Spring to summer northward migration of high O$_3$ over the western North Atlantic

Yunsoo Choi,1,2 Yuhang Wang,1 Qing Yang,1 Derek Cunnold,1 Tao Zeng,1 Changsub Shim,3 Ming Luo,3 Annmarie Eldering,3 Eric Bucsela,4 and James Gleason4

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Tropospheric O$_3$ columns retrieved from OMI and MLS measurements, NO$_2$ columns from OMI, and upper tropospheric O$_3$ concentrations from TES over North America and the western North Atlantic from April to August 2005 are analyzed using the Regional chEmical and trAnsport Model (REAM). Large enhancements of column and upper tropospheric O$_3$ over the western North Atlantic comparable to those over the eastern United States are found in the satellite measurements and REAM simulations. The O$_3$ enhancement region migrates northward from spring to summer. Model analysis indicates that the northward migration is driven by seasonal shifts of O$_3$ transported from the stratosphere and that produced through photochemistry from surface emissions and lightning NOx. As their uncertainties improve, satellite measurements of O$_3$ and its precursors will be able to provide more quantitative constraints on pollutant outflow from the continents.


1. Introduction

Many investigators have reported better agreement between the satellite measurements and the model results in tropical regions than at mid and high latitudes partly due to the difficulty in simulating cloud convection and lightning at mid latitudes [e.g., Chandra et al., 2004; Martin et al., 2006; Ziemke et al., 2006; Hudman et al., 2007]. Recent GEOS-CHEM studies have shown that lightning NOx production at higher latitudes in the standard global model needs to be increased by up to a factor of 4 [e.g., Li et al., 2005; Martin et al., 2006; Hudman et al., 2007]. Choi et al. [2005] demonstrated that the transient enhancements of lightning NOx and the convective transport of CO have large enough signals for satellites to detect.

In this work, we examine O$_3$ enhancements over the western North Atlantic during the transition from spring to summer in 2005 using measurements from recent satellite instruments, the Ozone Monitoring Instrument (OMI), the Microwave Limb Sounder (MLS), and the Tropospheric Emission Spectrometer (TES) onboard the NASA Aura satellite. These O$_3$ enhancements are direct evidence of continental outflow and the ability of models to simulate these features is necessary before applying the models to assess the impacts of pollutant emissions. We investigate how O$_3$ enhancements in the outflow region over the North Atlantic compare to the source region of its precursors and if the location of large O$_3$ outflow changes from spring to summer. Further, we apply the Regional chEmical and trAnsport Model (REAM) [Choi et al., 2005; also submitted manuscript, 2007] to investigate the contributions by surface pollutant emissions, lightning NO production, and transport from the stratosphere to the observed spring to summer migration of O$_3$ enhancements over the western North Atlantic.


The nadir horizontal resolutions of OMI, MLS, and TES are 13 × 24 km$^2$, 30 × 150 km$^2$, and 5 × 8 km$^2$, respectively. The tropospheric O$_3$ column is derived using a residual method by estimating the stratospheric O$_3$ column using MLS O$_3$ profiles above 215 hPa and subtracting that amount from OMI level 2 total column O$_3$ [Yang et al., 2007]. The mid-latitude stratospheric O$_3$ columns between 215 hPa and the tropopause are obtained by SAGE II mapping. The detailed validation of the OMI-MLS tropospheric column O$_3$ products was performed by Yang et al. [2007].

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1Department of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, Georgia, USA.
2Now at Jet Propulsion Laboratory, Pasadena, California, USA.
3Jet Propulsion Laboratory, Pasadena, California, USA.
4NASA GSFC, Greenbelt, Maryland, USA.

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[6] The retrieved tropospheric NO$_2$ columns from OMI measurements [Bosma et al., 2006] and their uncertainties are obtained from the NASA Goddard Earth Sciences Distributed Active Archive Center. We use only OMI tropospheric NO$_2$ column data with a cloud fraction of <20%. TES O$_3$ data are obtained from the NASA Langley Atmospheric Science Data Center. Only upper tropospheric TES O$_3$ retrieval data with degrees of freedom of >3.5 are used. REAM results for comparison are processed with TES retrieval averaging kernels for O$_3$ [Worden et al., 2007].

3. Model Description

[7] The REAM model was used previously by Choi et al. [2005; also submitted manuscript, 2007], Jing et al. [2006], and Wang et al. [2006] to simulate chemical transport over North America. We use the same setup here (with a horizontal resolution of 70 km). Spring and summer 2005 GEOS-CHEM global chemical transport model (version 7.2) simulations [Bey et al., 2001] are used to specify initial and boundary conditions for trace gases. Emission inventories for combustion and industrial sources and algorithms for biogenic sources are adopted from GEOS-CHEM, except for the fossil fuel NO$_X$ and CO emissions over the United States, which are taken from the 1999 US Environmental Agency National Emission Inventory (version 2). A production rate of 3.0 $\times$ 10$^{26}$ NO molecules per flash (K. Pickering, personal communication, 2005) is specified for both intracloud and cloud-to-ground flashes. More details of REAM set up and evaluations with surface, ozonesonde, aircraft, and satellite measurements are presented by Choi et al. (submitted manuscript, 2007).

