2.1 GLOBAL MODELING STUDIES OF POTENTIAL CLIMATE CHANGE EFFECTS ON U.S. AIR QUALITY – PART 1: HOW WELL CAN PCM DRIVE THE CHEMICAL TRANSPORT MODEL?

Lin, J.-T. [1], D. J. Wuebbles[1]*, K. Patten[1], K. Hayhoe[1,2], and X.-Z. Liang[3]
[1]Dept. of Atmospheric Sciences, Univ. of Illinois at Urbana-Champaign, Illinois, USA
[2]Dept. of Geosciences, Texas Tech University, USA
[3]Illinois State Water Survey, Illinois, USA

Abstract

Driven by NCAR-DOE Parallel Climate Model (PCM) meteorology, the global chemical transport model MOZART-2.4 (Model for OZone And Related chemical Tracers version 2.4) is used to simulate the global current and future summertime air quality, focusing on the continental U.S. It is shown here that MOZART-2.4 driven by the present-day PCM meteorology produces tropospheric ozone and precursors concentrations that are overall comparable to those driven by the NCEP/DOE AMIP II reanalysis (R-2). However, as compared with the R-2-driven run, the PCM-driven simulation yields higher NOx and ozone concentrations off the western coasts of the major continents and larger CO values over most of the globe. In North America, the main disagreement occurs in the areas with high anthropogenic emissions such as the New York area and California, where PCM tends to produce a photochemical environment that suppresses local air pollution. These disagreements due to PCM climate biases may have important consequences on the U.S. air quality study using a regional modeling system that requires the specification of lateral boundary conditions of the chemistry from the MOZART-2.4 output, especially the inflow CO, NOx and ozone background concentrations off the southwestern coast. In addition, the biofuels and biomass burning emissions of CO are scaled by a factor of 1.55 in order to comply with the IPCC (2001) budget analyses. A sensitivity experiment shows that this scaling has little impact on the MOZART-2.4 simulation of the CO, NOx and ozone concentrations outside the areas dominated by biofuels or biomass burning emissions.

1. Introduction

Air quality simulations are an important approach in evaluating the adverse effects of different pollutants to human health (Lippman 1993), agricultural and forest productivity (Heck et al. 1984; Friedman et al. 1988) and ecosystem (NRC 1991). A state-of-the-art integrated air quality modeling system, which couples global and regional meteorological and chemical models, is being used to simulate current and future air quality projection under global climate and emissions change scenarios (Liang et al. 2005a).

Air quality models rely on meteorological data to specify the atmospheric circulation that controls the photochemical variation. This reliance arises from the strong dependence of production and evolution of major pollutants (ozone and particulate matters) upon meteorological conditions, including temperature, wind, solar radiation, humidity and mixing height (Comrie 1996; Sillman 1999; Gebhart et al. 2001). Past model studies suggested that meteorology is one of the major sources of model uncertainty (Russell and Dennis 2000 and references wherein). Numerous studies have shown that the largest impact on daily pollutant variations comes from meteorology (e.g., Comrie 1996; Davis and Gay 1993; Wise and Comrie 2005). However, the uncertainty introduced by meteorological inputs has not been well quantified due to the lack of high-quality observations and insufficient model intercomparisons (Russell and Dennis 2000), especially given the nonlinear responses of different pollutants to the change of meteorology. The projection for future air quality will depend on that for future meteorology and the corresponding uncertainty needs to be quantified. This study evaluates the MOZART-2.4 sensitivity to the meteorology as generated by the fully-coupled model PCM (Washington et al. 2000) versus the observational proxy R-2 (Kanamitsu et al. 2002).

* *Corresponding author address:* Donald J. Wuebbles, Univ. of Illinois, Dept. of Atmospheric Sciences, Urbana, IL, 61801; email: Wuebbles@atmos.uiuc.edu.

2. Model Experiment Design

2.1 Model Description

MOZART-2.4 is a global chemical transport model widely used in studies of global distributions and seasonal variations of atmospheric trace gases and particles, plus effects on these resulting from climate variability and changes (e.g., Hauglustaine et al. 1999; Horowitz et al. 2003; Lamarque et al. 2004). MOZART-2.4 simulates the long-range transport of a comprehensive set of pollutants and precursors affecting air quality around the planet (Solomon et al. 2000; NRC 2001). MOZART-2.4 can be driven by various meteorological datasets with the flexibility to adopt the spatial resolution of the input data. Here we use the R-2 at a horizontal resolution of T62 (~1.875° or 210 km) with 28 vertical levels and the PCM at T42 (~2.8125° or 310 km) with 18 vertical levels. Three MOZART-2.4 simulations for the summer of 1996 are conducted and compared in this study. The first experiment is driven by the R-2 meteorology but uses the MOZART-3.0 emissions (see section 2.2), hereafter referred to as the Old-CO run. The other two parallel simulations are driven by the R-2 and PCM meteorology and use biofuels and biomass burning emissions of CO that are scaled up to comply with the IPCC budget analyses (see section 2.2), hereafter referred to as the R-2 and PCM run, respectively.

