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Impacts of long-range transport of global pollutants and precursor gases on U.S. air quality under future climatic conditions

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[1] The U.S. air quality is impacted by emissions both within and outside the United States. The latter impact is manifested as long-range transport (LRT) of pollutants across the U.S. borders, which can be simulated by lateral boundary conditions (LBC) into a regional modeling system. This system consists of a regional air quality model (RAQM) that integrates local-regional source emissions and chemical processes with remote forcing from the LBC predicted by a nesting global chemical transport model (model for ozone and related chemical tracers (MOZART)). The present-day simulations revealed important LRT effects, varying among the five major regions with ozone problems, i.e., northeast United States, midwest United States, Texas, California, and southeast United States. To determine the responses of the LRT impacts to projected global climate and emissions changes, the MOZART and RAQM simulations were repeated for future periods (2048–2052 and 2095–2099) under two emissions scenarios (IPCC A1Fi and B1). The future U.S. air quality projected by the MOZART is less sensitive to the emissions scenarios than that simulated by the RAQM with or without incorporating the LRT effects via the LBC from the MOZART. The result of RAQM with the LRT effects showed that the southeast United States has the largest sensitivity of surface ozone mixing ratio to the emissions changes in the 2095–2099 climate (−24% to +25%) followed by the northeast and midwest United States. The net increase due to the LRT effects in 2095–2099 ranges from +4% to +13% in daily mean surface ozone mixing ratio and +4% to +11% in mean daily maximum 8-h average ozone mixing ratios. Correspondingly, the LRT effects in 2095–2099 cause total column O_3 mixing ratio increases, ranging from +7% to +16%, and also 2 to 3 more days with the surface ozone exceeding the national standard. The results indicate that future U.S. air quality changes will be substantially affected by global emissions.

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1. Introduction

[2] The Fourth Assessment Report of the *Intergovernmental Panel on Climate Change* [2007] identified the observed increase in global average temperatures since the mid-20th century as likely due in large part to the observed increase in anthropogenic greenhouse gas concentrations. The report also indicated a high likelihood of a variety of future changes in the climate system if atmospheric concentrations of radiatively important gases like carbon dioxide and methane continue to increase. Because regional air quality is tied closely to the atmospheric circulation as well as regional emissions of pollutants and/or their precursor gases, any changes in wind patterns induced by anthropogenic forcing can impact future regional air quality [*Hogrefe et al.*, 2004; *Murazaki and Hess*, 2006]. Projections of climate change are affected by the uncertainty in climate forcing due to future emissions of the anthropogenic gases and particles, uncertainty in modeled responses to given forcing scenarios, and uncertainty due to missing or misrepresented physical processes in models [*Cubasch et al.*, 2001]. To address the uncertainties, H.-C. Huang et al. (Potential changes of continental U.S. air quality under future climatic conditions, submitted to *Journal of Geophysical*

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Research, 2008) (hereinafter referred to as Huang et al., submitted manuscript, 2008a) used different modeling configurations including two cumulus schemes, outputs from two global climate model (GCM), and four IPCC emissions scenarios to estimate the impact of climate changes on the U.S. air quality using a regional modeling system. This study showed that the simulated tropospheric ozone (O_3) mixing ratios in the period of 2095–2099 are very sensitive to the emissions scenarios as driven by the output of the Parallel Climate Model [*Washington et al.*, 2000; *Dai et al.*, 2001]; the results are less sensitive for simulations using the Hadley Center GCM output. The sensitivity to modeling configurations is found to be regionally dependent, similar to the results of *Murazaki and Hess* [2006]. The southeast United States is identified to be the area most sensitive to the projected climate, with the changes of regional and 5-summer mean ozone mixing ratios ranging from −30% to +59%. The northeast and midwest United States are the next most sensitive regions, with the ozone changes ranging from −24% to +42%, and −19% to +41%, respectively. The more polluted A1Fi and cleaner B1 emissions scenarios, in general, represent the upper and lower bound of the estimations, respectively. However, the impact of long-range chemical transport of pollutants and their precursor gases outside the United States on the U.S. air quality was not accounted for in the study.

[3] *Fiore et al.* [2002] has shown anthropogenic emissions from outside the United States can enhance afternoon surface ozone over the U.S. by 4–7 parts per billion by volume (ppbV). *Lin et al.* [2008a] showed that future changes of intercontinental transport of European and Asian pollutants can have significant impact on U.S. pollution. Under the polluted scenario, the changes in European and Asian anthropogenic emissions lead to increases of about 3–8 ppbV surface ozone mixing ratio over the western United States in 2049 and 2099. The Asian anthropogenic emissions changes alone contributed to a 2–6 ppbV increase of surface ozone mixing ratio. Under the clean scenario, the European and Asian anthropogenic emissions changes lead to decreases of about 1–2 ppbV ozone mixing ratio over the western United States. The European and Asian biogenic emissions changes were shown to have a minor impact with increases of about 1 ppbV ozone mixing ratio over the United States during the summertime in 2049 and 2099 under the polluted scenario. The corresponding increase of ozone mixing ratio was even smaller under the clean scenario. The IPCC Special Report on Emissions Scenarios [*Nakicenovic and Swart*, 2000] shows that the fractional global carbon dioxide emission in developing countries is expected to increase from 31% (in 1990) to 68–77% by 2100. For the cleaner B1 emissions scenarios, although by 2100 the nitrogen oxides (NO_x) and volatile organic compounds (VOC) emissions of the industrialized regions decrease from 12.8 to 2.2 MtN and 42.0 to 13.0 Mt, respectively, the developing countries in Asia only have a minor decrease of NO_x (VOC) emissions from 6.9 (32.7) to 5.5 MtN (29.3 Mt). The developing countries in the Africa, Latin America, and Middle East even have an increase of NO_x (VOC) emissions from 6.6 (48.3) to 36.2 MtN (83.5 Mt). The developing countries will produce around 2 to 16 times the NO_x and VOC emissions of industrialized countries by 2100. As discussed in section 3, Asia and Latin America are located upstream of two major inflow boundaries of the United States, the western and southern borders. *Berntsen et al.* [1999] and *Jaffe et al.* [1999] have shown that Asian emissions influence air quality over the northwestern United States, especially during the spring. *Jaffe et al.* [2003] has similarly shown that the background ozone is increased in the rural west coast of North America owing to Asian emissions. Even if the United States strengthens its emissions control strategy, the Jaffe et al. results indicate that it is likely that the long-range transport will impact future U.S. air quality, with increases of U.S. background ozone concentrations occurring through increased amounts of ozone and ozone precursor gases being transported across geographical boundaries.

[4] Huang et al. (submitted manuscript, 2008a) has shown that an enhanced control strategy on future North America anthropogenic emissions can greatly impact the U.S. air quality. Huang et al. (submitted manuscript, 2008a) estimated climate change impacts on future air quality projections from the regional air quality modeling system simulations with clean lateral boundary inflows. These clean lateral boundary conditions are derived from initial setting of clean background concentrations in the outmost grids at the four edges of the coarse modeling domain, and then replaced by the regional air quality model simulated values in the grids one cell width further from the edges after a spin-up integration for 3 days, and remain the same throughout the rest of the summer period. Thus the long-range transport of pollutants from remote origins is not incorporated into the regional domain. To explore this potential impact and provide an initial estimate of the long-range transport impact on future air quality changes, this study uses the same regional air quality modeling system as Huang et al. (submitted manuscript, 2008a) and a global chemical and transport model, both driven by the same GCM output. The effect of the long-range transport is introduced by the time-varying chemical lateral boundary conditions extracted from the global chemical and transport model output at the edges of the modeling domain. Section 2 briefly describes the modeling components and the experimental design. The results and discussions are in section 3, followed by the summary and conclusions in section 4.

2. Modeling System and Experiments

2.1. Modeling Components

[5] The Parallel Climate Model [*Washington et al.*, 2000] was used to drive the future regional air quality modeling system simulations. It consists of an atmospheric GCM, an ocean GCM, a land model, and a sea-ice model. It produces a present-day global climate that is comparable to observations [*Washington et al.*, 2000; *Dai et al.*, 2001]. The regional air quality modeling system consists of a regional climate model, an emission model, and a regional air quality model. The RCM is based on the fifth-generation Penn State University/National Center for Atmospheric Research mesoscale modeling system version 3 (J. Dudhia et al., PSU/NCAR mesoscale modeling system tutorial class notes and user's guide: MM5 modeling system version 3, 2000, http://www.mmm.ucar.edu/mm5/documents/MM5_tut_Web_notes/tutorialTOC.htm) with improvements including more realistic buffer zone treatment, ocean interface, and

cloud-radiation interactions [*Liang et al.*, 2001, 2004]. The Sparse Matrix Operator Kernel Emissions modeling system [*Houyoux et al.*, 2000] was the emission model to process the U.S. Environmental Protection Agency National Emissions Inventory data. The regional air quality model was developed from the SARMAP Air Quality Model [*Chang et al.*, 1997] with improvements including a faster and more accurate numerical scheme for solving gas-phase chemistry [*Huang and Chang*, 2001]. The Regional Acid Deposition Model Mechanism version 2 (RADM2) [*Stockwell et al.*, 1990] with updated reaction coefficients was used in this study. *Huang et al.* [2007, also submitted manuscript, 2008a] showed that the regional air quality modeling system is capable of simulating the present-day tropospheric ozone formation on the regional and monthly means. The diurnal cycles of observed ozone were realistically reproduced over the selected regions during 1996–2000 summers. The regional air quality modeling system tended to overestimate daily mean ozone mixing ratios in the Northeast (+3 to +10 ppbV) and Midwest (−1 to +5 ppbV) United States. It captured the diurnal ozone evolution very well in the Texas subdomain, but underestimated daily mean ozone mixing ratios in the California subdomain (−4 to −8 ppbV).