4. Results and Discussion

4.1. OMI-MLS Tropospheric O$_3$

[8] Figure 1 shows the monthly mean tropospheric column O$_3$ derived from OMI-MLS measurements and the corresponding model simulations from April to August 2005. REAM-simulated tropospheric O$_3$ columns are 1–11% lower than those derived by OMI-MLS measurements, and the correlation coefficients improve from 0.59 to 0.77 from April to August. Simulated vertical O$_3$ profiles compare well with ozonesonde measurements except for the underestimations in the upper troposphere particularly in spring due to a low bias in the specified upper boundary condition (Choi et al., submitted manuscript, 2007). The influence of the upper boundary condition decreases in July and August because the vertical gradient of upper tropospheric O$_3$ decreases.

[9] The OMI-MLS columns indicate high O$_3$ over the eastern United States and the western North Atlantic. Over the continent, the activation of photochemistry from spring to summer, driven in part by increasing solar insolation, is evident as high O$_3$ concentrations move northward. Continental O$_3$ enhancements are overestimated in July and August in regions affected by lightning (C. Zhao, unpublished results, 2007), implying that its effect is overestimated in the model.

[10] Despite large surface emissions of O$_3$ precursors over North America, O$_3$ enhancements over the western North Atlantic are as high as they are over the eastern United States. In fact, O$_3$ column enhancements over the oceanic regions are higher than they are over land in April and May. As the season progresses from spring to summer, the high O$_3$ regions over the western North Atlantic move northward from the coast off Florida to the coast off New England.

[11] REAM captures the observed O$_3$ enhancements over the eastern United States and the western North Atlantic and the northward migration of high O$_3$ outflow over the oceanic region. To examine the reasons for the migration from spring to summer, we conduct sensitivity simulations. O$_3$ concentrations without lightning and surface NO$_X$ emissions are subtracted from the standard simulation to compute the effects of these sources. In a third sensitivity study, we first run tagged tracer simulations using GEOS-CHEM to compute the fractions of O$_3$ transported from the stratosphere. The Synoz method is used in GEOS-CHEM to simulate cross-tropopause transport [Bey et al., 2001]. We remove the stratospheric contributions in the REAM sensitivity simulation by using only the tropospheric portions of initial conditions and lateral and upper boundary conditions for O$_3$. We estimate the stratospheric contribution by subtracting O$_3$ in the third sensitivity simulation from the standard one. The results are shown in Figure 1. Over the United States, the contributions of surface NO$_X$ emissions, stratospheric transport, and lightning during the summer-time in this study are 41–52%, 31–36%, and 15–22%, respectively, comparable to those (45–50%, 35–40%, and 15–20%) by Chandra et al. [2004].

[12] In April and May, the largest contribution to high column O$_3$ is due to transport from the stratosphere. The resulting O$_3$ enhancements are over the southeastern United States, adjacent Gulf of Mexico, and western North Atlantic regions. Transport of O$_3$ from the stratosphere decreases from spring to summer as wave propagation from the troposphere to stratosphere weakens in summer [e.g., Wang et al., 1998, and references therein]. Photochemical contribution from surface NO$_X$ emissions is small in April but becomes comparable to the stratospheric contribution in May as photochemical activity increases with larger solar insolation and more abundant water vapor [e.g., Wang et al., 2003; Choi et al., submitted manuscript, 2007]. The contribution from lightning NO$_X$ remains small during this period in 2005.

[13] In June, July, and August, the stratospheric contribution becomes much smaller (>a factor of 2 decreases over the western North Atlantic). At 300 hPa, for example, stratospheric contributions decrease from 20–40% in April to <20% in June. The contribution from surface NO$_X$ emissions is similar to that in May and the outflow region of high O$_3$ moves toward the coast of the northeastern United States. The contributions from lightning NO$_X$ are much larger than in May. The outflow region of high O$_3$ produced from lightning NO$_X$ is similar to that of surface NO$_X$-produced O$_3$, reflecting increasing convective activities in summer.

[14] Yang et al. [2007] has conducted comprehensive comparisons of the OMI-MLS tropospheric O$_3$ columns with ozonesonde measurements. As an illustration, we show the comparison of OMI-MLS tropospheric O$_3$ columns with ozonesonde measurements at Wallops Island (38°N, 75°W),
Goosebay (53.3°N, 60.4°W), Kelowna (49.9°N, 119.4°W), and Yarmouth (43.9°N, 66.1°W) from April to August 2005 (Figure 2). We also show the corresponding REAM simulation results. Compared with ozonesondes, both OMI-MLS and REAM underestimate tropospheric O3 columns, with mean biases of \(-2.3\) and \(-4.5\) DU for OMI-MLS and REAM, respectively. The correlation coefficients are 0.78 and 0.82, respectively. The model-simulated tropospheric O3 column is lower than the OMI-MLS because REAM-simulated upper tropospheric O3 concentrations tend to be low. REAM bias is propagated from GEOS-CHEM through the specified upper boundary condition (Choi et al., submitted manuscript, 2007). The bias in the upper boundary condition likely leads to some underestimates of the stratospheric contribution to the tropospheric O3 column shown in Figure 1.

