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# Attribution of future US ozone pollution to regional emissions, climate change, long-range transport, and model deficiency

H. He<sup>1</sup>, X.-Z. Liang<sup>1,2</sup>, H. Lei<sup>3</sup>, and D. J. Wuebbles<sup>4</sup>

<sup>1</sup>Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD 20740, USA

<sup>2</sup>Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD 20740, USA

<sup>3</sup>National Oceanic and Atmospheric Administration, Air Resource Laboratory, College Park, MD 20740, USA

<sup>4</sup>Department of Atmospheric Sciences, University of Illinois, Urbana, IL 61820, USA

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Correspondence to: X.-Z. Liang (xliang@umd.edu)

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# Abstract

A regional chemical transport model (CTM) is used to quantify the relative contributions of future US ozone pollution from regional emissions, climate change, long-range transport (LRT) of pollutants, and model deficiency. After incorporating dynamic lateral boundary conditions (LBCs) from a global CTM, the representation of present-day US ozone pollution is notably improved. This nested system projects substantial surface ozone trends for 2050's: 6-10 ppbv decreases under the "clean" A1B scenario and  $\sim$  15 ppbv increases under the "dirty" A1Fi scenario. Among the total trends, regional emissions changes dominate, contributing negative 20-50% in A1B and positive 20-40% in A1Fi, while LRT effects through chemical LBCs and climate changes account 10 for respectively 15-50% and 10-30% in both scenarios. The projection uncertainty due to model biases is region dependent, ranging from -10 to 50%. It is shown that model biases of present-day simulations can propagate into future projections systematically but nonlinearly, and the accurate specification of LBCs is essential for US ozone projections. 15

## 1 Introduction

A number of studies have made projections of future air pollution under changes in emissions and climate using chemical transport models (CTMs) at both global and regional scales (Jacob and Winner, 2009; Weaver et al., 2009). Global CTMs repre-<sup>20</sup> sent atmospheric chemistry and transport processes, including long-range transport (LRT), and resulting changes in atmospheric composition, across the planetary scale. However, lack of detailed emissions and inadequate spatial resolution in existing studies can cause substantial errors (e.g., Lin et al., 2008; Lei et al., 2013). Regional CTMs have smaller spatial grids, more detailed emissions, and more explicit chem-<sup>25</sup> ical mechanism, to resolve fine-scale characteristics of atmospheric chemistry and physical process, including transport, but require input of chemical lateral boundary



conditions (LBCs) to account for LRT of air pollutants and their precursors (e.g., Huang et al., 2008). Chemical LBCs can be prescribed from climatological statistics, measurements, or global CTM simulations. For instance, past studies with the US Environmental Protection Agency (EPA) Community Multiscale Air Quality (CMAQ) model have

- <sup>5</sup> used predefined time-invariant LBCs (Tagaris et al., 2007; Hogrefe et al., 2011), satellite retrievals and ozonesonde observations of chemical species (Tang et al., 2009; Pour-Biazar et al., 2011), or global CTM simulations (Akritidis et al., 2013; Hogrefe et al., 2011). These studies indicate that the selection of LBCs can substantially affect the performance of regional CTMs.
- <sup>10</sup> Domestic and international policy decisions to regulate US air quality need identification of individual and combined effects of projected changes in emissions, climate, and LRT (Hogrefe et al., 2004). Prior studies have usually focused on a single scenario for emissions change and/or climate change, and lack separation of future LRT effects (Weaver et al., 2009). In this study, we conducted an evaluation of projected changes in
- <sup>15</sup> US air quality based on multiple future pollution emissions and climate states coupled with fixed and dynamic chemical LBCs. It is a comprehensive study from the regional CTM perspective to separate individual effects of emissions change, climate change, and LRT on future US air pollution, also examined are relative contributions and corresponding uncertainties.

