1 Elevated Middle and Upper troposphere ozone observed downstream of Atlantic tropical
2 cyclones.

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Abstract

16 During the peak period of hurricane activity in the summer of 2010, vertical profiles of
17 ozone using ozonesondes were taken downstream of tropical cyclones in the Western and
18 Eastern Atlantic Ocean basin at Barbados and Cape Verde. Measurements are taken for
19 tropical cyclones Danielle, Earl, Fiona, Gaston, Julia and Igor. The measurements show
20 an increase in ozone mixing ratios with air originating from the tropical cyclones at 5-10
21 km altitude. We suggest that observed lightning activity associated tropical cyclones and
22 the subsequent production of NO_x followed by upper level outflow and subsidence ahead
23 of the tropical cyclones and aged continental outflow from West Africa thunderstorms
24 produced observed increases in ozone mixing ratios. Hurricane Danielle showed the
25 largest changes in ozone mixing ratio with values increasing from 25 ppb to 70 ppb
26 between 22 and 25 August in the middle troposphere, near 450 hPa; warming and drying
27 in the middle and lower troposphere. Measurements of ozone mixing ratios in Cape
28 Verde show higher ozone mixing ratios prior to the passage of tropical storm Julia but
29 low ozone mixing ratios and high relative humidity up to 300 hPa when the storm was in
30 close proximity. This is due most likely the vertically transported from the marine
31 boundary layer.

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1. Introduction

Continental landmasses in the tropics serve as the primary source of anthropogenic (biomass burning, industrial, transportation, cooking) or natural (lightning (LNO_x), biogenic soil NO_x) emissions of ozone precursors leading to the production of ozone in the troposphere. Tropical oceans in contrast serve as a significant sink of ozone in the marine boundary layer through surface deposition, chemical transformation or destruction (Read et al. 2008). Hence, low tropospheric column ozone (TCO) values are found over the tropical oceans, except for the Tropical Southern Atlantic, where ozone precursors from biomass burning, biogenic soil emissions and LNO_x and the resultant ozone from South America and Africa can be transported towards the tropical South Atlantic and undergo subsidence, producing the observed Wave-1 zonal pattern from September-November (Thompson et al. 2003; Ryu and Jenkins, 2005). Oceanic regions, such as the Tropical Pacific or Western Atlantic, that are distant from biomass burning or continental lightning sources have low observed TCO values and lower mid-tropospheric mixing ratios (Thompson et al. 2003; Ziemke et al 2011; Jenkins et al. 2013).

Increases in observed tropospheric ozone mixing ratios over the Northern Tropical Atlantic are due to either the long-range transport from continental areas or through the secondary production of ozone by lightning (LNO_x) from convection. Lightning is estimated to produce 2-8 Tg of nitrogen (N) annually (Martin et al., 2007) with some uncertainties related to the LNO_x production per flash for intra-cloud (IC) versus cloud to ground flashes (CG). Bucsela et al. (2010) show NO_x mixing ratio enhancement of 1.74-2.35 from lightning activity and 174 moles LNO_x per flash based on aircraft measurements of 4 oceanic convective systems during the Tropical

Composition, Cloud and Climate Coupling (TC⁴) experiment. Aircraft measurements over the continental US show increases in NO_x from lightning and model predicted downstream ozone production of 5-13 ppb O₃/day (Stith et al. 1999; DeCaria et al. 2005). In general, the Tropical Atlantic Ocean located downstream of lightning activity over continental Africa have been linked to elevated ozone mixing ratios based on observations (Jenkins and Ryu, 2004; Ryu and Jenkins 2005; Martin et al., 2007; Jenkins et al. 2008; Jenkins et al., 2012; Jenkins et al., 2013; Jenkins et al. 2014) and global chemical transport models (GCTMs) (Martin et al., 2007; Savaugue et al., 2008; Barret et al., 2010). Over the Southern Tropical Atlantic Ocean during its wet season, LNO_x production from convection over Central Africa and South America followed by subsidence may contribute to ozone production of 5-15 ppb O₃/day thereby elevating ozone mixing ratios in the middle troposphere (MT) during NH winter leading to the Tropical Atlantic Ozone Paradox (Thompson et al. 2000).

However, Thompson et al. (2003) using stations in the Southern Hemisphere Additional Ozonesondes (SHADOZ) network show that significant variations in ozone mixing ratios can occur on time-scales of 1-3 days indicating the strong influence of meteorological conditions on tropospheric ozone mixing ratios. In relationship to the Northern Tropical Atlantic, 3-5 day African Easterly Waves (AEWs) (Burpee, 1972) serve as a mid-tropospheric source of vorticity for tropical cyclones (TCs) and can be associated with convective activity and lightning prior to leaving the West African coast (Payne and McGarry, 1977; Jenkins et al. 2013). Jenkins et al. (2013) show that middle/upper tropospheric (MT/UT) ozone mixing ratios are higher by 5-10 ppb ahead of the AEW trough axis based on data from Dakar, Senegal. In addition, lower tropospheric

(LT) ozone mixing ratios are strongly altered by the Saharan Air Layer (SAL) at Dakar, Senegal and Sao Vicente, Cape Verde (Jenkins et al. 2012; Jenkins et al 2013a)

Upon exiting the West African coastline, ground based lightning observations show that AEWs, which develop into tropical cyclones (TCs) over the Eastern Atlantic Ocean are associated with lightning activity (Chronis et al. 2007). In the presence of Saharan dust, in which mineral dust aerosols can serve as ice nuclei, high lightning activity was found for Tropical Depression 4 (subsequently TS Debby) and Tropical Depression 8 (subsequently hurricane Helene) off the coast of West Africa during 2006 (Jenkins and Pratt, 2008).

