Spring to summer northward migration of high O₃ over the western North Atlantic

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Abstract

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

Major pathways for North American pollution outflow in the free troposphere include cloud convection [e.g., Dickerson et al., 1987; Pickering et al., 1988] and transport by the warm conveyor belt (WCB) [e.g., Cooper et al., 2002; Li et al., 2005]. The large impact of convection with associated lightning on O$_3$ and its precursors in the upper troposphere (UT) over North America are evident in the in situ measurements in two experiments: the SASS (Subsonic Assessment) Ozone and NO$_x$ Experiment (SONEX) [e.g., Singh et al., 1999] and the Intercontinental Transport Experiment-North America, Phase A (INTEX-A) [e.g., Singh et al., 2006]. The enhanced O$_3$ and its precursors in the UT can be quickly exported to the western North Atlantic by westerly
winds [Park et al., 2004; Choi et al., 2005, 2007]. Another mechanism, WCB, associated with mid-latitude cyclones, transfers air pollutants from the boundary layer to the free troposphere [e.g., Cooper et al., 2002; Li et al., 2005; Kiley et al., 2006]. The interplay between meso-scale cloud convection and synoptic-scale WCB strongly affects seasonal changes in the spatial distribution of pollutants [Hess, 2005].

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., Edwards et al., 2003; 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. Jing et al. [2006] showed that model-simulated transient tropospheric O$_3$ column changes are in general agreement with OMI-MLS-derived tropospheric O$_3$ column observations. In general, pollution enhancements due to convection and lightning are easier to detect over the ocean than over land at mid latitudes [e.g., Li et al., 2005; Choi et al., 2005, 2007].

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. 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 O₃ outflow changes from spring to summer. To further analyze the satellite measurements, we apply the Regional chEmical and trAnsport Model (REAM) [Choi et al., 2005, 2007]. We conduct sensitivity analyses to investigate the contributions by surface pollutant emissions, lightning NO production, and transport from the stratosphere to the observed spring to summer migration of O₃ enhancements over the western North Atlantic.

2. Satellite Measurements: OMI-MLS Tropospheric Column O₃, OMI Tropospheric Column NO₂, and TES O₃

OMI, MLS, and TES are onboard the NASA Aura satellite, which passes over the equator at 1:45 PM local time. The nadir horizontal resolutions of OMI, MLS, and TES are 13 × 24 km², 30 × 150 km², and 5 × 8 km², respectively. The tropospheric O₃ column is derived using a residual method by estimating the stratospheric O₃ column using MLS O₃ profiles above 215 hPa and subtracting that amount from OMI level 2 total column O₃ [Yang et al., 2007]. The MLS data (version 1.5) are used for the derivation of OMI-MLS tropospheric O₃ columns. The mid-latitude stratospheric O₃ columns between 215 hPa and the tropopause are obtained by SAGE II mapping; the altitudes of the tropopause are taken from the NCEP reanalysis data [Yang et al., 2007]. The coincidence criteria for MLS stratospheric columns with OMI total column measurements are ±1.25° longitude by ±1.25° latitude on the same day. The detailed validation of the OMI-MLS tropospheric column O₃ products was performed by Yang et al. [2007].

The retrieved tropospheric NO₂ columns from OMI measurements [Bucsela et al., 2006] and their uncertainties are obtained from the NASA Goddard Earth Sciences Distributed Active Archive Center (GES DAAC). We use only OMI tropospheric NO₂
column data with a cloud fraction of < 40%. TES O$_3$ data are obtained from the NASA Langeley Atmospheric Science Data Center (ASDC). Only upper tropospheric TES O$_3$ retrieval data with degrees of freedom of > 3.5 are used. When compared with the results of the observations, the REAM results are processed with TES retrieval averaging kernels for O$_3$ [Worden et al., 2007].

3. Model Description

The REAM model was used previously by Choi et al. [2005, 2007], Jing et al. [2006], Wang et al. [2006], and Zeng et al. [2003, 2006] to simulate chemical transport over North America. We use the same setup here. The model has a horizontal resolution of 70 km, with 23 vertical layers reaching 10 hPa. The National Center for Atmospheric Research/Penn State MM5 is used to simulate meteorological fields using four-dimensional data assimilation (FDDA) with the National Center for Environmental Prediction reanalysis, surface, and rawinsonde observations. Most meteorological variables are archived every 30 minutes. The variables related to convection and lightning are archived every 2.5 minutes. 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. Regional simulations were spun up during the last two weeks of March 2005. 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 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. [2007].

4. Results and Discussion

4.1 OMI-MLS Tropospheric O$_3$

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. The OMI-MLS columns indicate high O$_3$ over the eastern United States and the western North Atlantic. 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.

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 NOx 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 by McLinden et al. [2000] 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.