#### 20 2 Model simulations and observations

This study uses the National Center for Atmospheric Research (NCAR) Community Climate System Model version 3 (CCSM3) (Collins et al., 2006a, b) to generate global climate states at the present-day (1995–1999) and future (2048–2052) periods. Two future climate projections were made under the Intergovernmental Panel on Climate

<sup>25</sup> Change (IPCC) Special Report on Emissions Scenarios (SRES) A1B and A1Fi scenarios (Nakicenovic et al., 2000). Meteorological conditions over North America were downscaled using the Fifth-generation Penn State/NCAR Mesoscale Model based re-



gional climate model (CMM5) that refines US climate simulations (Liang et al., 2001, 2004, 2006, 2008).

Anthropogenic emissions of pollutants for the present-day simulation were based on the EPA 2002 National Emissions Inventory (NEI 2002, hereafter called present-day

- <sup>5</sup> emissions). All pollution emissions were processed using the Sparse Matrix Operator Kernel Emissions model (SMOKE) (Houyoux et al., 2000) driven by the CMM5 downscaled meteorology (see details in Tao et al., 2007). Future pollution emissions were specified according to the IPCC SRES A1B and A1Fi scenarios, consistent to CCSM3 climate projections. A1B depicts a balanced forecast for usage of fossil fuel and renew-
- <sup>10</sup> able energy, while A1Fi strongly assumes continued intensive consumption of fossil fuel (Nakicenovic et al., 2000). Therefore, compared with the present-day emissions affecting air quality, A1B and A1Fi emissions represent a "clean" and "dirty" outlook, respectively.

For CMAQ simulations, fixed LBCs were adopted from the predefined vertical profiles with species concentrations as a function of height, which are default in CMAQ representing relatively clean air conditions (CMAS, 2007). Dynamic LBCs were obtained from the NCAR Community Atmospheric Model with Chemistry (CAM-Chem) simulations driven by the same CCSM3 climate and anthropogenic emissions under the approaches as discussed above (see details in Lei et al., 2012, 2013).

The US summertime (June, July, and August) surface ozone was simulated by CMAQ version 4.6, with the Carbon Bond 2005 scheme (CB05) (Yarwood et al., 2005) as the chemical mechanism and driven by the CMM5 meteorology at 30 km grid spacing. Ten CMAQ and five CAM-Chem modeling experiments, using various combinations for three emissions scenarios (present-day, A1B future, A1Fi future), three cli-

<sup>25</sup> mate states (present-day, A1B future, A1Fi future), and two LRT schemes (fixed and dynamic LBCs), were conducted (Table 1). For robust statistics, all analyses of ozone were based on the maximum daily 8 h average (MDA8) ozone concentrations averaged over 5 years, i.e. 1995–1999 for present-day simulations and 2048–2052 for future pro-



jections. These experiments were compared to identify the individual and combined effects of emissions change, climate change, and LRT.

The model performance of present-day ozone simulations was evaluated, relative to surface ozone observations obtained from the EPA Air Quality System (AQS) database (www.epa.gov/ttn/airs/airsaqs). Since these model simulations were based on the NEI 2002 emissions, summertime MDA8 ozone concentrations from AQS were averaged during 2000–2004. The following analyses focused on key regions where high proba-

bility of air quality violations and large sensitivities to climate changes are anticipated, including California, Texas, the Southeast, Northeast, and Midwest. Observations and model simulations were averaged into these five subdomains for comparison (Fig. 1).

# 3 Results and discussion

# 3.1 Present-day ozone pollution and effects of dynamic LBCs

Table 2 compares the 5 year average summer MDA8 ozone concentrations from surface observations and model simulations sampled over the monitoring sites, as well as statistics of the normalized bias (NB) and normalized gross error (NGE), following Tao 15 et al. (2007). Generally, CMAQ with fixed LBCs (case 6) underestimates the summertime surface ozone, with NGE  $\sim$  15% in the Southeast, Northeast, and Midwest and  $\sim$  40 % in California and Texas. While incorporating LRT through dynamic LBCs (case 1), CMAQ simulations are improved in all five subdomains, reducing NGE by 2–10%. This may suggest that CAM-Chem provides more realistic LRT of air pollutants than 20 what the fixed LBCs imply, albeit CAM-Chem (case 11) itself overestimates surface ozone in the US, with NGE  $\sim$  15% in Texas and 30–50% in the other four subdomains. LBCs for regional CTM simulations control LRT of pollutants and their precursors crossing the modeling boundaries. Previous studies show that accurate boundary conditions are essential to model performance of regional CTMs (Pour-Biazar et al., 2011; 25 Tang et al., 2009). In summer, westerly winds in the middle and upper troposphere are



prevalent in the US, so LBCs on the western boundary are crucial to LRT of chemical species (Tao et al., 2007; Huang et al., 2008; Lin et al., 2008; Chen et al., 2009). Transport from the south is also important due to the summertime low level jet (Zhu and Liang, 2013).