[6] With the same meteorology conditions, the inclusion of the aerosol mechanism has a minimal impact on the chemical production of tropospheric ozone. The reason is that the present aerosol formation is an added product on the gas-phase chemical mechanism. It does not alter the oxidants and ozone-precursors fields unless the nighttime heterogeneous formation involving N_2O_5 is considered. The initial results show that the difference is up to 4 ppbV over U.S. continental area and mostly in the area of low daily maximum ozone mixing ratio. The aerosol could have both direct and indirect feedback on climate change. With changing meteorology, aerosols can impact tropospheric ozone formation by transport, photochemical reactivity, and emissions. The indirect feedback that involves the cloud effect could be large, but cloud feedbacks remain among the largest sources of uncertainty in climate models. The Parallel Climate Model simulation used in this study did not contain the feedback mechanism from aerosols that is also not included in the GCM simulations of *Hogrefe et al.* [2004] and *Murazaki and Hess* [2006]. Because the noninteractive approach or one-way forcing was adopted for the RCM meteorology driven by Parallel Climate Model simulations as well as the regional air quality model simulation driven by regional climate model meteorology, the aerosol module was turned off in the regional air quality model to accommodate aggressive regional air quality model simulations described in section 3. More detailed description of the regional air quality modeling system as well as the global reanalysis data set [*Kanamitsu et al.*, 2002] and Parallel Climate Model is given by *Huang et al.* [2007, also submitted manuscript, 2008a] (see also H.-C. Huang et al., Simulation of continental U.S. air quality on climatic timescales under present-day conditions, submitted to *Journal of Geophysical Research*, 2008) (hereinafter referred to as Huang et al., submitted manuscript, 2008b).

[7] The global chemical and transport model employed here is the Model for OZone And Related Chemical Tracers (MOZART-2.4) [*Horowitz et al.*, 2003], which simulates 63 species, 135 gaseous reactions and 26 heterogeneous processes at the global scale and includes a global emissions database for the species involved in ozone chemistry. The advection, surface emission/deposition, vertical diffusion, convection, cloud/precipitation and chemistry are integrated at 15 min steps. The model has 18 sigma-pressure (σ-p) hybrid levels. Simulations were conducted with both the T62LR horizontal resolution ($\sim$1.9° or 200 km) driven by global reanalysis and T42LR horizontal resolution ($\sim$2.8° or 300 km) driven by Parallel Climate Model for the present-day climate and T42LR horizontal resolution driven by Parallel Climate Model for the future climate. The midpoint of the top layer is about 3.0 hPa. The spin-up of MOZART started about 1.5 years before the first summer for investigation, for example, started at December 1994 for the 1996–2000 period. *Lin et al.* [2008b] showed that the MOZART simulated summer mean surface ozone mixing ratios are within 20 ppbV of observed ozone mixing ratio over most of the United States. The global chemical and transport model simulations are independent from the regional air quality modeling system and provide an assessment of the sensitivity of the local ozone-climate relationships to long-range transport of pollutants such as from the East Asia.

[8] Huang et al. (submitted manuscripts, 2008a, 2008b) have shown that future air quality changes are sensitive to the incorporated regional climate model cumulus parameterization scheme. In general, the *Grell* [1993] cumulus parameterization scheme produced a more realistic surface ozone distribution with fewer over-estimates of daytime ozone mixing ratio in the present-day climate, especially in the eastern United States (Huang et al., submitted manuscript, 2008a). In this study, only the Grell scheme was used by the regional climate model. Both the global chemical and transport model and regional climate model meteorology were driven either by the global observations (global reanalysis) or by the Parallel Climate Model output in the present-day climate simulations. For the future climate simulations, both the GTCM and regional climate model meteorology were driven by the Parallel Climate Model output.

2.2. Experiments

[9] The regional air quality modeling system integrated the chemical and transport processes in a nested computational domain (Figure 1). The coarse domain has a 90-km grid spacing and extends from the Pacific to the Atlantic Ocean, including much of Canada and Mexico. The four inner subdomains have a 30-km grid spacing that covers the northeast and midwest United States, California, and Texas. Subsequent comparisons between simulations were based on the nonoceanic grids within 30-km subdomain values. The southeast United States has been shown by Huang et al. (submitted manuscript, 2008a) to have the most substantial air quality changes. Therefore, the air quality changes due to the long-range transport in this area also will be discussed on the basis of the 90-km results.

[10] To compare with the results of Huang et al. (submitted manuscript, 2008a, 2008b), the simulation period is 1996–2000 (denoted as 2000s) for the present-day climate simulations, and 2048–2052 (denoted as 2050s) and 2095–2099 (denoted as 2090s) for the future climate simulations.

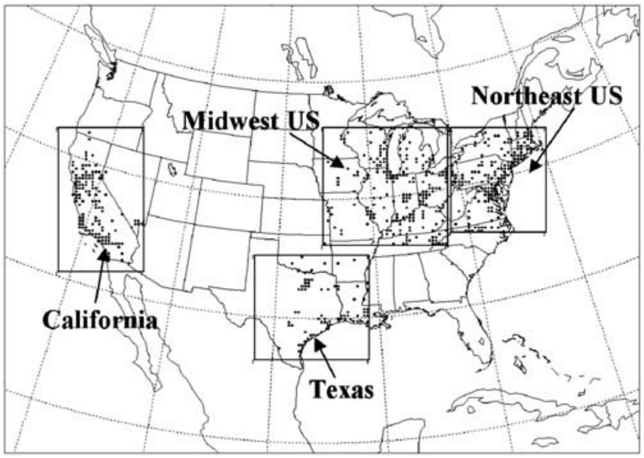

Figure 1. Nested modeling domains with 90-km grid size (outer mesh) and 30-km grid size boundary (inner boxes) for the northeast United States, midwest United States, California, and Texas as well as the selected area of the southeast United States. The dots represent the location of observations.

The regional air quality modeling system and global chemical and transport model were each run for five summers (June through August) in the present-day climate (2000s, control) and five summers for each of the 2050s and the 2090s. Summertime mean ozone mixing ratios were calculated for the selected regions over five summers. Each future simulation was compared with the control simulation to assess projected changes in ozone mixing ratios. For model evaluation purposes, surface hourly O_3 measurements were obtained from the USEPA Air Quality System (AQS; http://www.epa.gov/ttn/airs/airsaqs/detaildata/downloadaqsdata.htm). As detailed by Huang et al. (submitted manuscript, 2008b), model-simulated O_3 mixing ratios in each target area were compared with the corresponding observed values averaged for all monitoring stations in the regional air quality model grid cell containing the target area. A set of criteria defined by *Huang et al.* [2007] was also applied to select qualifying monitoring grid cells for comparison. The selected grids contain more than 95% of hourly records of each year. The selected qualifying grids are shown in Figure 1.

[11] Both the regional air quality modeling system and MOZART simulations examined the impact of climate change on ozone mixing ratios from the IPCC SRES emissions scenarios. In the future climate simulations, the Parallel Climate Model outputs were available for two emissions scenarios described in the IPCC SRES, the A1Fi and the B1. In general, the A1Fi is a more polluted scenario than the B1 scenario. They represent the upper and lower bounds of the most likely future scenarios projected by the IPCC. The A1Fi and B1 scenarios will be referred to as polluted and clean scenarios in the following discussion. The regional air quality modeling system projected the future anthropogenic emissions on the basis of the present-day values of anthropogenic emissions. The projected changes of NO_x and VOC in the 2050s and the 2090s were detailed by Huang et al. (submitted manuscript, 2008a). The NO_x (VOC) emissions changes of United States and Canada ranged from −58% to +33% (−42% to +3%) in the 2050s and −83% to 100% (−64% to +67%) in the 2090s. The projected future Mexico emissions generally had increases for both NO_x and VOC, except for the VOC in the clean scenario (Table 1). Future biogenic emissions from vegetation, based on land-use categories, were allowed to change in response to the simulated regional climate model-meteorology. Because of the complex issues that impact the land-use changes, the uncertainty of the existing land-use model projection is substantial. As discussed extensively by Huang et al. (submitted manuscript, 2008a), the land-use data were assumed to be the same both in the present-day and future climate simulations.

[12] Future emissions changes of the MOZART simulations include anthropogenic, biomass burning, and biogenic/soil sources. Changes in long-lived greenhouse gases and ozone precursors are driven explicitly by the changes in energy-related anthropogenic and biomass burning emissions as projected by the polluted and clean scenarios. Changes in anthropogenic emissions are based on the calculations of the socio-economic models, MiniClimate Assessment Model (MiniCAM; http://www.globalchange.umd.edu/models/minicam) and Integrated Model to Assess the Global Environment (IMAGE) model (http://www.mnp.nl/image). Changes in biogenic VOC emissions are primarily based on their dependence on temperature [*Guenther et al.*, 1994; *Guenther*, 1997] and the CO_2 level [*Potosnak*, 2002]. The projected increase of global mean surface temperature for Parallel Climate Model is about 2.1C in the 2090s. Therefore, it is expected that biogenic emissions will have an increase. Oceanic and aircraft emissions for all species and soil emissions for NO_x are not changed owing to the great uncertainties. The projected future emissions changes between the MOZART and regional air quality modeling system are different (Table 1). It is because the regional modeling system used the IPCC SRES standardized emission scenarios that utilize six modeling approach [see *Special Report on Emissions Scenarios*,

Table 1. Relative Changes of Future Emissions Related to Present-Day Values (2000s) for MOZART and Regional Air Quality Modeling System in the Areas of United States and Canada, Mexico, and Outside North America[a]

		MOZART			RAQM	
Species	Period	United States and Canada	Mexico	Outside North America	United States and Canada	Mexico
NO_x	2050s	−6 (4)	23 (31)	20 (32)	−58 (33)	125 (213)
NO_x	2090s	−11 (16)	7 (44)	1 (16)	−83 (100)	25 (313)
VOCs	2050s	12 (26)	17 (33)	19 (31)	−43 (3)	−15 (100)
VOCs	2090s	22 (69)	30 (81)	26 (67)	−64 (67)	−35 (116)

[a]Changes given as percent. RAQM, regional air quality modeling system. The values are for the IPCC B1 (cleaner) scenario while the values in the parentheses are for the IPCC A1Fi (more polluted) scenario.