4.2. OMI Tropospheric Column NO2

[15] NOx is a major precursor for tropospheric O3 production. Figure 3 shows the average tropospheric NO2 columns retrieved from OMI and simulated by REAM in May–August 2005. Overall, the model results are in reasonably good agreement with OMI retrievals. Compared with the OMI tropospheric NO2 columns, REAM overestimates NO2 columns by \(~1 \times 10^{15}\) molecules cm\(^{-2}\) over

**Figure 1.** Monthly mean tropospheric O3 columns from April to August 2005 derived from OMI-MLS satellite measurements (first column), REAM simulations (second column), produced due to lightning NOx (third column), produced due to surface emissions, and transported from the stratosphere (last column).

**Figure 2.** Tropospheric O3 columns measured by ozonesondes, derived from OMI-MLS and simulated by REAM at Wallops Island, Goosebay, Kelowna, and Yarmouth from April to August 2005. Coincidence criteria for OMI-MLS-derived column ozone are within ±3° by longitude, ±2.5° by latitude on the same day as the ozonesonde measurements.
the Ohio Valley, where power plant NOx emissions are large. Frost et al. [2006] found that NOx emissions from power plants in 2004 were 50% of those in 1999 over the region. This reduction was not taken into account in our simulations. Model results indicate that lightning NO2 contributions are largest over the southern part of the US. Simulated direct outflow enhancements of NO2 are off the coast of northeastern US. REAM also simulates higher NO2 columns over the ocean than OMI. Compared to GOME [Choi et al., 2005; also submitted manuscript, 2007] and SCIAMACHY [Martin et al., 2006], the NO2 columns retrieved from OMI over the western North Atlantic are lower. The reason for this difference is unclear; detailed studies of OMI and SCIAMACHY NO2 retrievals are needed. Simulated O3 enhancements due to lightning (Figure 1) are downwind from the NOx enhancements because of time needed for photochemical production of O3.

4.3. TES UT O3 Measurements
[16] Due to insufficient spatial coverage of TES O3 measurements over North America from April to June 2005, we use only TES observations in July and August. TES O3 distributions are patchy due in part to measurement uncertainties. Figure 4 shows the comparison of TES retrieved and model simulated O3 mixing ratios at 325 hPa (300–350 hPa). Simulated mean REAM O3 concentrations are \( \sim 5\% \) lower. Comparisons with in situ measurements such as ozonesondes show that the UT TES O3 measurements tend to be overestimated [Worden et al., 2007]. TES O3 enhancements are over similar regions of tropospheric O3 column enhancements shown in Figure 1, although the details of the spatial patterns are different. Different sampling frequencies and more importantly the measurement uncertainties of TES are contributors to the difference. The correlation between TES measurements and REAM simulations (\( R \approx 0.5 \)) is lower than those for tropospheric O3 column due likely to more patchy measurements from TES. The large enhancements of TES O3 in southern Canada are not simulated in the model. More analysis is needed to confirm the validity of these enhancements. Simulated UT O3 enhancements due to lightning are \( \geq 20 \) ppbv over the southeastern United States and the western North Atlantic.
These enhancements are generally larger than the mean uncertainties of TES O₃ retrievals of 5–20 ppbv.

5. Conclusions

[17] Tropospheric O₃ columns by OMI and MLS, tropospheric NO₂ columns by OMI, and upper tropospheric O₃ by TES from April to August 2005 are analyzed with REAM. Large enhancements of column O₃ over the western North Atlantic, comparable to those over the eastern United States, are found in the OMI-MLS satellite measurements and are simulated by the model. Both OMI-MLS measurements and REAM simulations show a northward migration of high O₃ outflow region from spring to summer. Model results indicate that springtime outflow is mainly driven by transport from the stratosphere and O₃ production from surface emissions, while summertime outflow is mainly driven by O₃ production from surface and lightning NOₓ emissions. The northward migration of the outflow over the western North Atlantic primarily reflect the location change from the region with large O₃ transport from the stratosphere to the region with large production by lightning NOₓ. Upper tropospheric O₃ enhancements due to lightning NOₓ enhancements are analyzed using TES measurements. While qualitative agreement is found, quantitative analysis is hindered by measurement and simulation uncertainties. Reducing the uncertainties and improving the temporal coverage of satellite measurements are needed to further constrain the factors controlling the continental pollution outflow identified in the model study.

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References


E. Bucsela and J. Gleason, NASA GSFC, Greenbelt, MD 20771, USA. (yunsoo.choi@jpl.nasa.gov)