2.2 Emissions

The emissions as used in the Old-CO run are derived from the MOZART-3.0 database, which is based on the one described in details by Horowitz et al. (2003). In the R-2 and PCM simulations, biofuels and biomass burning emissions of CO are scaled up by a factor of 1.55, i.e., from 673 to 1042 Tg/yr, based on the IPCC budget analyses (Houghton et al. 2001). This increases the global CO budget from 1162 to 1532 Tg/yr and puts biofuels and biomass burning emissions to the high end of the current estimate. The adjustment, however, has a relatively small impact over the continental U.S. where the primary CO emitter is fossil fuel burning, industry and transportation. For the same reason, the U.S. contributes almost 15% to global annual NOx emissions. Most organic carbon emissions in the U.S. come from isoprene, methanol and terpenes. Despite its strong emissions, methanol is not a chemically active species compared to isoprene and terpenes, thus its contribution to ozone formation is relatively insignificant. In contrast, NMV (anthropogenic hydrocarbons containing 4 carbons or more per molecule) are primary anthropogenic organic carbon species because of their high reactivity with hydroxyl radical (OH, the atmospheric cleanser) and strong emissions.

	Global	USA only
CO	1161.9	97.8
CO [a]	1531.6	102.4
NOx	45.3	6.5
Acetone	27.2	1.7
C2H4	17.5	0.9
C2H6	12.5	1.1
C3H6	6.8	0.3
C3H8	11.8	0.7
CH2O	4.2	0.2
CH3OH	238.7	17.7
NMV	65.5	9.3
Isoprene	568.6	26.2
Terpenes	144.2	9.8

Table 1. Annual emission budgets of ozone precursors. The units are Tg/yr for CO and oxidized hydrocarbons, TgN/yr for NOx and TgC/yr for hydrocarbons.
[a] Biofuels and biomass burning emissions are scaled up by 155%

2.3 Meteorology

Figure 1 compares the R-2 and PCM simulated temperature and wind distributions on the surface (averaged in the lowest 100 m layer in R-2 and 150 m layer in PCM) over North America averaged in the summer of 1996. The temperature differences over the surrounding oceans are generally within 1 ˚C. Both R-2 and PCM show high temperatures exceeding 24˚C in the southern U.S. PCM, however, produces systematically cold biases over the Great Plains, with 2-8˚C colder temperature than observations (Liang et al. 2005b). Cold biases are also found along the eastern coastal States (1-3˚C) and southwest U.S. (2-8˚C). The cold biases in the Great Plains and southwest U.S. are identified with a poor simulation of the lower-level jet stream that has a substantially weaker core in Texas and turns too quickly toward northeast at the northern extend. In addition, high temperatures in the southwest and the southeast are separated by a cool airmass. This cold tongue is too strong in PCM as compared with R-2. In the central U.S., the PCM biases are generally within 1˚C.

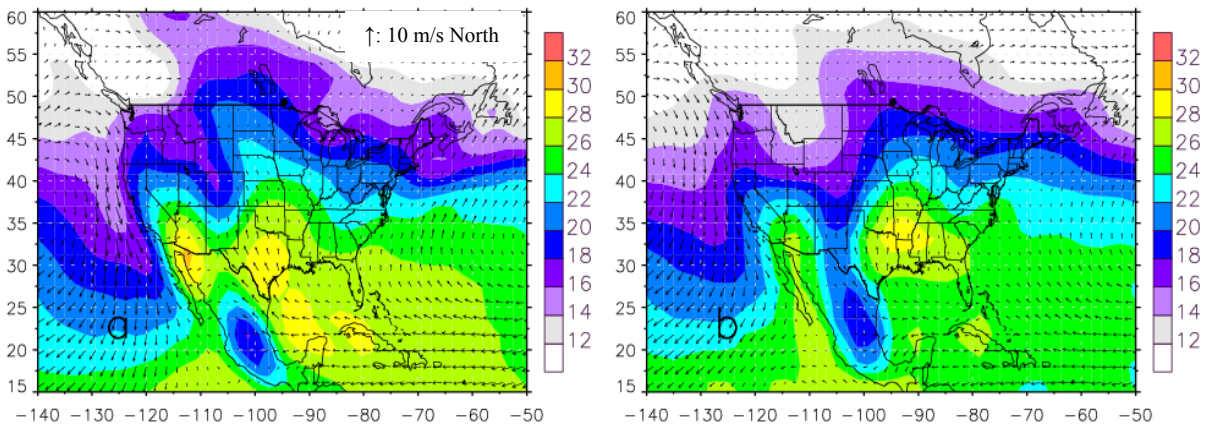

Figure 1. R-2 (a) and PCM (b) simulated surface (2-m) temperature and (20-m) wind distributions averaged in 1996 summer. The temperature (°C) is shown in colors, while the wind (m s^{-1}) is depicted by vectors.

