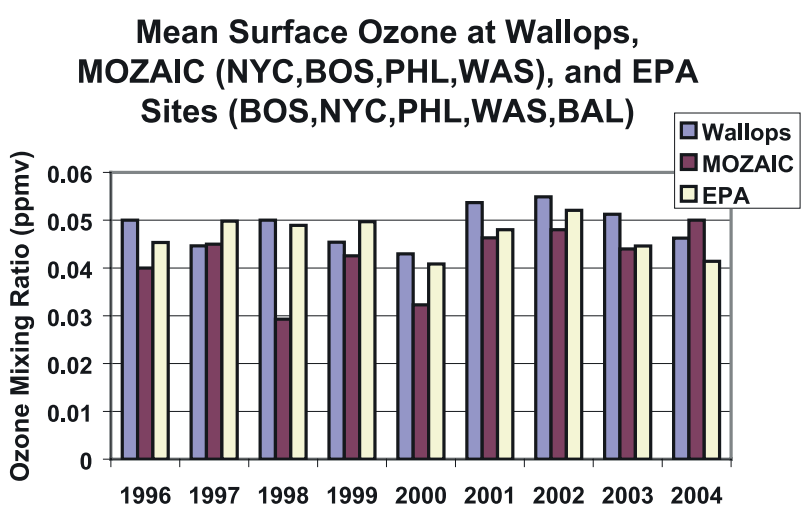

Figure 10. Surface O_3 (mean from surface to 0.1 km) for 1996–2004 (during JJA) from Wallops soundings (blue) and average from MOZAIC LTO profiles at four NENA airports (red). Same profiles are used as in Figure 8. Also displayed are daytime (0900–2100 local time, averaged, in yellow) surface O_3 readings from stations around five NENA cities (New York, Washington, Baltimore, Boston, and Philadelphia). Auxiliary Material gives city-by-city annual mean and median EPA O_3 values.

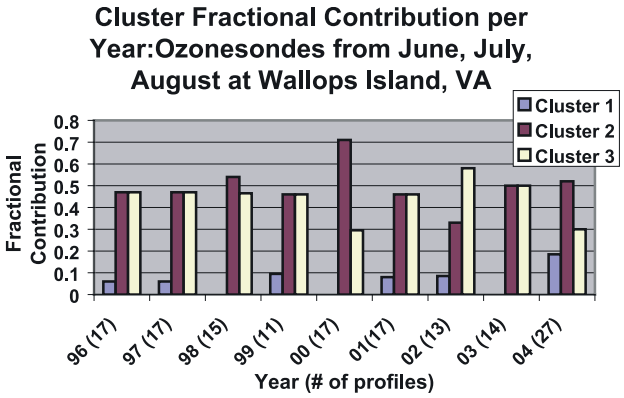

Figure 11. Histogram of the fractions of three statistically classified types of Wallops JJA O_3 profiles (from 0 to 10 km) for 1996–2004. The number of soundings for each JJA is at bottom. Profiles assigned to type 1 represent a low "background" marine-type profile in which O_3 declines monotonically above the surface. Classes 2 and 3 are discriminated by degree of O_3 pollution in the lower troposphere, with class 3 having greatest O_3 below 600 hPa. Class 2 fraction for 2004 is not exceptional but class 1 profiles are the highest fraction in 1996–2004. Class 3 fraction is a low value matched in only one other year. The years 2002 and 2004 represent an opposite distribution. Violations of the National Ambient Air Quality Standard for O_3 numbered 18 in Maryland in 2002 and only one in 2004. Similar statistics applied to New York, Rhode Island, and Massachusetts.

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 O_3 retrievals from INTEX-NA aircraft in situ measurements, unpublished manuscript, 2007], aircraft [*Pfister et al.*, 2006; 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], ship [*Fehsenfeld et al.*, 2006] and a remote ground site [*Val Martín et al.*, 2006]. 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 much UT O_3 measured in IONS soundings over eastern North America. Relevant source apportionments of summertime European O_3 soundings and aircraft profiles appear in *Colette et al.*, [2005]. Their summertime ST fractions average 15–20%, slightly less than the NENA average computed in this study.