- Figure 2 shows the mean nitrogen oxides  $(NO_x)$  and volatile organic compounds (VOCs) altitude profiles from fixed LBCs and dynamic LBCs on the western and southern boundaries of the CMM5-CMAQ domain. In the west, fixed LBCs have similar VOCs concentrations, but substantially overestimate the NO<sub>x</sub> concentrations, relative to using dynamic LBCs. In the south, fixed LBCs significantly underestimate both NO<sub>x</sub> and
- VOCs concentrations, by ~ 60 and ~ 90 %, especially near the surface. The reason is that the south boundary passes Mexico and the Caribbean islands, where significant amount of emissions exists. However, these emissions are missed when using fixed LBCs.

Figure 3 presents an example of CAM-Chem VOCs altitude profiles for the south boundary. The usage of dynamic LBCs substantially improves simulations of surface ozone under present emissions and climate, compared simulations with fixed LBCs. As such, the regional CMAQ nested with the CAM-Chem dynamic LBCs improves prediction of the present-day US ozone, providing higher credibility to study future ozone pollution than the CMAQ with fixed LBCs and the CAM-Chem standalone. In the fol-

<sup>20</sup> Iowing analysis, CMAQ simulations with dynamic LBCs are considered as the baseline reference for subsequent analyses to evaluate the contributions of emissions change, climate change, and LRT effect on future air quality.

#### 3.2 Model biases and uncertainty propagation

Table 3 summarizes regional ozone levels for the five subdomains among the ten <sup>25</sup> CMAQ and five CAM-Chem experiments. The model biases are defined as the differences between simulation results and AQS observations (left column in Fig. 4). Generally, CAM-Chem overestimates summertime MDA8 ozone, by 15–20 ppbv in the Southeast, Northeast, and Midwest, and ~ 2 ppbv in Texas, but underestimates the



value (~ 2 ppbv) in California. CMAQ with dynamic LBCs produces a general underprediction, with ~ 10 ppbv in Texas, ~ 5 ppbv in California, the Southeast and Midwest, and ~ 0.2 ppbv in the Northeast.

Note that the model differences between CAM-Chem and CMAQ with dynamic LBCs (right column in Fig. 4) are not entirely consistent with the model biases. For instance, in California, model biases for CAM-Chem and CMAQ are -2.4 and -5.4 ppbv, i.e. the difference of these two models over the monitoring sites are 3.0 ppbv. However, the model difference for present-day scenarios (defined as CAM-Chem minus CMAQ with dynamic LBCs) is -14.5 ppbv, which is much larger than 3.0 ppbv with opposite sign.

- <sup>10</sup> These discrepancies are likely caused by the sampling bias of monitoring sites. Figure 5 shows the locations of monitoring sites superposed on the present-day CMAQ simulations in California. Monitoring sites are concentrated in urban and suburban areas such as the Central Valley and the Los Angeles Basin, where high ozone levels are predicted; however limited monitoring sites exist in rural areas where low ozone levels
- <sup>15</sup> are predicted. As shown in Table 2, the 5 yr mean MDA8 ozone from EPA AQS observations is 53.6 ppbv, higher than the present-day prediction by CMAQ over these monitoring sites (48.2 ppbv), but there exists positive NB (12.8 %). These results suggest that models, both CAM-Chem and CMAQ, have large overestimates in areas where low ozone levels are observed, while small underestimates in areas where high ozone
- 20 levels are observed. These biases are caused by the uneven distribution of air quality monitoring sites, and cannot be eliminated in the model evaluation. The result suggests that more monitoring sites are needed in rural areas to better represent future ozone pollution.