The surface wind circulation simulated by the PCM is reasonable as compared with R-2, where the Bermuda subtropical Atlantic high and the Northeast Pacific high are well captured. Important differences, however, exist in their core position, strength and associated slows. In particular, the Great Plain lower-level jet in PCM is too weak, causing inadequate regulation over the Great Plains and central U.S. by the warm/moist airmass from the Gulf of Mexico. In addition, the wind in the northeast U.S. is weak, implying a more stagnant atmosphere and accumulation of pollutants there.

3. Results

3.1 CO

Figure 2 shows that in case Old-CO, the control run (Panel a), CO exceeds 90 ppb throughout the continental U.S. Over most of the western U.S., CO is consistently between 90 and 120 ppb, with the exception in California where the overall concentration is more than 200 ppb. The Los Angeles area has a CO level spike. In the eastern U.S., the Midwest, represented by the downstream areas of Chicago, and the Northeast, centered at New York City (NYC), are noticeable because of high CO levels, the result of high

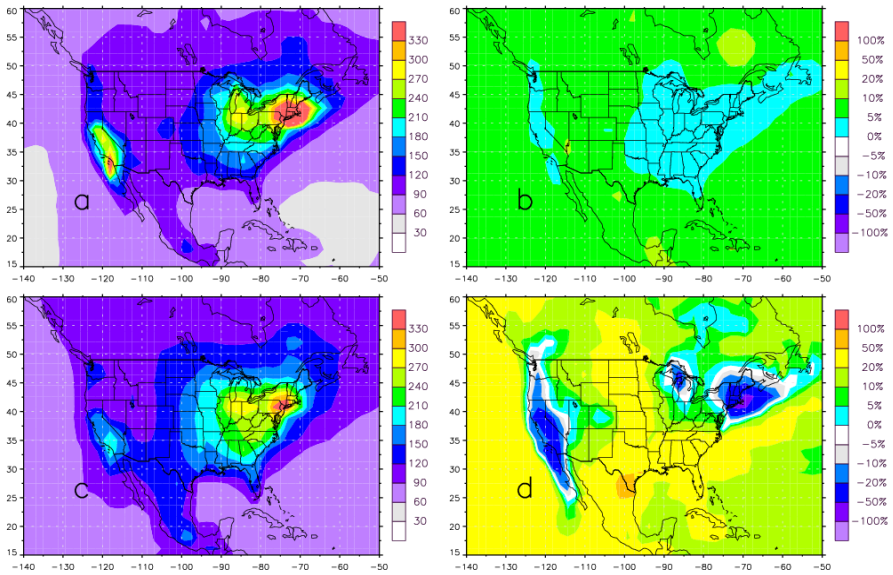

Figure 2. Daily mean surface CO (ppb) in 1996 summer. (a) the control run or Old-CO (with the R-2 meteorology and no CO emissions scaling), (b) percentage difference between Cases R-2 and Old-CO. (c) Case PCM, (d) percentage difference between Cases PCM and Old-CO.

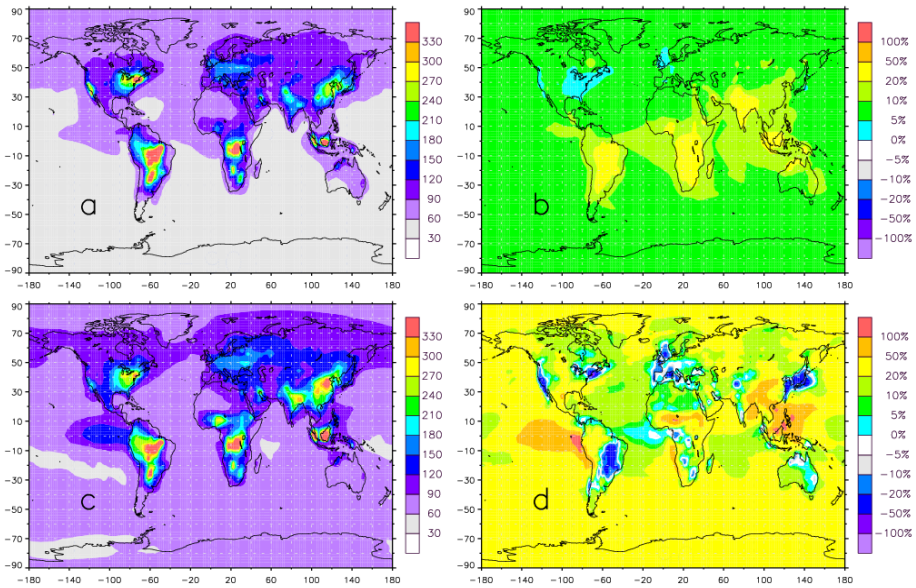

Figure 3. Same as Fig. 2 except over the globe..

