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Multi-site tropospheric ozone measurements across the North Tropical Atlantic during the summer of 2010

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HIGHLIGHTS

- Higher tropospheric column ozone in the Eastern Atlantic relative to the Western Atlantic.
- Higher TCO values during (IOP I) rather than (IOP II) for Barbados and Cape Verde.
- Higher ozone mixing ratios are found in the 925–850 hPa layer in Cape Verde.
- Pre-trough conditions have higher ozone mixing ratios than post-trough conditions.

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ABSTRACT

Ozone soundings are launched during two Intensive Observing Periods (IOPs) from Dakar, Senegal; Sao Vicente, Cape Verde; and St James, Barbados to investigate ozone variability across the North Tropical Atlantic during June/July and August/September 2010. Two objectives of the campaign are to compare background tropospheric ozone mixing ratios and its variability associated with the Saharan Air Layer (SAL) and African Easterly Waves (AEWs) at sites located in the Eastern and Western Tropical Atlantic Ocean. During IOP1 (June–July), reduced ozone mixing ratios are found in the SAL with elevated ozone levels at the SAL's base and above it. During IOP I, the tropospheric column ozone (TCO) is higher at Cape Verde (27.5 DU) when compared to Barbados (19.8 DU). During IOP II (August–September) ozone-rich air is found above 500 hPa prior to the passage of AEW or developing tropical cyclones. The observed larger mixing ratios of middle/upper tropospheric ozone are most prominent at Dakar prior to the passage of an AEW, which we attribute to lightning NO_x. During IOP II the tropospheric column ozone is highest at Dakar (30.5 DU) when compared to Cape Verde (20.2 DU) and Barbados (17.2 DU).

1. Introduction

Understanding the distribution of tropospheric ozone is of key importance because it controls the oxidizing capacity of the atmosphere; and it is also a greenhouse gas (Logan and Kirchhoff, 1986; Fishman et al., 2008). At present, Anthropogenic tropospheric ozone contributes approximately 0.35 W m^{-2} of positive radiative forcing on a global basis (Parchauri and Reisinger, 2007). The spatial and temporal variability of tropospheric ozone at locations across the Northern Hemisphere (NH) Tropics Atlantic and Africa (land and ocean) has been investigated through satellite, ship, aircraft and ground measurements (Thompson et al., 2000; Jenkins and Ryu, 2004; Ryu and Jenkins, 2005; Jenkins et al., 2008; Real et al., 2010; Saunois et al., 2009; Nalli et al., 2011). Tropospheric measurements of ozone variability in Africa, north of the Equator, are more problematic because of sparse and irregular ozone measurements relative to

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the Southern Hemisphere Additional Ozonesondes (SHADOZ) network (Thompson et al., 2003).

The recent African Monsoon Multidisciplinary Analysis (AMMA) field campaign (Redelsperger et al., 2006) produced several years of regular ozonesonde measurements in Benin (Thouret et al., 2009) along with a number of focused aircraft and ground measurements in West Africa (Stewart et al., 2008; Reeves et al., 2010). The measurements during the AMMA field campaign in particular helped so shed light on processes associated with ozone variability in West Africa. There are three primary sources of tropospheric ozone emissions during NH summer: (a) lightning; (b) NO emissions from soils; (c) biomass burning in the Southern Hemisphere (SH) (Mari et al., 2008; Real et al., 2010). Lightning over West Africa can serve as a source of NO_x (LNO_x) during the NH summer season potentially leading to an enhancement of middle/upper troposphere ozone mixing ratios. Jenkins and Ryu (2004); Ryu and Jenkins (2005) and Jenkins et al. (2008) suggest from satellite and ozonesonde data that the upper troposphere ozone mixing ratios are enhanced during June, July and August (JJA) as a result of lightning. Barret et al. (2010) show that ozone mixing ratios over the Eastern Tropical Atlantic are enhanced by 10–20 ppb contributing up to 6 Dobson Units (DU) to the Tropospheric Column Ozone (TCO) during August 2006 in the upper troposphere based on 4 chemical transport models (CTMs) using parameterized lightning.

Biogenic sources of nitrogen, which are released from soils, especially at the beginning of the wet season can enhance lower tropospheric ozone mixing ratios. Summer season enhancements of NO_2 from satellite observations and modeling studies support the influence of soils as likely sources of lower troposphere ozone (Jaegle et al., 2004; van der et al., 2008; Williams et al., 2009). Aircraft measurements during AMMA show significantly higher ozone mixing ratios over recently wetted areas in the Sahel when compared to dry areas (Stewart et al., 2008). Jenkins et al. (2012a), suggest that biogenic sources of NO_x are responsible for enhanced ozone mixing ratios in the lower troposphere during the transition between the pre-monsoon and monsoon periods in Senegal during 2008. They also suggest that elevated ozone mixing ratios at the base of the Saharan Air Layer (SAL) are associated with biogenic sources of NO_x, which may have been emitted in the presence of moist atmospheric conditions. Cross-equatorial transport of ozone and its precursors from biomass burning in Central Africa (5–15° S) can also enhance ozone mixing ratios over West Africa, but especially in regions near the Gulf of Guinea. Ozone and its precursors can be vertically transported into convection where it can be lifted to the middle/upper troposphere before being transported westward (Sauvage et al., 2007a,b; Mari et al., 2008; Real et al., 2010).