The differences between the driving CAM-Chem and its downscaling CMAQ under the present-day climate and emissions to those under future scenarios (i.e., case 1 vs. case 11 for present-day study; case 2/3 vs. case 12/13 for A1B/A1Fi scenarios) are similar in California (> -10 ppbv), Southeast (2–6 ppbv), and Northeast (1–3 ppbv). However, differences with opposite sign are shown in Texas (~ 3 ppbv for the presentday and ~ -2 ppbv under future scenarios). In the Midwest, the differences are similar



for the present-day and A1B scenarios (3–4 ppbv) but opposite under A1Fi (-1.0 ppbv). Given the large differences in the chemistry mechanism, spatial resolution and emissions, the contrasts between the global and regional CTMs are well preserved in going from the present to future cases. The results suggest that the principal characteristics

- of model biases in simulating the present-day ozone pollution are systematically propagated into the future projections, suppressed (A1B) and amplified (A1Fi) by CMAQ relative to CAM-Chem. The actual correspondences between present-day model biases and future projected changes are however highly nonlinear, and thus cannot be simply removed through subtracting the output of CAM-Chem itself. A similar conclusion was reached for regional climate change projection (Liang et al., 2008). Therefore
- sion was reached for regional climate change projection (Liang et al., 2008). Therefore regional nested CTM simulations for both present and future scenarios are needed to resolve the bias correction issue.

# 3.3 Trends of future US ozone

Tropospheric ozone is produced through photochemical reactions of precursors, mainly NO<sub>x</sub> and VOCs (EPA, 2006; Seinfeld, 1991). Table 4 summarizes the changes of projected ambient temperature and emissions of NO<sub>x</sub> and VOCs. Surface temperature increases by 1.3–2.0 °C and 1.5–3.4 °C in different subdomain under A1B and A1Fi scenarios, respectively. Anthropogenic NO<sub>x</sub> emissions decrease by 30–50 % under A1B emissions and increase by 30–40 % under A1Fi emissions. Because VOCs emissions,

especially biogenic VOCs, are enhanced by high ambient temperature (Camalier et al., 2007; Bloomer et al., 2009; Jacob and Winner, 2009), future VOCs emissions increase by 10–30 % and 15–50 % under A1B and A1Fi climate, respectively.

Figure 6 presents the future ozone trends projected by CAM-Chem and downscaled by CMAQ (i.e., case 11 vs. case 12/13 for CAM-Chem; case 1 vs. case 11/13 for CMAQ

with dynamic LBCs; case 6 vs. case 7/8 for CMAQ with fixed LBCs). Future ozone levels are mainly controlled by emissions changes over North America. For instance, CAM-Chem projects 4–11 ppbv decreases and 8–13 ppbv increases of summertime MDA8 ozone under A1B and A1Fi, respectively. CMAQ simulations with fixed LBCs



show similar trends, 4–14 ppbv decreases and 8–12 ppbv increases under A1B and A1Fi, respectively.

However, the CMAQ downscaling with dynamic LBCs produces different trends than the CMAQ with fixed LBCs. Under A1B, CMAQ simulates  $\sim 0.5\, ppbv$  increases (rather

- than decreases) in Texas, whereas 6–10 ppbv decreases in the other four subdomains are weaker than CAM-Chem. LRT of air pollutants into the US compensates partially for the effects of emissions reductions in CMAQ on the projected future ozone for the Southeast, Northeast, Midwest, and California, while increase of MDA8 ozone in Texas shows the importance of cross-border transport from Mexico. Under A1Fi, CMAQ with
- dynamic LBCs generates ~ 15 ppbv increase of future MDA8 ozone, higher than CAM-Chem, suggesting that the LRT effect and emissions increase are combined. Therefore, although emissions changes dominate the future US ozone pollution trend, dynamic LBCs from the driving global CTM can substantially influence prediction from the regional CTM.

