8.5 SOUNDER OBSERVATIONS AND WRF-CHEM MODEL SIMULATIONS: IMPACT STUDY ON TROPOSPHERIC OZONE INCREASES OBSERVED DURING THE 2010 AEROSE CAMPAIGN

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1. INTRODUCTION

Ozone is a critical tropospheric oxidant and a greenhouse gas. There is abundant tropospheric ozone over the eastern Atlantic Ocean. This ozone abundance is a maximum in magnitude of tropospheric column ozone known as the "ozone wave one." It is part of a series of minima and maxima in tropospheric ozone that extend zonally across the Equator and meridionally across the tropical latitudes (30°S-30°N, Thompson et al. 2003; Sauvage et al. 2006; Ziemke et al. 2006). The mean May 2010 maximum is 30-35 Dobson Units (DU, Figure 1) over the Gulf of Guinea and off the coast of Equatorial Guinea. The Aerosols and Ocean Science Expeditions (AEROSE, Morris et al. 2006; Jenkins et al. 2008; Nalli et al. 2011) detected abundant tropospheric ozone from 2006-2013. This data acquisition platform is beneficial for accumulating ozone data. All of these data provide increasingly more dense spatial and temporal observations to the relatively data-sparse regions of the eastern Equatorial Atlantic Ocean and the African continent.

While many sources of tropospheric ozone exist (i.e. anthropogenic pollution, biogenic, tropopause folding, and high solar zenith angle), the biomass burning emissions produce ozone in considerable quantities (Pickering et al. 1996; Jenkins et al. 1997) and are intense and widespread over continental Africa (Jenkins et al.1997; Smith 2012; Smith et al. 2014). Specifically, for June 2006, the WRF-Chem model depicts ozone increases up to 33 ppbv in the lower troposphere (surface-700 hPa, LT), over the SH Atlantic Ocean downwind of the biomass burning in Central Africa.

This study will examine carbon monoxide (CO) as a biomass burning tracer. A tracer meaning that it is a smoke emissions constituent whose flow is highly correlated with the synoptic, mesoscale and convective flow. Although CO is an ozone precursor and is generated by combustion, the time-scale on which it produces ozone is longer than the two-week period of this study.

This primary goal of this study is to examine how much tropospheric ozone is produced in this "ozone wave one" maximum using the AEROSE ozonesonde data, NOAA-Unique sounder CO and ozone trace gas products from the Sounder. This study uses NOAA operational IR/MW sounder trace gas products because they provide synoptic scale context to their suite of meteorological and trace gas products. Moreover, sounders will be flown on the polar orbiting satellites (i.e. Met-Op A and NOAA-19, Hilton et al. 2013) for years to come. To study ozone generation over the eastern Equatorial Atlantic Ocean and examine the transport of CO, this study will use the high-resolution model WRF-Chem. The second goal of this study is to identify the similarities in the quantitative and spatial similarities in CO transport and ozone generation amongst the three products.

Section 2 describes the sounder data and the high resolution WRF-Chem Model parameter settings. Section 3 displays the sounder results and model simulation output. Section 4 is the discussion and Section 5 summarizes and provides future work.

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2. DATA AND MODEL FRAMEWORK

2.1 *WRF-Chem model parameters*

The WRF-Chem model (Version 3.5.1) is a three-dimensional, mesoscale, model where chemical trace gases and meteorology are coupled (Grell et al. 2005). The model parameters are set so that each model experiment has identical meteorology and biogenic and anthropogenic chemical emissions. The MOZART-4 global transport chemistry model (Emmons et al. 2010) output is embedded in the WRF-Chem structure, hence this study used it as a chemical mechanism. In addition to using MOZART-4 as a chemical mechanism, it is also used for chemical and lateral boundary conditions.

The meteorological initial and lateral boundary conditions are driven by the 1° x 1° 6-hourly Global Forecasting System Final Analyses (GFS-FNL, NCEP, 1999) output. This study uses the Lin Microphysics Scheme (Lin et al. 1983) and the Grell convective scheme (Grell and Devenyi, 2002). The WRF-Chem model simulations are run for the period: 0000 UTC 08 April to 0000 UTC 12 May 2010. The simulations have a 12 km resolution; our spatial domain is 20°S-20°N and 40°W-25°E, a model top pressure level of 50 hPa, and 30 vertical sigma levels.

Two WRF-Chem model simulations are run with the aforementioned parameters: 1) control and 2) biomass burning. The control simulation includes the MOZART-4 chemical mechanism and MEGAN biogenic emissions (Guenther et al. 2006). The biomass burning simulation adds Fire Inventory of NCAR (FINN, Wiedinmyer et al. 2011), a 1.0 km by 1.0 km global fire emission daily estimates model.

