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Linking global to regional models to assess future climate impacts on surface ozone levels in the United States

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[1] We investigate the impact of climate change on future air quality in the United States with a coupled global/regional scale modeling system. Regional climate model scenarios developed by dynamically downscaling outputs from the GISS GCM are used by CMAQ to simulate present air pollution climatology, and modeled surface ozone mixing ratios are compared with recent observations. Though the model accurately simulates ozone in the northeast U.S. and in central California, a positive bias of 10–15 ppb exists throughout most of the central and southeast U.S. The model is also applied to a simulated 2050 climate based on the IPCC A1B greenhouse gas scenario. Two future simulations are conducted, one with anthropogenic emissions held at 2001 levels, and one with anthropogenic emissions reduced in accordance with the A1B scenario. Without ozone precursor emissions changes, increases from 2–5 ppb in summer mean 8-h ozone mixing ratios are projected in Texas and parts of the eastern U.S., while high ozone episodes become more frequent. Increases of 2–8 ppb during the autumn are predicted over a large area in the central and southwest U.S., suggesting a lengthening of the ozone season. These increases within the regional modeling domain are predicted despite large decreases in the future global background ozone mixing ratio. Substantial decreases exceeding 15 ppb during the summer are predicted for the future reduced emissions case. A sensitivity test conducted with 30% higher methane mixing ratio yields widespread ozone increases of 0.5–2 ppb, an effect larger than that of climate-induced increases in isoprene emissions, demonstrating the need to consider changes in methane levels alongside climate change when simulating future air quality.

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1. Introduction

[2] Though global-scale models have been used to explore the impact of alternative emission scenarios on future ozone levels, regional-scale models are more commonly used by regulatory agencies to test the impact of specific emission controls or proposed industrial facilities. These air quality management applications typically have been based on present meteorological conditions, so that potential changes in climate are not included in the assessment. In cases where emission controls are implemented over several decades (e.g., U.S. Clean Air Interstate Rule), there is potential for different model predictions depending on which future climate scenarios are considered.

[3] To assess whether climate change should be taken into account in emission control strategy tests, modeling studies are needed to test the sensitivity of air quality to future climate scenarios. Because future emissions are highly uncertain, the Intergovernmental Panel on Climate Change (IPCC) has developed a number of emission scenarios intended to span a range of possible future socioeconomic conditions [*IPCC*, 2000]. These scenarios can then be used to provide a consistent set of drivers across multiple models, resulting in an ensemble of global/regional climate simulations and air quality simulations. However, as noted by *Dentener et al.* [2005], none of the IPCC scenarios take into account existing air quality legislation, with mandated reductions of emissions of ozone precursors.

[4] A few studies have demonstrated that climate change can result in higher future ozone levels over polluted areas such as the eastern United States [*Hogrefe et al.*, 2004; *Mickley et al.*, 2004a], California [*Steiner et al.*, 2006], and southern Germany [*Forkel and Knoche*, 2006], even when ozone decreases globally due to enhanced loss rates asso-

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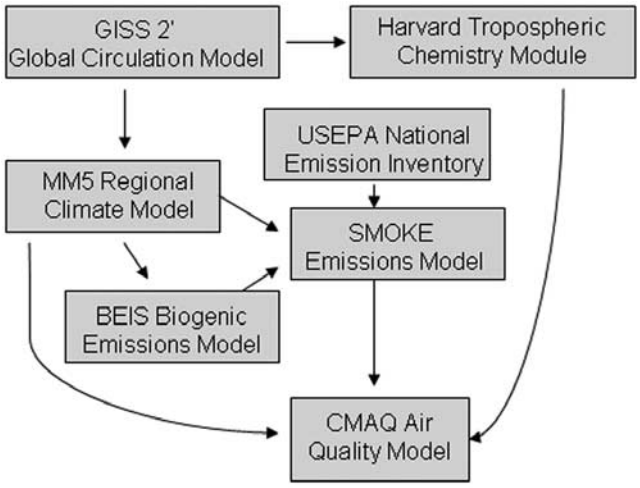

Figure 1. Schematic diagram showing linkages between global and regional modeling components.

ciated with increased absolute humidity [*Stevenson et al.*, 2000; *Murazaki and Hess*, 2006; *Liao et al.*, 2006; *Racherla and Adams*, 2006]. *Hogrefe et al.* [2004] presented modeling predictions that suggest ozone would increase under the IPCC A2 greenhouse gas emissions scenario. Their modeling study relied on a dynamic downscaling approach wherein a global climate model was used to drive regional scale meteorological simulations, which were then used as input to the air quality model to introduce climate forcing into the predictions. *Hogrefe et al.* [2004] used fixed chemical boundary conditions.

[5] The IPCC A2 scenario used in the study of *Hogrefe et al.* [2004] assumes a regionally heterogeneous world with a low rate of economic integration and slower convergence of fertility patterns, resulting in high population growth; by contrast, the A1B scenario assumes a higher rate of economic growth and integration, low population growth, and a balance between fossil-fuel and renewable energy sources [*IPCC*, 2000]. To test further the sensitivity of ozone and aerosols to climate on a regional scale, a series of meteorology and air quality simulations has been developed based on the A1B scenario. *Leung and Gustafson* [2005] developed 10-year simulations of regionally downscaled climate under present and future (ca. 2050) A1B scenarios for the continental United States. Some similar research efforts have simulated air quality in the nearer-term future, such as 2030 [*Dentener et al.*, 2005, 2006; *Forkel and Knoche*, 2006; *Stevenson et al.*, 2006; *Unger et al.*, 2006]. For this application, however, the decision was made to focus the future climate simulation on the year 2050 as a compromise between having a sufficient climate change signal and the difficulty in projecting emission scenarios too far into the future.

[6] This paper presents the air quality model predictions of ground-level ozone under present and future climatological conditions using the downscaled regional climate simulations of *Leung and Gustafson* [2005]. Our work builds on the approach of *Hogrefe et al.* [2004] by following a more optimistic scenario and by applying dynamic chemical boundary conditions. We also conducted continuous 5-year simulations for the present-day and future, and examined a larger region of the United States than did *Hogrefe et al.* [2004]. Both an evaluation of the current period ozone simulation and a comparison to future predictions will be presented.

2. Modeling System

[7] The modeling system used for this study is composed of a global climate model (GCM) coupled to regional scale climate, emissions, and chemical transport models, with linkages as depicted in Figure 1. Each component of this modeling system is described in further detail below.

2.1. Global Climate Simulations

[8] The GCM used is derived from the Goddard Institute for Space Studies (GISS) 2′ model as described by *Mickley et al.* [2004a], coupled to a tropospheric ozone-NO_x-hydrocarbon model as in *Mickley et al.* [1999]. The GCM version used here has a horizontal resolution of 4° latitude and 5° longitude and nine vertical layers in a sigma coordinate system extending from the surface to 10 hPa. It has been evaluated for the present-day by *Rind et al.* [1999] and *Rind and Lerner* [1996]. We have applied a "qflux ocean" [*Hansen et al.*, 1988], which allows sea surface temperatures to respond to climate change, but keeps the ocean heat transport fluxes fixed. Doubling CO_2 in the GISS model relative to the present-day yields a climate sensitivity of 0.8°C m^2 W^{-1}, within the range of sensitivity reported for current GCMs [*Mickley et al.*, 2004b; *IPCC*, 2007]. The global climate simulation covers the period 1950–2055, with greenhouse gas concentrations updated annually using observations for 1950–2000 [*Hansen et al.*, 2002] and the A1B scenario from the IPCC for 2000–2055 [*IPCC*, 2000]. The radiation scheme assumes present-day climatological values for ozone and aerosol concentrations and has no feedbacks due to future pollutant concentration changes. The tropospheric ozone-NO_x chemistry module provides chemical boundary conditions (BCs) for the chemical transport model, described further below.