#### **3.4** Relative contributions on future US ozone pollution

As discussed above, the CAM-Chem model projects different future ozone changes than the regional CMAQ model (Table 3). This difference can be defined as model deficiency due to different model structures, including chemical and physical schemes between the global CAM-Chem and regional CMAQ. The individual contributions from emissions, climate, LRT, and model deficiency, to the future ozone projection, are determined following Tao et al. (2007). The relative contribution from factor  $X_i$  is defined as  $X_i % = \frac{X_i}{\sum |X_i|} \times 100 \%$ , where  $X_i$  stands for emissions, climate, LBCs, and model deficiency. These values are calculated as follows:

1. Effects of emissions are calculated through comparing results from CMAQ with dynamic LBCs under future emissions and CMAQ with dynamic LBCs under present emissions, e.g., case 2 minus case 4.



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- 2. Effects of climate changes are calculated through comparing present and future simulations from CMAQ with dynamic LBCs under present emissions, e.g., case 4 minus case 1.
- 3. Effects of LBCs are calculated through comparing results of CMAQ with dynamic LBCs and CMAQ with fixed LBCs under the same climate and emissions, e.g., case 2 minus case 7.
- 4. Effects of model deficiency are calculated through comparing results from CMAQ and CAM-Chem under the same climate and emissions, e.g., case 1 minus case 11.
- <sup>10</sup> 5. The total effects are defined as sum of all these 4 factors.

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Figure 7 indicates that the future ozone projection is non-linear to emissions change, climate change, LRT effect, and model deficiency. Changes of regional emissions substantially influence future ozone levels, contributing negative 20–50% under A1B decreased emissions and positive 20–40% under A1Fi increased emissions, to the total effects. Contributions of climate changes are all positive (10–30%) because of the en-

hanced ambient temperature and biogenic emissions as discussed above.

The LRT effect through incorporation of dynamic LBCs also boosts the future ozone levels, but shows strong regional dependence under both A1B and A1Fi. It contributes 40–50% of the total effects in California and Texas while only 15–30% in the South-

- $_{20}$  east, Northeast, and Midwest. These results suggest that California and Texas are more vulnerable to LRT of air pollutants, which likely origin from Asia and Mexico, respectively. The relative contribution of model deficiency to the total effects has large regional variations, from around  $-10\,\%$  in California and Midwest,  $\sim 10\,\%$  in Northeast, and 30–50% in Southeast, under A1B and A1Fi. In Texas, model deficiency contributes
- around –15 % under A1B and ~ 5 % under A1Fi, which could be caused by the change of ozone precursor emissions discussed above. The relative contributions of model deficiency (positive in Northeast and Southeast, negative in California and Midwest) are



generally consistent with the CAM-Chem ozone biases having overestimates (underestimates) in the eastern (western) US (Fig. 7). The CAM-Chem present-day model biases are propagated systematically but nonlinearly at regional scales into the future ozone projections in CMAQ through incorporating dynamic LBCs. It is thus possible to qualitatively estimate the potential influence of dynamic LBCs from a global CTM on the future regional CTM projection by examining their differences in simulating the presentday condition. A complete removal of the influence due to model biases is however not feasible.

# 4 Conclusions and discussion

- The CMM5-CMAQ system can successfully reproduce summertime ozone pollution and its incorporation of dynamic LBCs from CAM-Chem can improve the overall model performance. The results suggest that effects of LBCs for driving the regional CTM are comparable, if not larger, to effects of climate changes, both of which are regionally dependent. As such, for the future air quality study the accuracy of LBCs or the realistic
- LRT effects represented is as important as the accuracy of future climate projections. It is also found that the CAM-Chem biases can propagate systematically but nonlinearly from the present-day simulation into the future change projection, resulting in large uncertainties due to model structure errors.

In conclusion, simulations with current global CTMs alone are not optimal to project future ozone pollution in the US because of their inability to resolve both emissions and climate as accurate as regional CTMs. Therefore, regional CTMs refinements as nested with dynamic LBCs from global CTM predictions are necessary to incorporate both more realistic LRT effects and more detailed local emissions changes to more credibly project the future US air quality. Since major biases of global CTMs can propagate from the present-day into future simulations and also into the North American domain through dynamic LBCs, cautions must be taken in interpreting the US ozone



trends in 2050s resulted from this study that assumes the nested system as the baseline proxy for future ozone projection.