2000, Appendix 4] while only two of the six models (MiniCAM and IMAGE) were used in the MOZART emissions projection. These six models used in IPCC SRES scenarios are different in modeling emissions scenarios and integrated assessment framework.

[13] The U.S. fossil fuel emissions increase under the polluted scenario and decrease under the clean scenario. However, the biofuel emissions that are related to the VOC emissions increase under the clean scenario. The MOZART future emissions projections are similar to those of *Lin et al.* [2008a] and are summarized in Table 1. The U.S. and the Canada NOx emissions have increases of −6% to +4% in the 2050s and −11% to +16% in the 2090s under the clean and polluted scenarios, respectively. Both the Mexico and the countries outside North America have general increases ranged from 20% to 32% in the 2050s and 1% to 44% in the 2090s under the clean and polluted scenarios, respectively. The global VOC emissions increase substantially in future climate both in the 2050s (26% to 33%) and the 2090s (67% to 81%) under the polluted scenario. The United States, Canada, and the countries outside North America have a similar increase while Mexico has the highest increase. Under the clean scenario, the VOC emissions from the United States and Canada increase from 12% in the 2050s to 22% in the 2090s. For Mexico and countries outside of North America, VOC emissions increase up to 19% in the 2050s and to 30% in the 2090s.

[14] The MOZART emissions were based on the global emissions data sets that are slightly different from those of the regional air quality modeling system which are based on the regional emissions reported by the United States, Mexico, and Canada. It is noted that the differences would lead to different ozone production levels within the modeling domains. However, these emissions differences mainly impacted the ozone production within the United States and should have more impact on the outflow grids along the eastern boundary than others (because of eastward synoptic systems movement). Because of the longer timescale simulation conducted in this study, it is likely that the recirculation of MOZART North America emissions changes can impact the inflow lateral boundary conditions of North America domain, especially from the western and southern boundaries. Because the North America outflow immediately mixes with air parcels in downstream areas and is subsequently modified by the local emissions, it is difficult for MOZART to distinguish the magnitude between the influence of emissions changes within and outside of North America. Therefore, the long-range transport impact discussed in this study should be considered as the integrated impact of intercontinental transport on the lateral boundary conditions of regional air quality modeling system including the global changes of emissions, chemical compositions and reactivities, and loss. The pollutant and precursor gases produced outside the United States still can be detected, as discussed in section 3, from the inflow along model boundaries. This global chemical and transport model data set provides us an opportunity to study the impacts of the pollutant productions and/or emissions outside the United States on the U.S. air quality.

[15] The output values of MOZART, available at the 3-h interval, were mapped from its coarse grids (∼200 km as driven by the global reanalysis and ∼300 km using the Parallel Climate Model meteorology) onto the regional air quality model 30-km grids. First, the fractional values based on the overlapping area of each MOZART coarse grid in a regional air quality model grid were generated. Then, the mixing ratio of the regional air quality model grid was obtained by adding the contributions of overlapped MOZART grid(s). This is to conserve the mass especially near the boundary of two coarse grids. The mass-weighted averaging in the vertical was used to map the species mixing ratios from the MOZART 18 layers to the regional air quality model 15 layers. The top boundary of the regional air quality model is at about 100 hPa. For the organics, based on the chemical characteristics, the MOZART species are either grouped or partitioned into the regional air quality model species.

3. Results and Discussions

3.1. Present-Day Ozone Simulation

[16] MOZART is designed to simulate global transport of chemically reactive species and their reactions. It uses a coarser spatial resolution than the regional air quality model and thus its representation of chemical characteristics is expected to be less detailed. By design, the MOZART provides the lateral boundary conditions for the regional air quality model to downscale air quality simulation over the United States. Before this coupling takes place, it is imperative to examine the consistency between the MOZART and regional air quality model simulations under the same meteorological conditions. The present-day climate MOZART simulations, as driven by both the global reanalysis and Parallel Climate Model meteorology, produces higher ozone mixing ratios than the regional air quality modeling system over most of the regional air quality model domain except for the northwest corner (not shown). The differences are especially large (> 20 ppbV) in the Northeast corridor, areas around Lake Erie, Michigan, Florida, Texas and Louisiana, and southern California. Hot spots with the largest differences (> 30 ppbV) are near the major metropolitan areas. There is a difference between emissions used in regional air quality model simulations and that in MOZART simulations over the area of continental United States. The comparison showed that regional emissions had 57% more NO_x emissions and 44% less isoprene emissions than the MOZART emissions over the area of continental United States. The differences of CO (+11%) and other VOC emission budgets (−10%) between the two data sets are relatively small. The local emissions influence can be masked owing to the coarse MOZART grid resolution where the dominant urban NO_x emissions and the higher rural VOC emissions are mixed together. The artificial mixing can produce the favorable NO_x to VOC ratio for higher ozone production. Given the finer spatial resolution, the regional air quality modeling system is believed to correctly allocate more urban NO_x emissions than the global MOZART emissions database. This allows the regional air quality model to generate lower urban ozone mixing ratios than the MOZART owing to the O_3 titration effect.

[17] Figure 2 illustrates the vertical profiles of the clean lateral boundary conditions for O_3, NO_x, and non-methane volatile organic compounds (NMV). The control regional air quality model integration with such clean lateral boundary

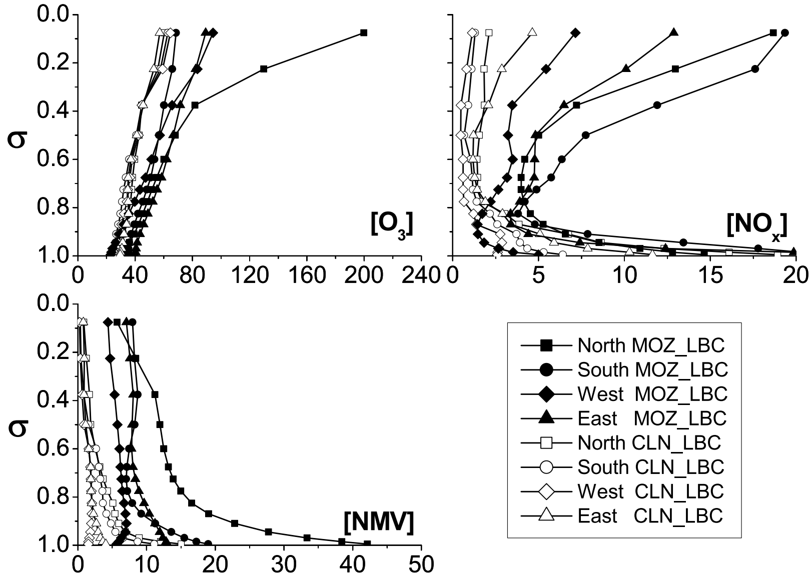

Figure 2. Vertical profiles (in sigma coordinate) of ozone (O_3, in ppbV), nitrogen oxides (NO_x, in 10^{-2} ppbV), and nonmethane volatile organic compounds (NMV, in ppbC) of the clean (CLN) and MOZART (MOZ) lateral boundary conditions averaged along individual lateral boundaries.

conditions is driven by the regional climate model downscaled meteorology from the global reanalysis in the 2000s. To investigate the impacts of long-range transport on the U.S. air quality, parallel regional air quality model runs are conducted where the time- and space-varying lateral boundary conditions are constructed from the MOZART outputs driven by the same global reanalysis.

[18] The vertical distributions of MOZART O_3, NO_x, and NMV mixing ratios averaged within each boundary and during the entire simulation period also are shown in Figure 2. Substantial differences exist for all boundaries. The MOZART lateral boundary conditions generally have higher O_3 mixing ratios than the clean lateral boundary conditions in vertical, especially in the upper troposphere at the northern boundary. This is partially because the mapped MOZART upper layers are already in the stratosphere in the northern portion of the regional air quality model modeling domain. The MOZART lateral boundary conditions have substantially higher NO_x mixing ratios in the upper troposphere, especially at the southern and western boundaries. This is the strong indication of the long-range transport from the anthropogenic pollution originating in Latin America and Asia. Similar phenomena also were found by *Jaffe et al.* [2003], *Parrish et al.* [2004], *Liang et al.* [2007], and field campaigns such as the Transport and Chemical Evolution over the Pacific (TRACE-P; http://hyperion.gsfc.nasa.gov/Missions/TRACEP/index.html) and Intercontinental Chemical Transport Experiment (INTEX-B; http://www.espo.nasa. gov/intex-b/index.html).