emissions and modest chemical reactivity. In comparison, for the R-2 case, when CO emissions are scaled but the meteorology is kept unchanged, no significant increase of CO concentration is seen, and the impacts in the western coastal area and the eastern U.S. are within 5% (Panel b). This is consistent with the insignificant emissions increase through the scaling in these areas. When the PCM meteorology is used, however, the most noticeable feature is the decrease in such polluted areas as California and NYC (Panels c and d). Note that the increase of CO especially off the western coast, though partially contributed by the CO emissions scaling, is consistent with a stronger Northeast Pacific High in the PCM meteorology and suggests more CO is transported from the upstream sources such as Asia in the corresponding simulation. Nevertheless, PCM overall offers a fairly good meteorological field for MOZART-2.4 and the differences between Case PCM and Case Old-CO are much better than 50% in most parts of North America.

Throughout the globe, high CO concentrations are marked by high anthropogenic (fossil fuel, industrial, and transportation) emissions. This includes the U.S., South America, the southern Africa and Southeast Asia (also marked by high biomass burning emissions) (Figure 3). The maximum concentration exceeds 300 ppb in most of the highly polluted areas. In China, daily average CO exceeds 180ppb and is even higher in the northeastern coastal areas due to fast industrial development and modest emission control. Northern India is facing similar problems. In comparison, Western Europe, although well developed, does not produce as much CO pollution partially due to the stricter emission control. Over most of the oceans, CO levels are below 60ppb. However, the northern Atlantic Ocean is an exception due to surrounding strong emitters and climate conditions. It is also noted that in the Old-CO control run (Panel a), there is an obvious north-south downward hemispheric gradient of CO. The scaling of biofuels and biomass burning emissions do significantly increase CO concentration where those emissions dominate such as in South America, the southern Africa, India and Southeast Asia, followed by their downstream areas. However, the increase is less than 10% outside those regions (Panel b). In comparison, driven by PCM, MOZART-2.4 cannot capture the extreme CO levels found in many areas (Panels c and d). It is also noted that PCM leads to much higher CO over the southern oceans so that the hemispheric gradient disappears. Investigation of daily mean wind fields suggests that stronger anti-cyclones off the southeastern coast of South America in PCM brings more pollutant from the strong CO source into the southern ocean regions and contributes to the difference with the R-2 case. Such differences tend to hold globally under the PCM meteorology.

3.2 NOx

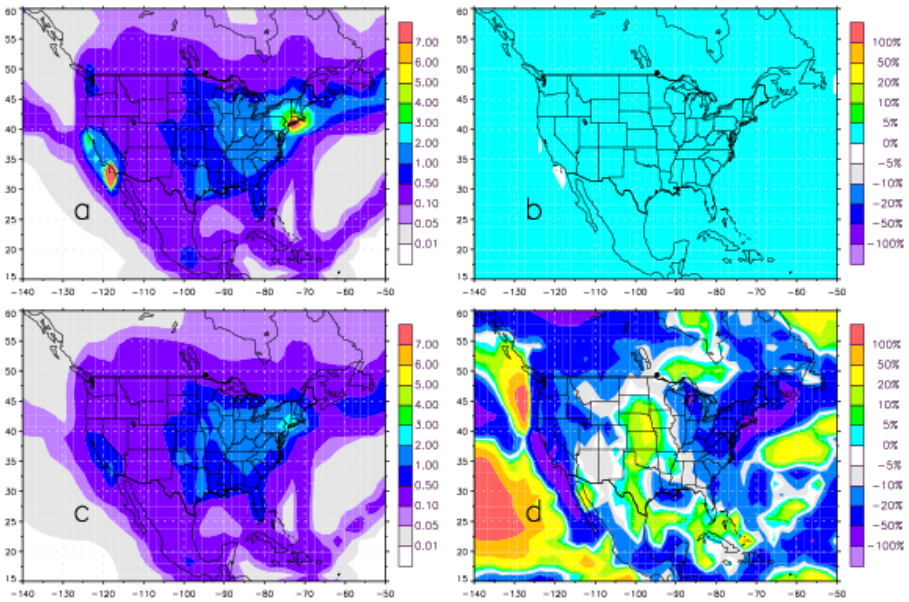

Figure 4. Same as Fig. 2 except for NOx at 2100 GMT.