Climatologically, the North Atlantic hurricane season (1 June - 30 November) has 11-12 named storms which includes: 6 hurricanes and 2-3 major hurricanes generated. During 2010, there were 19 named storms, 12 hurricanes and 5 becoming major hurricanes. The 2010 hurricane season ranked 10th in terms of Accumulated Cyclone Energy (ACE) corresponding to 190% of the 1950-2000 median (Bell et al. 2011). During the period of 14 August through 16 September there were six named tropical storms (TS) or hurricanes in the eastern or western Atlantic: Danielle (Category 4), Earl (Category 4), Fiona (TS), Gaston (TS), Igor (Category 5), Julia (Category 4). Lightning which is an indicator of convective intensity has been connected to hurricane activity with lightning activity upstream, as far away as eastern Africa, serving as a possible precursor for Atlantic TC activity (Price et al. 2007).

The use of ground-based lightning networks provides spatial/temporal lightning characteristics of Atlantic TCs. In addition, the relationship between lightning activity

and TC intensity changes has also been examined. Abarca et al. (2011) show the highest flash densities are found with tropical depressions (TDs) and TSs relative to hurricanes under non-intensifying and intensifying conditions; Demaria et al., (2012) show a similar result for the Atlantic and N. Pacific basins. The spatial distribution of lightning from ground and satellite based lightning detection systems shows high flash densities near the eyewall of the storm (0-40 km), and outer rainbands (> 200 km from the center), but low flash densities between the eyewall and outer rainbands (Molinari et al. 1999; Cecil and Zipser, 2002; Abarace et al., 2012; Demaria et al., 2012).

There is also evidence to support an increase in lightning frequency preceding the maximum intensity of TCs (Molinari et al. 1999; Price et al. 2009; DeMaria et al. 2012). However, Demaria et al. (2012) also show that there is an increase in lightning activity in rapidly decaying systems near the eyewall. While vertical wind shear tends to inhibit tropical cyclone development, there is evidence that the lightning densities in the inner core region are higher in sheared systems (Demaria et al. 2012); this relationship does not hold for the outer rainbands.

A vertical depiction of tropical cyclones suggests an uncertain picture for the horizontal and vertical distribution of ozone mixing ratios. Because the marine boundary layer (MBL) is the primary source of air for tropical cyclones, rising motions will distribute MBL air horizontally and vertically throughout the tropical cyclone. MBL air, however, contains very low ozone mixing ratios due to negligible sources of pollutants, surface deposition, abundant hydroxyl radicals and Halogen destruction (Read et al. 2008). Conversely, thunderstorm activity associated with TCs, will produce elevated LNO_x ratios in the MT/UT, where it is likely detrained into the mean easterly flow and

carried downstream away from the tropical cyclone, especially if wind shear is present; the photochemical production of ozone will follow during daytime hours. Consequently, the vertical distribution of ozone mixing ratios should be low throughout the tropical cyclone except downwind of lightning activity where higher mixing ratios would be found. During the peak of the 2010 hurricane season (August/September) low TCO values suggest that other sources of secondary ozone production (biomass burning in the Southern Hemisphere, soil emissions from West Africa) have a negligible impact on ozone over the Tropical North Atlantic (Jenkins et al., 2013).

Limited aircraft measurements of ozone mixing ratios for two strong hurricanes (Floyd and Georges) near and within the eye-wall show low ozone mixing ratios (< 20 ppb) below 4 km (Carsey and Willoughby, 2005). Minimal lightning activity was observed in Hurricanes Floyd and Georges. A second study of Super-Typhoon Mireille found the lowest ozone mixing ratios near the surface within the MBL, with higher ozone mixing ratios (< 50 ppb) at a flight level of 11.3 km above the eye (Newell et al. 1996). To the northeast of the center, higher concentrations of ozone and NO_x were found at 6-7 km altitudes potentially suggesting lightning and the production of LNO_x. Further, measured ozone and water vapor mixing ratios in the middle and upper troposphere were anti-correlated. The presence of sinking stratospheric air and high ozone mixing ratios were not found within the eye or other parts of the tropical cyclones in the aircraft measurements (Newell et al. 1996; Carsey and Willoughby, 2005).

During 2010, a US multi-agency field campaign was conducted from Florida and the Caribbean to examine the genesis and intensification processes of tropical cyclones (Montgomery et al. 2012; Braun et al. 2013; Rogers et al. 2013). In support of the

campaign ground measurements (thermodynamic, dynamic) were taken in Barbados with additional vertical profiles of ozone taken from mid-August through mid-September in the Western Atlantic at Barbados and the Eastern Atlantic at Cape Verde and Senegal (Jenkins et al. 2013). During the period, ozone measurements for 6 tropical cyclones (5 Western Atlantic, 1 Eastern Atlantic) are presented. The primary objective of this work is to show observations associated with outflow from the tropical cyclones as it relates to ozone enrichment in the MT. We also show that ozone mixing ratios are significantly reduced near the time of TC passage when moist environmental conditions (presumably MBL air) are found throughout the troposphere; which is consistent with aircraft measurements (Newell et al., 1999; Carsey and Willoughby, 2005).

2. METHODS

To investigate the variability of ozone mixing ratios associated with tropical cyclones in the western Atlantic a total of 16 ozonesondes are launched from Barbados (13.1°N, 59.1°W) during the period of 14 August through 16 September at 1800 UTC 2010. The ozone measurements targeted five tropical cyclones where Barbados was located downstream of these systems. The five systems are hurricane Danielle, hurricane Earl, tropical storm Fiona, tropical disturbance Gaston and hurricane Igor. The locations of tropical cyclones are determined from the National Hurricane Center (NHC) 6-hour “best track” analysis (Landsea and Franklin, 2013). In addition to the two radiosondes launched by the meteorological services, we launched 2 radiosondes each day increasing atmospheric measurements to 4 per day in Barbados. We also show ozone measurements related to the passage of tropical storm Julia near Sao Vicente, Cape Verde (16.4°N, 24.8°W). Ozonesondes at Barbados were prepared 3-7 days prior to their launch and

described in Jenkins et al. (2013). Ozone mixing ratio data for the case studies at the two sites are averaged over 100 meter layers from the surface through 150 hPa. We define the lower troposphere (LT) as the first 3 km (1000-700 hPa), the middle troposphere (MT) as 3-10 km (700-300 hPa) and the upper troposphere (UT) as 10-14.5 km (300-150 hPa). We also show a limited number of ozone measurements taken during the PREDICT field campaign aboard the G-V aircraft using the fast-O₃ instrument which has a range of 0-1000 ppb (Ridley et al. 1992; Montgomery et al. 2012).