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|      |          | Experiment Design    |                        |                  |
|------|----------|----------------------|------------------------|------------------|
| Case | СТМ      | Climate <sup>a</sup> | Emissions <sup>b</sup> | LRT <sup>℃</sup> |
| 1    | CMAQ     | Present              | Present                | Dynamic          |
| 2    | CMAQ     | A1B                  | A1B                    | Dynamic          |
| 3    | CMAQ     | A1Fi                 | A1Fi                   | Dynamic          |
| 4    | CMAQ     | A1B                  | Present                | Dynamic          |
| 5    | CMAQ     | A1Fi                 | Present                | Dynamic          |
| 6    | CMAQ     | Present              | Present                | Fixed            |
| 7    | CMAQ     | A1B                  | A1B                    | Fixed            |
| 8    | CMAQ     | A1Fi                 | A1Fi                   | Fixed            |
| 9    | CMAQ     | A1B                  | Present                | Fixed            |
| 10   | CMAQ     | A1Fi                 | Present                | Fixed            |
| 11   | CAM-Chem | Present              | Present                | Global           |
| 12   | CAM-Chem | A1B                  | A1B                    | Global           |
| 13   | CAM-Chem | A1Fi                 | A1Fi                   | Global           |
| 14   | CAM-Chem | A1B                  | Present                | Global           |
| 15   | CAM-Chem | A1Fi                 | Present                | Global           |

 Table 1. Experiment Design of regional CMAQ and global CAM-Chem experiments.

<sup>a</sup> Present climate: 1995–1999; future climate: 2048–2052.

<sup>b</sup> Present emissions: NEI 2002.

 $^{\rm c}$  Fixed LBCs are from the time-invariant predefined profiles, dynamic LBCs are from CAM-Chem simulations.



|         | Data/Model |   | California           | Texas                 | Southeast             | Northeast            | Midwest               |
|---------|------------|---|----------------------|-----------------------|-----------------------|----------------------|-----------------------|
| Obs.    | EPA AQS    | Site No. <sup>a</sup><br>Mean, ppbv                     | 148<br>53.6          | 91<br>46.2            | 110<br>53.9           | 160<br>53.4          | 220<br>52.1           |
| Case 11 | CAM-Chem   | Mean, ppbv<br>NB, % <sup>b</sup><br>NGE, % <sup>c</sup> | 51.2<br>13.3<br>42.0 | 48.5<br>5.4<br>14.4   | 69.6<br>30.0<br>30.0  | 73.9<br>48.7<br>48.7 | 68.5<br>32.4<br>32.4  |
| Case 1  | CMAQ       | Mean, ppbv<br>NB, %<br>NGE, %                           | 48.2<br>12.8<br>37.4 | 35.0<br>-23.9<br>24.1 | 48.3<br>-9.6<br>11.6  | 53.2<br>6.9<br>14.8  | 48.0<br>-6.9<br>10.1  |
| Case 6  | CMAQ       | Mean, ppbv<br>NB, %<br>NGE, %                           | 43.2<br>2.0<br>39.6  | 30.2<br>-34.6<br>34.6 | 44.8<br>-16.2<br>17.0 | 49.8<br>-0.1<br>16.8 | 44.8<br>-13.2<br>15.1 |

#### **Table 2.** Comparison of present-day ozone simulations with EPA AQS observations.

<sup>a</sup> Number of EPA AQS site in the selected domain. <sup>b</sup> NB: normalized bias, NB =  $\frac{1}{N} \sum_{i=1}^{N} \frac{CMAQ_i - Obs_i}{Obs_i} \times 100\%$ . <sup>c</sup> NGE: normalized gross error, NGE =  $\frac{1}{N} \sum_{i=1}^{N} \frac{[CMAQ_i - Obs_i]}{Obs_i} \times 100\%$ .

