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Application of satellite data in a regional model to improve long-term ozone simulations

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Abstract To investigate an alternative technique of providing background and transboundary transport inputs for ozone (O_3) simulations on a regional scale, the EPA's Community Multi-scale Air Quality (CMAQ) model was integrated with high spectral resolution data from the Tropospheric Emission Spectrometer (TES) aboard the NASA's Aura satellite. This study presents a comprehensive model evaluation of O_3 for the entire year of 2009 over the contiguous United States with a focus on the State of Texas using both ozonesonde and ground measurements. While improving model performance in the upper atmosphere, CMAQ's initial and boundary conditions (IC/BC) derived from the original TES data do not improve model performance in the troposphere because the satellite data exaggerated concentration of tropospheric O_3. With a 10-ppb deduction of O_3 concentration from TES, the IC/BC derived from the adjusted TES improves model performance from ground level through the upper atmosphere. The mean bias of daily maximum 8-h average concentration of O_3 (MDA8) from the ground monitored in Texas decreased from 7 ppb to 4 ppb. Model results also show small influences of O_3 from the upper troposphere on the concentrations at the ground level. With a complete exclusion of stratospheric layers, changes of annual mean MDA8 of O_3 concentrations at ground-level were smaller than 1.1 % in Dallas and Houston. In addition, limitations of satellite data are discussed and recommendations are provided regarding the future application of satellite data in regional O_3 simulations.

Keywords CMAQ · Initial and boundary conditions · Ozonesonde · Satellite data · Tropospheric Ozone

1 Introduction

Ozone (O_3) is one of six criteria air pollutants listed in the National Ambient Air Quality Standards (NAAQS) set by the United States Environmental Protection Agency (EPA). It causes a variety of chronic problems to human health and the environment even in low concentrations (Brunekreef and Holgate 2002; Chappelka and Samuelson 1998; Brook et al. 2004; Lippmann 1989; Fuhrer et al. 1997). Interactions of solar radiation with nitric oxide

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and nitrogen dioxide (NO_x) and volatile organic compounds (VOCs) emitted from anthropogenic and natural sources promote ground-level O_3 formation. In the United States, the O_3 standard has been revised many times since it was first established in 1971. The primary standard was designed to protect public health, while the secondary standard was designed to protect the environment. The EPA has been using the same values for both primary and secondary standards. In 1997, EPA replaced 1979 O_3 standard for 1-h average of 0.12 ppm (ppm) with an 8-h standard set at 0.08 ppm. The 8-h concentration is calculated from a 3-year average of annual fourth-highest of daily maximum 8-h average (MDA8). In March 2008, the EPA tightened the 8-h standard to 0.075 ppm (or 75 parts per billion, ppb). The current NAAQS for O_3 is being reviewed and the limit is expected to be further lowered to 0.06–0.07 ppm in the near future. The impending primary NAAQS is rather comparable with the existing O_3 guideline set by the World Health Organization (WHO). The WHO suggests a guideline for public health protection using MDA8 at 100 μg m^{-3} (0.05 ppm), and the exposure to this level of O_3 is associated with an estimated 1–2 % increase in daily mortality (Krzyzanowski and Cohen 2008). The EPA is also considering of introducing a new cumulative, seasonal secondary standard in a range of 7 to 15 ppm-hours to provide better protection against O_3-related impacts on agriculture, plants, and sensitive ecosystems. Not only will the new standards increase the burden on emission control under the State Implementation Plans (SIPs), it will also broaden O_3 nonattainment areas which are determined using the concentration measured by a ground monitoring network. For the EPA, an attainment area is justified by 3-year average of the fourth-highest MDA8 O_3 concentrations measured at each monitor within an area over each year and the average must not exceed the standard. Based on the 2008 standard at 75 ppb, there are 39 out of 73 (53 %) regulatory Continuous Ambient Monitoring Stations (CAMS) in the State of Texas that exceed the 8-h O_3 standard for a 3-year period of 2010, 2011, and 2012. In this same period, changing the standard will have a drastic impact on the number of nonattainment areas. The number of violating CAMS stations would leap to 57 out of 73 (78 %) if the NAAQS limit was tightened to 70 ppb (only a 5-ppb difference). Under the WHO guideline, the nonattainment areas would cover most part of Texas, if not entirely. When the new EPA standards become effective, the problem of nonattainment will escalate from a local problem (several neighboring counties) to a regional problem. The Texas nonattainment problem and various O_3 standards can be illustrated as in Fig. 1. Even though the recent economic recession might have contributed to a better air quality, the concentrations in many Texas metropolitan areas still far exceed the impending NAAQS for O_3.

Air quality modeling is one of major components in preparing the SIP to rationalize how Texas will attain or maintain NAAQS. To assist policy makers and researchers, the EPA developed the Community Multi-scale Air Quality (CMAQ) modeling system (Byun and Schere 2006; Foley et al. 2010) to help establish air pollution control measures and to study atmospheric sciences. The protection of human health and the environment, as well as cost effectiveness of the SIP depends on accuracy of the model. CMAQ simulations require initial conditions (IC) and boundary conditions (BC). IC data provide chemical species representing the background concentrations of the modeling domain at the beginning of the simulation, while BC data are needed to provide concentrations of chemical species at the lateral boundaries of the modeling domain representing transboundary transport. The standard IC/BC use time-invariant profiles that do not correspond to long-range transport dynamics, and do not efficiently reflect current conditions (especially during the recent economic recession as illustrated in Fig. 1 for O_3). In addition, the standard O_3 profile usually understates O_3 in upper troposphere and lower stratosphere in order to counteract excessive vertical mixing in previous CMAQ versions. It is customary to substitute the

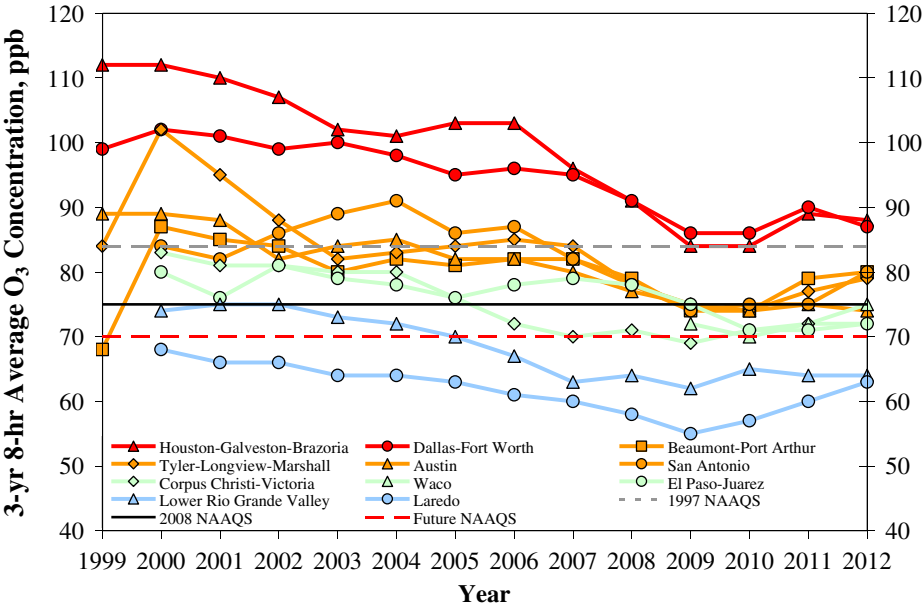

Fig. 1 Average ozone (O_3) concentrations represent air quality of the areas in Texas and various 8-h O_3 standards. Data used to make this figure are available at http://www.tceq.state.tx.us/cgi-bin/compliance/monops/8hr_attainment.pl

standard IC/BC with dynamic IC/BC derived from concentration results of global models such as GEOS-Chem (Bey et al. 2001) and MOZART (Model for OZone And Related chemical Tracers) (Emmons et al. 2010). However, use of global models to provide IC/BC in CMAQ over the United States domains has shown mixed results in previous studies. Appel et al. (2007) reported that their CMAQ simulation with BC derived from GEOS-Chem in July 2001 improved model performance over the simulation with the standard profile for ground O_3 predictions. On the contrary, Hogrefe et al. (2011) shown that model O_3 results from CMAQ with time-invariant static profile were more comparable to observed concentrations than the results from CMAQ with BC derived from MOZART which overestimated concentrations of O_3 and its precursors, while Tong and Mauzerall (2006) concluded that their CMAQ simulation during summer of 1996 with BC derived from MOZART underestimated O_3 in the upper troposphere and it had a negligible effect on O_3 near ground level when compared with the simulation with the standard BC. There are also some issues regarding the use of global models. First, simulation results from different global models can be very different. For example, Tang et al. (2007) reported that differences in mean CO concentrations in the summer of 2004 among BC derived from three different global models were as large as 40 ppb, and the effects of the BC on CO were significant at surface and throughout the troposphere. They also demonstrated that regional simulations with the different global models were sensitive to the BC especially for CO (carbon monoxide) and O_3. Second, some global models (e.g. MOZART) do not have a complete stratospheric chemistry. Many atmospheric species including CO and O_3 are constrained in the stratosphere with data from model and measurement, so it is usually inappropriate for studies of the dynamics and detailed structure of the upper troposphere and stratosphere (Emmons et al. 2010).