Summer mean afternoon NOx concentration (at 2100 GMT, Figure 4) is greatly heterogeneous over the continental U.S. in that the highs and lows differ by a factor of 50 (in the control run, Panel a). Consistent with NOx emissions, the highs are in the northeast, represented in this discussion as NYC, and California, represented as Los Angeles (LA), and exceed 7 ppb despite the fast depletion of NOx during the daytime when ozone is formed. The CO emissions scaling does not have significant impact on NOx (Panel b). Under the PCM meteorology, MOZART-2.4 does not reproduce the extreme NOx concentration in both NYC and Los Angeles, consistent with the CO simulations in the same areas. The disagreement in the LA area is caused by the coarser PCM resolution, smaller extreme temperature and stronger westerly. The positive difference off the western coast is partly because NOx levels in these areas are very low and consequently very sensitive to meteorological conditions. Nevertheless, in most parts of the

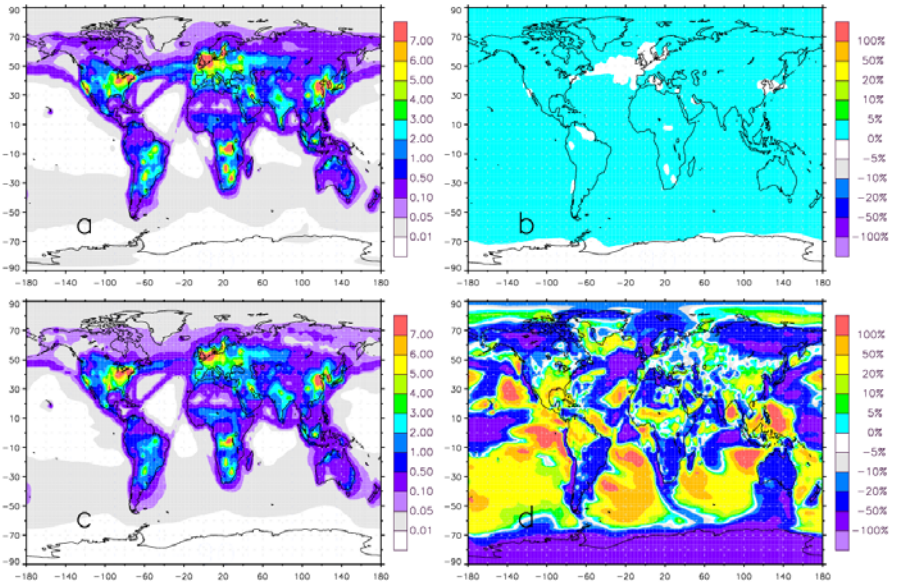

Figure 5. Same as Fig. 2 except for NOx over the globe.

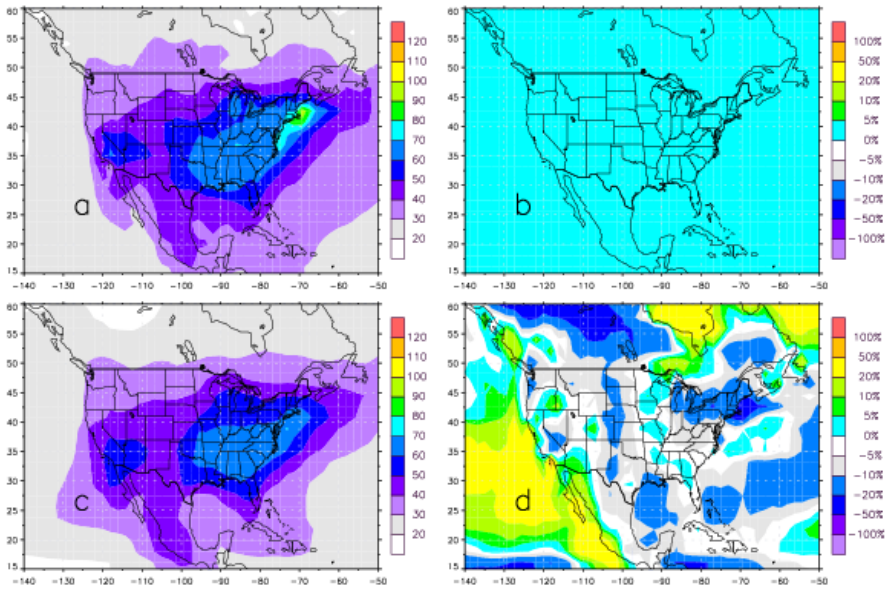

Figure 6. Same as Fig. 2 except for ozone.

inland continent, the differences between the R-2 and the PCM cases are within 20% (Panel d), which suggests that PCM overall is highly representative outside those highly polluted areas, especially given the short lifetime of NOx and the resulting sensitivity to meteorology.

On the global scale, summer mean NOx levels spread by several orders of magnitude because of its short surface lifetime (Figure 5, Panel a and c). The extreme concentrations in the U.S., Europe and China correspond to high anthropogenic emissions. The CO emissions scaling slightly increase NOx concentration in most regions, while a decrease is also seen over the northern Atlantic Ocean and Western Europe. Nevertheless, the difference is within 5% globally (Panel b). Comparison of the R-2 and PCM cases suggests that PCM meteorology leads to much higher NOx over the southern oceans and much lower NOx over the southern continents. The difference over the northern oceans, on the other hand, is greatly variable. In many populated areas such as the U.S., Europe and the China-India area, the PCM case also gives larger values but the difference is within 50% (Panels c and d).