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**Table 3.** Summary of summertime average MDA8 ozone concentrations from present-day and future simulations in each subdomain.

|      | Regional Mean MDA8 ozone (ppbv) |       |           |           |         |
|------|---------------------------------|-------|-----------|-----------|---------|
| Case | California                      | Texas | Southeast | Northeast | Midwest |
| 1    | 64.7                            | 49.3  | 56.7      | 67.7      | 60.1    |
| 2    | 56.0                            | 50.0  | 51.1      | 57.3      | 52.5    |
| 3    | 77.8                            | 62.8  | 70.7      | 83.0      | 75.9    |
| 4    | 62.4                            | 55.1  | 63.8      | 70.1      | 62.6    |
| 5    | 67.9                            | 54.2  | 62.0      | 73.7      | 67.5    |
| 6    | 56.5                            | 45.3  | 53.4      | 64.3      | 56.9    |
| 7    | 45.1                            | 41.5  | 43.8      | 50.0      | 46.0    |
| 8    | 64.3                            | 54.9  | 64.4      | 75.4      | 68.7    |
| 9    | 55.6                            | 44.7  | 55.0      | 63.8      | 57.8    |
| 10   | 59.0                            | 48.7  | 57.4      | 68.5      | 62.7    |
| 11   | 50.1                            | 52.5  | 62.7      | 70.2      | 64.4    |
| 12   | 44.8                            | 48.0  | 53.0      | 58.8      | 55.8    |
| 13   | 58.2                            | 60.2  | 74.2      | 83.4      | 74.9    |
| 14   | 53.4                            | 53.7  | 64.3      | 73.2      | 66.8    |
| 15   | 53.5                            | 54.3  | 69.0      | 78.1      | 70.8    |



**Table 4.** Summary of summer mean ambient temperature,  $NO_x$  and VOCs emissions, from present and future CMAQ simulations in each subdomain.

| Case  | Temperature, °C          | $NO_x$ , mole s <sup>-1</sup> | VOCs, mole $s^{-1}$ |
|-------|--------------------------|-------------------------------|---------------------|
| 1, 2  | 22.9/27.4/27.0/23.7/23.6 | 1.0/1.8/2.0/3.1/2.5           | 4.1/3.3/6.2/3.6/2.5 |
| 3, 4  | 24.9/29.3/28.9/25.0/25.1 | 0.6/1.2/1.1/1.6/1.4           | 4.5/4.4/7.7/3.6/3.4 |
| 5, 6  | 26.3/30.0/28.7/25.2/25.7 | 1.4/2.4/2.7/4.2/3.3           | 5.0/4.2/7.2/4.2/3.9 |
| 7, 8  | 24.9/29.3/28.9/25.0/25.1 | 1.0/1.8/2.0/3.1/2.5           | 4.1/3.3/6.2/3.6/2.5 |
| 9, 10 | 26.3/30.0/28.7/25.2/25.7 | 1.0/1.8/2.0/3.1/2.5           | 4.1/3.3/6.2/3.6/2.5 |

Values for California, Texas, Southeast, Northeast, and Midwest are separated by "/", respectively.



**Figure 1.** The CMM5-CMAQ domain (the outmost box) and subdomains. CA: California; TX: Texas; SE: Southeast; NE: Northeast; MW: Midwest.











Figure 3. Averaged altitude profiles of VOCs on the south boundary of the CMM5-CMAQ domain (data below  $\sim$  200 hPa are showed).





**Figure 4.** Present-day bias, model difference, and trends of MDA8 ozone. Bias is defined as the discrepancy between model simulations and surface observations. Model differences are calculated through CAM-Chem results minus CMAQ with dynamic LBCs.





**Figure 5.** Comparison of EPA AQS ozone observations and model simulations. 5 yr averages of summertime MDA8 ozone are calculated between 2000 to 2004. Filled cycles show the locations and MDA8 ozone levels of EPA AQS observations; background color shows the present-day MDA8 ozone predictions from CMAQ with dynamic LBCs.



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**Figure 6.** Trends of MDA8 ozone. Trends are defined as the future cases minus the present-day cases under A1B (left column) and A1Fi (right column) scenario.