The available NOAA-Unique sounding product during the time period of the 2010 AEROSE 2010 was the IASI. The IASI spectral resolution enables retrieval of CO and ozone in the troposphere. The raw IASI profiles are processed to 100 tropospheric and stratospheric layers with two Equator-crossing

ascending and descending orbits per day. The swaths of these orbits span 2132 km zonally and meridionally extend to 15°S. At this southern latitude, there is not full spatial coverage over the Equatorial Atlantic Ocean and African continent. For IASI, CO is sensitive in the MT and ozone is sensitive in the UT.

3. RESULTS

3.1 *AEROSE ozonesonde measurements*

The 2010 AEROSE Campaign launched 19 ozonesondes, twelve of which were downwind of the biomass burning emissions and LNO_x along the ITCZ (Figure 1) which stretched along the Gulf of Guinea and West African coast. The first of the ozonesondes were launched on the 29 April 2010 and the last for this particular study period is 12 May 2010 (Figure 3). The first six of these ozonesonde launches occurred along the Equator, the next six were launched along 23°W and the 12 May ozonesonde was launched during the left turn of the NOAA R/V *Ronald H. Brown* Ship along 20°N. An analysis of all of the ozonesondes show a propensity for middle tropospheric ozone increases of 20-80 ppbv. Several ozonesonde launches depict an increase in ozone centered around 500 hPa. One of these cases occurred between 29 April-5 May 2010. Three of these cases occurred between 6 May and12 May 2010. There is one incidence of ozone mixing ratios exceeding 120 ppbv just below 500 hPa. With the exception of three incidents during the 29 April – 12 May 2010 of ozone mixing ratios above 70 ppbv, generally there are fewer ozone increases in the upper troposphere (UT) above 400 hPa (Figure 3).

3.2 *Satellite environmental data record observations*

The IASI CO mixing ratios at 639 hPa exceed 130 ppbv over the Sahel of Africa and this plume extends off of the coast of Guinea-Bissau (Figure 4). There are also high CO mixing ratios emerging from Central Africa, originating from the biomass burning in southern Democratic Republic of Congo (DRC) and sporadic biomass burning in Ghana, Nigeria, Cameroon, Central African Republic, Congo and remainder of the DRC. The IASI CO product detects CO mixing ratios lower than 100 ppbv over the Gulf of Guinea and the Equatorial Atlantic Ocean.

The IASI ozone mixing ratios (Figure 5) over the Sahel are in the 50-60 ppbv. Along the immediate Gulf of Guinea coast ozone mixing ratios are in the 30-40 ppbv range. These increase to 40-50 ppbv by the 10 May 2010. Ozone mixing ratios range from 40-50 ppbv over the SH Atlantic Ocean and appears to be originating from the east and southeast (Smith and Nalli, 2014).

3.3 Late April/early May WRF-Chem model results

The CO mixing ratios in the model (Figure 6) over the Sahel are in the 130-160 ppbv range which are similar IASI CO mixing ratios. The difference in the biomass burning simulation and the control run depicts 20-80 ppbv rise in CO off of the coast of Liberia and Guinea-Bissau throughout the 29 April-12 May 2010 period. These CO mixing ratio differences of 20-80 ppbv emerge from southern Democratic Republic of Congo (DRC), northern Cameroon, Nigeria, southern Mali, and Benin but still in the 20-80 ppbv range.

For ozone the biomass burning ozone mixing ratios (Figure 7) are similar to IASI at 273 hPa. Over the Sahel of Africa the ozone mixing ratios are greater 50-60 ppbv range. The model shows the ozone mixing ratios decreasing along the coast with the deep convection lofting lower ozone mixing ratios from the LT to the 273 hPa. In the model, the low ozone mixing ratios has a spatial bias over the Sahel as opposed to the Gulf of Guinea Coast.

The model depicts an increase of 20-80 ppbv in CO mixing ratios (Figure 8) when the control simulation CO mixing ratios are subtracted from those in the biomass burning simulation. These CO mixing ratio increases scattered over Nigeria and Cameroon in the form of periodic and distinct plumes of CO. The biomass burning simulation is showing what IASI depicts: there is a gradual rise in the ozone mixing ratios along the Gulf of Guinea Coasts. The increase in biomass burning ozone mixing ratio over the control simulations (Figure 9) shows that there is a significant 40-50 ppbv increase off shore of the Guinea-Bissau and Liberia in late April 2010. Then there is a widespread area of 10-20 ppbv ozone mixing ratio increase off of the coast of the West Africa and the Gulf of Guinea for much of the 29 April to 12 May 2010 period.

4. DISCUSSION

GTCM models have been used for atmospheric chemistry studies, but the high-resolution WRF-Chem model that we use in our study can resolve more meteorological features that affect the chemistry.