2.2. Regional Climate Simulations

[9] A regional climate model (RCM) based on the Penn State/NCAR Mesoscale Model (MM5) [*Grell et al.*, 1994] was used to downscale the GCM output for 1995–2005 and 2045–2055. A two-way nested configuration was used in the RCM, with 108 km and 36 km horizontal resolution for the outer and inner domains, respectively, and 23 vertical layers [*Leung and Gustafson*, 2005]. Lateral boundary conditions from the GCM outputs were applied at 6 h intervals without assimilation of observational data. MM5 options used included the Grell cumulus parameterization scheme with shallow convection, Reisner1 mixed phase cloud microphysics, the Medium Range Forecast Model (MRF) PBL scheme, the NOAH land-surface model, and the Rapid Radiative Transfer model (RRTM). Further discussion of the physics parameterizations used are provided by *Leung et al.* [2003]. In general, choices were made to preserve the features of the GCM simulation rather than attempting to match present observed climatological patterns. The RCM outputs were archived hourly and used to provide meteorological conditions for the air quality model.

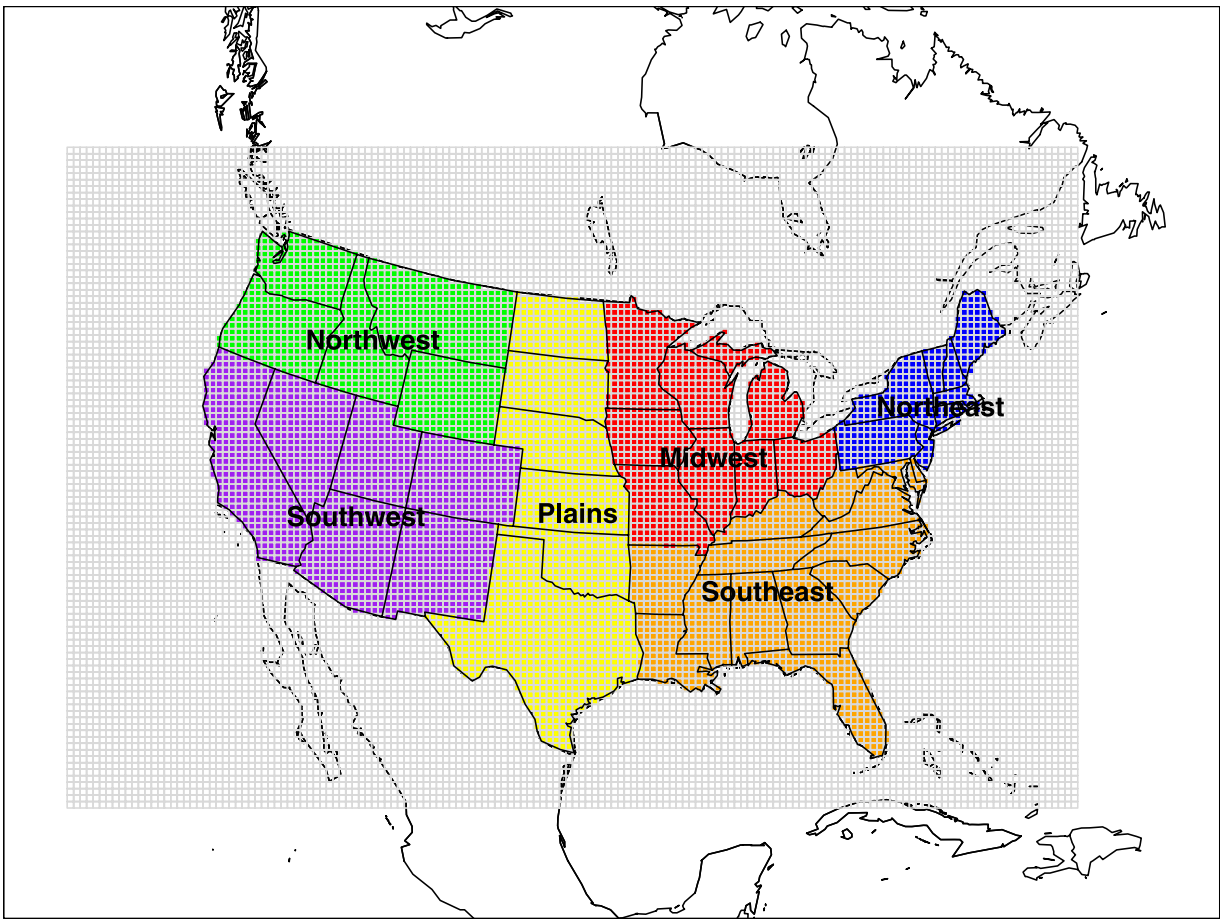

Figure 2. Modeling domain and region definitions.

2.3. Air Quality Modeling

2.3.1. Model Configuration

[10] Air quality simulations were conducted using the Community Multiscale Air Quality (CMAQ) chemical transport model (*Byun and Ching* [1999]; *Byun and Schere* [2006], available at http://www.cmascenter.org) version 4.5. CMAQ is an Eulerian chemical transport model which simulates the transport, chemical transformation, and deposition of gas and aerosol pollutants, and has been used successfully in a number of retrospective modeling studies [e.g., *Appel et al.*, 2007; *Eder and Yu*, 2006] and forecast applications [*Otte et al.*, 2005; *Eder et al.*, 2006; *Hogrefe et al.*, 2007]. However, these types of modeling studies differ from the present work in that they are designed to attain better agreement with observed air quality data by employing four-dimensional data assimilation (FDDA) of meteorological observations or initializing short-term ($\sim$24–48 h) forecasts with reanalysis results. The studies cited above also differ from the regional climate simulations used in this study in the physics options used within the meteorological model. CMAQ options used here included the SAPRC chemical mechanism [*Carter*, 2000], the Rosenbrock chemical solver [*Sandu et al.*, 1997], and the Regional Acid Deposition Model (RADM) cloud scheme. Chemical dry deposition velocities were calculated using the "M3dry" dry deposition scheme [*Pleim et al.*, 2001]. The domain encompassed the entire continental U.S., parts of Canada and Mexico, and the surrounding oceans at a horizontal resolution of 36 km and with 14 vertical layers from the surface to 100 hPa (see Figure 2). With only 14 vertical layers, CMAQ does not resolve the tropopause, and applies a zero-flux vertical boundary condition for all species. Accordingly, we do not simulate stratospheric ozone chemistry, the injection of stratospheric ozone into the troposphere, or how those processes might change under a future climate. Air quality feedbacks on climate (e.g., localized heating by ozone and cooling by sulfate aerosol) are also not simulated in the current version of CMAQ.

[11] One feature that distinguishes the modeling system used in the present study from that of other studies using global scale meteorology to drive regional climate and air quality simulations [*Hogrefe et al.*, 2004; *Tagaris et al.*, 2007] is the use of monthly averaged species tracer outputs from the GCM-coupled chemistry module to provide time-varying chemical boundary conditions. Transported species included O_3, NO_x, PAN, CO, isoprene, and 15 other VOCs. The relatively coarse time resolution is sufficient to represent seasonal influences and long-term changes in the transported species. It will not, however, capture the episodic intercontinental transport of pollution, as that was not the intention of this research.

2.3.2. Simulations

[12] One design goal of this study was to assess the effect of changing climate, which we take to encompass the

Table 1. Description of Air Quality Simulations

Name	Modeling Period	Emissions Year[a]
CURR	1999–2003	2001
FUT1	2048–2052	2001
FUT2	2048–2052	2050
CURBC[b]	Jul 2048	2001
ISOP25[c]	Jul 1999	2001
METH[d]	2048	2001
CHES[e]	2001–2003	2001
CB-IV[e,f]	2001	2001

[a]2001: Anthropogenic emissions based on U.S. EPA 2001 National Emission Inventory; 2050: Anthropogenic emissions based on scaling factors in Table 2. In all cases, biogenic emissions are computed from the meteorology for the particular modeling period.

[b]Global model predictions for July 1999 were used for chemical boundary conditions.

[c]Isoprene emissions increased by 25%.

[d]Methane mixing ratio increased from 1.85 ppm to 2.40 ppm.

[e]Meteorological simulation used assimilation of observational data and different MM5 configuration; see text.