This paper presents an alternative method of using remote sensing data to create IC/BC for CMAQ simulations in order to improve model performance of O_3 concentrations from ground-level through the stratosphere. The use of remote sensing (especially satellite) in air quality research has

progressed considerably over the last decade. Three major applications of satellite data in air quality modeling include estimates of emissions, analyses and forecasts of events that affect air quality (transport of air pollutants), and inference of air quality (Martin 2008). For this study, the applications of satellite data are extended by using the data from the Tropospheric Emission Spectrometer (TES) (Beer et al. 2001) to provide IC/BC of O_3 and CO for CMAQ. The TES is an imaging infrared Fourier-transform spectrometer aboard the Aura satellite from the National Aeronautics and Space Administration (NASA) for studying air quality and climate. The TES was chosen because of its ability to generate vertical profiles of the atmosphere from the ground surface to the stratosphere. The importance of IC and BC in chemical transport models have been addressed in numerous studies (Pongprueksa et al. 2008; Jimenez et al. 2007; Ritter et al. 2013; Samaali et al. 2009; Hogrefe et al. 2011; Tang et al. 2009). However, these previous studies used time-invariant or model-derived (usually from global models) profiles for IC and BC which may be different from the actual ones. A few recent studies have incorporated satellite data into their IC and BC of CMAQ and those results have shown some model improvements over the standard or default IC and BC (Pour-Biazar et al. 2011; Shi et al. 2012; Shi et al. 2010), but their simulation episodes only last for days or weeks. The short-term simulations in previous studies might differ from a longer simulation, which the influence of IC can be disregarded while the influence of BC can be effectively evaluated. For this study, a comprehensive model evaluation of O_3 for the entire year of 2009 is performed by using both ozonesonde (balloon-borne) and ground-based measurements. This year was chosen because of the coverage and availability of TES data, as well as a drastic reduction of O_3 concentration during the recent economic recession. The State of Texas is focused in this study due to its vast size and diverse geography and availability of ozonesonde data in 2009, in addition to its location near the center of the model domain which should yield a representative baseline result (minimum effect) from the boundary conditions.

2 Model configurations and model input data

2.1 Model configurations

Stable versions of meteorological model (WRF3.4) and chemical transport model (CMAQ4.7.1) were used in this study. There are numerous configuration options available for both models. Brief model configurations are described in the following sections. For the modeling domain, the physical descriptions of the two models are provided in Table 1.

2.1.1 Chemical transport model configurations

The EPA's Community Multi-scale Air Quality (CMAQ) modeling system (Byun and Schere 2006; Foley et al. 2010) is a state-of-the-science, community, Eulerian photochemical model. It is the model of choice by the EPA for policy makings in air quality. Model application of CMAQ has been extensively used for O_3, aerosol and mercury studies ranging from local or urban scale through hemispheric scale (Appel et al. 2007; Lin et al. 2005, 2008, 2010; Pirrone et al. 2010; Pongprueksa et al. 2008, 2011; Wang et al. 2009; Bullock and Brehme 2002; Eder and Yu 2006). CMAQ is assembled with diverse science process modules to provide one-atmosphere and community multiscale modeling capability. The model was generally configured with default options for this study. In brief, Yamartino's scheme is used for mass-conserving advection, the Asymmetric Convective Model version 2 (ACM2) scheme (Pleim 2007) for vertical diffusion, 2005 version of lumped-structure Carbon-Bond mechanisms (CB05) for gas-phase mechanism (Sarwar et al. 2008; Yarwood et al. 2005) with chlorine chemistry (Tanaka et al. 2003), Rosenbrock chemistry solver

Table 1 Domain Descriptions of CMAQ, WRF, and TES

Parameter	CMAQ4.7.1	WRF3.4	TES L3
Map Projection	Lambert Conformal Conic	Same as CMAQ	Latitude/Longitude
Domain center	40°N, 97°W	Same as CMAQ	0°N, 0°E
Standard parallel 1	33°N	Same as CMAQ	–
Standard parallel 2	45°N	Same as CMAQ	–
Single-level cell number (X×Y)	152×116	164×128	90×83
Vertical level number (Z)	32	Same as CMAQ	16 (15+surface)
Cell size (dX×dY)	36 km×36 km	Same as CMAQ	4°×2°
Longitudinal coverage	222° to 303°	217° to 308°	0° to 360°
Latitudinal coverage	18°N to 58°N	16°N to 60°N	82°S to 82°N
Top pressure (hPa)	50	Same as CMAQ	4
Data frequency	hourly	Same as CMAQ	daily

(Sandu et al. 1997; Rosenbrock 1963), and CMAQ aerosol model (Binkowski and Roselle 2003) for aerosol dynamics. The photolysis rates required for the simulations were calculated from the Total Ozone Mapping Spectrometer (TOMS) satellite data (McPeters 2009). In this study, CMAQ was configured to cover the contiguous United States (CONUS, in Fig. 2) with a grid resolution of 36 km (Lambert conformal projection centered at 40°N and 97°W, 152×116 cells). The model vertical structure was extended to the lower stratosphere (top pressure at 50 hectopascals, hPa) in order to account for the entire troposphere across the domain. Although CMAQ4.7.1 has been corrected for excessive vertical transport to the ground surface level (Emery et al. 2011), a 32-layer of model vertical structure was used as an additional prevention while maintaining a favorable model computational speed.

2.1.2 Meteorological model configurations

The Weather Research and Forecasting (WRF) model (Grell et al. 2005; Wang et al. 2012; Weisman et al. 2008) was used to provide three-dimensional (3-D) meteorological conditions for CMAQ chemical transport model. The model input data (Global Forecast System, GFS) needed for WRF were taken from the National Centers for Environmental Prediction (NCEP) with 1° spatial and 6-h temporal resolution. Meteorological data were generated using the Advanced Research WRF (ARW) version 3.4. The key configurations include Kain-Fritsch scheme cumulus parameterization (Kain 2004), WRF double-moment 6-class microphysics (Hong et al. 2010), Rapid Radiative Transfer Model (RRTM) for long and short wave radiation (Clough et al. 2005), Pleim-Xiu land surface model and surface layer (Xiu and Pleim 2001; Pleim and Xiu 1995), and ACM2 (Asymmetric Convective Model) (Gilliam and Pleim 2010) planetary boundary layer. The WRF outputs were processed through the Meteorology-Chemistry Interface Processor (MCIP4) (Otte and Pleim 2010) to generate model-ready meteorology for chemical transport simulations. A model top of 50 hPa and 32 vertical layers were used to resolve vertical transport (same vertical structure as CMAQ).

2.2 Model input data

2.2.1 Emissions data

Global emission inventories (EIs) used in this study including the components of CO, NO_x, SO_2, VOCs, ammonia (NH_3), methane (CH_4), and particulate matter (PM) etc. Most of the inventories

are available for the year 2009 with a relatively fine-grid resolution (0.5°×0.5°). The four major categories of the EIs include anthropogenic, biomass burning, biogenic, and oceanic emissions. The anthropogenic emissions are categorized into sectors (e.g. aviation, ships, transportation, energy, solvents, waste, industries, residential, and agriculture etc.). The anthropogenic and biomass burning emissions were projected from the Representative Concentration Pathways (RCPs) database (van Vuuren et al. 2011; Riahi et al. 2007). The RCPs are four independent pathways developed by four individual modeling groups to represent radiative forcing scenarios (2.6, 4.5, 6.0, and 8.5) used in the Intergovernmental Panel on Climate Change (IPCC) studies. All of the emission cases are similar for the year 2009; therefore, RCP8.5 is selected in this study. The RCPs data are available at "ftp://ftp-ipcc.fz-juelich.de/pub/emissions/gridded_netcdf". For anthropogenic emissions, the US EIs from EPA and RCPs are comparable because both are derived from the same national emissions inventory (NEI). The biogenic emissions were derived from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al. 2006; Stavrakou et al. 2011) while the oceanic emissions, which have a coarser grid resolution (1°×1°), were derived from the Precursors of Ozone and their Effects in the Troposphere (POET) database (Granier et al. 2005). These data were combined, speciated, re-gridded, and interpolated into daily files for CMAQ simulation. The VOCs emissions were speciated according to emission source activities and the EPA's SPECIATE database (a repository of VOCs and PM speciation profiles of air pollution sources) (Simon et al. 2010), and then were converted to lumped-structure species in the CB05 mechanism.