Lightning locations and activity are determined from the Worldwide Lightning Location Network (WWLLN) using Very Low Frequency (VLF) signals (Virts et al., 2013). The WWLLN detection frequency is generally less than 10% over land when compared to the space-borne Lightning Imaging Sensor (LIS) because it detects primarily cloud to ground (CG) lightning. Rudlosky and Shea (2013) show that the detection efficiency is higher over the Tropical Atlantic, which may be due to less frequent but more intense CG lightning when compared to land-areas. Daily lightning activity related to the tropical disturbances is computed by considering all WWLLN lightning locations within 5° to the north, south, east or west of the tropical disturbance center identified by the NHC. The National Centers for Environmental Prediction (NCEP)/ Global Reanalysis data (Kalnay et al., 1996) are used for middle tropospheric (400 hPa) wind flow and daily Outgoing Longwave Radiation (OLR) values at 2.5° x 2.5° are used to identify deep convection for case studies (Liebmann and Smith, 1996). The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model is used to compute 3-day back trajectories (Draxler and Hess, 1988).

3. RESULTS

3.1 Western Atlantic Ozone Measurements

Figure 1a shows the location of tropical disturbances examined in this study, based on best track NHC locations, passing to the north of Barbados during the measurement period with Fiona and Gaston making the closest approach. We found 12 out of 16 ozone measurements influenced by tropical cyclones during the period of 14 August to 16 September. Each of these tropical disturbances are associated with significant numbers of lightning strikes (Figure 1b, Table 1) with hurricane Earl producing the greatest lightning activity as it intensified from a tropical storm on 28 August (day 2) to a hurricane on 29 August (day 3). The most intense TC of the season, Hurricane Igor, produced increasing numbers of lightning flashes as a major hurricane on 12 September (day 1) through 15 September (day 4) in Figure 1b. Tropical Storm Fiona and Tropical Low Gaston also produced significant amounts of lightning activity as the storms moved westward (Table 1).

Barbados, because of its relatively isolated location in the Western Atlantic is subject to low surface ozone mixing ratios (< 25 ppb) due to ozone destruction in the lower troposphere from Halogen and HO_x destruction, surface deposition and limited sources of surface emission (Figure 2) (Petrovavlovskikh, and Oltzman, 2012; Jenkins et al., 2013). However, Figure 3a shows that in general when tropical systems are approaching Barbados, there is an increase in ozone mixing ratios primarily in the MT (500-300 hPa) except for Hurricane Earl. Ozone mixing ratios in the LT (< 750 hPa), in contrast, show very little change in response to approaching tropical systems.

Increases in ozone mixing ratios are anti-correlated to relative humidity in the middle troposphere in association with air flowing from tropical disturbances, similar to earlier aircraft measurements. We observe a dry atmosphere, with relative humidity values less than 30% , and increases in ozone mixing ratios (Figure 3b). The mean wind directions at 6-10 km altitude have an easterly component with higher ozone mixing ratios, but in some cases, such as hurricane Igor where the size of the storm is very large, westerly winds also transported higher ozone mixing ratios into Barbados (Figure 3c). Next we provide a summary of ozone mixing ratio changes associated with each tropical disturbance in the Western Atlantic during 14 August- 16 September 2010.

3.11 Hurricane Danielle (23-27 August)

Figure 4a shows Hurricane Danielle located upstream of Barbados on 25 August 2010, with lightning locations primarily at outer rain-bands on the poleward side of the TC center where low OLR values are found (Figure 4a). Middle troposphere wind streamlines as depicted by NCEP Reanalysis show the arriving air at Barbados during the period of 23-25 August 2010 emerging from Hurricane Danielle. Figure 4b shows vertical profiles of measured ozone during the period of 22-27 August and also for non-tropical cyclone measurements during the period of 14 August - 16 September. Prior to Danielle approaching Barbados on 22 August, ozone-mixing ratios are less than 30 ppb in the MT (500-300 hPa). However, 2 days later on 24 August, we observed ozone mixing ratios approaching 50 ppb between 550 and 400 hPa. On 25 August, ozone mixing ratios increase to 70 ppb at 450 hPa, nearly 40 ppb higher than on 22 August. As, hurricane Danielle moves into higher latitudes, ozone-mixing ratios are reduced to less than 25 ppb in the MT on 27 August.

Figure 5a,b show the differences in relative humidity and ozone in the soundings for 22 August and 24 and 25 August when large increases in ozone mixing ratios are found. Large reductions in relative humidity of 30-50% are found in the 3000-9000 meter layer on the 24 and 25 of August relative to 22 August. In this same layer ozone mixing ratios increase by 20 ppb on 24 August and by more than 40 ppb on 25 August relative to 22 August. Figures 5c, d show that warming is also found at altitudes below 10,000 m with the warming pronounced near 9000 and 6000 meters on both days. Warming temperatures of 2-3° C are found relative to the 22 August. Much smaller changes (increase/decreases in) in relative humidity, ozone mixing ratios and temperature are found above 10 km with the most pronounced changes in the MT.