[f]Used CB-IV chemical mechanism [*Gery et al.*, 1989]; all other simulations used SAPRC [*Carter*, 2000].

associated changes in biogenic emissions and background chemical composition, on simulated air quality in relation to the effect of projected changes to anthropogenic emissions of ozone precursors, recognizing that emissions projections for the future are highly uncertain. CMAQ simulations were conducted for two 5-year subset periods "1999–2003" and "2048–2052" out of the two 10-year intervals for which the RCM simulation was run. These CMAQ simulations are climatological representations of present and future air quality under the A1B scenario, and are not intended to represent or predict the actual day-to-day variations in pollutant levels for either the present or future modeling periods. Three sets of 5-year simulations were conducted (see Table 1). The first simulation (CURR) used chemical BCs and downscaled meteorology from 1999–2003, while FUT1 and FUT2 used chemical BCs and downscaled meteorology from 2048–2052. To assess the relative impact of the changing chemical background, a one-month sensitivity study (CURBC) was conducted using chemical BCs from July 1999 and meteorology from July 2048.

[13] The Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system [*Houyoux et al.*, 2000] version 2.2 was used to prepare hourly emissions inputs consistent with the meteorology, as both evaporative emissions and plume rise are functions of temperature. The Biogenic Emissions Inventory System (BEIS) [*Hanna et al.*, 2005] version 3.13 was used to compute emissions of 33 VOCs from vegetation, including isoprene, methanol, methyl butanol, and 14 monoterpenes, as well as nitric oxide emissions from soils. The Biogenic Emissions Land use Database version 3 (BELD3) for North American vegetation [*Kinnee et al.*, 1997] was used to compute hourly emissions with a resolution of 1 km, which were then lumped and aggregated in SMOKE according to the chemical mechanism and modeling grid used for this application. In our chemical mechanism, isoprene nitrate is quickly deposited and serves as a sink of NO_x. As shown in Table 2, isoprene emissions estimates increase by an average of 23% under the future meteorology. To isolate the effect of the increased isoprene emissions from other climate variables, a sensitivity study (ISOP25) was conducted for July 1999 with isoprene emissions increased by 25%.

[14] In preparing emissions for the future simulation, no changes were made to the BELD3 database. The assumption that land use patterns will not change over the next few decades represents a major source of uncertainty in the future emission estimates. Also, because BEIS does not include the negative feedback rising CO_2 levels may have on isoprene emissions, the 23% relative increase between the future and current periods shown in Table 2 may be overestimated [*Arneth et al.*, 2007]. Finally, lightning as a source of NO_x in the upper troposphere was not considered for either the present or future climate simulations.

[15] For the CURR and FUT1 simulations, anthropogenic emissions were based on the U.S. Environmental Protection Agency (EPA) 2001 National Emission Inventory, i.e., held constant except to the extent that they are influenced by meteorological conditions. For the FUT2 simulation, anthropogenic emissions were changed according to the Asian Pacific Integrated Model (AIM) projections for the A1B scenario, as shown in Table 2. While global emissions projections of VOCs and NO_x at 2050 under the A1B scenario are higher than current levels, projected emissions are substantially lower for the countries belonging to the Organization for Economic Cooperation and Development as of 1990 (i.e., the "OECD90" region, which includes the U.S. and Canada but not Mexico [see *IPCC*, 2000]). These OECD90 region-specific estimated changes were applied throughout the domain, including in Mexico. Emissions of ammonia and primary particulate matter for FUT2 are unchanged from those of FUT1. This scaling approach is admittedly simplistic and is intended to be a minimal sensitivity test of the range of impacts that could result from the A1B scenario. Efforts are ongoing to develop detailed future emission scenarios for 2050 that will be used for an additional series of simulations for this project. However, it should be noted that the large reductions in Table 2 are in line with reductions mandated for the eastern U.S. by 2015 under the EPA's Clean Air Interstate Rule.

[16] Because methane is a slow-reacting, relatively well-mixed chemical species, it is not dynamically simulated in regional-scale air quality models. However, on a global scale methane plays an important role in determining tropospheric ozone concentrations; recent research has

Table 2. Emissions of Biogenic and Anthropogenic Ozone Precursors

Species	2001	2050[a]
Isoprene, Tg C a^{-1}	10.8	+22%
CO, Tg C a^{-1}	52.1	−46%
VOCs,[b] Tg C a^{-1}	33.6	−21%
NO_x, Tg N a^{-1}	7.2	−45%
SO_2, Tg S a^{-1}	8.4	−63%
NH_3, Tg N a^{-1}	3.3	unchanged

[a]Change in A1B scenario emissions for OECD90 region for the year 2050 relative to 2000, as computed by AIM model [*IPCC*, 2000].

[b]Excluding isoprene and CO.

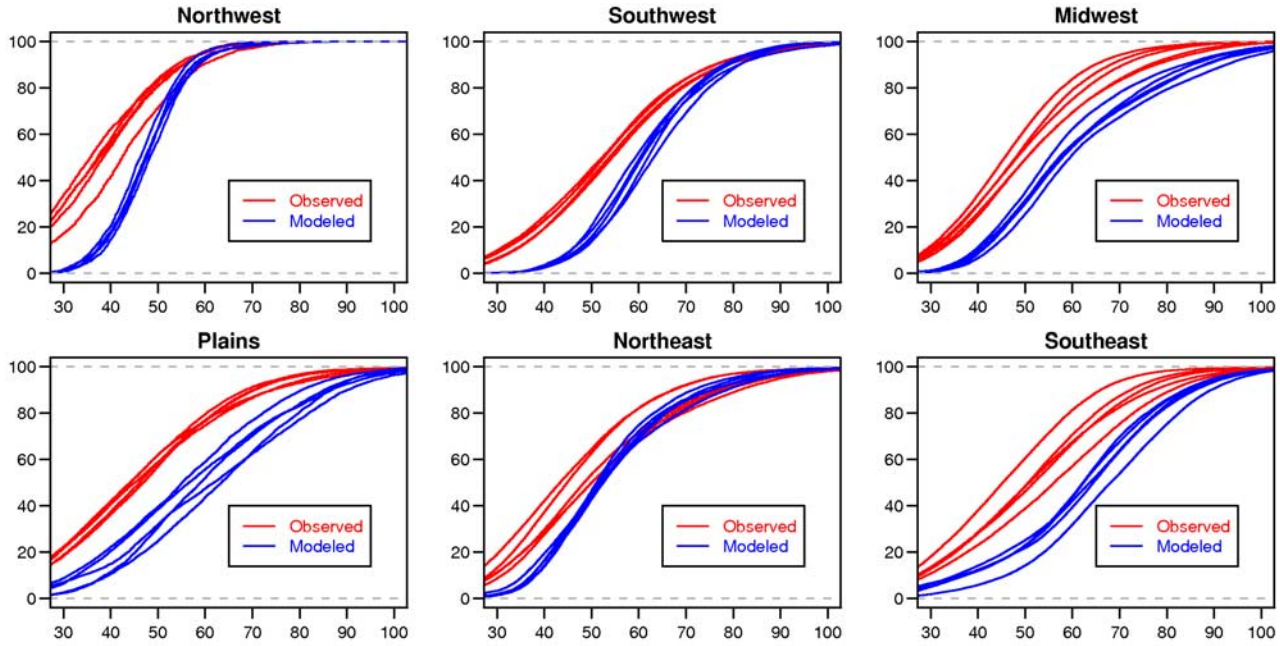

Figure 3. Empirically determined cumulative distribution functions (CDFs) of observed and CMAQ modeled maximum daily 8-h average (MDA8) ozone mixing ratios (ppb), 1 May to 30 September, for each year in 1999–2003. Results broken down regionally as shown in Figure 2.

linked future increases in methane concentrations to increased ozone concentrations and has proposed control of methane emissions as an efficient way to reduce both tropospheric ozone and radiative forcing [*Fiore et al.*, 2002; *Unger et al.*, 2006; *Dentener et al.*, 2005; *West and Fiore*, 2005; *West et al.*, 2007]. By default, methane in CMAQ is assigned a spatially and temporally uniform mixing ratio of 1.85 ppm, a value somewhat higher than the *IPCC* [2000] global mean value of 1.76 ppm but appropriate for the midlatitude northern hemisphere. To investigate the effect of increased ambient methane levels on ozone, a sensitivity simulation (METH) was conducted for one future summer with the same emissions as the FUT1 case. In the METH run, the methane mixing ratio was increased to 2.40 ppm, which is the projected globally-averaged abundance at 2050 under the A1B scenario [*IPCC*, 2000].