Dry deposition of ozone on vegetation is the primary sink during NH summer season while dry and wet deposition of HNO_3 can limit NO and hence ozone mixing ratios. Over land, this process leads to a latitudinal gradient in ozone mixing ratios in West Africa (Saunois et al., 2009; Reeves et al., 2010) with surges in the monsoon flow associated with the wet season transporting ozone poor air poleward into the Sahelian region (Jenkins et al., 2012a). Hence, ozone mixing ratios in the planetary boundary layer should be relatively low because of ozone destruction in the absence of significant sources of CO or CH_4 from biomass burning or other anthropogenic sources of pollution, which may exist in urban areas of West Africa (Ancellet et al., 2011).

Over the ocean's marine boundary layer (MBL), there are several chemical sinks in addition to surface deposition that limit ozone mixing ratios under low NO_x concentration (<10 pptv): Photolysis, ozone-HO_x reactions and halogen destruction of ozone (Read et al., 2008). Measurements from the Eastern Atlantic at Cape Verde show ozone destruction up to 5 ppbv day^{-1} at the surface and throughout the MLB through 500 m (Read et al., 2008). In general, ozone destruction occurs through most of the year at Cape Verde under low NO_x mixing ratios (Lee et al., 2009). Some entrainment from the free troposphere and elevated NO_x concentrations from the African Continent can lead to the enhancement of ozone mixing ratios but these factors are smaller than those associated with ozone destruction (Lee et al., 2009).

Downstream over the Western Atlantic at Barbados within the MBL, low ozone mixing ratios (20–30 ppbv) are found throughout the year, with the lowest values (10–20 ppbv) found during the summer months (Oltmans and Levy, 1994). Savoie et al. (1992) show that low surface ozone mixing ratios at Barbados are often associated with air masses that originate from Africa, while elevated ozone mixing ratios are associated with air masses that originate from higher latitudes and from altitudes above the MBL. Hence ozone destruction likely occurs across the entire Tropical North Atlantic Ocean (TNAO) under low NO_x conditions and destructive chemical processes similar to measurements at Cape Verde (Read et al., 2008).

Three summer season weather features which occur on mesoscale or synoptic time scales can also influence tropospheric ozone: African Easterly Waves (AEWs); Mesoscale Convective Systems (MCSs) and; the Saharan Air layer (SAL). AEWs can indirectly serve as a potential source for tropospheric ozone because convection with lightning is often found ahead of the trough axis (Payne and McGarry, 1977). However, convective precipitation can limit background NO_x concentrations (via wet deposition of HNO_3), potentially offsetting LNO_x. The lack of tropospheric ozone measurements associated with AEWs has not clarified which process is dominant. Because of the 3–5 day frequency of AEWs (Burpee, 1972), tropospheric ozone mixing ratios could undergo regular variations. This pattern of tropospheric ozone variations can be determined by sampling air ahead of and behind the trough axis.

MCSs in West Africa are often associated with large concentrations of ice and lightning and have a distinct diurnal pattern with the highest frequency of MCSs found between 1800 and 0000 UTC (Mathon and Laurent, 2001; Cecil et al., 2005). Squall lines (Houze, 1977), which can be long-lived and travel long distances are often associated with lightning and the rear-inflow jet can transport mid-troposphere air into the lower troposphere, potentially enhancing ozone mixing ratios near the ground. Grant et al. (2008) observed increases in surface ozone mixing ratios due to downdrafts from MCSs in Senegal during the summer of 2006. However, the stratiform region of a squall line can serve as an ozone sink if low NO_x concentrations occur because of wet deposition with steady, lighter and longer duration rain when compared to the leading convective towers. Limited measurements of MCSs during August 2006 in association with AMMA suggest that the reduced upper troposphere ozone mixing ratios are associated with the vertical transport of ozone poor air from the boundary layer with some enhancement from LNO_x (Huntrieser et al., 2011).

Saharan air and dust can also influence tropospheric ozone mixing ratios in the lower to middle troposphere. The SAL is characterized by a stable and dry air and located between 2 and 6 km (Carlson and Prospero, 1972; Karyampudi et al., 1999; Leon et al., 2003, 2009). Studies show that dust outbreaks reduce ozone mixing ratios due to heterogeneous chemistry (Zhang et al., 1994; Bonasoni et al., 2004; De Reus et al., 2000, 2005). However, Jenkins et al. (2012a,b) show that while there is ozone depletion in the SAL, there is also an enhancement of ozone mixing ratios near the peak of the SAL's low-level inversion just above the marine MBL, which they attribute to biogenic sources of NO on dust aerosols, which may have been released in the presence of moist conditions.

The objective of this paper is to provide an overview of measured tropospheric ozone variability during the summer of 2010