[19] Figures 3a, 3b, and 3c compare the regional and 5-summer mean ozone diurnal cycles observed and simulated by the regional air quality model and the MOZART as driven by the global reanalysis over the qualified grids where the observations are sufficient (see Huang et al., submitted manuscript, 2008b). The data point under each year represents the domain-average summer mean surface ozone mixing ratio for each of 24 h. For the 30-km subdomains, the regional air quality model with the clean

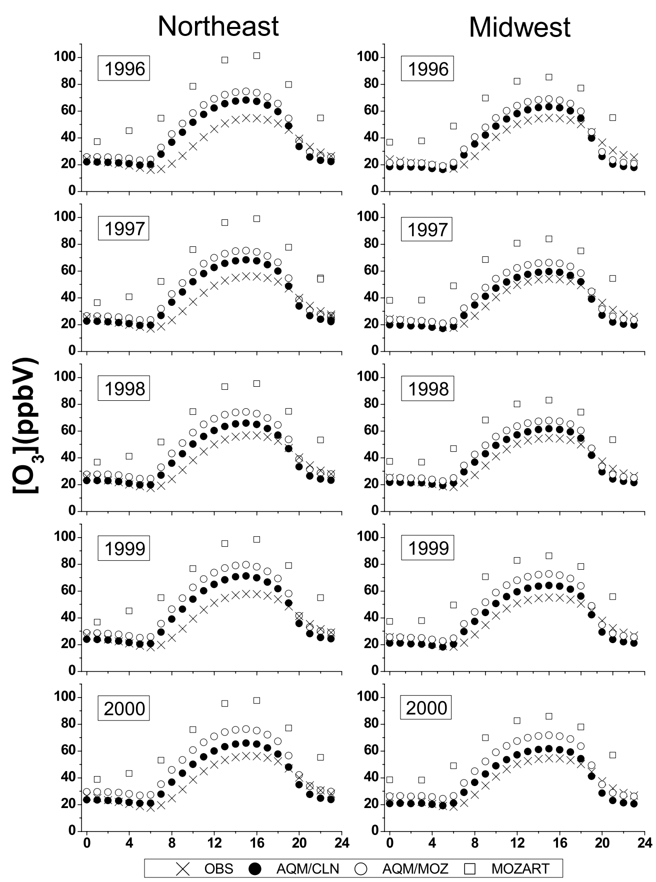

Figure 3a. Summer mean surface ozone diurnal cycles of observed (OBS) and simulated by the regional air quality model with the clean lateral boundary conditions (AQM/CLN), regional air quality model with the MOZART lateral boundary conditions (AQM/MOZ), and MOZART as driven by the global reanalysis during 1996–2000 for the (left) northeast and (right) midwest United States. All calculations are done over the selected grids where observations are sufficient.

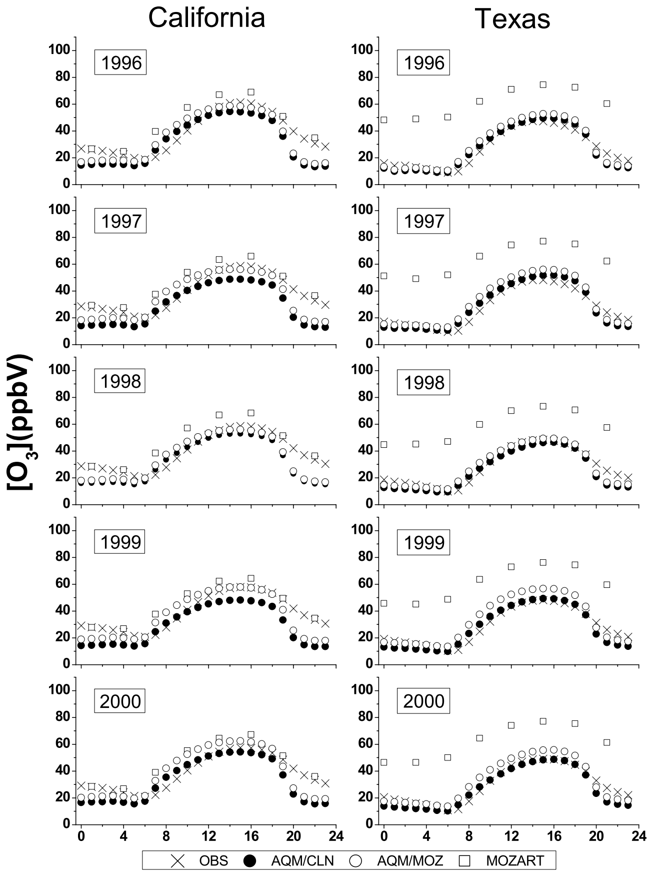

Figure 3b. Similar to Figure 3a but for (left) California and (right) Texas, United States.

lateral boundary conditions captures very well the phase of the diurnal cycle for all subdomains. It overestimates daily mean ozone in the northeast United States while underestimating that in California, with an overall bias of +5 and −8 ppbV, respectively. The regional air quality model simulation with the clean lateral boundary conditions is fairly realistic in the midwest United States and Texas, with an overall bias of +2 and −1 ppbV, respectively. On the other hand, the MOZART simulation is much less realistic, producing a general overestimation of substantially larger magnitudes, with overall positive biases of 26, 31, and 33 ppbV respectively in the midwest United States, northeast United States, and Texas subdomains; one exception is in the California subdomain where the MOZART simulation is slightly better than the regional air quality model simulation with the clean lateral boundary conditions. The regional air quality model simulations with the clean lateral boundary conditions in the southeast United States, based on the 90-km results, also show a substantially higher daytime overestimation similar to the MOZART. However, the regional air quality model with the clean lateral boundary conditions simulated more realistic nighttime values.

[20] Figures 4a, 4b, and 4c show the results when both models are driven by the Parallel Climate Model meteorology. As compared with Figures 3a–3c, the Parallel Climate Model-driven models simulate lower ozone mixing ratios in the northeast, midwest, and southeast United States as well as the Texas subdomain, while higher values in the California subdomain. This is a systematic feature as illustrated in Figure 5, where the tendency is consistent between the regional air quality model with the clean lateral boundary conditions and MOZART. The result suggests that the meteorology influence on the ozone simulation is regionally dependent. Overall the regional air quality model with the clean lateral boundary conditions produces smaller biases in the northeast United States, California, and southeast United States as driven by the Parallel Climate Model meteorology than the global reanalysis, while the performance in the midwest United States and Texas subdomains remain excellent for both cases. Although a similar improvement occurs in the MOZART as driven by the Parallel Climate Model meteorology, the model biases are still substantially large, ranging from +11 ppbV in the California subdomain to +31 ppbV in the southeast United States.

[21] The difference between the MOZART and regional air quality model simulations with the clean lateral boundary conditions is systematic and substantial. With the finer resolution and better simulated regional circulation, the regional air quality model is able to improve substantially the ozone simulation over most of the continental United States. Even with the inclusion of long-range transport estimated by the MOZART output, as described later in this section, the result remains the same. The small differences in both model results as driven by the Parallel Climate

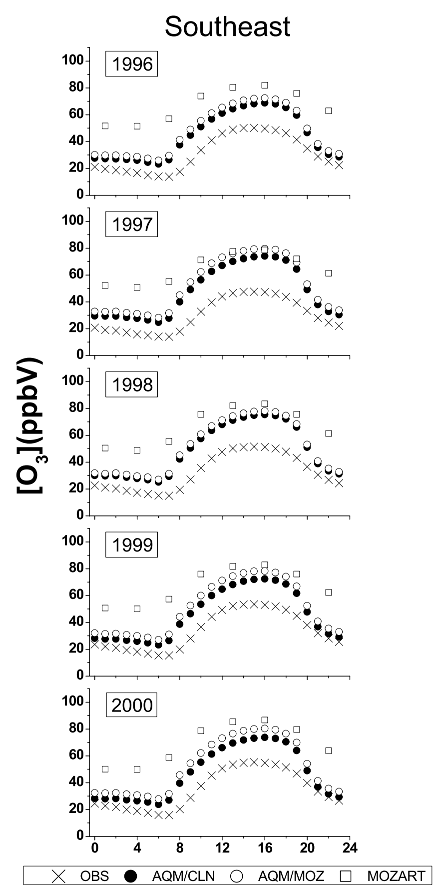

Figure 3c. Similar to Figure 3a, but for the southeast United States.

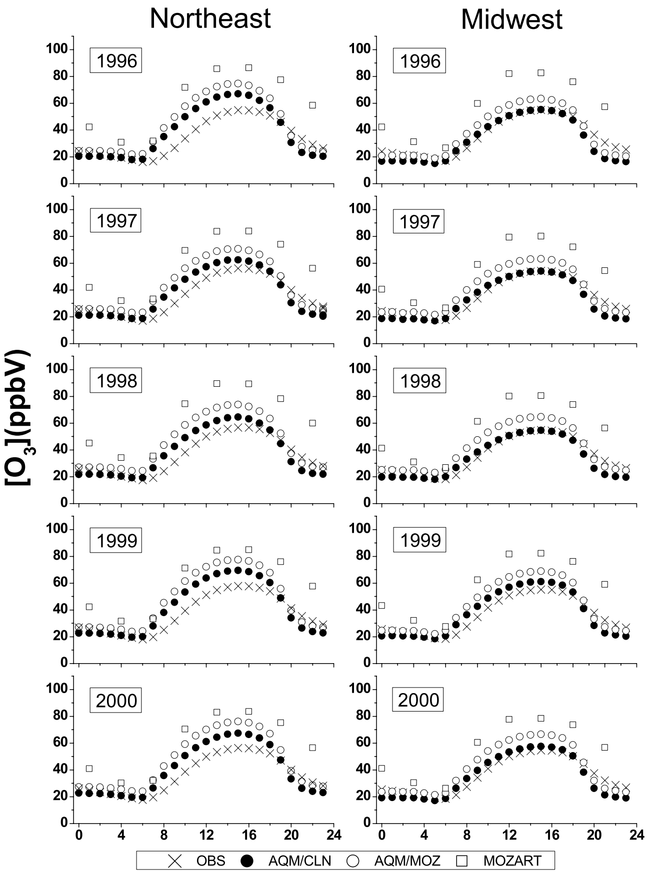

Figure 4a. Similar to Figure 3a, but for simulations driven by the Parallel Climate Model meteorology.