[17] As noted previously, the meteorology used within the present work differs in several respects from the way that meteorology is simulated in retrospective modeling with CMAQ. These include driving MM5 with lateral boundary conditions from a GCM rather than using FDDA within the interior of the modeling domain, conducting a continuous 5-year simulation rather than reinitializing the model every few days, and various different model physics options within MM5. Though it is beyond the scope of this study to compare exhaustively the effect of each individual difference on simulated air quality outcomes, we choose two other retrospective MM5-CMAQ modeling studies for comparison with the CURR simulation. These two studies both used a similar configuration of MM5, whose differences from the present modeling system include the Kain-Fritsch2 cumulus parameterization without shallow convection, the Asymmetric Convective Model PBL scheme, and the Pleim-Xiu land-surface model, as well as using FDDA of observed meteorology. One study (CHES) is a 2001–2003 simulation also using the SAPRC chemical mechanism; the other (CB-IV) is a 2001 simulation using the Carbon Bond-IV chemical mechanism [*Gery et al.*, 1989]. Both of these comparison simulations use a very similar emissions inventory to that used for the CURR simulation, with the primary difference being that the CB-IV simulation incorporated continuous emissions monitoring data from electrical power plants. We do not restrict the comparison across these three simulations to the single year 2001 because the CURR meteorology for "2001" is not the same as the "actual" meteorology used for the other two studies.

3. Results and Discussion

3.1. Present Period Evaluation

[18] Hourly ozone observations were retrieved from EPA's Air Quality System (AQS) database for 1999–2003. Only stations having annual data completeness of 40% or greater for each year were retained for analysis, resulting in 876 sites. In comparing model simulations against observations, model values are extracted for the grid cells containing monitoring stations without any spatial interpolation. Because the meteorology model is driven at the boundaries by a global climate model, it would be inappropriate to evaluate the air quality results by comparisons of model values with observations paired in time and space. Instead, we adopt an approach in which we compare means and the upper tails of the distributions. However, for all cases the observational data are aggregated over the days and years corresponding to the modeling period. While other years may represent similar climate, emission changes related to the Acid Rain Program and the NO_x State

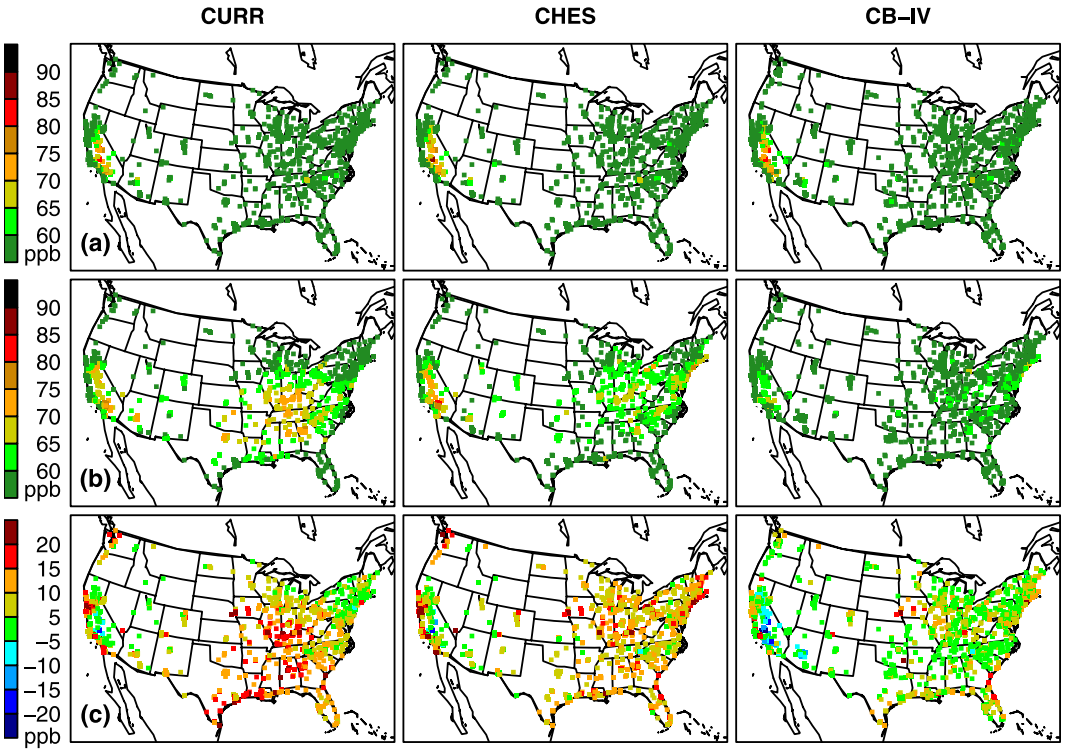

Figure 4. (a) Mean of daily maximum 8-h average (MDA8) ozone mixing ratios (ppb) at AQS observational monitoring sites, 1 May to 30 September for the years corresponding to each modeling period; (b) mean of CMAQ modeled MDA8 ozone (ppb) over the same time period for three different model configurations: CURR, CHES, and CB-IV; (c) difference between mean modeled and observed MDA8 ozone (ppb). For CURR, the modeling years are 1999–2003; for CHES, 2001–2003; and for CB-IV, 2001.

Implementation Plan (SIP) Call would introduce changes in the ozone levels during the late 1990s and 2004 that are not representative of the emission inventory used for these simulations.

[19] The air quality metric used throughout this paper is the maximum daily 8-h average (MDA8) ozone mixing ratio, because that is the quantity on which the U.S. National Ambient Air Quality Standards are based. The empirically determined cumulative distribution functions (CDFs) of observed and modeled maximum MDA8 ozone for the "ozone season" (defined here as 1 May to 30 September) for each year 1999–2003 are broken down regionally and shown in Figure 3. Each panel shows the percentage of MDA8 ozone observations and model predictions at the observational monitoring sites that exceed a given value. The CURR MDA8 ozone distributions agree quite well with observations in the Northeast U.S., particularly in the upper end of the distribution, when the mixing ratio exceeds about 60 ppb. The lower end of the distribution of modeled ozone mixing ratios is too high, consistent with past studies which have shown that CMAQ typically overestimates O_3 on days with low O_3 concentrations [*Eder and Yu*, 2006]. The spread between the individual modeled CDFs in the Northeast is somewhat less than the spread between the observed CDFs, indicating that the modeling system is underestimating the interannual variability. The relationship between the observed and modeled CDFs is similar for the Southwest region, except that the model's positive bias is even larger and extends up to the 80th percentile, or around 80 ppb. For the Northwest, Midwest, Plains, and Southeast U.S. regions, however, the modeled CDFs lie almost entirely outside the range of the observed CDFs, though the extent of the interannual variability in regional ozone levels is captured better than in the Northeast.

[20] Means of the observed and modeled MDA8 ozone mixing ratios from May–September for the 5-year period 1999–2003 are presented in the left column ("CURR") of Figure 4, along with model biases at each site. As can be seen from Figure 4c and Table 3, moderate to large overpredictions are evident in much of the eastern U.S. Modeled mixing ratios are on average 1.4 ppb higher than observations at sites in the Northeast, while large positive biases

Table 3. Bias in Region-Wide Average of Modeled Maximum Daily 8-h Average Ozone Mixing Ratios (ppb) Relative to Observations at AQS Sites, May–Sept 2001

Region[a]	CURR	CHES	CB-IV
Northwest	10.3	13.0	3.4
Southwest	7.6	8.2	0.3
Midwest	9.0	11.7	5.6
Plains	13.0	9.1	3.2
Northeast	1.4	9.9	5.1
Southeast	11.4	9.4	5.2
All sites	8.6	9.7	3.8

[a]As defined in Figure 2.