2.2.2 Initial and boundary conditions (IC/BC)

Initial Conditions (IC) and Boundary Conditions (BC) are required for CMAQ simulations. IC data provide chemical species representing background concentrations for the modeling domain at the beginning of the simulation. In general, the influences of IC can be weakened by using a proper model "spin-up" period or launching the model simulation prior to the interested time frame. A model spin-up of 2 weeks for reducing the influence of IC has been suggested for a long-term model simulation of O_3 in a similar CONUS domain (Samaali et al. 2009). In this study, a model spin-up period of 30 days is used in order to further minimize the influences of IC. BC data are needed to provide concentrations of chemical species at the lateral boundaries of the modeling domain representing transboundary transport. The standard CMAQ approach uses a time-independent set of vertical concentration profiles of chemical species coming with the CMAQ model for IC and BC data. The IC and BC data are similar, which the BC slightly varies from the IC at each side of the boundaries. For O_3, the standard CMAQ profile range is from 30 ppb at the ground level up to 70 ppb at the model top layer (upper troposphere to lower stratosphere depending on latitude). A typical sigma (σ) structure of 1.00, 0.98, 0.93, 0.84, 0.60, and 0.30 representing IC and BC includes O_3 concentrations of 30, 35, 40, 50, 60, and 70 ppb, respectively. The formula of sigma pressure can be defined as

$$\sigma = \frac{P_A - P_T}{P_G - P_T}, \tag{1}$$

where P_A is the air pressure at the layer of interest, P_G is the pressure at the ground, and P_T is the model top pressure. The typical model concentrations are much lower than the measurement data at upper layers, especially for mid- to high-latitudes where O_3 concentrations are up to a ppm level. It should be noted that the standard profile does not represent accurate O_3 concentrations of the troposphere in both spatial and temporal terms.

To compare with the standard approach and to improve CMAQ model simulation, the Tropospheric Emission Spectrometer (TES) level 3 or L3 (3-D satellite data) (Kusterer 2012)

were used in this study for providing IC and BC of O_3 and CO. The algorithm for generating daily L3 data is Delaunay triangulations on a latitude/longitude plane followed by 2-D interpolations for a fixed TES pressure level (Luo et al. 2007). The TES L3 concentration profiles (see Table 1 for grid description) from "ftp://l4ftl01.larc.nasa.gov/TES" were remapped to the CONUS domain and then linearly interpolated into IC and BC using the mid-layer height of each vertical layer. The model simulation results of the standard and TES profiles are further evaluated against ozonesonde data and ground measurements.

3 Model evaluation methods and observational data

3.1 Model evaluation methods

The performance of the CMAQ model was evaluated at in situ monitoring stations (ground-level and ozone sounding) using top five statistical metrics for model evaluation as recommended by Simon et al. (2012). These performance metrics include: Mean Bias (MB), Mean Error (ME), Normalized Mean Bias (NMB), Normalized Mean Error (NME), and correlation coefficient (r). Formulae for calculating these metrics along with detailed descriptions have been provided elsewhere (Boylan and Russell 2006; Simon et al. 2012), so they are not repeated here. The observed O_3 concentrations were paired with the hourly concentrations extracted from model cells where the monitoring stations are located.

3.2 Ozonesonde

The ozonesonde or ozone sounding device is a balloon-borne instrument that measures O_3 concentration at various altitudes (providing O_3 profiles in the troposphere and the stratosphere) along with some meteorological parameters (e.g. temperature and pressure etc.) and broadcasts the data back to a receiving station on Earth. Ozonesonde data have been widely used in satellite validation (Boynard et al. 2009; Nassar et al. 2008; Petelina et al. 2004; Randall et al. 2003; Sasano et al. 1999) and model evaluation (Bey et al. 2001; Jonson et al. 2010; Pfister et al. 2008; Rotman et al. 2004; Tarasick et al. 2007; Teyssèdre et al. 2007; Tong and Mauzerall 2006; Yu et al. 2007; Zhang et al. 2008). Sources of ozonesonde data used in this study include the World Ozone and Ultraviolet Radiation Data Centre (WOUDC), the National Oceanic and Atmospheric Administration's (NOAA) Earth System Research Laboratory (ESRL), and the Tropospheric Ozone Pollution Project (TOPP). The ozone sounding data of WOUDC, NOAA, and TOPP used for validation of satellite data and model evaluation are from "http://www.woudc.org", "ftp://ftp.cmdl.noaa.gov/ozwv", and "http://physics.valpo.edu/ozone", respectively. A map of the ozonesonde stations and description of the monitoring sites are shown in Fig. 2 and Table 2, respectively.

To evaluate O_3 profiles from two different sets of model input, observed O_3 profiles from the sounding are paired with profiles from the typical standard initial and boundary conditions (IC/BC) and profiles derived from the Tropospheric Emission Spectrometer (TES). To prepare an O_3 profile, all of the concentration values throughout the year of 2009 at a sounding station are sorted into 167 bins of associate pressure from model top pressure (50 hPa) to the pressure at the ground, with an even increment about 4 to 6 hPa (hectopascals) depending upon the ground pressure. All of the concentration data in each pressure bins are then averaged to make pairs of pressure and concentration representing the profile at the monitoring site. Figures 3 and 4 show observed O_3 profiles at the sounding stations against the corresponding profiles from TES and the standard IC/BC. Figure 3 shows ozonesonde data from the WOUDC stations which are

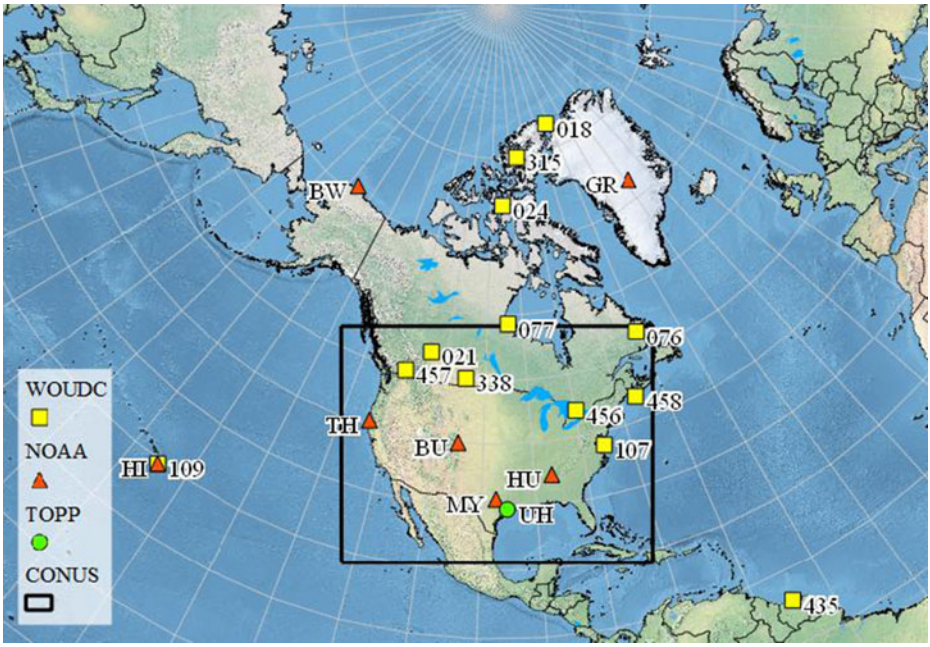

Fig. 2 Map of ozonesonde stations and the contiguous United States (CONUS) domain. Red triangles symbolize sites from the National Oceanic and Atmospheric Administration (NOAA). Yellow squares represent sites from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC). The green circle is ozonesonde location in Houston, Texas from the Tropospheric Ozone Pollution Project (TOPP). The area inside the rectangular frame is the CONUS domain

mainly located in Canada, while Fig. 4 shows the data from the NOAA and the TOPP stations that are located in the United States except one station in Greenland (GR). In the upper troposphere and lower stratosphere, the mean TES profiles are similar to the profiles of ozonesonde, while the standard IC/BC profiles are very different from the observed data. Concentration of O_3 at 100 ppb is used to estimate the tropopause, the boundary layer between stratosphere and troposphere, since this method has been efficiently utilized by Prather et al. (2011). Correlations of both TES and IC/BC with ozonesonde profiles are acceptable at lower troposphere. It is interesting to note that TES data are positive bias at lower troposphere (especially near the ground). In previous TES validation with aircraft measurements, Richards et al. (2008) also reported a positive bias of 7 ppb (5-15 %) on average in troposphere.