Model simulation versus global reanalysis assimilation suggest that the Parallel Climate Model meteorology is adequate for the regional air quality modeling system to produce realistic downscaling of the U.S. air quality. Given the general overestimation of ozone mixing ratio of the MOZART simulations, the selected global chemical and transport model that provides chemical lateral boundary conditions to the regional air quality model should be carefully studied to properly understand the impacts of long-range transport on the U.S. air quality.

[22] To study the roles of pollutant transports across the individual and combined boundaries, the clean lateral boundary conditions were replaced by the MOZART lateral boundary conditions separately in individual and all four boundaries. Table 2 summarizes the regional air quality model-simulated (driven by the global reanalysis) changes of daily mean ozone mixing ratio averaged during the summers of 2000s over the selected areas. These changes are calculated relative to the control values, i.e., the clean lateral boundary conditions. Clearly, the air quality simulations in the northeast and midwest United States are greatly influenced by the northern lateral boundary conditions, whereas that in Texas (California) is dominated by the southern (western) lateral boundary conditions. Although the magnitude of the gross mean mixing ratio at the lateral boundaries is important, specific locations of where the species are actually transported into the regional domain is critical. Figure 6 shows the regional air quality model-simulated differences in daily mean O_3 mixing ratio caused by replacing the clean with the MOZART lateral boundary conditions in the southern boundary only and in all four boundaries. They are averaged during the summers of present-day climate. Clearly, most of the long-range pollutant transport is from Canada across the northern boundary and from South America along the western Gulf coast and Central America toward Texas across the southern boundary. When the MOZART lateral boundary conditions are incorporated, the regional air quality model produced a general increase in daily mean O_3 mixing ratio over North America, especially along the northern United States and southern Canada where the substantially higher upper tropospheric O_3 mixing ratio is suspected to mix downward into the surface layer.

[23] Figures 3a–3c and 4a–4c also compare the summer mean O_3 diurnal cycle averaged during the 2000s over all qualified grids within the four subdomains and the Southeast United States between observed values and values simulated by the regional air quality model using the regional climate model meteorology driven by the global reanalysis and Parallel Climate Model with the clean and MOZART lateral boundary conditions across all boundaries, respectively. Over all selected areas, the MOZART lateral boundary conditions elevate the O_3 mixing ratio throughout the day, especially in the afternoon. Figure 7 shows that the regional air quality model ozone biases are substantially enhanced in the northeast, midwest, and

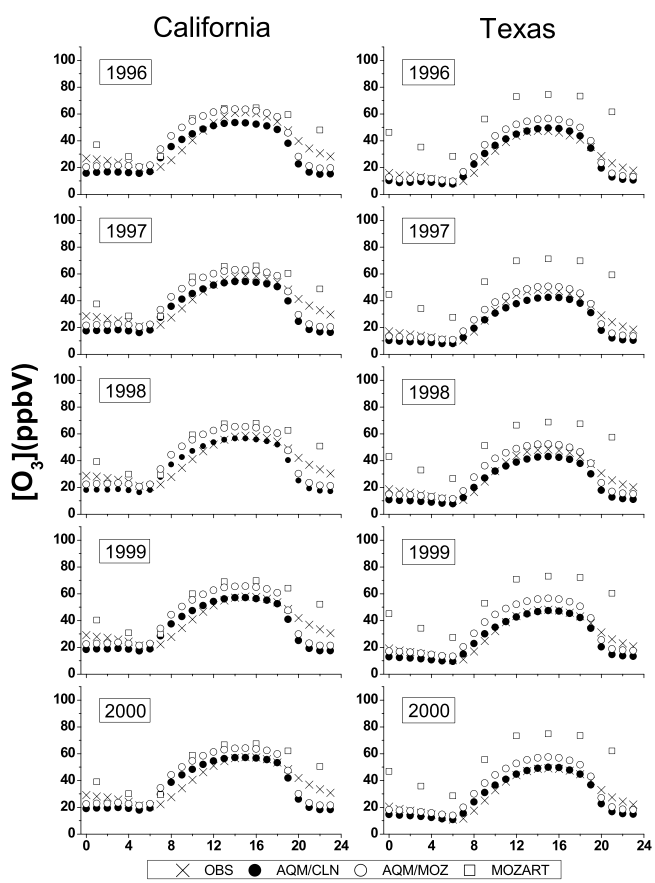

Figure 4b. Similar to Figure 3b, but for simulations driven by the Parallel Climate Model meteorology.

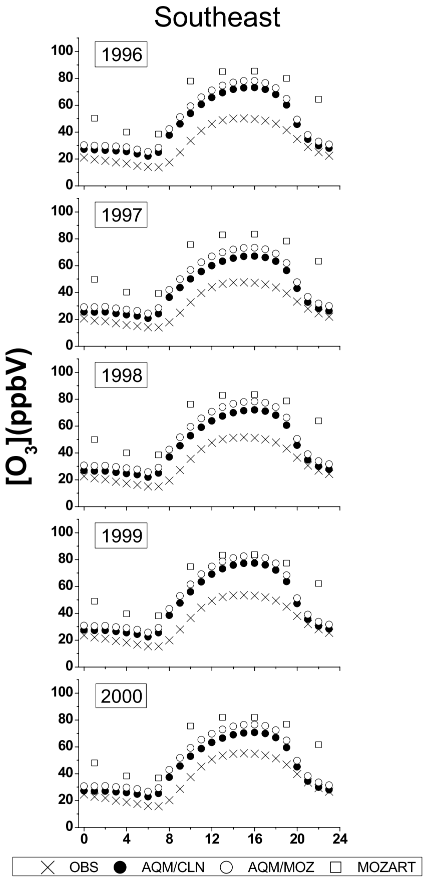

Figure 4c. Similar to Figure 3c, but for simulations driven by the Parallel Climate Model meteorology.

southeast United States by using the MOZART versus clean lateral boundary conditions. The enhanced regional air quality model overestimation results mainly from the much higher upper tropospheric ozone mixing ratio in the MOZART northern lateral boundary conditions, which may likely be unrealistic. In Texas, the small underestimation by the regional air quality model simulations with the clean lateral boundary conditions becomes an overestimation in the regional air quality model simulations with the MOZART lateral boundary conditions. Only in California, the regional air quality model simulation with the MOZART lateral boundary conditions is better, where the large underestimation is reduced by a factor of 3, from −8 to −3 ppbV. The regional increase due to the long-range transport (regional air quality model with the MOZART lateral boundary conditions minus regional air quality model with the clean lateral boundary conditions) is similar among all regions, ranging from +4 to +6 ppbV (+5 to +7 ppbV) with the average of 5 ppbV (6 ppbV) driven by the global reanalysis (Parallel Climate Model) meteorology. We speculate that the MOZART may capture the observed ozone distribution (higher than the clean assumption) in the western boundary that helps to reduce the regional air quality model underestimation in the California subdomain. The mean increase of daily maximum 8-h average ozone mixing ratio in 2000s ranges from +6 to +10 ppbV. The magnitude of impact is similar to those found by *Fiore et al.* [2002].

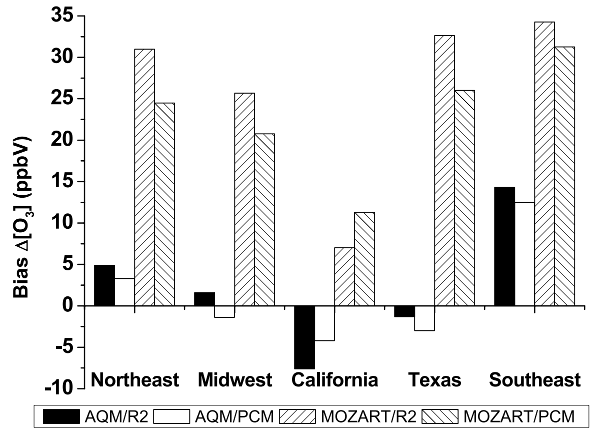

Figure 5. Regional air quality model (AQM) with clean lateral boundary conditions and MOZART biases (modeled minus observed) of summer daily mean ozone (ppbV) averaged during 1996–2000 over each of the five subdomains as driven by the global reanalysis (R2) and Parallel Climate Model (PCM) meteorology.

[24] Figure 8 shows the vertical profile of regional and 5-summer mean ozone mixing ratio of the regional air quality model simulations with the clean lateral boundary conditions and with the MOZART lateral boundary conditions using the regional climate model meteorology driven by the global reanalysis and Parallel Climate Model output. In general, the regional air quality model simulations with the MOZART lateral boundary conditions repeatedly produced more ozone mixing ratio than the regional air quality model with the clean lateral boundary conditions in the entire column with substantial increases in the upper troposphere. This is attributed to the persistent influence of boundary condition differences in the upper troposphere if there are no modeled sources in the upper troposphere [*Liu et al.*, 2001]. The regional air quality model with the MOZART lateral boundary conditions simulations produced similar profiles between the global reanalysis and Parallel Climate Model meteorology.