Figures 6 a, b show HYSPLIT 3-day back trajectories in the middle troposphere (5500, 6500 and 7500 meters) on 24 and 25 August at 1800 UTC. On both days the air arriving to Barbados had undergone subsidence as it is transported towards the southwest during its first two days. On 24 August, 3 day back trajectories originate to the northeast of Barbados in the range of 15-21° N and 35-40° W (Figure 6a). Additional analysis of 500 hPa streamline flow corresponding to 1800 UTC back trajectories shows that the flow arriving at Barbados on 24 August emerged from a trough at 500 hPa associated with TC Danielle beginning on 22 August (Supplemental Figure 1b). TC Danielle had a westward tilt with height and the 500 hPa trough is located to the west of the TC center by approximately 3 ° longitude and also extends poleward. Hence, air from TC Danielle in the MT was directed toward Barbados by the 500 hPa trough beginning on 22 August and was more pronounced on 23 August (Supplemental Figure 1c).

Back trajectories on 25 August show the source of air arriving at Barbados emerging from latitudes between 18 and 24° N and further westward (46-50° W), which is closer to the track of TC Danielle (Figure 6b). The back trajectories correspond closely to the position of the 500 hPa trough on 23 and 24 August (Supplemental Figures 2b, 2c) which is located westward of the TC center. Further, the back trajectories on 25 August, which have a more northerly position corresponds to the increased amplitude of the 500 hPa trough on 23 and 24 August (Supplemental Figures 2b, 2c). In summary, TC Danielle is the probable source of air arriving at Barbados 48 hours prior to measured higher MT ozone mixing ratios on 24 and 25 August.

3.12 Tropical Cyclone Earl (27-29 August)

Figure 3a shows only modest increases in ozone mixing ratios with the approach of TS Earl primarily on 29 August in the 350-250 hPa layer. Lightning associated with TS Earl increased by an order of magnitude between 28 and 29 August, but ozone-mixing ratios do not substantially increase. We attribute this to the vertical transport of humid, ozone poor air from the MBL along with limited northerly flow (except in the 300-250 hPa layer) from TC Earl on 29 August. High frequency radiosonde measurements depict winds with a northerly component prior to the 29 August 18Z ozonesonde measurement, when there is a shift to westerly and then southerly winds throughout the atmosphere (Figure 7a). Dry conditions are found between 500 and 300 hPa between 0000 UTC 27 August and 0000 29 August followed by moist conditions from the surface through 300 hPa on 29 August (Figure 7b). Dropsondes from the NASA DC-8 within hurricane Earl also show moist air (RH > 50%) from the surface to near 400 hPa north of Barbados on

the 29 August, consistent with moist conditions found from radiosondes at Barbados (Figures 8 a, b).

3.13 Tropical Storm Fiona

High lightning activity is associated with Tropical Storm Fiona as it is approaching Barbados (Table 1) on 31 August and associated elevated ozone mixing ratios, dry air and northeasterly winds at pressure levels less than 600 hPa on 31 August (Figures 3a, b) as reported in Jenkins et al. (2013) prior to its passage. The elevated ozone is suggestive of lightning and possible stratospheric ozone contributions. TS Fiona shows the highest ozone mixing ratios in the UT with moderate increases in ozone mixing ratio values in the MT. This is the only tropical cyclone that showed such a pattern suggesting a possible stratospheric contribution based on the ozone sounding on 31 August. The passage of TS Fiona on 1 September is associated with a large reduction in ozone mixing ratios, a significant moistening of the atmospheric column from the MLB and a shift in winds with a southerly component based on Figure 2a and from high frequency radiosondes (not shown).

Limited aircraft measurements from the PREDICT field campaign on 31 August show increasing O₃ mixing ratios (50 – 70 ppb) in the 6-10 km altitude range during the ascending and descending periods after takeoff and prior to landing in Barbados ahead of tropical storm Fiona (Figures 9a, 9b), which is consistent with vertical profiles of ozone mixing ratios from ozonesondes on 31 August (Figure 3a). During the flight on 31 August, the aircraft had the opportunity to fly in the vicinity of TS Fiona at a flight altitude of 13.8 km in the UT, when deep convection and lightning were present. Figures

10a-d show a significant increase in ozone mixing ratios at approximately 1300-1400 UTC with high ozone mixing ratios (~100-120 ppb) found along the southern flight path at 58° W, which was to the northeast and southwest of lightning activity on 31 August at approximately 1300-1400 UTC. Most of the lightning activity along 58° W occurred between 1130 and 1400 UTC on 31 August. Dropsondes between 1330 and 1410 UTC along 58° W show a dry atmosphere (RH < 20%) between 6-12 km (Figure 11a, 11b) ahead of TS Fiona, similar to the humidity measurements from Barbados on 31 August (Figure 3b)

3.14 Tropical Low Gaston (5-6 September)

Tropical Low Gaston, although north of Barbados, shows evidence of influencing the weather at Barbados on 6 September with winds from the northeast throughout much of the 800-300 hPa layer and drier air (Figure 3b) . Lightning activity increased from 5 through 7 September as TD Gaston moved into the Caribbean (Table 1). Ozone mixing ratios of 40-60 ppb are found in the MT (550-300 hPa layer) as shown in Figure 3a. High frequency radiosondes show a shift to somewhat drier conditions beginning by 0000 UTC 6 September with a shift in wind directions to the northeasterly winds relative to 5 September when winds are blowing from the east with a southerly component (Figure 3c). On 6 September, 1800 UTC at the time of the ozonesonde measurement, moist air is found near 450 hPa.