Figure 5 shows comparisons between all of ozonesonde profiles in 2009 with the O_3 profiles from the typical standard initial and boundary conditions (IC/BC) of CMAQ and the Tropospheric Emission Spectrometer (TES). The data from ozonesonde profiles are on the x axis and the data from the standard IC/BC or TES are on the y axis. For the entire vertical structure of the model (top figures, pressure≥50 hPa), TES data provided a better representation of ozone profiles than the standard IC/BC. The profiles from the TES correlate well with the ozonesonde data (correlation coefficient, r=0.95) and have better statistical metrics than the profiles from the standard IC/BC. In the troposphere (bottom figures, O_3≤100 ppb), the performance of the standard IC/BC profiles improves for all metrics, while the performance of TES improves in terms of MB, ME, and NME but slightly drop for NMB and r when compared with the metrics from the entire model's vertical layers. The standard IC/BC concentrations are somewhat

Table 2 Description of ozonesonde monitoring stations used in this study in 2009

Site ID	Location	Country	Latitude (degree)	Longitude (degree)	Elevation (meter)	Number of Flight	Sampling Period	Source
STN018	Alert	Canada	82.49	−62.34	75	33	07 Jan. to 30 Dec.	WOUDC
STN021	Stony Plain	Canada	53.54	−114.1	766	48	07 Jan. to 30 Dec.	WOUDC
STN024	Resolute	Canada	74.71	−94.97	46	33	07 Jan. to 23 Dec.	WOUDC
STN076	Goose Bay	Canada	53.3	−60.36	36	49	08 Jan. to 30 Dec.	WOUDC
STN077	Churchill	Canada	58.74	−94.07	30	30	07 Jan. to 23 Dec.	WOUDC
STN107	Wallops Island	USA	37.93	−75.47	13	53	08 Jan. to 30 Dec.	WOUDC
STN315	Eureka	Canada	79.98	−85.94	10	71	09 Jan. to 31 Dec.	WOUDC
STN338	Bratts Lake	Canada	50.2	−104.7	580	36	07 Jan. to 30 Dec.	WOUDC
STN435	Paramaribo	Suriname	5.81	−55.21	7	39	14 Jan. to 28 Dec.	WOUDC
STN456	Egbert	Canada	44.23	−79.78	252	54	15 Jan. to 16 Dec.	WOUDC
STN457	Kelowna	Canada	49.94	−119.4	456	49	07 Jan. to 30 Dec.	WOUDC
STN458	Yarmouth	Canada	43.87	−66.11	9	41	14 Jan. to 30 Dec.	WOUDC
BU	Boulder, CO	USA	40	−105.25	1743	42	13 Jan. to 31 Dec.	NOAA
BW	Burrow, AK	USA	71.32	−156.6	10	28	12 Mar. to 15 Apr.	NOAA
GR	Summit	Greenland	72.57	−38.48	3211	41	06 Jan. to 31 Dec.	NOAA
HI/ STN109	Hilo, HI	USA	19.43	−155.04	11	47	06 Jan. to 30 Dec.	NOAA/ WOU-DC
HU	Huntsville, AL	USA	35.28	−86.59	196	50	03 Jan. to 26 Dec.	NOAA
MY	Moody, TX	USA	31.32	−97.33	251	19	25 Aug. to 30 Sep.	NOAA
TH	Trinidad Head, CA	USA	40.8	−124.16	20	47	15 Jan. to 29 Dec.	NOAA
UH	Houston, TX	USA	29.7	−95.3	19	66	18 Mar. to 05 Dec.	TOPP

overestimated at the lower troposphere (low O_3 region) and underestimated at the upper troposphere (higher O_3 region). Similar to Richards et al. (2008), TES profiles show positive biases across the troposphere (7.9 ppb in average) with biases gradually reduce from the ground to tropopause. Relatively high biases of O_3 concentration near the ground can affect the interpretation of the model results; therefore, an additional CMAQ simulation with adjusted

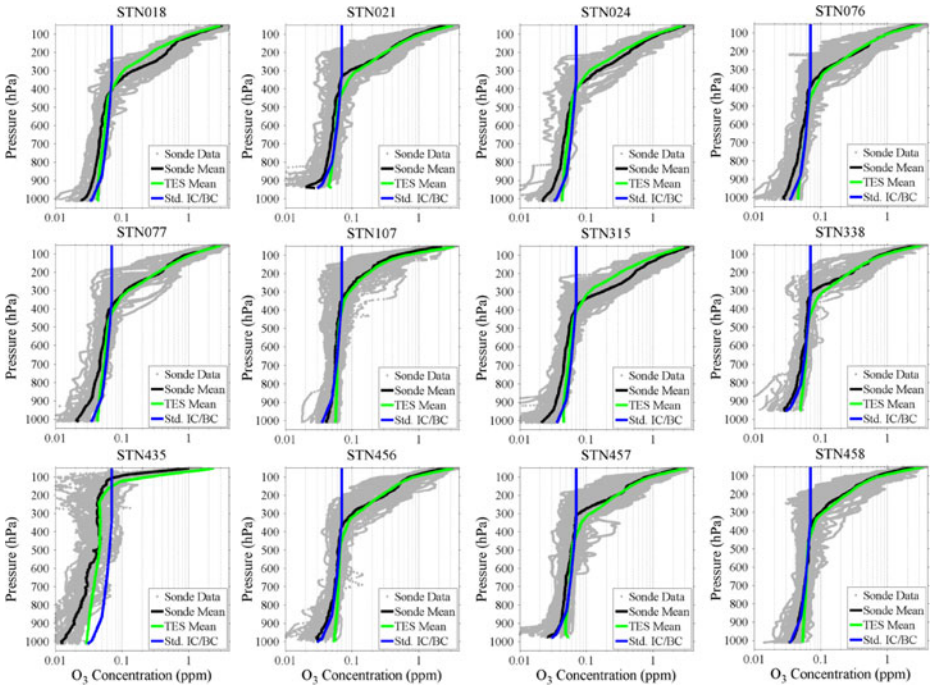

Fig. 3 Typical standard O_3 concentration profiles of initial and boundary conditions (IC/BC) and observed profiles that are primarily located in Canada from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) stations and the Tropospheric Emission Spectrometer (TES) in 2009. The blue lines represent IC/BC profiles. Ozonesonde data are indicated by gray dots. The black and green lines represent the annual mean profiles of ozonesonde data and TES data, respectively. 12 WOUDC sites for TES validation include Alert, Canada (STN018); Stony Plain, Canada (STN021); Resolute, Canada (STN024); Goose Bay, Canada (STN076); Churchill, Canada (STN077); Wallops Island, United States (STN107); Eureka, Canada (STN 315); Bratts Lake, Canada (STN338); Paramaribo, Suriname (STN435); Egbert, Canada (STN456); Kelowna, Canada (STN457); and Yarmouth, Canada (STN458)

TES data was also investigated. For the additional case, TES ozone profiles with a deduction of 10 ppb were used as an alternative IC/BC in CMAQ. The fixed deduction value was used to simplify both data processing and interpreting the model outputs. Since previous studies by Fiore et al. (2002, 2003) reported surface background O_3 of the United States between 15 ppb and 35 ppb, the O_3 concentration at 35 ppb was chosen to represent a clean background in this study. The deduction is estimated from the linear regression parameters in troposphere ($y=0.844x+15.79$) as shown in Fig. 5 (bottom-right) that TES data are about 10 ppb (10.33 ppb) higher than ozonesonde data at 35 ppb.