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 and 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 NOx 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., 2007]. The contribution from lightning NOx remains small during this period in 2005.

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 NOx 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 NOx are much larger than in May. The outflow region of high O$_3$ produced from lightning NOx is similar to that of surface NOx-produced O$_3$, reflecting increasing convective activities in summer.
REAM-simulated tropospheric O$_3$ columns are 1-11% lower than those derived by OMI-MLS measurements, and correlation coefficients increase from 0.59 to 0.77 from April to August. 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) 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 O$_3$ columns, with mean biases of -3 and -4 DU for OMI-MLS and REAM, respectively. The correlation coefficients are 0.81 and 0.82, respectively. The more extensive evaluation of 8 ozonesonde stations by Yang et al. [2007] shows a low bias (~4 DU) at 35-60°N in the summer. The model-simulated tropospheric O$_3$ column is not higher than the OMI-MLS because REAM-simulated upper tropospheric O$_3$ concentrations tend to be low. REAM bias is propagated from GEOS-CHEM through the specified upper boundary condition [Choi et al., 2007]. The bias in the upper boundary condition likely leads to some underestimates of the stratospheric contribution to the tropospheric O$_3$ column shown in Figure 1.

4.2 OMI Tropospheric Column NO$_2$

NOx is a major precursor for tropospheric O$_3$ production. Figure 3 shows the average tropospheric NO$_2$ columns retrieved from OMI and simulated by REAM in the summer of 2005. Overall, the model results are in reasonably good agreement with OMI retrievals. Compared with the OMI tropospheric NO$_2$ columns, REAM overestimates NO$_2$ columns by $\sim 1 \times 10^{15}$ molecules cm$^{-2}$ over the Ohio Valley, where power plant NO$_x$...
emissions are large. Frost et al. [2006] and Kim et al. [2006] found that NO_x 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 NO_2 contributions are largest over the southern part of the US, although lightning contribution is overestimated over northern Mexico. Simulated direct outflow enhancements of NO_2 are off the coast of northeastern US. REAM also simulates higher NO_2 columns over the ocean than OMI. Compared to GOME [Choi et al., 2005, 2007] and SCIAMACHY [Martin et al., 2006], the NO_2 columns retrieved from OMI over the western North Atlantic are lower. The reason for this difference is unclear. Simulated O_3 enhancements due to lightning (Fig. 1) are downwind from the NO_x enhancements because of time needed for photochemical production of O_3.

4.3 TES UT O_3 Measurements

Due to insufficient spatial coverage of TES O_3 measurements over North America from April to June 2005, we use only TES observations in July and August. TES O_3 distributions are patchy due in part to measurement uncertainties. Figure 4 shows the comparison of TES retrieved and model simulated O_3 mixing ratios at 325 hPa (300-350 hPa). Simulated mean REAM O_3 concentrations are ~5% lower. Comparisons with in situ measurements such as ozonesondes show that the UT TES O_3 measurements tend to be overestimated [Worden et al., 2007]. TES O_3 enhancements are over similar regions of tropospheric O_3 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 is lower than those for tropospheric O₃ column (R≅0.5) due likely to more patchy measurements from TES. Simulated UT O₃ enhancements due to lightning are larger than 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, which range from 5 to 20 ppbv.

5. Conclusions

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 uncertainties and sampling coverage. Improvements in satellite measurements are needed to further constrain the factors controlling the continental pollution outflow identified in the model study.
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References


Figure 1. Monthly mean tropospheric O$_3$ 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 O$_3$ columns measured by ozonesondes, derived from OMI-MLS and simulated by REAM at Wallops Island from April to August 2005. Coincidence criteria for OMI-MLS-derived column ozone are at ±3° by longitude, ±2.5° by latitude on the same day as the ozonesonde measurements. Ozonesonde data are obtained from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC).
Figure 3. Mean tropospheric NO$_2$ columns during summer 2005 from OMI satellite measurements (first column), those from REAM simulations (second column), and those contributed by lightning production in the model (third column). Only OMI data with cloud fractions of < 40% are used. Corresponding model results are taken at the same time and locations as the OMI measurements. Lightning contribution is estimated by subtracting simulated NO$_2$ without lightning production from the standard simulation.

Figure 4. Mean tropospheric O$_3$ mixing ratios at 325 hPa (300-350 hPa) for July and August 2005 from the TES satellite measurements (first column), those from REAM (second column), and REAM estimated lightning contribution (third column). 27 global survey and 4 special observation products are used. Lightning contribution is estimated by subtracting simulated O$_3$ without lightning NO$_x$ from the standard simulation. Only TES O$_3$ data with a degree of freedom of > 3.5 are used. Corresponding model results are sampled at the same time and locations as the TES measurements. The model results shown here have been processed using TES O$_3$ retrieval averaging kernel.