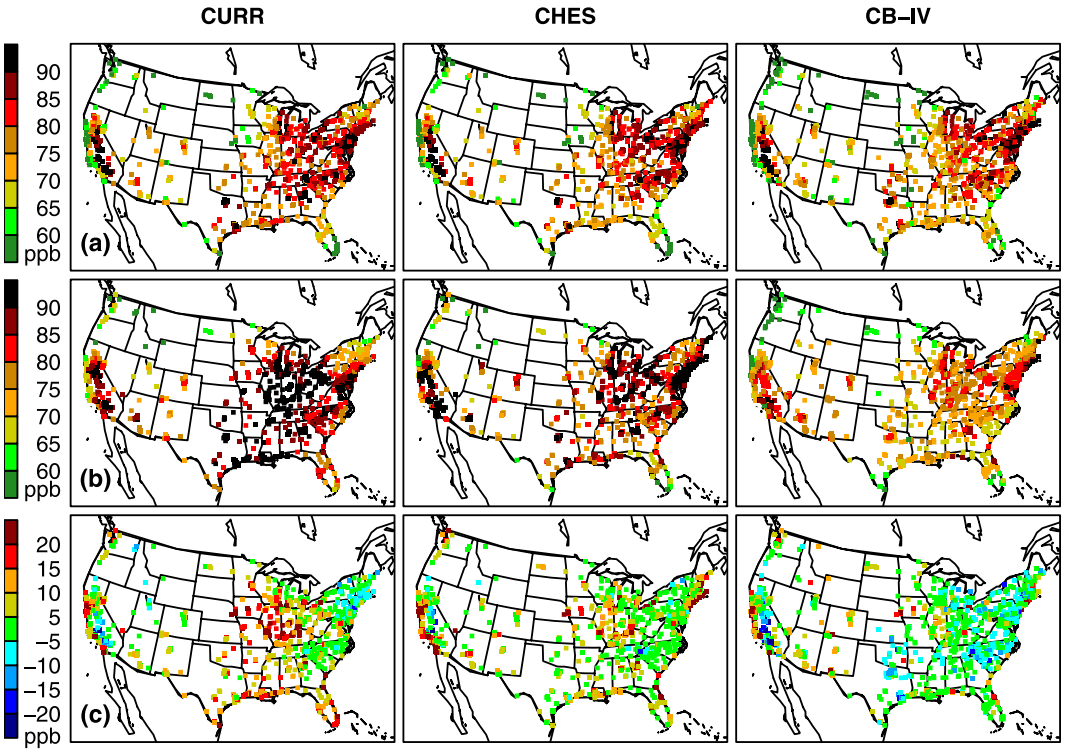

Figure 5. (a) Average of extreme (≥90th percentile) daily maximum 8-h average (MDA8) ozone mixing ratios observed at AQS monitoring sites, 1 May to 30 September for the years corresponding to each modeling period; (b) average of extreme CMAQ modeled MDA8 ozone (ppb) over the same time period for three different model configurations: CURR, CHES, and CB-IV; (c) difference between modeled and observed extreme MDA8 ozone (ppb). For CURR, the modeling years are 1999–2003; for CHES, 2001–2003; and for CB-IV, 2001.

exceeding 15 ppb exist at some sites in the south central and southeast U.S. Results are more mixed in California, with the model being positively biased on the coast and unbiased or even negatively biased at inland sites.

[21] There are numerous meteorological factors that may be contributing to these biases. Some of these positive biases in ozone levels may be attributable to previously documented surface temperature biases in the RCM, generally about 1–2°C, but reaching as high as 4°C in parts of the Southeast during summer [*Leung and Gustafson*, 2005]. Though the RCM realistically simulates precipitation levels over the western U.S., *Leung and Gustafson* [2005] reported dry biases over the eastern and southeastern U.S. of 50–80% during summer. Various factors associated with less frequent precipitation, including reduced cloudiness and accompanying increased photolysis rates as well as less frequent pollutant removal by wet deposition, could contribute to elevated ozone predictions. The planetary boundary layer (PBL) height west of 100°W is also underpredicted [*Gustafson and Leung*, 2007], which would be expected to cause positive biases in pollutant concentrations. In addition, the synoptic scale Bermuda High pressure system is not simulated correctly (Cooter et al., The effects of large-scale simulated climate errors on regional ozone anomalies ca. 2000, submitted manuscript, 2008), with subtropical high pressure displaced further northeast off the U.S. coast and higher pressure over the south central and southeastern U.S., resulting in higher ozone over those regions.

[22] Results from the two other MM5-CMAQ modeling studies described in section 2.3.2 are also presented in Figure 4. The CHES simulation is also biased high relative to the observations, while the bias in the CB-IV simulation is much lower. Shown for comparison in Figure 5 are the average of the mixing ratios at or above the 90th percentile, i.e., the highest 10% of the distribution. The positive bias seen in the means is also evident in Figure 5, though the CURR and CHES simulations compare somewhat more favorably with observations for days with high ozone than they do on average. Table 3 shows regionally-averaged biases in MDA8 ozone mixing ratios for the three different 2001 simulations. Because the CB-IV and CHES simulations use the same meteorology and virtually the same emissions, the differences between them (ranging from 4.0–9.5 ppb and averaging 5.6 ppb) is primarily due to their differing chemical mechanisms.

[23] The SAPRC mechanism was used for this work both because it is generally considered to be more scientifically accurate than CB-IV and because it is more compatible with the tropospheric ozone chemistry model of *Mickley et al.* [1999] used for boundary conditions, which was regarded as a critical element for this study. The CB-IV mechanism, which includes fewer reactions and thus runs somewhat faster than the SAPRC mechanism, has been used more often for regulatory applications in the U.S. The CB-IV mechanism's superior performance statistics within CMAQ are likely due to compensating errors elsewhere in the

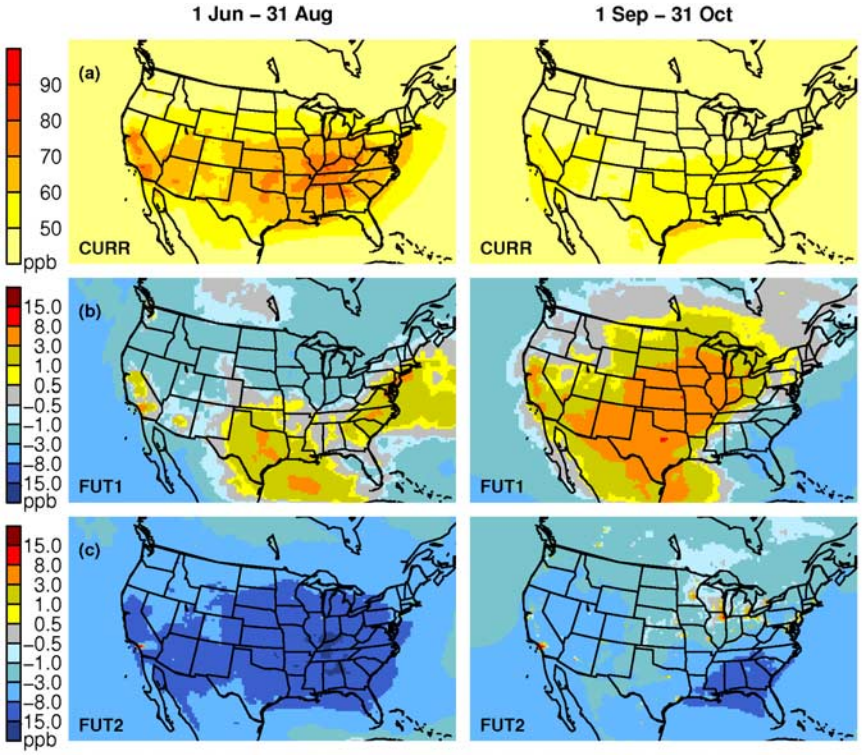

Figure 6. (a) Mean of CMAQ modeled daily maximum 8-h average (MDA8) ozone mixing ratios (ppb), June–August (left) and September–October (right), CURR; (b) difference (FUT1-CURR) in mean MDA8 ozone, JJA and SO; (c) difference (FUT2-CURR) in mean MDA8 ozone using anthropogenic emissions scaled according to Table 2 JJA and SO.

modeling system rather than to it being intrinsically more correct than SAPRC. Recent work has highlighted the importance of the chemical representation of organic nitrates from isoprene oxidation [*von Kuhlmann et al.*, 2004; *Fiore et al.*, 2005; *Wu et al.*, 2007], and it is worth noting that the formation of isoprene nitrates is reversible in SAPRC but irreversible in CB-IV. The bias in the CURR results, ranging regionally from 1.4–13 ppb and averaging 8.6 ppb overall, is somewhat higher than the bias typically seen in retrospective modeling applications with CMAQ. A similar overprediction of surface ozone over the United States has been reported by other modeling studies [e.g., *Murazaki and Hess*, 2006; *Wu et al.*, 2008]. The overprediction we present here is within the range of that of other models and points to limitations of current understanding of ozone photochemistry.