3.3 Ground monitoring stations

In order to evaluate model performance of ground-level O_3, this study uses hourly O_3 data in 2009 from the Air Quality System (AQS). The AQS contains site information, meteorological data, and ambient air quality data collected by the EPA and state agencies. The AQS database is available at "http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm". Data from the AQS are the standard data used to assist attainment/non-attainment designations and to evaluate State Implementation Plans. In Texas, the Texas Commission on Environmental Quality (TCEQ) collects air quality data from the Continuous Ambient Monitoring Stations (CAM) and sends the

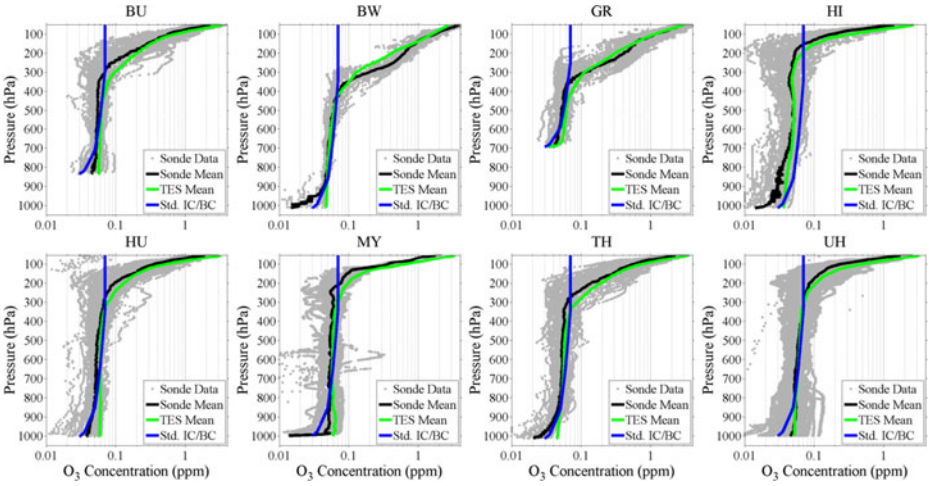

Fig. 4 Same as Fig. 3 but for observed profiles from the National Oceanic and Atmospheric Administration (NOAA) and the Tropospheric Ozone Pollution Project (TOPP). These stations are mostly located in the United States. For validation of TES satellite data, 7 NOAA sites and 1 TOPP (Houston, TX; UH) were used in this study. NOAA sites include Boulder, CO (BU); Burrow, AK (BW); Summit, Greenland (GR); Hilo, HI (HI); Huntsville, AL (HU); Moody, TX (MY); and Trinidad Head, CA (TH)

data to the EPA to incorporate into the AQS database. The locations of AQS/CAM monitoring stations in Texas for model evaluation in this study are presented in Fig. 6.

4 Results and discussion

4.1 Evaluation with ozonesonde measurements

The O_3 sounding data from the monitoring stations (WOUDC, NOAA, and TOPP) located within the Contiguous United States (CONUS) domain (see Fig. 2) were paired with the modeled O_3 concentrations by using air pressure data from the measurements and the meteorological model. To simplify the model data processing, the released ozonesonde points were used to determine modeling columns being extracted throughout the balloon flights (releasing columns). Since the model's grid-size is relatively large and wind speed is fairly weak in lower troposphere, a typical ozonesonde balloon is not expected to travel away from the releasing column. While a much stronger wind speed in the upper atmosphere can carry the balloon out of the column, O_3 concentration in this layer does not fluctuate much. Therefore, it is acceptable to compare ozonesonde data with O_3 concentrations extracted from CMAQ simulation cases by using this simplified method. The three simulation cases include: (1) CMAQ with the standard IC/BC, (2) CMAQ with IC/BC derived from TES data, and (3) CMAQ with IC/BC derived from adjusted TES data (10 ppb deduction). The measured data and model data were sorted and averaged to make average O_3 vertical profiles as described earlier in section 3.2.

Fig. 7 shows O_3 profiles from ozonesonde and CMAQ simulation cases in the CONUS domain. The profiles from CMAQ are similar to profiles from the model input data in Fig. 3 and Fig. 4. In the lower troposphere, the profiles from CMAQ simulations are closer to the observed ozonesonde profiles than the profiles from the standard IC/BC and TES, especially near ground

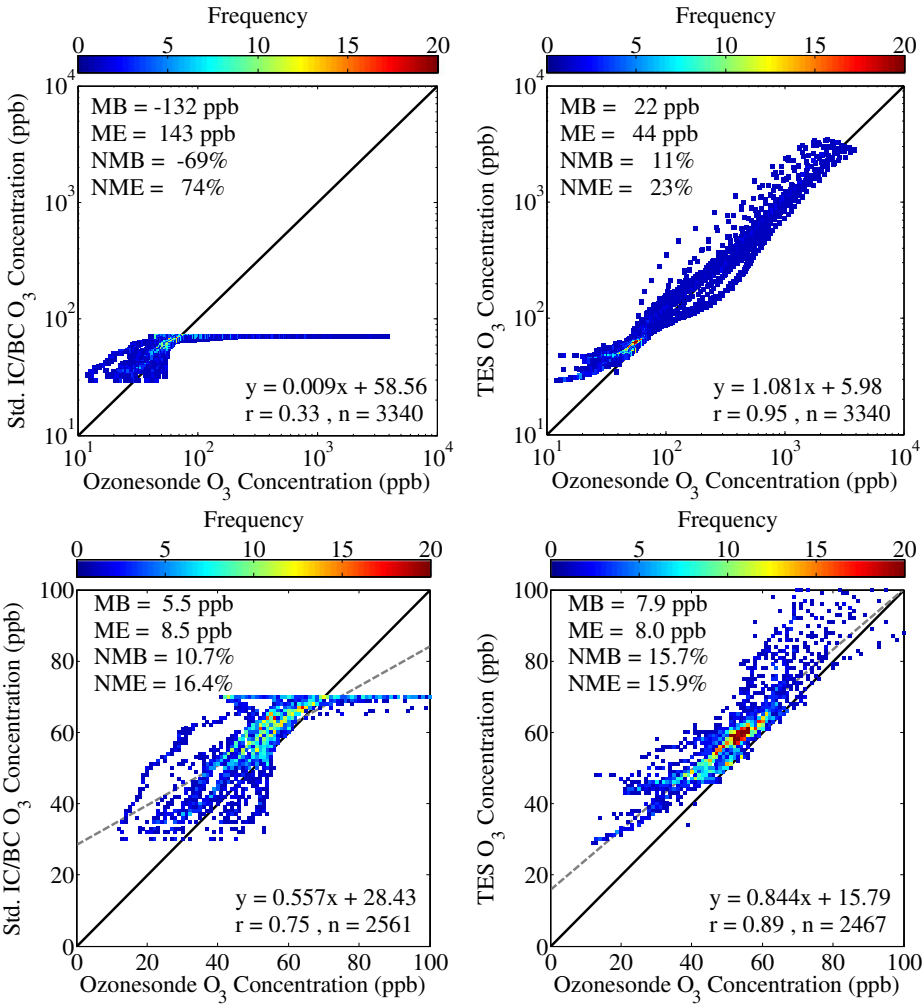

Fig. 5 Scatter density plots with linear regression and statistical metrics of the ozonesonde concentration profiles versus the profiles from (left) typical standard initial and boundary conditions (IC/BC) of CMAQ and from (right) Tropospheric Emission Spectrometer (TES). The comparisons of the entire model's vertical structure (pressure≥50 hPa) are shown on the top while the results within troposphere (O_3≤100 ppb) are shown at the bottom

level. This suggests stronger interactions between emissions and chemical reactions in lower troposphere than in upper troposphere and stratosphere. At top layers of the model, O_3 concentrations from CMAQ are heavily affected by the selections of IC/BC. O_3 profiles of CMAQ using standard IC/BC are different from the profiles of CMAQ using IC/BC derived from TES. The standard IC/BC profiles are comparable with the profiles of CMAQ using the standard IC/BC, while the TES profiles are comparable with the profiles of CMAQ using IC/BC derived from TES. Apart from missing lightning NO_x emission in the upper atmosphere (Jourdain et al. 2010; Wang et al. 2013; Allen et al. 2010), this may due to the fact that most current regional models including CMAQ do not incorporate stratospheric chemical reactions that are included in some global models. It is worth noting that a recent model developement

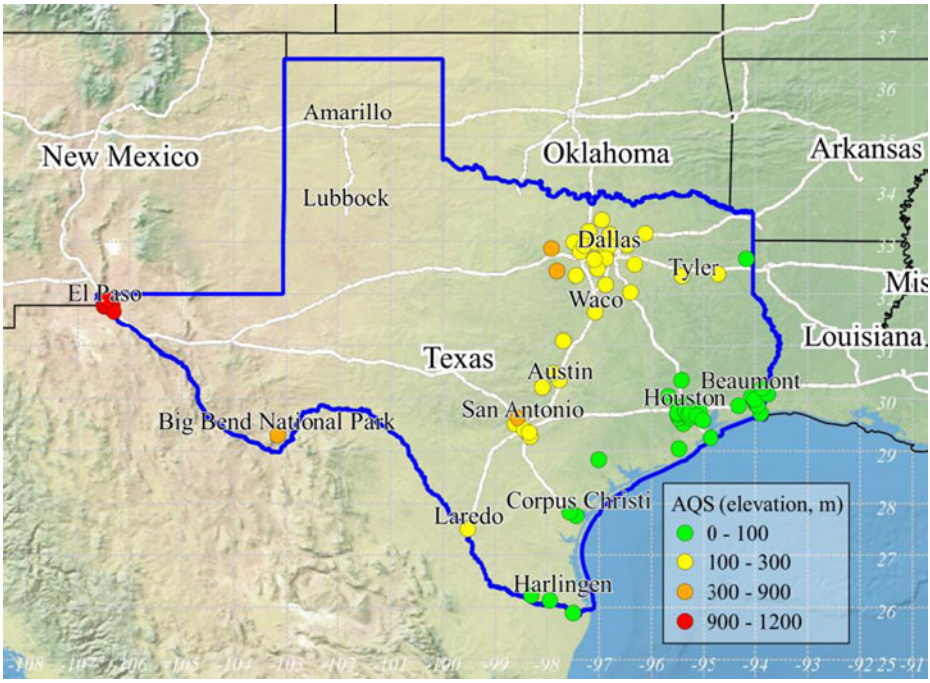

Fig. 6 Map of the Air Quality System (AQS) monitoring stations in Texas. Circles represent the locations with colors being varied by the site elevations

incorporates chemical reactions of both urban and global (Karamchandani et al. 2012; Zhang et al. 2012) which may reduce the impact of IC/BC on the upper atmosphere.