[25] The aforementioned sensitivity study showed that long-range pollutant transport clearly plays an important role on the regional U.S. air quality. The ozone mixing ratio simulated by the MOZART is more realistic than the regional air quality model in California but much worse with substantially larger overestimations in the other regions. Much of the degradation results from the long-

Table 2. Relative Changes of Daily Mean Ozone Mixing Ratio Averaged During the 1996–2000 Summers Over the Selected Subdomains as Compared to the Results Using Clean Lateral Boundary Conditions at Four Model Boundaries[a]

MOZART LBC	Northeast United States	Midwest United States	California	Texas	Southeast United States
Northern only	+9	+9	+4	−2	+2
Southern only	−2	−2	−4	+5	−1
Western only	+2	+2	+10	0.	+1
Eastern only	−2	−3	−5	−1	+1
All four boundaries	+15	+15	+19	+14	+9

[a]Relative changes given as percent. LBC, lateral boundary conditions.

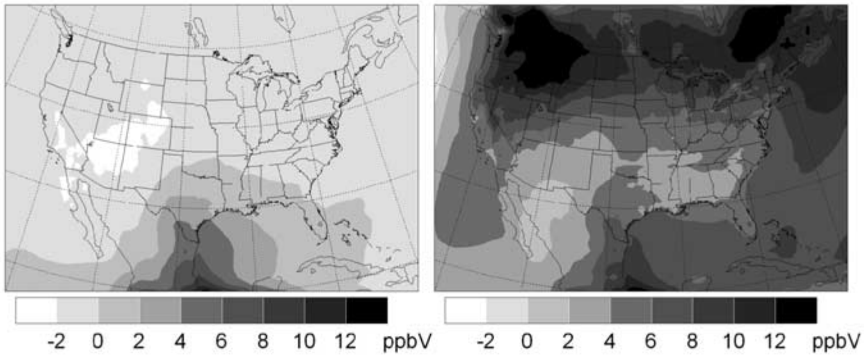

Figure 6. Regional air quality model-simulated differences in summer daily mean ozone mixing ratio averaged during 1996–2000 caused by replacing the clean lateral boundary conditions with the MOZART lateral boundary conditions (left) only in the southern boundary and (right) for all four boundaries.

range transport across the northern and southern boundaries, where pollutants originated in Canada and Mexico are important contributors. More rigorous validation is warranted for the global chemical and transport models in reproducing the observed vertical distributions along the boundaries before their outputs can be used credibly to provide chemical lateral boundary conditions driving the regional air quality model for downscaling. The results identified an additional uncertainty of using the global chemical and transport model in climatic air quality studies. The baseline (present-day climate) simulations must include the same global chemical and transport model used in the future climate projections. For the same reason, extra caution must be taken in conducting sensitivity experiments and interpreting the results in terms of how the long-range transport affects the U.S. air quality under the future projection scenarios of global climate and emissions changes.

3.2. Future Ozone Change Projection

[26] Given the substantial biases and inter-model difference in simulating the present-day ozone mixing ratio, it is prudent to examine the surface O_3 change against its own control in future projection. Followed the approach detailed by Huang et al. (submitted manuscript, 2008b), the regional air quality model simulations with the MOZART lateral boundary conditions driven by the Parallel Climate Model meteorology in the present-day climate were used as the

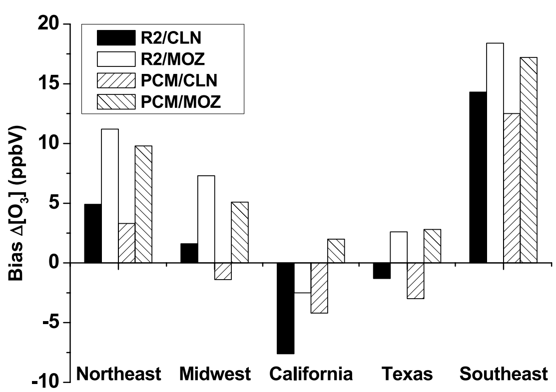

Figure 7. Regional air quality model biases (modeled minus observed) of summer daily mean ozone (ppbV) averaged during 1996–2000 over the five subdomains using the clean (CLN) and MOZART (MOZ) lateral boundary conditions with regional climate model meteorology driven by the global reanalysis (R2) and Parallel Climate Model (PCM) output.

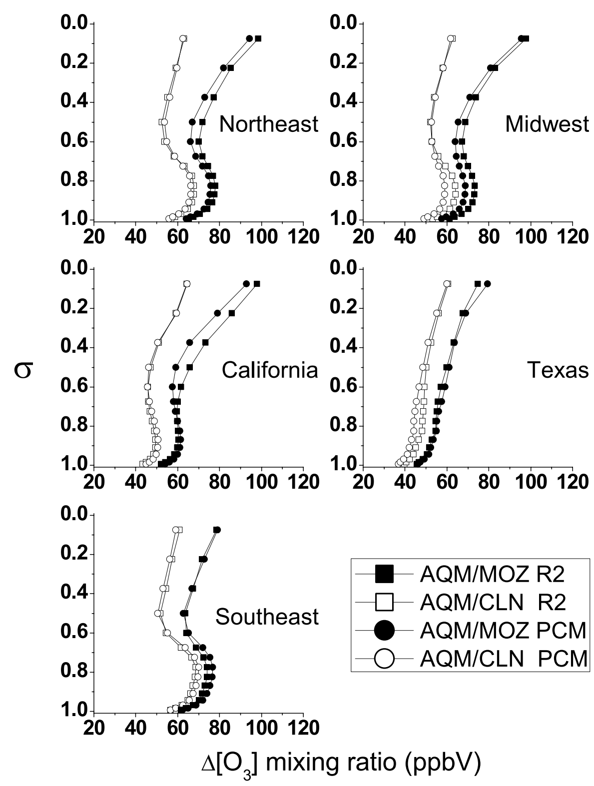

Figure 8. Vertical distribution (in sigma coordinate) of regional five-summer mean ozone mixing ratio over the five selected regions of regional air quality model (AQM) simulations using the regional climate model meteorology driven by the Parallel Climate Model (PCM) output and global reanalysis (R2) with clean (CLN) and MOZART (MOZ) lateral boundary conditions in the present-day climate.

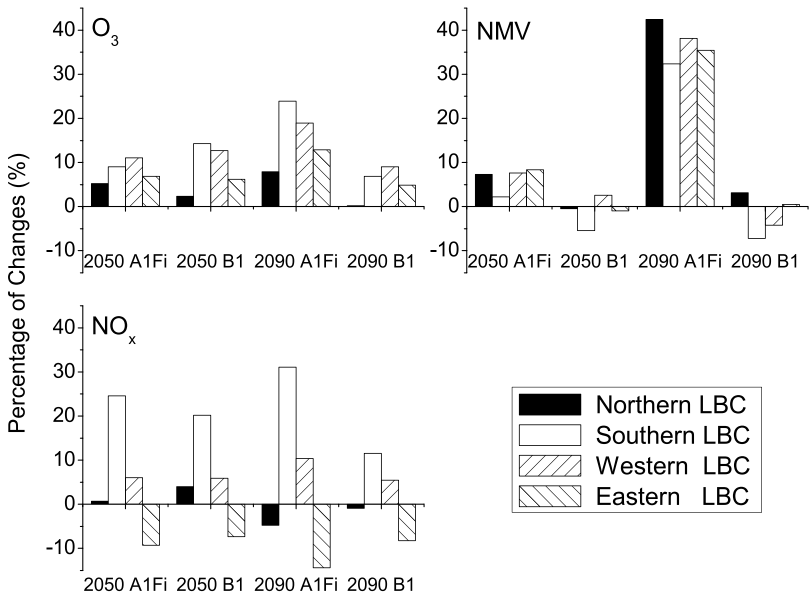

Figure 9. Changes of MOZART lateral boundary conditions from the 2000s to the 2050s and the 2090s under the polluted (A1Fi) and clean (B1) emissions scenarios for ozone (O_3), nitrogen oxides (NO_x), and nonmethane volatile organic compounds (NMV).

control. The relative changes of long-range transport impact were derived from the comparison between the control and the regional air quality model simulations with the MOZART lateral boundary conditions in the future climate. Figure 9 shows the transition of the MOZART lateral boundary conditions from the 2000s to the future climate (the 2050s and the 2090s) under the polluted and clean emissions scenarios for species of O_3, NO_x, and NMV. The air masses transported across all boundaries generally are more polluted in the polluted scenario than in the clean scenario. The air masses were dirtier (cleaner) in the 2090s than the 2050s for the polluted (clean) scenario. The MOZART O_3 lateral boundary conditions for both emissions scenarios in the future climate increased, especially on the western and southern boundaries. The increase can be as high as +24% (southern boundary) under the polluted scenario in the 2090s. It indicates that foreign countries may have a trend of net ozone increase regardless of the U.S. emissions control strategy in the future climate. This will elevate the U.S. background ozone concentration. The NO_x lateral boundary conditions showed a more substantial increase in the southern boundary (+12% to +31%), and it mainly comes from the Mexico and Central America. The NMV lateral boundary conditions show a huge increase (~ 37%) under the polluted scenario in the 2090s at all boundaries. Changes in NO_x and NMV lateral boundary conditions indicate that enhanced U.S. ozone production can be anticipated with the increased transport of ozone precursor gases across the boundaries, especially from the south and west.