3.2. Future Period Modeling Results

[24] Spatial distributions of the summer (1 June to 31 August) and early autumn (1 September to 31 October) mean MDA8 ozone mixing ratios for the three simulations are given in Figure 6 The top row (a) shows the mean MDA8 mixing ratios for the CURR simulation, i.e., during the 5 years representing present climate (1999–2003); the middle row (b) shows differences between the FUT1 and CURR simulations, i.e., considering just climate change alone, and the bottom row (c) shows differences between the FUT2 and CURR simulations, i.e., combining the effect of climate change with changes in emissions of ozone precursors. For the summer months of the FUT1 simulation, increases of 3–5 ppb are predicted in eastern Texas and along much of the east coast, while decreases of 1–3 ppb are predicted in the Northwest and the northern Midwest, and 3–8 ppb off the Pacific coast. The increase is much larger and more widespread during September and October, suggesting a possible lengthening of the ozone season over most of the central and southwest U.S. as previously proposed by *Fiore et al.* [2002]. By contrast, substantial decreases are evident throughout most of the U.S. in the 2050 reduced emissions case FUT2. These decreases are most dramatic in the summer (12–16 ppb), but are also substantial for the September/October period, especially in the Southeast. The large reductions in NO_x emissions in FUT2 lead to predictions of localized ozone increases in southern California during the summer and in several other urban areas during September and October.

[25] Analogous plots of the 95th percentile MDA8 ozone mixing ratios are presented in Figure 7. The difference between the FUT1 and FUT2 simulations is even more pronounced when it comes to these extreme events. Considering just climate change alone, with future anthropogenic emissions held at current levels (i.e., FUT1), the 95th percentile peak 8-h ozone mixing ratio increases by 3–8 ppb in much of the eastern U.S. This is consistent with the findings of *Hogrefe et al.* [2004], who reported an average increase in summertime 4th-highest ozone mixing ratios of 6.5 ppb for the 2050s as a result of climate change alone under the A2 greenhouse gas scenario. For the future

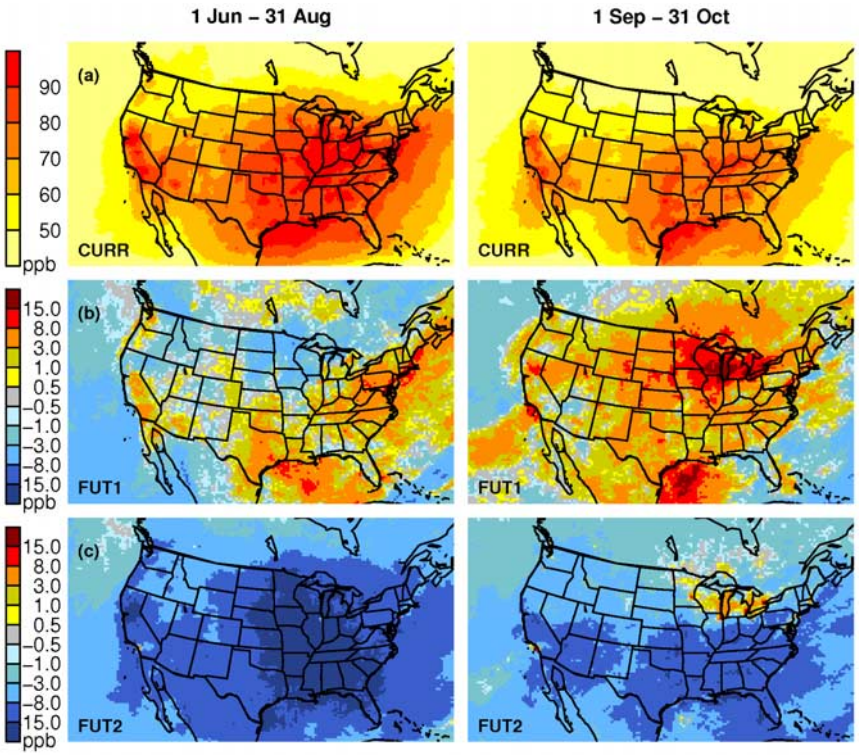

Figure 7. (a) 95th percentile of CMAQ modeled daily maximum 8-h average (MDA8) ozone mixing ratios (ppb), June–August (left) and September–October (right), CURR; (b) difference (FUT1-CURR) in 95th percentile MDA8 ozone, JJA and SO; (c) difference (FUT2-CURR) in 95th percentile MDA8 ozone, JJA and SO.

reduced emissions case FUT2, however, the 95th percentile summertime 8-h ozone mixing ratio decreases by at least 5 ppb nearly everywhere in the continental U.S., and by as much as 15 ppb in large portions of the eastern and central U.S. These results imply that, at least for the magnitude of emissions control assumed for the U.S. under the IPCC A1B scenario, the decrease in surface ozone due to the change in emissions outweighs the increase due to climate change. This conclusion is contrary to that of *Hogrefe et al.* [2004], who found that climate change and future emissions changes under the A2 scenario would both lead to enhanced surface ozone levels in the eastern U.S. Under the A2 scenario, NO_x and VOC emissions increase in the United States, which accounts for their dramatically differing results. Therefore, it is worth emphasizing that although the difference in temperature change at 2050 predicted by a range of global climate models in response to the A1B and A2 scenarios is small at about 0.25°C [*IPCC*, 2001] and as a consequence the impact of climate change on regional air quality is similar between these two scenarios, the simulated ozone changes are substantially different, and indeed of opposite sign, if changes in ozone precursor emissions associated with these two scenarios are considered as well.

[26] *Leung and Gustafson* [2005] previously analyzed two 10-year data sets of downscaled meteorology outputs, of which the two 5-year periods used for air quality modeling in this work are subsets. In examining changes in temperature, precipitation, surface radiation, PBL ventilation, and the frequency of stagnation events, *Leung and Gustafson* [2005] predicted worsened air quality in Texas and slight improvement or no impact in the Midwest during the summer, and worsened air quality throughout most of the U.S. during the fall. Figure 8 shows the summertime difference (FUT1-CURR) for four meteorological or meteorologically-influenced variables: surface temperature, precipitation, surface solar radiation, and isoprene emissions, which increase in response to increased temperature and photosynthetically active radiation. It is not possible to attribute definitively the climate-induced changes in air quality (i.e., the difference in ozone between FUT1 and CURR) to any one parameter, because the interaction between them is quite complex, and different factors will predominate in different regions and different years. However, the spatial pattern of the change in five-summer average MDA8 ozone mixing ratios (Figure 6) are in closest agreement with the changes in surface radiation (Figure 8c), with the greatest increases in southern and central Texas and decreases in the north and northwestern U.S. This suggests that the change in surface solar radiation (which is itself due to changed daytime cloud cover) is a primary driver of the climate-only effect on air quality. Our results concur with earlier studies that show the importance of cloud cover and solar insolation on the photochemistry of ozone [e.g., *Sillman and Samson*, 1995; *Dawson et al.*, 2007].