3.15 Hurricane Igor

In the case of major Hurricane Igor, the MT center as depicted by NCEP is located southeast of the surface center (Figure 12a). The wind trajectories arriving at

Barbados in the MT originate from Hurricane Igor; lightning activity was elevated but relatively constant over the ozone measurement period and was preferentially found in the outer rain-bands with Hurricane Igor. Figure 12b shows increasing elevated ozone mixing ratios on 12 and 13 September in the middle troposphere between 450 and 300 hPa, however by 14 September elevated ozone mixing ratios extend downward to 600 hPa at Barbados with the approach of Hurricane Igor. The ozone mixing ratio values are 20-30 ppb larger than the non-TC average. During the closest approach to Barbados on 14, 16 September, O₃ mixing ratios near 550 hPa increase by approximately 20 ppb compared to previous days.

Figures 13 a,b show the vertical profile differences in relative humidity and ozone for 12 September and 14 and 16 September when large increases in ozone mixing ratios are found. Large reductions in relative humidity of 20-40% are found below 9000 meters on 14 September relative to 12 September. Reductions in relative humidity in the 5000-9000 meter layer correspond to increases in ozone mixing ratios for 14 September. The largest reductions in relative humidity of 20-30% are found in the middle troposphere (4000-7500 m) and associated with increases in ozone mixing ratios up to 20 ppb.

Figures 13c, d show that warming (approximately 2° C) is also found throughout the troposphere when comparing 14 and 12 September. The largest warming of more than 3° C is found on 16 September at approximately 7000 m (Figure 13 d).

HYSPLIT 3-day back trajectories at 5500, 6500 and 7500 meters for 13 September and 16 September are shown in Figures 14 a, b. The origin of the air on 13 September is located between 15-20° N and extends eastward to near 43°W on 10 September. Additional analysis of 500 hPa streamline flow corresponding to the back

trajectories shows that the flow on the 13th emerges from a trough at 500 hPa associated with TC Igor beginning 10 September (Supplemental Figure 3a, b, c). Similar to TC Danielle, the 500 hPa trough was located west of the TC center by approximately 3-5 ° longitude and extends poleward. Thus, air from TC Igor in the MT is directed toward Barbados by the 500 hPa trough beginning on 10 August and continued through 12 September (Supplemental Figure 3c).

The HYSPLIT back trajectories show MT air on 16 September having a northerly (5500, 6500 meters) or westerly origin (at 7500 meters) in Figure 14b. The shift of winds from a northeasterly direction to a northerly or westerly direction is tied to the closed cyclonic circulation of 500 hPa flow on 14 September as TC Igor move towards the northwest (Supplemental Figures 4a-c). The back trajectories are most influenced by air that is within 2 days of Barbados because of the size of TC Igor. In summary, TC Igor is the source of air arriving at Barbados 48 hours prior to measured higher MT ozone mixing ratios on 13 and 16 September.

High frequency sounding measurements for Hurricane Igor show northeasterly winds throughout much of the troposphere until 14 September when westerly winds began to influence the lower and eventually most of the troposphere by 1800 UTC 16 September as wrap-around flow from this large system affected Barbados (Figure 15a), similar to the HYSPLIT trajectories (Figure 14 b). Low values of relative humidity (< 25 %) were initially limited to the 450-300 hPa levels on 11 September, but by 0000 UTC 14 September extended downward to 600 hPa (Figure 15b) and anti-correlated to increasing ozone mixing ratios in Figure 11b. The change in wind direction from easterly to westerly winds had no influence on the dry air associated with Hurricane Igor.

3.2 Eastern Atlantic Ozone Measurements

3.2.1 Tropical Storm Julia

Next, we show vertical profiles of ozone mixing ratios from Sao Vicente, Cape Verde (16.26° N, 24.8° W) associated with TS Julia, which passes south of the Cape Verde Islands. Deep convection and lightning are associated with the tropical disturbance and found over Senegal on 11 September and it passed near the Cape Verde Islands on 12 September (Figures 16a). On 13 September, deep convection is located over the Cape Verde Islands and the lightning activity is displaced northeast of Cape Verde, with MT flow coming from the Tropical Atlantic (Figure 16b). By 15 September major Hurricane Julia had moved northwest of the Cape Verde Islands with horizontal transport at 400 hPa from West Africa (Figure 16c). A new area of convection and lightning is found over Guinea Bissau and Southern Senegal on 15 September.

Figure 17a shows that prior to the storm impacting Cape Verde on 11 September, ozone mixing ratios of 40-55 ppb are found at pressure levels less than 500 hPa. However, on 13 September, ozone mixing ratios were nearly uniform with a maximum value of 30 ppb found between the surface and 200 hPa. This implies the vertical transport of ozone poor air from the MBL as TS Julia passed over Cape Verde. On 15 September, ozone mixing ratios in the middle troposphere increased some 15-20 ppb relative to 13 September. We suggest that the increase in ozone mixing ratios on 15 September are caused by aged enriched MT ozone air leaving West Africa where LNO_x serves as a source of elevated ozone mixing ratios (Jenkins et al. 2012). During the three days, ozone mixing ratios at pressure levels less than 500 hPa were negatively correlated to

relative humidity, with humid conditions and low ozone mixing ratios from the surface through 300 hPa found on 13 September (Figure 17b). Relative humidity values are generally less than 20% on 11 September and generally below 50 % on 15 September when higher ozone mixing ratios are observed. HYSPLIT 3-day back trajectories (Figure 15c) from 13 September at Sao Vicente show the source of air being West Africa in the MT (5500, 6500 and 7500 m). However, the trajectories show the air originating in the lower troposphere between 850-750 hPa and being lifted to the MT, with the largest increase in elevation occurring over the last 36 hours (after 12 Sept 0000 UTC). The trajectories show an abrupt turn towards the northwest in the last 24 hours as it enters into the MT, consistent with the southeasterly flow associated with the trough at 400 hPa in Figure 16b.