The higher mixing ratios of CO (see Figs. 6 and 8) that are in a westward plume extending from the Guinea-Bissau Coast and westward are likely the result of the concentrated biomass burning emissions there. These biomass burning emissions were lofted convectively to the middle troposphere (700-400 hPa, MT) where there is concentrated biomass burning smoke (Pickering et al.1996; 2015). Over Nigeria and areas further east along the Sahel there is scattered biomass burning. Some of the CO mixing ratio increases could be the result of pollution advected westward from the Middle East (Barret et al. 2010). Some of the CO from the Middle East pollution is likely interacting with the biomass burning emissions and causing the 40-80 ppbv increases.

The lower values in the LT are likely the result of wet deposition. Nitrogen dioxides are likely destroyed by the HNO_3 which lead to reduced ozone. The low ozone mixing ratios are convectively lofted to the UT. The lower values are then advected westward and likely influence the UT chemistry in the vicinity of 23°W. Hence, there is not as noticeable of an increase in

ozone in the UT as opposed to MT as detected by the ozonesondes (see Fig. 7).

The CO mixing ratios inside the WRF-Chem model (see Fig. 7) are similar in magnitude and encompass much of the NH of continental Africa. The CO takes the form of plumes that are the outflow of convection at 639 hPa. These plumes originated from the smoke at the surface, thus implying convective transport. The CO plumes (20-80 ppbv) intersect R/V *Ronald H. Brown* Ship track along the Equator and 23°W, suggesting that some of ozone increases could be the result of biomass burning in West Africa. In the model, the ozone generally increases 20-30 ppbv over the western two-thirds of the Gulf of Guinea (see Fig. 2). On 29 April the ozone increases were over 50 ppbv. These ozone mixing ratio increases geographically align with the May 2010 OMI Tropospheric Column Ozone is 30-35 DU and a relative maxima in tropospheric column ozone globally across the Equator (see Fig. 1, Ziemke et al. 2006). The cyclonic gyres north of the Equator direct the ozone precursors and ozone to this region where collects and becomes abundant over the two-week period. This broad ozone increase of 20-30 ppbv extends into the SH. Despite this not being the Northern Hemispheric summer season, a time in which biomass burning and convection are much more widespread and intense, ozone increases in late April/early May are similar in magnitude and location to the ozone increases found in the regional modeling studies of Bouarar et al. (2010) and Smith et al. (2014).

5. CONCLUSION AND FUTURE WORK

The IASI products showed ozone mixing ratios of 20-30 ppbv (see Fig. 5) over the western and equatorial portions of the African continent which generally matched the AEROSE ozonesondes at 273 hPa and aligned geographically with the May 2010 Tropospheric Column Ozone maximum. Nitrogen oxides ($NO + NO_2 = NO_x$) produce ozone rapidly, so it is likely that the ozone increases over the Gulf of Guinea aare a direct result of the NO_x generated by the biomass burning.

The WRF-Chem model ozone mixing ratio output at 275 hPa is also in the same range as the ozonesondes and the IASI product. Air containing low mixing ratios of ozone is convectively lofted to the UT. However, there is a northern spatial bias of the low ozone mixing ratio air. The CO mixing ratios are 130-150 ppbv over the Sahel and southern DRC and the model mixing ratios (see Fig. 4) align with the magnitude.

The 500 hPa pressure level is not analyzed in this study, but should be as a next step. The ozonesondes suggest significant ozone increases centered at this level. Information about the synoptic and mesoscale regimes at 500 hPa will be useful as well. In addition to viewing additional layers, we plan to compute back trajectories from the NOAA-Air Resources Laboratory-HYSPLIT model. This product will be a verification of the flow for the model and can provide information on the sources of these flows.

Data assimilation of the CO mixing ratios from the IASI product will be helpful for the community because of the synoptic/global scale coverage and sampling frequency (2/day for a single satellite). But data assimilation of satellite sounder trace gases is still a largely unexplored and underutilized use of sounder products. In the future, IASI offline model averaging kernels will be applied. However, the results presented here are sufficient because this study chose model pressure levels where IASI is sensitive.

Intertropical convergence zone convection is prevalent over the African continent throughout the year, hence lightning-induced nitrogen oxides (LNO_x) produce considerable ozone and are significant contribution to the ozone wave one maximum. The global LNO_x emissions estimates have been refined to a range of 3.0-6.3 Tg N yr^{-1}. But these estimates are based on global chemistry transport model studies (i.e. Schumann and Huntrieser, 2007; Murray et al. 2012; Stravruku et al. 2013, and Miyazaki et al 2014). High resolution, mesoscale models found ozone increases of 11-15 ppbv throughout the troposphere over the Equatorial Atlantic Ocean in both the Northern and Southern Hemispheres downwind of convection (Smith et al. 2015, in prep). Hence, further analysis with the finer scale models with subsequent AEROSE ozonesonde data is necessary (Smith et al. 2015, in prep).