3.3 Ozone

On the summer mean basis, the background surface ozone over most of the continental U.S. is more than 40 ppb, except in the northwest. The highs over the eastern U.S. exceed 60 ppb (Figure 6, Panel a and c). Of more concern are the extremes just off of the northeastern coast, i.e., more than 70ppb, in the control run (Panel a). Given the strong daily variation of ozone so that the maximum 8 hour average, if calculated, would be higher than the daily average. Such an ongoing (or potential) violation of the National Ambient Air Quality Standards (NAAQS) for 8-hour average ozone (80ppb) could have resulting adverse impacts on ecosystem, agriculture and human health. It is noted that no significant difference is introduced by the CO emissions scaling (Panel b). In contrast, the extreme ozone case no longer exists under the PCM meteorology, which is consistent with the CO and NOx simulations and suggests a tendency in the PCM meteorology to reduce extreme pollutant concentrations. Note that ozone concentration in Los Angeles is much larger using the PCM meteorology, although the corresponding NOx level is much lower compared to that using the R-2 meteorology. Although the coarse resolution in either case, together with the topography, may undermine the strength of the simulations in this area, the comparison is indeed consistent with the air quality issue in Los Angeles that ozone production is hydrocarbon limited, and thus the residual NOx diminishes the ozone concentration. The positive difference off the southwestern coast in the PCM case is consistent with positive differences of CO and NOx in the same area. Overall, however, the PCM case gives comparable results to the Old-CO case, with differences within 5-10%, in many areas.

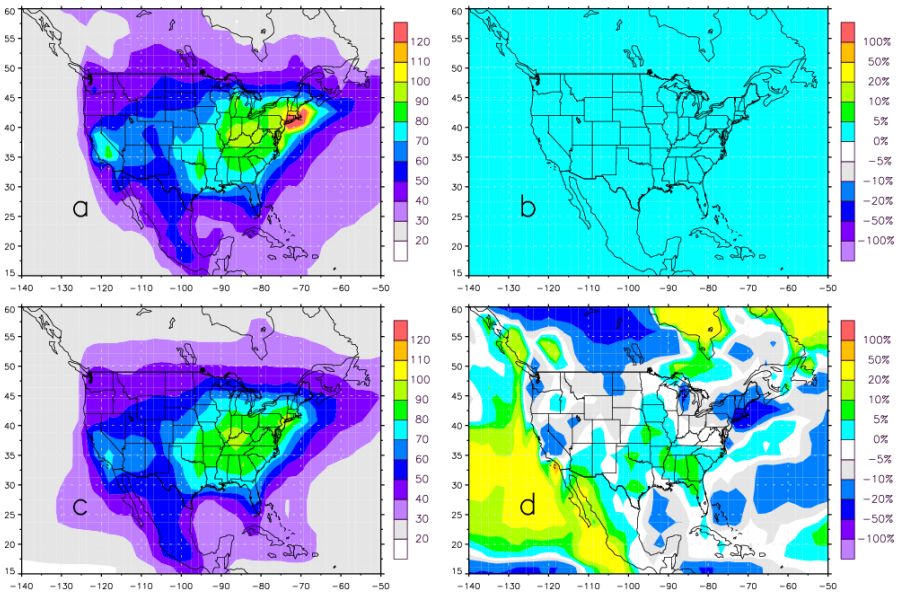

Figure 7. Same as Fig. 2 except for ozone at 2100 GMT.

Figure 7 shows that in the Old-CO control run (Panel a), the summer mean afternoon ozone concentration (i.e., at 2100 GMT) well exceeds 120 ppb (NAAQS for 1-hour ozone) in the New York area. Ozone levels are also high over the eastern U.S. and California. The lowest ozone level is located in the northwest due to both lower emissions of precursors, especially NOx, and cooler weather (see Figure 2). The scaling of CO emissions is negligible (Panel b). Using the PCM meteorology, however, the extreme ozone in the New York area no longer exists (Panel c). Instead, the ozone level throughout the continental U.S. is below 100ppb. Another important feature is in California where the ozone level is decreased by about 10 ppb almost everywhere compared to the control run. The exception in Los Angeles is due to lower NOx, which does not consume ozone as much as under the R-2 meteorology. In addition, the use of PCM meteorology results in slightly higher ozone in the southeast and slightly lower ozone in the northwest and the Midwest with the