To further evaluate the model, scatter plots with linear regression parameters and statistical metrics of the ozonesonde profiles and profiles from CMAQ cases are shown in Fig. 8. Top subfigures show relationships between ozonesonde and model data for the entire model vertical layers while subfigures at the bottom show only relationships within the troposphere. In general, the case of CMAQ with IC/BC derived from the adjusted TES is slightly better than the CMAQ case with raw TES data in terms of MB, ME, NMB, NME, and slope of the regression. In the meanwhile, the profiles of CMAQ cases with IC/BC derived from both TES and adjusted TES are better than the profiles of the CMAQ with standard IC/BC. In the troposphere, CMAQ with IC/BC from TES perform better than the standard CMAQ in the slope but worsen in MB, ME, NMB, NME, and r. On the contrary, the CMAQ case with adjusted TES outperforms the standard CMAQ in terms of the regression slope, MB, and NMB while they are comparable in terms of ME, NME, and r. In conclusion, the CMAQ with IC/BC derived from TES performs better than the standard CMAQ only in the upper model layers, while the case with IC/BC derived from adjusted TES performs better than the standard case in the troposphere and across all vertical layers.

4.2 Evaluation with AQS surface measurements

In this section, three CMAQ cases are evaluated with ground measurement data from the AQS database in the State of Texas. To be relevant to the NAAQS for O_3, statistical metrics of daily maximum 8-h average (MDA8) were used to evaluate the model. Fig. 9 shows annual mean of

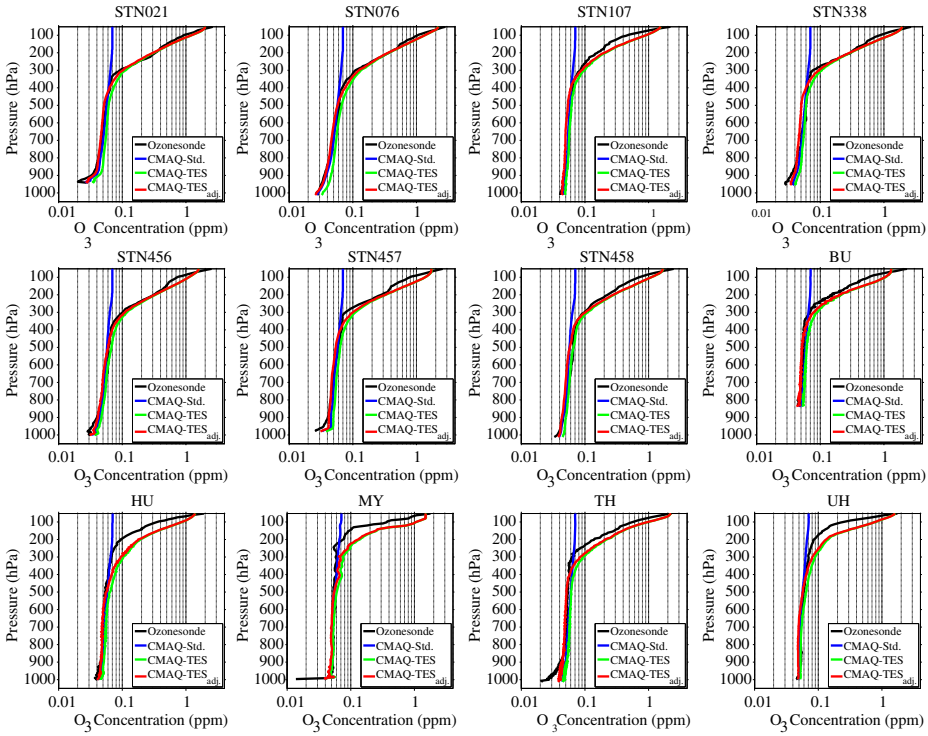

Fig. 7 O_3 profiles in 2009 at ozonesonde stations (black line) within the model domain (US and Canada) and the profiles from CMAQ simulation cases: Standard IC/BC (blue line), TES IC/BC (green line), and Adjusted TES IC/BC (red line)

MDA8 O_3 of the cases in 2009 on the left side of the figure and scatter plots of MDA8 of all Texas sites on the right side with the AQS data on the x axis and the data from CMAQ simulations cases on the y axis. The spatial distributions of mean MDA8 O_3 concentration of these CMAQ cases are similar with higher O_3 at high-elevation areas and lower O_3 at low-level areas. However, the case of CMAQ with IC/BC derived from TES (middle of Fig. 9) shows the highest mean of MDA8 among the other cases while the case of CMAQ with IC/BC derived from adjusted TES (bottom of Fig. 9) gives the lowest mean of MDA8. In overall, the statistical metrics (Fig. 9, right-side) from the entire model cases are comparable. The case of CMAQ with IC/BC from TES (middle of the figure) performs a little better than the standard CMAQ (top of the figure) in the regression slope and the correlation coefficient but relatively worsens in MB, ME, NMB, and NME. The CMAQ case with adjusted TES (bottom of the figure) performs slightly better than the other two cases in all of the metrics.

Model performances of the three CMAQ cases at each Texas area are summarized in Table 3. The Texas areas include: (1) Austin, (2) Beaumont, (3) Big Bend, (4) Corpus Christi, (5) Dallas, (6) El Paso, (7) Harlingen, (8) Houston, (9) Laredo, (10) San Antonio, (11) Tyler, and (12) Waco. A map of these areas was shown in Fig. 6. The CMAQ case with IC/BC derived from TES performs poorer than the standard case in terms of MB, ME, NMB, and NME at all the areas but shows better correlation coefficients. The CMAQ case with IC/BC derived from adjusted TES performs better than the standard case and the case with IC/BC derived from original TES in all of the statistical metrics except biases (MB and NMB) at the Big Bend National Park.

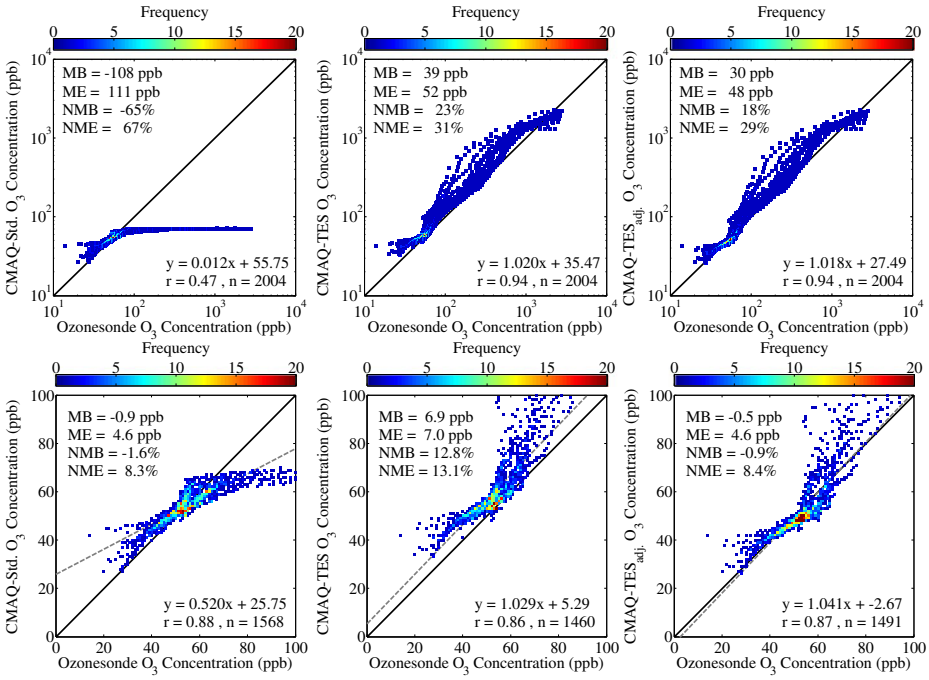

Fig. 8 Scatter density plots with linear regression and statistical metrics of the ozonesonde concentration profiles in CONUS domain versus the profiles from CMAQ simulation cases: (left) Standard IC/BC, (center) TES IC/BC, and (right) Adjusted TES IC/BC. The comparisons of the entire model's vertical structure (pressure≥50 hPa) are shown on the top while the results within troposphere (O_3≤100 ppb) are shown at the bottom

4.3 Influence of O_3 from upper model layers

Model performance of CMAQ at the ground-level does not improve by the utilization of original TES data in providing initial and boundary conditions. The errors and biases of O_3 at ground-level were slightly increased, even though TES data helped improve the performance in upper model levels (upper troposphere and lower stratosphere). The performance at ground-level can be slightly improved with the calibrated TES data. In spite of the fact that the O_3 concentrations of the TES data are subtracted by 10 ppb in average across the model vertical layers for the IC/BC of the adjusted TES case, the adjusted concentrations in the upper atmosphere are still much higher than the concentrations of the standard IC/BC. The simulation results suggest that there is a small influence of O_3 from the upper atmosphere to the ground-level, and the performance improvement of ground-level O_3 concentrations is due to a better representative of tropospheric O_3 which is similar to a conclusion drawn by Appel et al. (2007).