The AEROSE ozonesonde data is beneficial for studying tropospheric ozone, but is also beneficial for validating the NOAA-Unique sounder products, including (Nalli et al. 2011; Hilton et al. 2012). Just as water vapor and temperature retrievals are valuable for the National Weather Service (NWS),National Centers for Environmental Prediction (NCEP), trace gas retrievals are critical for air quality forecasting and global climate change studies. The AEROSE dedicated ozonesonde data will be used for validation of the NOAA-unique trace gas products.

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7. ILLUSTRATIONS

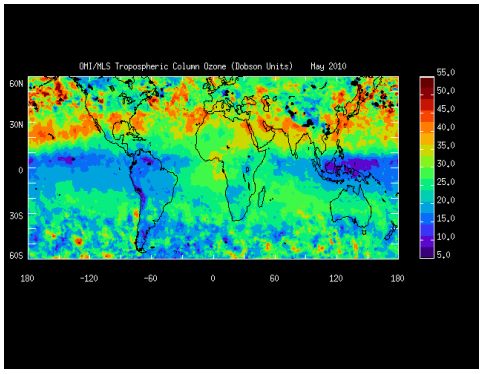

Figure 1. Global map of OMI/MLS Tropospheric Column Ozone (DU) averaged for the month of May 2010.

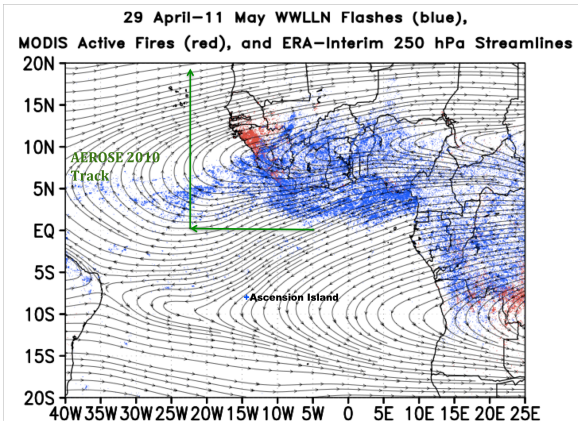

Figure 2. Map depicting the mean 250 hPa ERA-Interim streamlines for 29 April 2010 to 11 May 2010. The map includes fire locations (red dots) observed by MODIS, lightning flash locations (blue dots) from WWLLN, and the AEROSE 2010 track (green arrows) for the period of 29 April – 12 May 2010 and the domain of 40°W-25°E and 20°S-20°N.

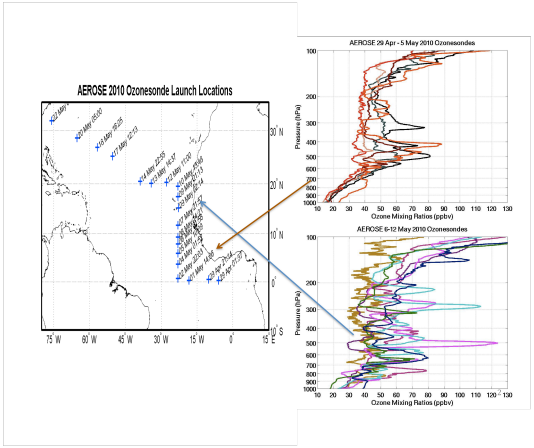

Figure 3. Depictions of a) locations ozonesonde launches during the AEROSE 2010 Campaign, and ozonesonde vertical profiles from the periods of b) 29 April – 5 May 2010 and c) 6–12 May 2010.

Figure 4. Animation of IASI Sounder CO mixing ratio (ppbv) granules at 639 hPa for the period of 29 April – 12 May 2010 and the domain of 40°W-25°E and 20°S-20°N. Found in Supplementary File 2.

Figure 5. Same as Figure 4 but for ozone mixing ratio (ppbv) at 273 hPa. Found in Supplementary File 3.

Figure 6. Animation of WRF-Chem biomass burning simulation of CO mixing ratio (ppbv) granules at 650 hPa for the period of 29 April – 12 May 2010 and the domain of 40°W-25°E and 20°S-20°N. Found in Supplementary File 4.

Figure 7. Same as Figure 6 but for ozone mixing ratio (ppbv) at 275 hPa. Found in Supplementary File 5.

Figure 8. Animation of IASI Sounder the CO mixing ratio (ppbv) increase of the WRF-Chem control simulation subtracted from the biomass burning simulation at 650 hPa for the period of 29 April – 12 May 2010 and the domain of 40°W-25°E and 20°S-20°N. Found in Supplementary File 6.

Figure 9. Same as Figure 7 but for ozone at 275 hPa. Found in Supplementary File 7.