[27] Figure 10 shows the projected changes of regional and 5-summer mean ozone mixing ratios in the 2050s and the 2090s. Figure 10 includes the results of the regional air quality model simulations with the clean lateral boundary conditions and with the MOZART lateral boundary conditions under the polluted and clean scenarios, MOZART

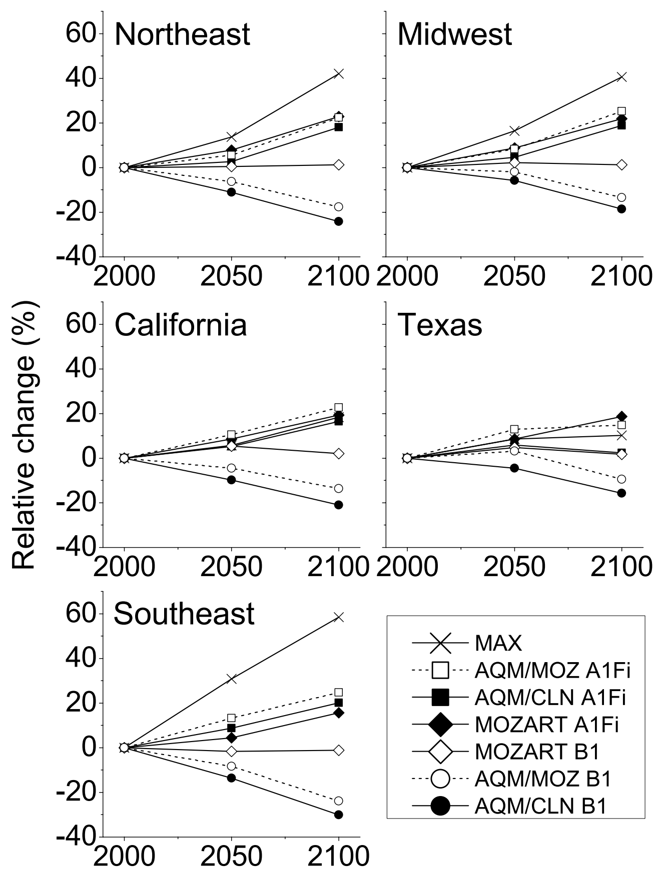

Figure 10. Changes in daily mean ozone mixing ratio from the present to future (the 2050s and the 2090s) climate over the five areas as simulated by the regional air quality model (AQM) using clean (CLN) and MOZART (MOZ) lateral boundary conditions as well as MOZART under the polluted (A1Fi) and clean (B1) emissions scenarios. The upper bound of changes in regional air quality model simulations of Huang et al. (submitted manuscript, 2008a) is marked as MAX.

Table 3. Net Long-Range Transport Impact, Difference of the Projected Changes (From the 2000s to the 2050s and the 2090s), in Five-Summer Mean Surface Ozone Mixing Ratio Under the Polluted and Clean Emissions Scenarios for the Selected Regions[a]

	2050s Polluted	2050s Clean	2090s Polluted	2090s Clean
Northeast United States	3	5	4	7
Midwest United States	4	4	6	5
California	5	5	6	7
Texas	7	8	13	6
Southeast United States[b]	4	5	5	6

[a]Difference is defined as the regional air quality model with the MOZART lateral boundary conditions minus the regional air quality model with the clean lateral boundary conditions, in percent.
[b]Use the interpolated 90-km results.

simulations under the polluted and clean scenarios, and the upper bound of the future projected increase from Huang et al. (submitted manuscript, 2008a) (denoted as MAX). The modeling setting of case MAX is similar to regional air quality model with the clean lateral boundary conditions and polluted scenario, except it used the Kain-Fritsch cumulus scheme [*Kain and Fritsch*, 1993]. The projections of regional air quality model with the clean lateral boundary conditions and clean scenario generally represent the lower bound of changes from Huang et al. (submitted manuscript, 2008a). The MOZART simulations show a lesser sensitivity (−1% to +23%) to emission scenarios than the regional air quality model with the clean lateral boundary conditions (−30% to +20%) and the regional air quality model with the MOZART lateral boundary conditions (−24% to +25%). Different from the regional air quality model, the sensitivities of MOZART in different regions are similar. While both models are driven by the Parallel Climate Model meteorology, the MOZART 5-summer mean ozone mixing ratio increase under the polluted scenario (+4% to +9% in the 2050s and +16% to +23% in the 2090s) is larger than the respective regional air quality model with the clean lateral boundary conditions (+3% to +9% and +2% to +20%) and similar to the regional air quality model with the MOZART lateral boundary conditions (+6% to +13% and +15% to +25%) throughout most of the United States (except for the southeast United States). Under the clean scenario, the regional air quality model simulations with the clean lateral boundary conditions and with the MOZART lateral boundary conditions projected a large ozone reduction over the selected regions in the 2050s, and even a deeper reduction into the 2090s. On the other hand, the MOZART projected a similar present-day air quality condition in the 2050s (−1% to +5%) and the 2090s (−1% to 2%). The projected increase of the regional air quality model with the MOZART lateral boundary conditions polluted remains smaller than the case MAX in the northeast, midwest, and southeast U.S. areas, but is slightly larger in the California and Texas subdomains.

[28] Table 3 shows the net impacts of the long-range transport (the changes of regional air quality model with the MOZART lateral boundary conditions minus the changes of regional air quality model with the clean lateral boundary conditions) generally increase from the 2050s to the 2090s. The long-range transport impact generally contributes positive changes to the 5-summer mean ozone mixing ratio under the emissions scenarios ranging from +3% to +8% in the 2050s and +4% to +13% in the 2090s. The long-range transport impacts under the clean scenario are greater than those under the polluted scenario. The only exception is the Texas region where a large increase of long-range transport impacts from the 2050s to the 2090s in the polluted scenario compares to a slightly decrease in the clean scenario, leading to a much greater long-range transport impacts under the polluted scenario in the 2090s. As projected in the IPCC SRES, even for the clean scenario, the Latin America regions have substantial anthropogenic emissions increase in 2100. This projection leads to the increase of O_3 and NO_x mixing ratio in the southern model boundary (Figure 9). Thus, the Texas was projected by the regional air quality model with the MOZART lateral boundary conditions to have worse air quality than the regional air quality model with the clean lateral boundary conditions in the 2090s. The net increase in mean surface ozone mixing ratio ranges from +7 to +10 ppbV in the 2050s and from +6 to +12 ppbV in the 2090s. The magnitude of impacts is greater than those given by *Lin et al.* [2008a], but it is likely attributed to the stronger future emissions variation projected by the regional air quality modeling system than that of *Lin et al.* [2008a] within the North America region.

[29] The net impacts of the long-range transport on the daily mean 8-h average ozone mixing ratio resemble that of the 5-summer mean ozone mixing ratio under the two emissions scenarios (Table 4). The long-range transport impact resulted in increases ranging from +2% to +8% in the 2050s and +4% to +11% in the 2090s. Tables 3 and 4 show the California and Texas subdomains endure greater long-range transport impact than other regions. As discussed above, California (Texas) is sensitive to the western (southern) lateral boundary conditions and there was an increase of precursor gases mixing ratio of ozone (NO_x and NMV) and ozone itself transported across the southern and western boundaries in the 2090s. Huang et al. (submitted manuscript, 2008a) shows that the case MAX generally projected a higher increase than the regional air quality model with the clean lateral boundary conditions. By replacing the clean lateral boundary conditions in the case MAX with the MOZART lateral boundary conditions, it is realistically expected that the case MAX with the MOZART lateral boundary conditions will have similar long-range transport impacts in ozone concentrations as the regional air quality model with the MOZART lateral boundary conditions under the future emissions scenario. Thus, although the regional sensitivity of regional air quality model with the MOZART lateral boundary conditions to emissions changes may be similar to that of Huang et al. (submitted manuscript, 2008a), the air quality in the future climate can be worse (estimated by case MAX with the MOZART lateral boundary conditions) under polluted scenarios and less improved (estimated by the regional air quality model with the MOZART lateral boundary conditions) under clean scenarios.

[30] The increase of daily mean ozone can adversely impact human health. As described by Huang et al. (submitted manuscript, 2008a), the number of exceedance days (days when the maximum 8-h average ozone mixing ratio exceeds the EPA standard of 84 ppbV) is used as a measure for the potential of health impacts. Following the approach

Table 4. Similar to Table 3 but for the Daily Maximum 8-h Average Ozone

	2050s Polluted	2050s Clean	2090s Polluted	2090s Clean
Northeast United States	2	5	4	7
Midwest United States	3	4	5	6
California	5	6	6	8
Texas	6	8	11	7
Southeast United States[a]	4	5	4	6

[a]Use the interpolated 90-km results.

of Huang et al. (submitted manuscript, 2008a), Table 5 shows the projected number of exceedance days in the future climate based on the relative change in Figure 10 and the AQS observations. In general, the threshold value of the probable health impacts is 4 days. For the polluted scenario, MOZART projected a higher risk for the northeast U.S., midwest U.S., and California areas. The probabilities are on the edge of health risk for the Texas and southeast U.S. regions. The long-range transport impact generally produced an average increase of 2 to 3 exceedance days for most areas under the polluted scenarios. California has a substantial increase of ~5 days on top of the 16 days predicted by the regional air quality model with the clean lateral boundary conditions. This will dramatically elevate the potential health risk. For the clean scenario, the projected future air quality situation is similar to the present-day climate for the MOZART while the regional air quality model projected a substantial reduction on the potential health risk.