To further examine the impact of the upper atmosphere to the ground-level O_3, the CMAQ with IC/BC derived from adjusted TES was used as base case. The model levels were gradually reduced from 32 levels (model top pressure=50 hPa) to 26, 22, and 16 levels while preserving the same meteorological conditions and emissions for the remaining levels (except some aircraft emissions were excluded along with the removed levels for the 22-level and 16-level cases). The model vertical structure and annual mean of zonal O_3 profile in 2009 is shown in Fig. 10. The TES data were used for displaying tropospheric and stratospheric O_3 from Northern and

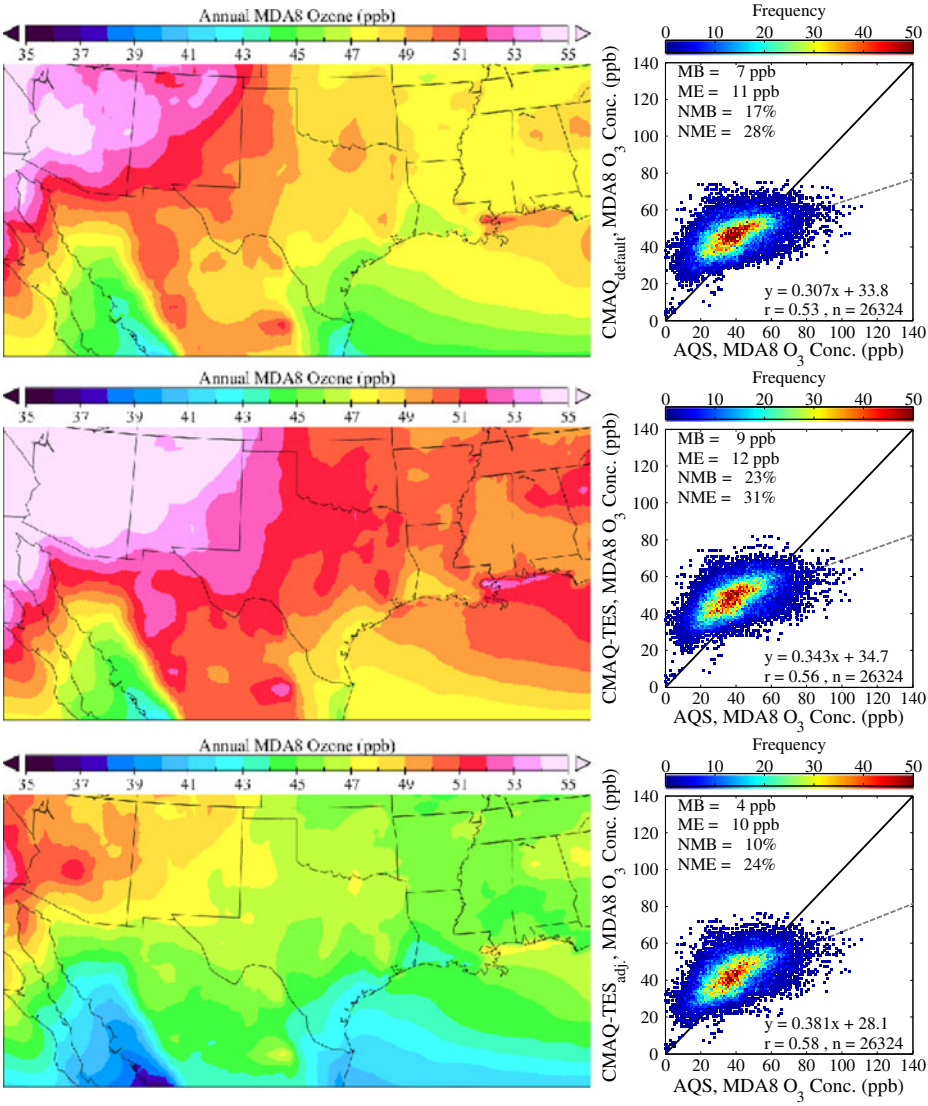

Fig. 9 Maps (left) of mean daily maximum 8-h average (MDA8) O_3 of Texas in 2009 and scatter plots (right) of MDA8 between observed data from the Air Quality System (AQS) monitoring sites in Texas and data from model simulation cases: (top) CMAQ with standard initial and boundary conditions (IC/BC), (middle) CMAQ with IC/BC from the Tropospheric Emission Spectrometer (TES), and (bottom) CMAQ with IC/BC from adjusted TES

Southern Hemispheres. The model domain encloses all of tropospheric O_3 with some stratospheric O_3 being excluded for the case with 26 levels. For the case with 22 levels, some of tropospheric and stratospheric O_3 were excluded from the model domain. The entire stratospheric O_3 was excluded from the model domain for the case with 16 model levels.

The differences of the base case (32-level) for the annual MDA8 O_3 and the three reduced level cases in CONUS domain are shown in Fig. 11. The subfigure on the top-left shows annual mean of MDA8 of ozone concentration in ppb while the other subfigures show the

Table 3 Model Performance of CMAQ Cases for MDA8 of O_3 at Texas AQS Sites in 2009

TCEQ Region	Number of Site	CMAQ-Standard					CMAQ-TES					CMAQ-Adjusted TES				
		MB (ppb)	ME (ppb)	NMB (%)	NME (%)	r	MB (ppb)	ME (ppb)	NMB (%)	NME (%)	r	MB (ppb)	ME (ppb)	NMB (%)	NME (%)	r
Austin	2	5	10	13	24	0.52	8	11	19	27	0.55	3	8	7	20	0.57
Beaumont	8	7	11	19	31	0.46	9	12	25	34	0.51	4	10	11	27	0.51
Big Bend National Park	1	1	6	1	14	0.30	3	7	6	14	0.46	−3	6	−6	14	0.48
Corpus Christi	3	8	11	22	32	0.51	10	13	29	36	0.57	5	9	14	27	0.57
Dallas/Fort Worth	19	4	10	9	23	0.61	6	11	15	25	0.62	2	9	4	21	0.64
El Paso	6	7	11	17	25	0.42	9	12	22	28	0.48	3	8	8	19	0.56
Harlingen	3	10	12	29	35	0.50	12	13	37	39	0.60	6	9	19	27	0.60
Houston	21	9	13	25	35	0.48	11	14	31	39	0.52	6	11	17	30	0.52
Laredo	1	12	13	38	40	0.46	15	15	45	46	0.53	9	10	28	32	0.53
San Antonio	5	7	10	17	26	0.49	9	11	23	29	0.55	4	9	9	22	0.55
Tyler	3	6	10	14	24	0.54	8	11	19	26	0.58	3	9	8	21	0.58
Waco	1	3	9	8	21	0.62	6	10	13	23	0.63	1	8	2	18	0.66