[41] The IONS network is the first time that NENA sondes have been distributed with sufficient temporal and spatial coverage to permit evaluation of day-to-day O_3 budgets [*Thompson et al.*, 2007]. The data have been used to evaluate regional and global models [*Pierce et al.*, 2007; *Pfister et al.*, 2006; *Tarasick et al.*, 2007] and to assess O_3 contributions with the FLEX-PART trajectory approach [*Cooper et al.*, 2006]. In the latter study, combined MOZAIC and IONS profiles for some sites make direct comparison between the ST O_3 analyses from the PT (lamina-labeling) method and the FLEXPART budgets impractical. However, for NENA sites where comparisons are straightforward (Sable, Narragansett and the R/V *Ronald H. Brown*), FLEXPART assigns a 10–15% fraction of below-tropopause O_3 to stratospheric influence. The FLEXPART budget is based on an effective tropopause height that averages 1–2 km lower than OTP in Table 3. This would be equivalent to 3–4 DU less ST O_3 (Table 3) or 10% less absolute ST O_3, bringing the lamina-labeling and FLEXPART estimates into agreement with one another.

5.2. Summary

[42] Using several hundred O_3 soundings collected through the IONS network in July-August 2004, the present study evaluates two terms in the budget, BL and ST O_3. The emphasis is on ST O_3 determined with lamina-labeling and tracers. Over six NENA sites (Wallops, Beltsville, Narragansett, Yarmouth, Sable and the R/V *Ronald H. Brown*) the mean ST O_3 fraction is 23 (±3%). This value is ~20% fractionally greater than the stratospheric attribution of thousands of summertime O_3 profiles collected over Europe [*Collette and Ancellet*, 2005]. The latter analysis was based on a more comprehensive set of correlative parameters though the final criteria (humidity and back trajectories) resembled the ones used in the present study.

[43] For the IONS period our ST O_3 amounts are equivalent to stratospherically influenced tropospheric O_3 determined from FLEXPART [*Cooper et al.*, 2006] when tropopause differences between the two methods are taken into account. Other pertinent analyses are those of *Pierce et al.* [2007] and Fairlie et al. (submitted manuscript, 2006).

[44] BL column O_3 and surface O_3 were also determined from the 2004 IONS soundings. A climatological perspective on IONS is given by Wallops sondes, EPA data from the NE United States and NENA MOZAIC landing/takeoff O_3 profiles from 1996 to 2004. The BL in 2004 over Wallops showed a very low frequency of O_3 surface pollution for JJA in the 1996–2004 period. However, on average, Wallops surface O_3 in 2004 was close to the 9-year average. The MOZAIC and EPA surface O_3 records for 2004 were not unusual either.

[45] For Wallops, the O_3 profiles and pv tracer showed that stratospheric influences in 2004 were typical. Potential vorticity statistics for IONS locations during JJA 1996–2004 suggest that a 15–30% summertime ST O_3 value over NENA could be a general result. Although episodes of midlatitude ST O_3 in summer [*Moody et al.*, 1995; *Merrill et al.*, 1996] or fall [*Cho et al.*, 1999; *Thompson et al.*, 1999] are known over eastern North America and the North Atlantic, there has not been enough profile data to evaluate the O_3 budget. The statistics presented here show that persistent summertime stratospheric influences in the North American troposphere may be more prevalent than previously considered. This conclusion is based on the unique temporal and spatial coverage afforded by the IONS network, MOZAIC and the long-term sounding record at Wallops.

[46] **Acknowledgments.** We are grateful to chief scientist T. S. Bates, J. E. Johnson, D. Hamilton (NOAA/PMEL), and the crew of the R/V *Ronald H. Brown*. Students J. F. Liesch, V. Davis, M. Taylor, O. Hylton, and L. M. Zamora and the NASA/Goddard Directors Discretionary Fund provided the Beltsville soundings. Thanks to J. L. Moody and A. Klepac (University of Virginia) for the Pellston sondes. Useful comments were made by R. R. Dickerson (University of Maryland), K. E. Pickering (NASA/Goddard), and J. L. Moody. NASA's Tropospheric Chemistry Program, NOAA's CMDL and Aeronomy Labs, 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, is supported by NASA Aura Validation and Penn State's Meteorology Department. Thanks to C. A. Piety (University of Maryland) and M. P. McEvoy (Penn State) for EPA data and graphics assistance. All IONS data and images reside at the ICARTT/INTEX archive: http://cloud1.arc.nasa.gov.

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