[31] Figures 11 and 12 show the vertical profiles of regional and 5-summer mean ozone mixing ratio difference of the regional air quality model simulations with the clean lateral boundary conditions and with the MOZART lateral boundary conditions under the polluted and clean scenarios in the 2050s and the 2090s, respectively. The difference is computed from the present-day simulations (Figure 8) to the future climate simulations under same modeling setting, for example, regional air quality model with the MOZART lateral boundary conditions. The regional air quality model with the clean lateral boundary conditions profiles show that the increase (decrease) of the surface ozone can lead to changes in upper troposphere with the same sign due to vertical transport and mixing under polluted (clean) scenario. Comparison between the regional air quality model with the clean lateral boundary conditions and with the MOZART lateral boundary conditions profiles also indicates that the middle to upper troposphere ozone mixing ratio are not only influenced by the changes in surface ozone but also the changes in upper troposphere ozone that are strongly influenced by the upstream upper troposphere lateral boundary conditions. It is evident that the changes in upper troposphere ozone mixing ratio can be mixed down at least to midtroposphere and possibly lower. It indicates the vertical mixing treatment of regional air quality model can be an additional source of uncertainty in climatic air quality projections. The magnitude of the difference intensified from the 2050s to the 2090s. The long-range transport increases the ozone mixing ratio in the entire troposphere with substantial differences in the upper troposphere attributed to the MOZART lateral boundary conditions. The changes of the mass-weighted total column ozone mixing ratio in the future climate (2090s) for the polluted (clean) scenario are +25% (−7%), +25% (−4%), +23% (+4%),

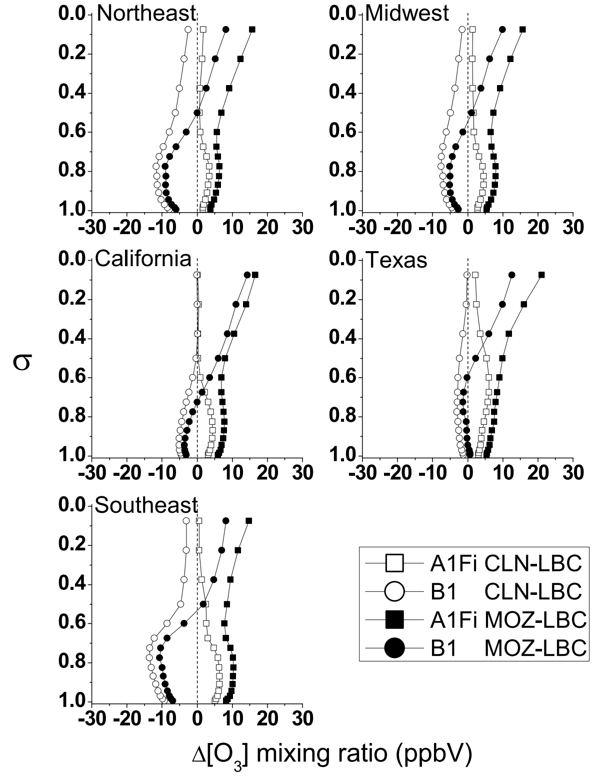

Figure 11. Vertical distribution (in sigma coordinate) of five-summer mean ozone mixing ratio difference from the period of 2000s of the regional air quality model simulations with the clean (CLN) and MOZART (MOZ) lateral boundary conditions under the polluted (A1Fi) and clean (B1) emissions scenarios for the period of 2050s.

Table 5. Number of Exceedance Days in the Present-Day and the Future Climate (2090s) Regional Air Quality Model Simulations With the Clean and the MOZART Lateral Boundary Conditions, as Well as the MOZART Simulations Under the Polluted and Clean Emission Scenarios[a]

	Present Climate	AQM/CLN Polluted	AQM/MOZ Polluted	AQM/CLN Clean	AQM/MOZ Clean	MOZART Polluted	MOZART Clean
Northeast United States	1	7	11	0	0	11	2
Midwest United States	1	6	9	0	0	8	1
California	5	16	21	0	1	18	6
Texas	1	1	3	0	0	4	1
Southeast United States	1	8	10	0	0	6	1

[a]Exceedance days given in days/summer/observed grid. AQM, air quality model; CLN, clean; MOZ, MOZART.

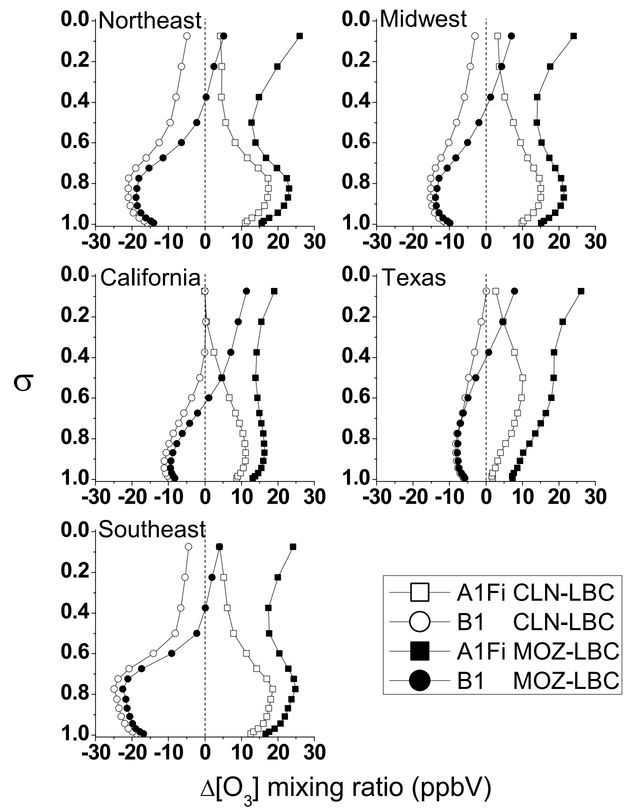

Figure 12. Similar to Figure 11, but for the period of 2090s.

+28% (−2%), and +29% (−10%) for the northeast U.S., midwest U.S., California, Texas, and southeast U.S. areas, respectively. The southeast United States shows the largest sensitivity of total column ozone mixing ratio to the emissions changes followed by the northeast United States and Texas. The relative changes in total column NO_x and NMV mixing ratio generally followed the trend of ozone mixing ratio (not shown). The range of NO_x changes is larger than that of NMV, for example, −74% to 81% versus 3% to 41% in the northeast United States. The net impacts of long-range transport on the total column ozone mixing ratio changes generally have increases of 10% (13%), 10% (11%), 13% (10%), 16% (7%), and 12% (11%) in the northeast U.S., midwest U.S., California, Texas, and southeast U.S. areas, respectively, under the polluted (clean) emissions scenario.

4. Summary and Conclusions

[32] The regional air quality modeling system, with finer spatial resolution and better-resolved regional circulation, produced better tropospheric ozone simulations than the global chemical and transport model for most of the selected areas, except for the California region. Both the global chemical and transport model and regional air quality modeling system respond to the driving global observed data set (global reanalysis) and GCM output in a similar manner for the present-day climate simulations. It is critical to apply appropriate lateral boundary conditions to account for the long-range transport in regional air quality model simulations. The lateral boundary conditions normally changed both in time and space. By applying the lateral boundary conditions extracted from the MOZART results, the bias of regional air quality model O_3 simulations relative to observations increases for the northeast U.S., midwest U.S., Texas, and southeast U.S. areas. The bias is reduced for the California region. The transported concentrations of pollutants in the global chemical and transport model clearly have impact on the regional air quality modeling system results through the imposed lateral boundary conditions. Uncertainties associated with the lateral boundary conditions along with other uncertainties should be considered in studies of the effects of future climate change on air quality.

[33] In this study, the baseline simulations (the controls) were established using the same modeling components including global chemical and transport model in the present-day climate simulations. Then the projected air quality changes were derived from the comparison between the control and future climate simulations under different emissions scenarios. The results showed that the MOZART simulations have a smaller sensitivity to emission scenarios than the regional air quality model simulations with the clean lateral boundary conditions and with the MOZART lateral boundary conditions. The southeast United States continues to have the biggest sensitivity (−24% to +25%) followed by the northeast (−18% to +22%) and midwest (−14% to +25%) United States. The results generally show that the net impact of long-range transport is an increase in the future tropospheric ozone concentration regardless of emissions scenarios. This is attributed to the rising ozone precursor gas emissions projected in future climate for developing countries as well as changes in global natural emissions that leads to the increased transport of precursor species across model boundaries. The regional air quality model simulations with the MOZART lateral boundary conditions produced increases of daily mean surface O_3 mixing ratio over that of the regional air quality model with the clean lateral boundary conditions ranging from +4% to +13% among the selected areas in the future (the 2050s and the 2090s), +7% to +16% on total column O_3 mixing ratio in the 2090s, and as high as 5 exceedance days in the 2090s. This study shows that the future regional U.S. air quality may not be entirely determined by the U.S. emissions control because of the long-range transport of chemical species. Air quality will be closely connected to the changes of future emissions on the global scale. The coupling of the global chemical and transport model and regional air quality modeling system is a vital tool to have a better estimation on the future U.S. air quality and should be used in climatic air quality studies. On the basis of the results presented by Huang et al. (submitted manuscript, 2008a) and the regional air quality model simulations with the MOZART lateral boundary conditions shown in this study, it is reasonable to anticipate that the upper bound of the tropospheric ozone changes estimated by Huang et al. (submitted manuscript, 2008a) should be higher if the long-range transport impact is incorporated.

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