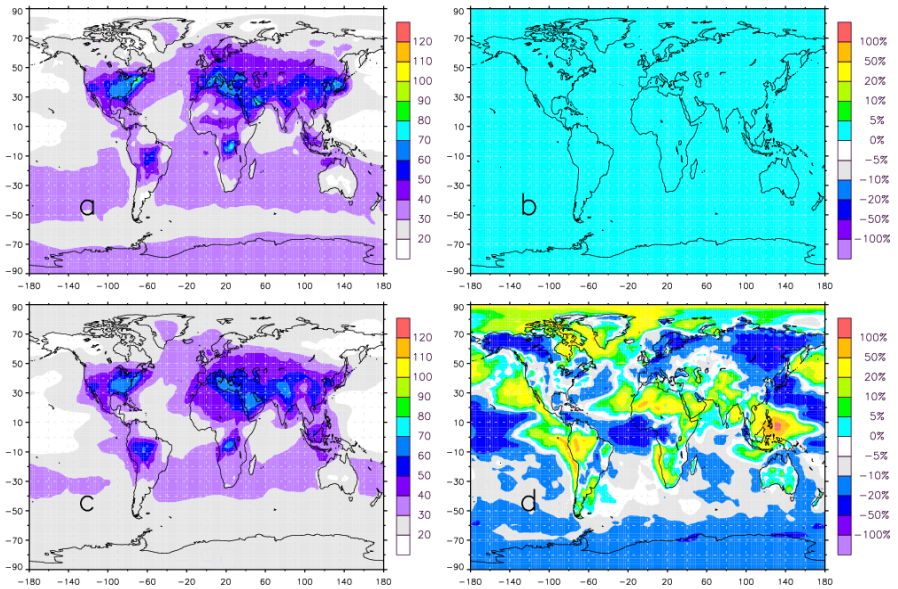

Figure 8. Same as Fig. 2 except for ozone over the globe.

differences within 10%. Off the southwestern coast, however, great positive difference is seen due to the enhanced Pacific High in PCM.

Globally, the distribution of summer mean ozone (Figure 8) exceeds 40 ppb in most regions of interest at mid-latitudes in the northern hemisphere (Panel a). The CO emissions scaling does not produce significant effects (Panel b). In comparison, the simulation is more sensitive to the meteorological conditions (Panels c and d). In the PCM case, large differences are a global phenomenon and are not limited to North America. However, the positive differences are generally consistent with CO and NOx simulations while the negative differences are not. In general, however, MOZART-2.4 is able to reproduce the primary global highs and lows patterns with the PCM meteorology.

4. Conclusions and Future Work

Driven by PCM, MOZART-2.4 is being used to simulate future U.S. air quality under climate and emission change conditions. As part of present study, this paper quantifies the impacts of scaling of CO biofuels and biomass burning emissions and, more important, the use of PCM meteorology on global air quality simulations for a current year (1996), using the R-2 meteorology without CO emissions scaling as the control run. In general, the CO emissions scaling does not have significant impacts on CO outside the regions dominated by strong biofuels or biomass burning emissions. The impacts on NOx and ozone are negligible on the global scale due to modest chemical reactivity of CO.

Comparisons between the control run and the PCM meteorology case, however, suggest that PCM tends to generate an atmosphere that favors the occurrence of more CO, NOx and ozone off the western coasts of most continents. In addition, the positive difference of CO expands globally except in those areas with strong anthropogenic emissions such as the U.S. and Western Europe. NOx is biased for PCM relative to R-2 more heterogeneously over the northern oceans due to its short lifetime on the surface, although a positive difference is generally seen over the southern oceans.

In North America, MOZART-2.4 driven by PCM does not capture the extreme anthropogenic pollution in California and the New York area. The consistency of this negative difference in CO, NOx and ozone suggests that the difference is originated from the internal property of PCM, thus implies a likely negative bias when PCM is used for the projection of future U.S. air quality. The uncertainty of the potential bias increases the difficulty in generating future projections. At the same time, the positive difference relative to R-2 of ozone and precursors off the southwestern coast is a result of the enhanced Pacific High in PCM, which brings more pollutants from such upstream regions as Asia into the U.S. While state-of-the-art integrated air quality modeling systems use global chemical transport model driven by PCM to offer regional models chemical boundary conditions to project future U.S. air quality, this difference implies a potential positive bias if the inflow lateral boundary is chosen in this region. This difference also suggests additional uncertainties in the contribution of upstream regions to U.S. air quality and the associated supplemental uncertainty in U.S. air quality simulation if upstream pollutants are not simulated well.

In order to quantify the model uncertainties resulting from emissions inputs, simulations for current years (1998-1999) using regional emissions datasets (EPA 1999 National Emission Inventory, etc.) over North America to replace the corresponding portions in global emissions dataset are also being generated under both R-2 and PCM meteorology. Current and future U.S. background pollutions will also be simulated to account for the individual contributions of Canada/Mexico as well as remote emissions sources such as Asia to U.S. air quality, and for the potential impacts of emissions policies in other countries on future U.S. air quality under climate change conditions. Together, these studies together help to quantify the uncertainties in the projection of future U.S. air quality.

Acknowledgement

The research was supported by the United states Environmental Protection Agency under award number EPA RD-83096301-0. The views expressed are those of the authors and do not necessarily reflect those of the sponsoring agencies or the Illinois State Water Survey.

Reference

Comrie, A. C., 1996: An all-season synoptic climatology of air pollution in the US-Mexico border, *Science of the Total Environment*, **276**, 49-68.