[27] Ozone boundary conditions from the global model averaged over the summer (JJA) and early fall (September–October) months and the 1999–2003 (CURR) and 2048–2052 (FUT) periods are shown in Figure 9, along with

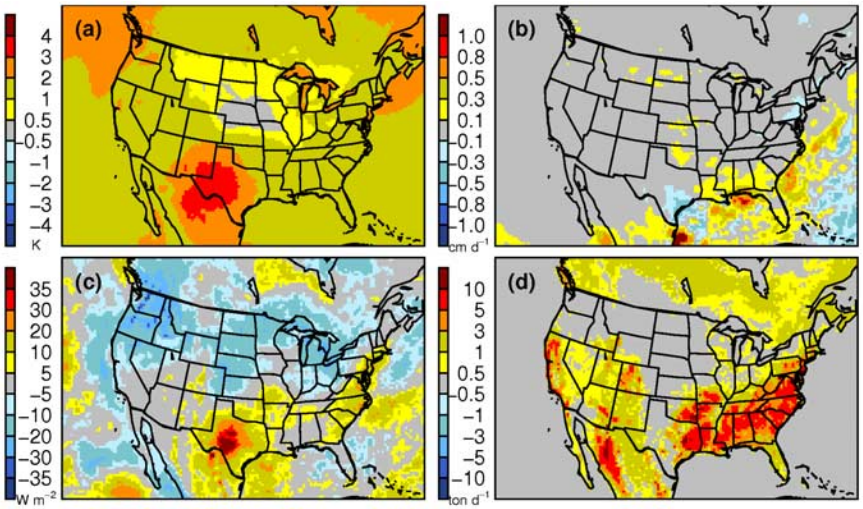

Figure 8. JJA differences (FUT1-CURR) in (a) surface temperature, (b) total precipitation, (c) surface solar radiation, and (d) isoprene emissions.

difference plots of the changes between the two periods. For both seasons, background ozone levels decrease in the future, with the largest decreases of 6.3 ppb (JJA) and 7.4 ppb (September–October) near the surface over the western boundary. Using a global chemistry model, *Wu et al.* [2008] have shown that ozone in remote regions generally decreases in the future climate due to increasing humidity. Water vapor is a sink of ozone. *Wu et al.* [2008] show that surface ozone mixing ratios increase over continents during summertime, largely driven by increased temperature and decreased ventilation. The substantial decreases in ozone coming into the CMAQ modeling domain account for the

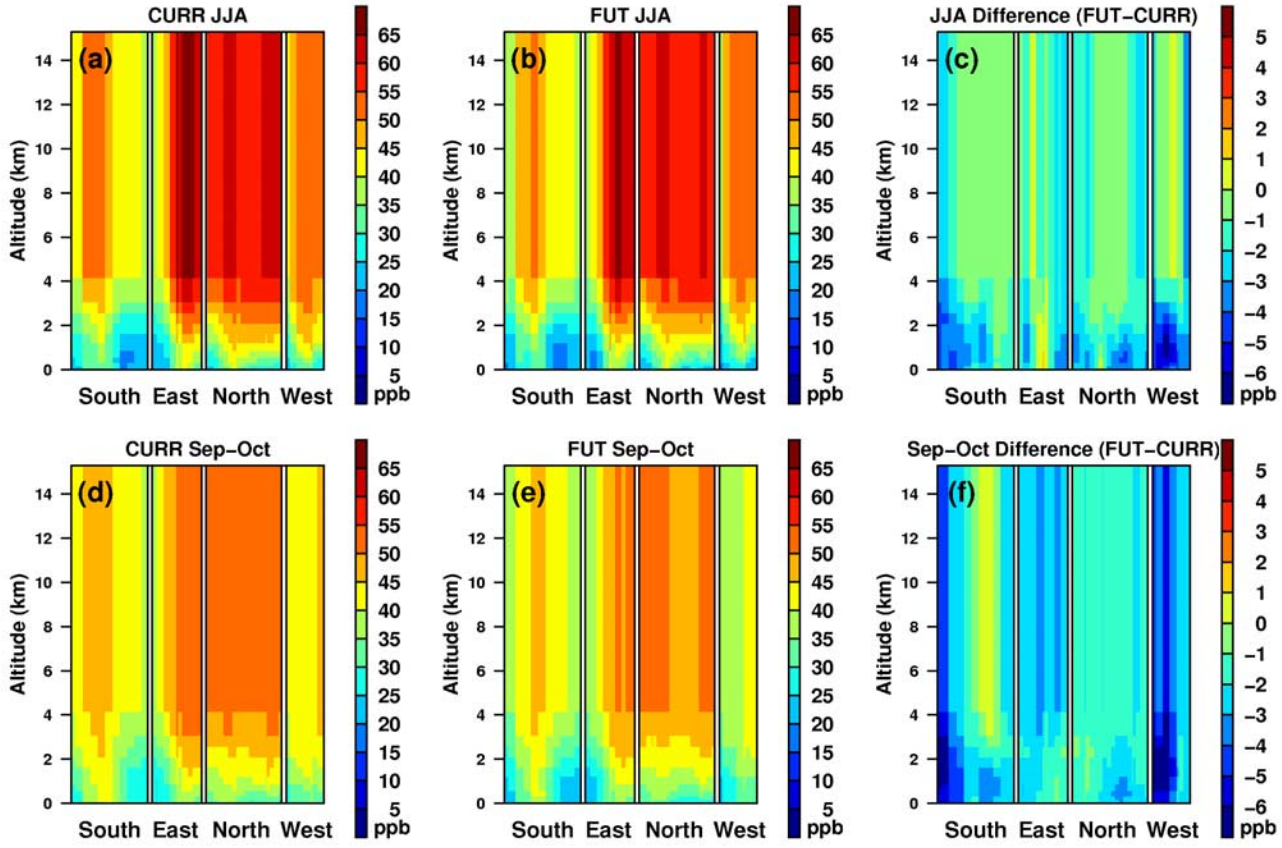

Figure 9. Vertical profile of average ozone boundary conditions (ppb) from global tropospheric chemistry module for 1999–2003 (CURR), 2048–2052 (FUT), and the FUT-CURR difference.

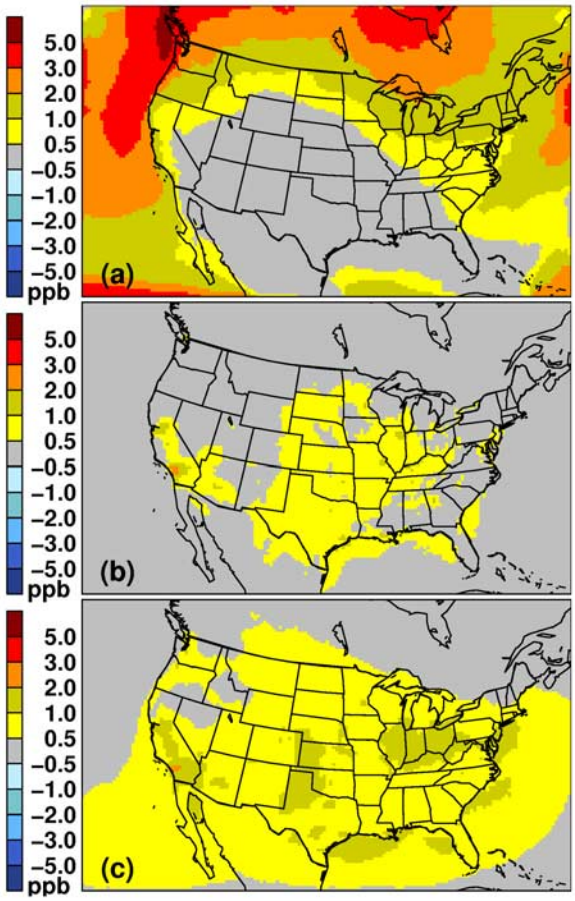

Figure 10. Sensitivities of MDA8 ozone. (a) Difference (CURBC-FUT1) for July 2048 when using global model chemical boundary conditions for July 1999, (b) difference (ISOP25-CURR) for July 1999 when isoprene emissions are increased by 25%; (c) difference (METH-FUT1) for JJA 2048 when methane mixing ratio is increased from 1.85 to 2.40 ppm.

decreases over the Pacific in Figure 6b. The increases seen over parts of the U.S. in the FUT1 simulation occur in spite of the reduced amount of ozone being advected into the domain via the boundary conditions. Figure 10a shows the difference between the CURBC and FUT1 simulations, which gives an indication of how much higher future ozone mixing ratios would be without considering the change in chemical background from the global model.

[28] The result from the isoprene sensitivity study ISOP25, which isolates the effect of isoprene emissions changes from other climate changes, is shown in Figure 10b. Although in the future climate isoprene emissions increase the most over the southeastern U.S. (Figure 8d), there is no significant accompanying change in predicted ozone mixing ratios there. This is consistent with the findings of *Wu et al.* [2008] that ozone in the Southeast is relatively insensitive to climate change. In that study, however, the formation of isoprene nitrate was irreversible and served as a sink of NO_x, while in our study isoprene nitrate was allowed to cycle NO_x back into the atmosphere. In our case, increasing isoprene emissions did not increase ozone significantly in the Southeast since ozone production in this region is NO_x-limited. With the 25% increase in isoprene emissions, MDA8 ozone increases by 0.5–1.0 ppb in most of the central U.S., while somewhat larger increases up to 2.5 ppb are seen in portions of California.