4. DISCUSSION AND CONCLUSION

During the summer season of 2010, the tropospheric ozone column is at its lowest value during the months of August and September over the Northern Tropical Atlantic, which is the peak of the Atlantic hurricane season (Jenkins et al. 2013). Limited aircraft studies have shown that MBL air limits ozone mixing ratios near the center of a tropical cyclone and there is no evidence of a stratospheric influence near the eye (Carsey and Willoughby, 2005; Newell et al. 1996). However, this does not rule out the possibility of stratospheric mixing in other parts of the tropical cyclone (i.e. outer rain-bands).

We found elevated ozone mixing ratios in the MT downstream of tropical disturbances, between 6-10 km, and low water vapor amounts in the Western and Eastern Tropical Atlantic (Barbados and Cape Verde). Conversely, with the passage or being in

close proximity to a tropical cyclone, we found low ozone mixing ratios and high water vapor content for Hurricane Earl, TS Fiona and TS Julia. A similar relationship has also been found by Newell et al. (1996) and Avery et al. (2010) show that ozone mixing ratios and condensed cloud water content are negatively correlated with oceanic deep convective systems (Avery et al. 2010). A similar pattern of anti-correlated relative humidity and ozone mixing ratios has been observed in the MT at Cape Verde (Jenkins et al. 2012), Senegal (Jenkins et al. 2014) and over the South Tropical Atlantic downstream of continental outflow from Africa (Thompson et al. 2000) and has been linked to aged continental air where there was significant amounts lightning activity.

The source of MT dry air and elevated ozone mixing ratios could have a stratospheric origin but we suggest that it is associated with subsidence related to the detrainment of LNO_x enriched air from convective cells near the eye-wall or outer rain-bands; Warming and drying patterns in the middle/lower troposphere were found with TC Danielle and Igor during their approach towards Barbados. TCs Danielle and Igor show a tilted circulation with height as the 500 hPa trough was located to the west of the TC center by 3 to 5 ° longitude and extended poleward. Jenkins et al. (2013) also show higher MT ozone mixing ratios ahead of the AEW trough at Dakar, Senegal due to LNO_x, which should persist downstream over the Atlantic Ocean (because of ozone's extended lifetime in the MT). We expect additional ozone production in addition to the continental contributions because of LNO_x generated within tropical disturbances.

We suggest that the air is detrained from the tropical cyclone convective zones and carried downstream of surface center by faster easterly winds aloft and MT trough which is located westward of the TC center. The enriched LNO_x air undergoes

subsidence, drying and warming and there is a production of ozone in cloud free regions as it moves westward, similar to processes over continental areas (DeCaria et al. 2005), thereby explaining the anti-correlation between elevated ozone mixing ratios and relative humidity. A similar process may occur over the South Atlantic Ocean (Thompson et al. 2000).

The ozone profiles downstream of tropical disturbances have a S-shape, suggestive of convective transport from the lower troposphere followed by detrainment (Folkins et al. 2002). This profile is found in the tropics and has been associated with biomass burning with convective transport being responsible (Reeves et al. 2010). Folkins et al. (2002), suggest the chemical production of ozone between 6-11 km as the primary source driving ozone mixing ratio tendencies relative to vertical advection and convective transport of ozone. The S-shaped profile has been observed in Senegal and Cape Verde with the observed ozone mixing ratio maximum in the 6-10 km altitude range; the MT peak has been linked to aged air from West African thunderstorm activity (Jenkins et al. 2012; Jenkins et al. 2014). Hence, given the lack of a surface source of ozone, we suggest that the chemical production of ozone via LNO_x from tropical disturbances or further upstream from West Africa to produce the S-shape.

We also found high lightning activity associated with weaker tropical disturbances Fiona and Gaston along with an increase in ozone mixing ratios, which is consistent with observations of Abarca et al. (2011) and Demaria et al., (2012). Because high lightning activity can occur at the outer rain bands (250 km from the center), LNO_x and the photochemical production of O_3 can occur well ahead of the tropical cyclone center, in addition to any LNO_x production near the eyewall. Cecil and Zipser (2002)

suggest that outer rain-bands likely contain more super-cooled water allowing for greater charge separation, thereby producing more lightning activity. Over the North Tropical Atlantic Ocean the production of LNO_x and O₃ may also be favored in the outer rain-bands where high concentrations of aerosols like Saharan dust would act as ice nuclei increasing lightning activity (Rosenfeld et al. 2012); as observed by Jenkins and Pratt (2008) off the West Africa coast in two developing tropical cyclones associated with Saharan air.

We also observed daily fluctuations in ozone mixing ratios that were associated with the tropical disturbances, which is consistent with SHADOZ observations that meteorological factors drive ozone-mixing ratios locally. These meteorological factors include the vertical transport of MBL poor air to the LT/MT, the horizontal transport of dry air from tropical disturbances and the secondary production of ozone from LNO_x. Large fluctuations of ozone mixing ratios, were found in the lower troposphere in association with (SAL) events, especially where large amounts of dust occurred (Jenkins et al. 2012; Jenkins et al. 2014).

There are 80-100 tropical cyclones and numerous weaker tropical disturbances (depressions) globally on an annual basis, which should lead to anomalous signals of increased ozone mixing ratios from the production of LNO_x downstream of tropical disturbances when lightning activity is present. But there is a considerable amount of variability in lightning activity with tropical cyclones and possibly for different ocean basins (Molinari et al. 1999; Demaria et al 2012). Any production of LNO_x by tropical cyclones should lead to an extended lifetime of NO_x by a factor of 5-10 in the MT/UT (Bucsela et al. 2010) enhancing the photochemical production of ozone. Further,

detrainment followed by subsidence and the drying of enriched NO_x air would limit losses through HNO₃ production. Future field campaign studying tropical cyclones should include NO_x and O₃ measurements to quantify the linkages between lightning, LNO_x, water vapor and ozone mixing ratios.