Davis, R. E., and D. A. Gay, 1993: A synoptic climatological analysis of air quality in the Grand Canyon National Park, *Atmospheric Environment*, **27A**, 713-727.

Friedman, R. M., J. Milford, R. Rapoport, N. Szabo, K. Harrison, S. V. Van Aller, R. W. Niblock, and J. Andelin, 1988: *Urban Ozone and the clean Air Act: Problems and Proposals for Change*, 160 pp., Office of Technology Assessment, United States Congress, Washington, D. C.

Gebhart, K. A., S. M. Kreidenweis, W. C. Malm, 2001: Back-trajectory analyses of fine particulate matter measured at Big Bend national Park in the historical database and the 1996 scoping study, *Science of the Total Environment*, **276**, 185-204.

Hauglustaine, D. A., G. P. Brasseur, S. Walters, P. J. Rasch, J. -F. Muller, L. K., Emmons, and M. A. Carroll, 1998: MOZART-2.4: a global chemical transport model for ozone and related chemical tracers, Part 2. Model results and evaluation, *J. Geophys. Res.,* **103**, 28,291-28,335.

Heck, W. W., W. W. Cure, J. O. Rawlings, L. J. Zaragoza, A. S. Heagle, H. E. Heggestad, R. J. Kohut, L. W. Kress and P. J. Temple, 1984: Assessing impacts of ozone on agricultureal crops, II, crop yield functions and alternative exposure statistics, *J. Air Pollut. Control Assoc.,* **34,** 810-817.

Horowitz, L. J., S. Walters, D. L. Mauzerall, L. K. Emmons, P. J. Rasch, C. Granier, X. Tie, J. -F. Lamarque, M. G. Schultz, G. S. Tyndall, J. J. Orlando, and G. P. Brasseur, 2003: A global simulation of tropospheric ozone and related tracers: description and evaluation of MOZART-2.4, version 2, *Journal of Geophysical Research*, **108** (D24), 4784, doi:10.1029/2002JD2853.

Houghton et al., 2001: *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the intergovernmental Panel on Climate Change,* Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881pp.

Kanamitsu, M., W. Ebisuzaki, J. Woollen, S. -K. Yang, J. J. Hnilo, M. Fiorino, and G. L. Potter, 2002: NCEP-DOE AMIP-II Reanalysis (R-2), *Bull. Am. Meteoro. Soc.* **83**, 1631-1643.

Lamarque, J.-F. and P. G. Hess, 2004: Arctic Oscillation modulation of the Northern Hemisphere spring tropospheric ozone, *Geophys. Res. Lett.*, **31**, L06127, doi:10.1029/2003GL019116.

Liang, X.-Z., H.-C. Huang, A. Williams, M. Caughey, K. Kunkel, D. J. Wuebbles, J.-H. Zhu, Z.-N. Tao, K. Patten and J.-T. Lin, 2005a: Impacts of Global Climate and Emission Changes on U.S. Air Quality: The Second Annual Report for the UIUC EPA STAR Project, University of Illinois, Urbana, IL, 46 pp.

Liang, X.-Z., J. Pan, J. Zhu, K.E. Kunkel, J.X.L. Wang, and A. Dai, 2005b: Regional climate model downscaling of the U.S. summer climate and future change. *J. Geophys. Res.* (submitted).

Lippman, M., 1993: Health effects of tropospheric ozone: Review of recent research findings and their implications to ambient air quality standards, *J. Exposure Anal. Environ. Epidemiol.,* **3**, 103-128.

NRC (National Research Council), 1991: *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, Natl. Acad. Press, Washington, D.C..

____, 2001: *Global Air Quality: An Imperative for Long-Term Observational Strategies*. National Academy Press, Washington, D.C., 56 pp.

Russell, A., and R. Dennis, 2000: NARSTO critical review of photochemical models and modeling, *Atmospheric Environment*, **34**, 2283-2324.

Sillman, S., 1999: the relation between ozone, NOx, and hydrocarbons in urban and polluted rural environments, *Atmospheric Environment*, **33**, 1821-1845.

Solomon, P., E. Cowling, G. Hidy, and C. Furiness, 2000: Comparison of scientific findings from major ozone field studies in North America and Europe. *Atmos. Environ.*, **34**, 1885-1920.

Washington, W.M., J.W. Weatherly, G.A. Meehl, A.J. Semtner Jr., T.W. Bettge, A.P. Craig, W.G. Strand Jr., J.M. Arblaster, V.B. Wayland, R. James, and Y. Zhang, 2000: Parallel climate model (PCM) control and transient simulations. *Clim. Dyn.*, **16**, 755-774.

Wise, E. K., and A. C. Comrie, 2005: Extending the KZ filter: application to ozone, particulate matter, and meteorological trends, *Journal of the Air and Waste Management Association*, **55 (8)**, 1208-1216