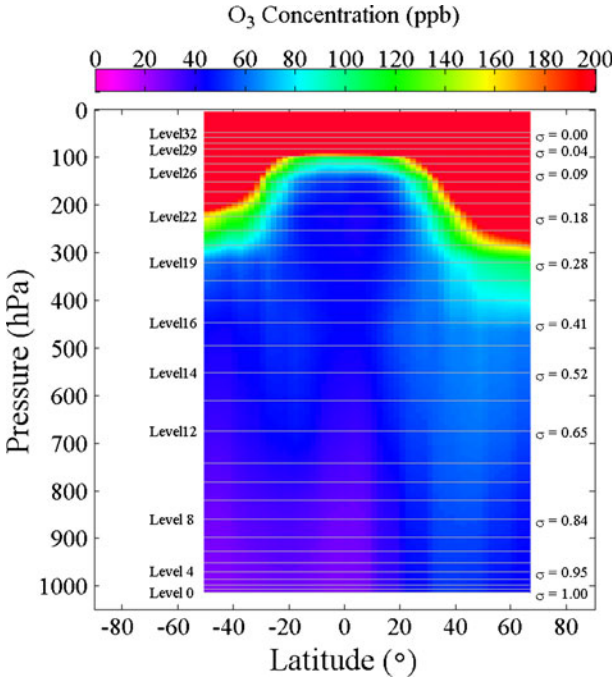

Fig. 10 Model vertical structure and profile of zonal mean O_3 from TES data in 2009

differences (in ppb) from reduced level cases. By removing top 6 levels from the base case (26-level, top-right), the changes of MDA8 O_3 concentrations at the ground appear at high-elevation areas but can be ignored (less than 0.03 ppb, 0.06 %). For the 26-level case, the emissions were not changed because all of the emissions were still contained within this model vertical structure. The differences are still negligible (less than 0.17 ppb, 0.39 %) when the model level was further reduced to 22 levels (bottom–left), even though some aircraft emissions were excluded along with the cut levels. When stratospheric layers were completely excluded as in the case of 16-level (bottom–right), the differences of ground-level O_3 from the base case are much higher than the other level removal cases but are relatively small (less than 2.5 ppb, 5.4 %) for the entire CONUS domain. For Texas, the differences are less than 1.4 ppb (3.1 %) in high elevation areas (Big Bend and El Paso) and smaller than 0.51 ppb (1.1 %) in low elevation areas such as Dallas and Houston (notorious O_3 nonattainment areas). From this additional experiment, the data confirm that the influence of O_3 from the upper atmosphere on the ground-level O_3 is generally small in CONUS domain, especially in Texas.

4.4 Prospective application of TES data

This study presents a utilization of satellite data as model inputs in order to improve model performance. However, there are three major limitations for using the Aura's TES satellite data. First of all, many atmospheric chemicals needed for chemical transport models are not currently measureable by remote sensing. Second, satellites usually cover large areas but their data are not temporally resolved as required in regional models. The TES only provides daily average data while the models generally require hourly data. Third, the Aura satellite

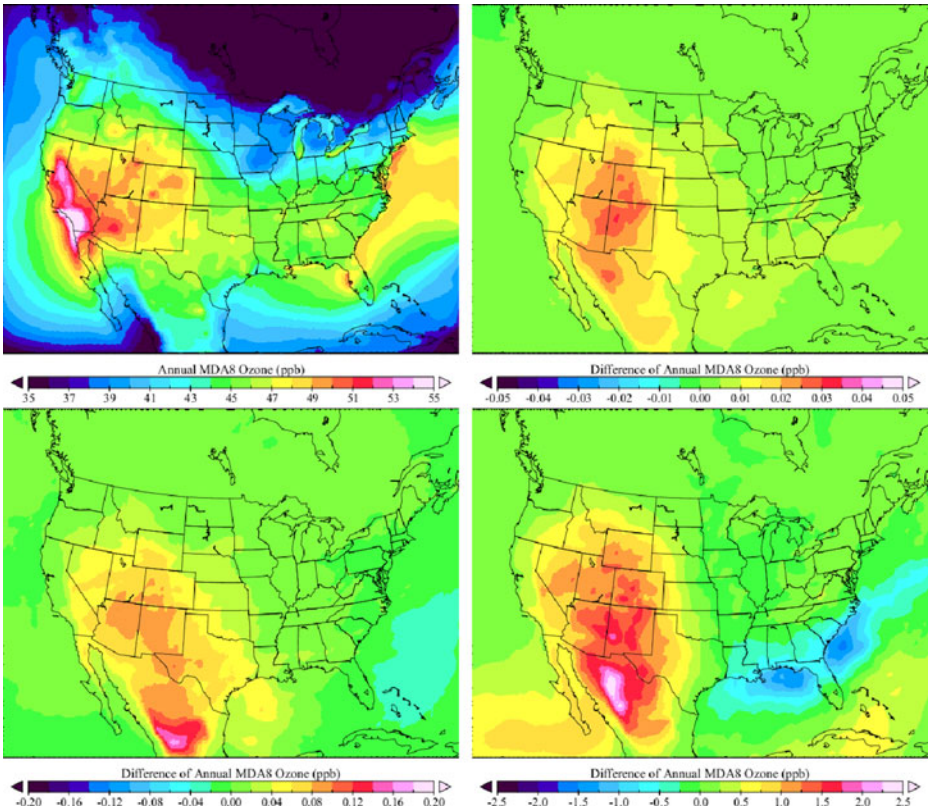

Fig. 11 Annual mean MDA8 ozone (ppb) at ground-level in CONUS domain (top left) and the differences between 32-level and the cases with reduced level: (top right) 26-level, (bottom left) 22-level, and (bottom right) 16-level

has gradually reduced zonal coverage from 82°S-78°N in 2008 to 32°S-48°N in 2010 and beyond in order to extend longevity of its instruments. The reduced coverage does not include some parts of the CONUS domain in the North.

To overcome these limitations, a regional model such as CMAQ needs to be extended to cover a larger domain in order to effectively utilize TES data. A domain as large as the Northern Hemisphere is suitable because the domain covers most of the major sources, and transport of atmospheric chemicals between the Northern and the Southern Hemispheres is much slower than the transport between the Eastern and the Western Hemispheres. TES data at or near the equator can provide initial and boundary conditions (IC/BC) of CMAQ for a northern hemispheric domain. Model results of the hemispheric CMAQ can further provide IC/BC with required temporal resolution, compatible chemical mechanisms, and all atmospheric chemical species needed for a smaller domain such as CONUS. The application of CMAQ with TES on a hemispheric scale comes with a caveat that CMAQ can not accurately generate stratospheric O_3. As described earlier, CMAQ currently does not incorporate stratospheric chemical reactions and it relies on stratospheric O_3 contributed from IC/BC. Around the Equator, the tropopause and lower stratosphere reside in upper model layers as shown in Fig. 10. High O_3 from these layers at the Equator will not efficiently transport down to lower model layers at higher latitudes of the

hemispheric domain, so the model results will underestimate O_3 concentrations in lower stratosphere. Fortunately, stratospheric O_3 has a marginal impact on the ground-level O_3 as demonstrated in the previous section. Ozone studies in the future should focus on lower troposphere where the precursors are concentrated and abundant. A future study should examine the utilization of hemispheric CMAQ results with IC/BC derived from TES data in providing IC/BC for subsequent regional CMAQ modeling. The author anticipates that the techniques and data presented in this study can be further used to refine model performance of ground-level O_3.

5 Conclusion

Model studies of O_3 in 2009, especially in the US, are challenging due to sudden changes in O_3 concentrations, which may be linked to better air pollution controls and the recent economic recession. The standard O_3 profiles normally used in CMAQ are time-invariant and may be outdated. In addition, lower O_3 concentrations can affect model performance by increasing statistical metrics such as NMB and NME. The author has demonstrated a long-term CMAQ simulation with the initial and boundary conditions (IC/BC) derived from data of the Tropospheric Emission Spectrometer (TES) aboard the NASA's Aura satellite. The satellite data can improve model performance in the upper model layers (upper troposphere and lower stratosphere). However, IC/BC derived from TES cannot improve model performance at ground-level because the satellite data overstated tropospheric O_3. With a calibration of O_3 concentration, the IC/BC of O_3 derived from adjusted TES improves model performance from ground level through the upper atmosphere. The mean bias of MDA8 from all of the Texas AQS sites was reduced from 7 ppb in the standard CMAQ case to 4 ppb (43 % reduction) in the case with IC/BC derived from the adjusted TES data.

The author also demonstrated the influence of O_3 from upper atmosphere to the ground-level O_3. With a complete exclusion of stratosphere, changes of annual mean MDA8 of O_3 concentrations at ground-level were smaller than 0.51 ppb (1.1 %) at O_3 nonattainment areas in Texas (Dallas and Houston). The result also included the impacts from exclusion of partial tropospheric air and some aircraft emissions due to model layer removal. These small changes in ground-level O_3 suggest a small influence of upper tropospheric O_3.

Satellite data can help improve model performance in air quality models. However, the data need to be validated especially in lower troposphere where measurement interferences may be very high. In addition, not all atmospheric chemicals used in the models are measureable from remote sensing. To be independent from global models, available satellite data can be used to create IC/BC for CMAQ model on a hemispheric scale, and the concentrations of modeled atmospheric chemicals can be used to further provide IC/BC for smaller domains.

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