Acknowledgments

We thank the Caribbean Institute of Meteorology and Hydrology in Barbados and the Cape Verde Atmospheric Observatory in Sao Vicente Cape Verde for providing facilities for the research. This work was funded by National Science Foundation under the Grant number AGS# 1013179. Ozonesonde data for Barbados will be made available upon request to the corresponding author, while ozonesonde data for Cape Verde can be found at the center for environmental data archival (<http://badc.nerc.ac.uk/data/capeverde/>)

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618

619 **Figure Captions**

620 Figure 1 (a) Locations of tropical disturbances based on NHC best track data; (b)
621 lightning activity associated within a 1-5 day locations in (a) for the tropical disturbances.
622 Name of tropical disturbances denoted by the letters D (Danielle), E (Earl), F (Fiona), G
623 (Gaston), I (IGOR).

624 Figure 2. 14 August-16 September 2010, daily surface ozone measurements (ppb) at
625 Raged Point, Barbados.

626 Figure 3. (a) ozone mixing ratio; (b) relative humidity; (c) wind direction for all ozone
627 measurements from 14 August – 16 September at Barbados at 1800 UTC.

628 Figure 4 (a) lightning locations, 400 hPa streamlines and outgoing longwave radiation for
629 TC Danielle on 25 August 2010. Barbados located in Red; (b) measured ozone mixing
630 ratios for hurricane Danielle. Non tropical cyclone data is also included for reference.
631 Units of ozone in ppb.

632 Figure 5 (a) Relative humidity differences between 24 August and 22 August 2010; (b)
633 Relative humidity differences between 25 August and 22 August 2010; (c) temperature
634 differences between 24 August and 22 August 2010; (b) Temperature differences
635 between 25 August and 22 August 2010.

636 Figure 6. Three-day back trajectories from Barbados at altitudes 5500, 6500 and 7500
637 meters for (a) 1800 UTC 24 August 2010; (b) 1800 UTC 25 August 2010

638 Figure 7. High frequency soundings during 0000 UTC 27 August - 0000 UTC 30 August
639 at Barbados. (a) Meridional winds (m-s^{-1}); (b) Relative humidity (percent).

640 Figure 8. Relative humidity profiles from the NASA DC-8 dropsondes. (a) 1841 UTC ;
641 (b) 2001 UTC, 15.94° N, 59.2° W.

642 Figure 9. Ozone mixing ratio from during the (a) ascending and (b) descending parts of
643 the PREDICT G-V flight on 31 August 2010. Units are ppb.

644 Figure 10. (a) 31 August, 2010 PREDICT G-V Flight path around TS Fiona with 1200-
645 1700 UTC WWLLN lightning locations and 1400 UTC Outgoing longwave radiation
646 (OLR) W-m^{-2} ; (b) Leg 1 ozone mixing ratios; (c) Leg 2 ozone mixing ratios; (d) Leg 3
647 ozone mixing ratios (in ppb).

648 Figure 11. 31 August, 2010 PREDICT G-V relative humidity from dropsondes at: (a)
649 1334 UTC; (b) 1400 UTC. Units are percent

650 Figure 12. lightning locations, 400 hPa streamlines and outgoing longwave radiation for
651 hurricane Igor on 13 September 2010. Barbados located in Red; (b) measured ozone
652 mixing ratios for hurricane Igor. Non tropical cyclone data is also included for reference.
653 Units of ozone in ppb.

654 Figure 13 (a) Relative humidity differences between 24 August and 22 August 2010; (b)
655 Relative humidity differences between 25 August and 22 August 2010; (c) temperature

656 differences between 24 August and 22 August 2010; (b) Temperature differences
657 between 25 August and 22 August 2010

658 Figure 14. Three-day back trajectories from Barbados at altitudes 5500, 6500 and 7500
659 meters for (a) 1800 UTC 13 Sept. 2010; (b) 1800 UTC 16 Sept. 2010

660 Figure 15. High frequency soundings during 1200 UTC 11 September - 1200 UTC 17
661 September at Barbados. (a) Meridional winds (m-s^{-1}); (b) Relative humidity (percent).

662 Figure 16. Lightning locations, 400 hPa streamlines and outgoing longwave radiation
663 for TC Julia on (a) 11 September 2010; (b) 13 September; (c) 15 September.

664 Figure 17. Ozone mixing ratios for (a) 11, 13 and 15 September from Cape Verde. (b)
665 Relative Humidity for (a) 11, 13 and 15 September from Cape Verde. Three day back
666 trajectories from Sao Vicente at altitudes 5500, 6500 and 7500 meters for (a) 1200 UTC
667 13 Sept. 2010.

668 Supplemental Figures

669 Figure 1. 3-day back trajectories from Barbados at altitude 6500 m (blue circles) starting
670 on (a) 1800 UTC 24 August 2010 with 22 August 500 hPa streamline and hurricane
671 location on 22, 23, 24 August at 1800 UTC; (b) same as (a) but with 500 hPa streamlines
672 at 1800 UTC 23 August.

673

674 Figure 2. 3-day back trajectories from Barbados at altitude 6500 m (blue circles) starting
675 on (a) 1800 UTC 25 August 2010 with 23 August 500 hPa streamline and TC location
676 on 23, 24, 25 August at 1800 UTC; (b) same as (a) but with 500 hPa streamlines at 1800
677 UTC 24 August.

678

679 Figure 3. 3-day back trajectories from Barbados at altitude 6500 m (blue circles) starting
680 on (a) 1800 UTC 13 Sept. 2010 with 11 Sept. 500 hPa streamline and TC location on 11,
681 12, 13 Sept. at 1800 UTC; (b) same as (a) but with 500 hPa streamlines at 1800 UTC 12
682 Sept.

1

2 **Table 1.** NHC Best Track locations (Figure 1a), intensity and daily WWLLN lightning
3 activity (Figure 1b) within 5° north, south, east and west of the NHC center \ at 1800
4 UTC. Ozonesonde measurements are underlined and bold.