[29] Figure 10c shows the result of the METH sensitivity study, in which a 30% increase in assumed methane mixing ratio results in MDA8 ozone increases that are comparable in magnitude to the increases due to isoprene (0.5–2.4 ppb), but much more widespread. The sensitivity of ozone to methane levels found in this work is somewhat lower than reported by *Fiore et al.* [2002], who obtained a decrease of 3 ppb in afternoon ozone levels and a 54% decline in exceedances of 80 ppb in response to a 50% reduction in atmospheric methane. There are numerous differences between the present study and that of *Fiore et al.* [2002], including the chemical mechanism, spatial resolution, emission inventory, and the magnitude and sign of the methane perturbation. Our work supports the conclusion that it is also important to consider changing methane concentrations when assessing the impact of climate change on air quality, and indeed that methane may have a bigger impact at 2050 than climate-induced changes in isoprene emissions.

[30] Figure 11 shows the number of 36 × 36 km model grid cells where the predicted MDA8 ozone mixing ratio in a given region exceeds 80 ppb. Each bar represents one year; in each panel, the first grouping is for the CURR simulation, the second grouping is for the FUT1 case, and the final grouping is for the FUT2 simulation. In light of the model's positive bias in comparison to observations, these exceedance frequencies are also likely to be biased high, particularly for the Plains and Southeast regions. However, they serve to illustrate the significant interannual variability in frequency of extreme ozone events, demonstrating the necessity of conducting multiyear simulations.

4. Summary and Conclusions

[31] Previous studies have demonstrated that regional-scale air quality could worsen in a future climate [*Mickley et al.*, 2004a; *Hogrefe et al.*, 2004; *Unger et al.*, 2006; *Forkel and Knoche*, 2006]. Given that there are obvious uncertainties in both the future scenarios and models, additional modeling studies are needed to develop robust conclusions about climate impacts on air quality. To this end, a series of present and future simulations of regionally downscaled climate and air quality has been developed. The regional climate simulations have been presented and evaluated in previous papers, and this paper presents the ozone predictions developed from this study.

[32] Evaluation of the ozone predictions for the present climate shows a substantial positive bias, which may be influenced both by the choice of chemical mechanism and by meteorological prediction uncertainties to a lesser extent. Comparison of the present and future air quality model predictions shows increases in ozone in Texas and portions of the eastern United States on the order of 2–5 ppb as a result of climate change alone. A larger increase in ozone is predicted for the September and October future months, suggesting an extension of the ozone season, which may have implications for air quality management applications. These increases within the regional modeling domain are

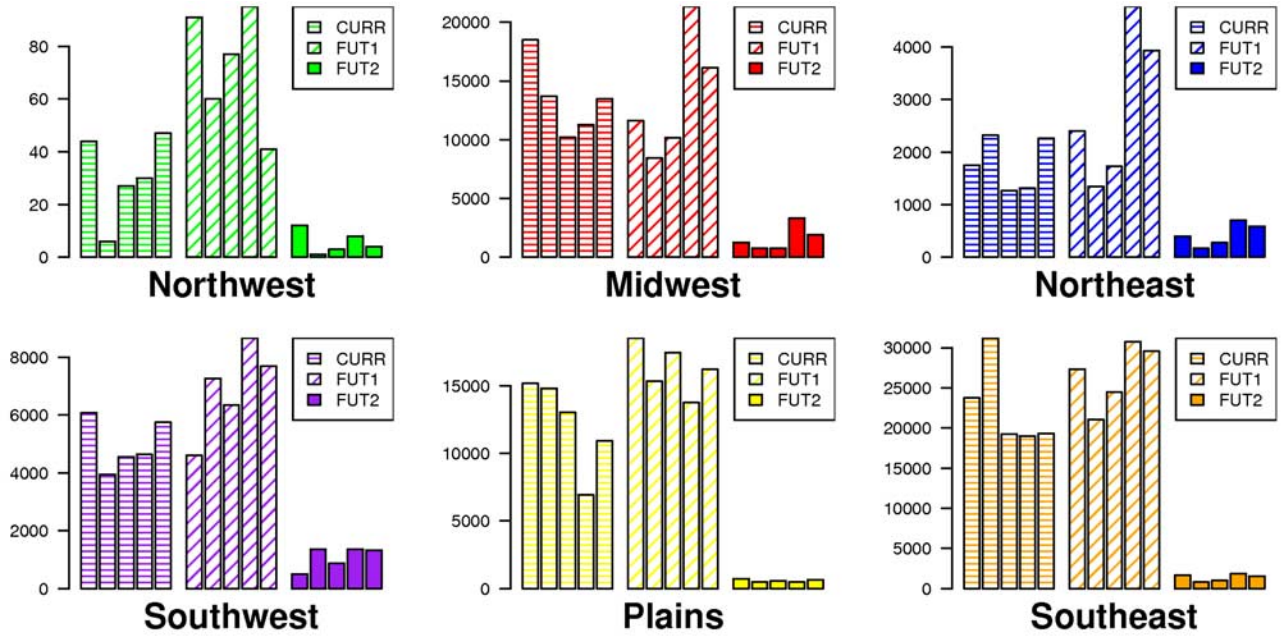

Figure 11. Frequency (grid cell d a^{-1}) in which CMAQ modeled daily maximum 8-h average (MDA8) ozone mixing ratio exceeds 80 ppb. Results broken down by scenario and region as shown. Each bar represents results from 1 year.

predicted despite large decreases in ozone boundary conditions from the global model. Our work shows the importance of including dynamic ozone boundary conditions in regional modeling studies of future air quality.

[33] In contrast to the case considering climate change alone without anthropogenic emissions changes, very large decreases in ozone are predicted with future emissions consistent with A1B AIM; however, the uncertainty in the future emission scenarios for ozone precursors is quite large. *Hogrefe et al.* [2004] found large increases in ozone when adapting the emission inputs to be consistent with the IPCC A2 scenario. While these results lend additional evidence to previous findings that climate change alone could lead to an increase in regional-scale ozone levels in the eastern U.S., they also highlight that it is critical to include a range of future emission scenarios for both greenhouse gases and ozone precursors. We also show that changes in methane concentrations need to be considered alongside changes in climate for assessments of future ozone, and that increased methane may have a larger impact than climate-induced changes in isoprene emissions. Our work is the first we know of to explore the effect of changing methane mixing ratios with the CMAQ model, or any regional scale air quality model.

[34] Avenues for further research include exploring the sensitivity of these findings to alternative emission scenarios, changes in land use patterns, using a more advanced GCM to drive the regional climate and air quality simulations, examining the impact of changes in humidity and clouds on the rates of ozone chemical production and destruction, as well as analyzing the particulate matter concentration predictions from these simulations. Limitations of these simulations include the absence of feedbacks from atmospheric pollutants on the simulated climate and the neglect of stratosphere-troposphere exchange. This study provides one series of model sensitivity studies and should be considered in light of previous and ongoing parallel studies [*Mickley et al.*, 2004a; *Leung and Gustafson*, 2005; *Huang et al.*, 2007; *Liao et al.*, 2006; *Racherla and Adams*, 2006; *Tagaris et al.*, 2007; *Wu et al.*, 2008]. While outside the scope of this individual paper, another forthcoming product of this research will be an intercomparison of findings from several ongoing studies to better understand the range of outcomes that could be anticipated based on climate scenarios and model uncertainties.

5. Disclaimer

[35] The research presented here was performed under the Memorandum of Understanding between the U.S. Environmental Protection Agency (EPA) and U.S. Department of Commerce's National Oceanic and Atmospheric Administration (NOAA) and under agreement number DW13921548. This work constitutes a contribution to the NOAA Air Quality Program. Although it has been reviewed by EPA and NOAA and approved for publication, it does not necessarily reflect their policies or views.

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