Name	Date	Latitude	Longitude	Cat	# WLLUN strikes
Danielle	23 Aug	15.2	40.9	1	716
	<u>24 Aug</u>	17.0	47.5	1	596
	<u>25 Aug</u>	20.7	52.6	1	2895
	26 Aug	24.8	56.4	2	3351
Earl	<u>27 Aug</u>	15.9	45.2	TS	172
	<u>28 Aug</u>	16.3	53.0	TS	1725
	<u>29 Aug</u>	17.3	59.3	1	17576
	30 Aug	19.0	64.2	4	15954
Fiona	30 Aug	14.9	47.7	TS	851
	<u>31 Aug</u>	16.3	56.9	TS	12102
	<u>1 Sept</u>	19.5	62.5	TS	10397
Gaston	4 Sept	16.5	46.6	T Low	1739
	<u>5 Sept</u>	17.0	51.2	T Low	2822
	<u>6 Sept</u>	17.5	55.5	T low	8757
	7 Sept	16.8	64.6	T low	11711
Igor	<u>12 Sept</u>	17.7	46.1	3	1345
	<u>13 Sept</u>	17.6	50.2	4	2721
	<u>14 Sept</u>	18.6	52.8	5	4868
	15 Sept	19.8	55.3	4	5238
	<u>16 Sept</u>	21.4	57.8	3	3834
Julia	12 Sept	13.1	21.3	TD	3806
	<u>13 Sept</u>	14.5	25.4	TS	2547
	14 Sept	15.9	29.2	1	217
	<u>15 Sept</u>	17.7	32.2	4	837

5

(a) TC Tracks August 20–16 September

(b) WLLUN Lightning locations within 10° lat, 10° of NHC TC position

NOAA Surface Ozone at Ragged point Barbados 14 Aug-Sept 16 2010

(a) Lightning locations, OLR and 400 hPa Streamlines 25 Aug

(b) Ozone Mixing ratio at Barbados (Hurricane Danielle)

(a) 72 hr Backtrajectory from 1800 UTC August 24, 2010

72 hr Backtrajectory from 1800 UTC August 25, 2010

(a)

Meridional Winds (V) Earl

(b)

Relative Humidity Earl

(a) 31 Aug. Lightning locations 12–17 UTC and 1400 UTC OLR (W/m²)

(b) PREDICT GV ozone mixing ratios at 57.907° W
Alt. 13.8 km (1324–1409 UTC) Leg 1

(c) PREDICT GV ozone mixing ratios at 13° N
Alt. 13.8 km (1410–1443 UTC) Leg 2

(d) PREDICT GV ozone mixing ratios at 53.6° W
Alt. 13.8 km (1443–1524 UTC) Leg 3

PREDICT Dropsonde 31 Aug. 1334 UTC Relative humidity

(b) PREDICT Dropsonde 31 Aug. 1409 UTC Relative Humidity

(a) Lightning locations, OLR and 400 hPa Streamlines 13 Sept

(b) Ozone Mixing ratio at Barbados (Hurricane Igor)

(a) 72 hr Backtrajectory from 1800 UTC 13 September, 2010

(a) Meridional Winds (V) Igor

(b) Relative Humidity Igor

(a) Lightning Locations, OLR (W/m^2) and 400 hPa Streamlines 11 Sept 2010

(b) Lightning Locations, OLR (W/m^2) and 400 hPa Streamlines 13 Sept 2010

(c) Lightning Locations, OLR (W/m^2) and 400 hPa Streamlines 15 Sept 2010

(c) **72 hr Backtrajectory from 1200 UTC Sept. 13 2010**

Key Highlights

- Ozone profiles are taken downstream of 5 tropical cyclones during the GRIP field campaigns in 2010.
- Elevated ozone mixing ratios are found in the mid-troposphere, with dry air and warming.
- Hurricanes Danielle and Igor show the largest increases in ozone mixing ratio.
- Lightning produced NO_x is suggested as the primary source of elevated ozone mixing ratios.
- ozone enriched air is transported ahead of the tropical cyclone in the mid-troposphere.

(a) 3-day back trajectories at 1800 UTC 24 Aug.,
& 1800 UTC 21 Aug. 500 hPa Streamlines

(b) 3-day back trajectories at 1800 UTC 24 August,
& 1800 UTC 22 August 500 hPa Streamlines

3-day back trajectories at 1800 UTC 24 Aug.,
& 1800 UTC 23 Aug. 500 hPa Streamlines

**(a) 3-day back trajectories at 1800 UTC 25 Aug.,
& 1800 UTC 22 Aug. 500 hPa Streamlines**

**(b) 3-day back trajectories at 1800 UTC 25 August,
& 1800 UTC 23 August 500 hPa Streamlines**

**(c) 3-day back trajectories at 1800 UTC 25 Aug.,
& 1800 UTC 24 Aug. 500 hPa Streamlines**

(a) 3-day back trajectories at 1800 UTC 13 Sep.,
& 1800 UTC 10 Sept. 500 hPa Streamlines

(b) 3-day back trajectories at 1800 UTC 13 Sept,
& 1800 UTC 11 Sept. 500 hPa Streamlines

(b) 3-day back trajectories at 1800 UTC 13 Sep.,
& 1800 UTC 12 Sept. 500 hPa Streamlines

(a) 3-day back trajectories at 1800 UTC 16 Sep.,
& 1800 UTC 13 Sept. 500 hPa Streamlines

(b) 3-day back trajectories at 1800 UTC 16 Sept,
& 1800 UTC 14 Sept. 500 hPa Streamlines

(c) 3-day back trajectories at 1800 UTC 16 Sept,
& 1800 UTC 15 Sept. 500 hPa Streamlines