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**Effects of 2000-2050 Global Climate Change on Ozone
and Particulate Matter Air Quality in the United States
Using Models-3/CMAQ System**

A Dissertation Presented for the Doctor of Philosophy Degree
The University of Tennessee, Knoxville

Yun Fat Lam
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DEDICATION

I dedicate this dissertation to my parents AhPing Lam and SuetYuk Chan; my sister ChoiPing Lam; and my wife WingChun Siu, for their unending love, and encouragement.

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ABSTRACT

The Models-3/Community Multi-scale Air Quality modeling system (CMAQ), coupled with Goddard Institute for Space Studies (GISS) atmospheric General Circulation Model (GCM), fifth Generation Mesoscale Model system (MM5), and Goddard Earth Observing System-CHEMistry (GEOS-Chem), was used to simulate atmospheric concentration of ozone and particulate matter over the continental United States 12-km and 36-km (CONUS) domains at year 2000 and year 2050. In the study, GISS GCM model outputs interfaced with MM5 were utilized to supply the current and future meteorological conditions for CMAQ. The conventional CMAQ profile initial and boundary conditions were replaced by time-varied and layer-varied GEOS-Chem outputs. The future emission concentrations were estimated using year 2000 based emissions with emission projections suggested by the IPCC A1B scenario. Multi-scenario statistical analyses were performed to investigate the effects of climate change and change of anthropogenic emissions toward 2050. The composite effects of these changes were broken down into individual effects and analyzed on three distinct regions (i.e., Midwest, Northeast and Southeast). The results of CMAQ hourly and 8-hour average concentrations indicate the maximum ozone concentration in the Midwest is increased slightly from year 2000 to year 2050, as a result of increasing average and maximum temperatures by 2 to 3 degrees Kelvin. In converse, there is an observed reduction of surface ozone concentration in the Southeast caused by the decrease in solar radiation. For the emission reduction scenario, the decline of anthropogenic emissions causes reductions of both ozone and $PM_{2.5}$ for all regions. The emission reduction has compensated the effect of increasing temperature. The overall change on the maximum daily 8-hr ozone and average $PM_{2.5}$ concentrations in year 2050 were estimated to be 10% and 40% less than the values in year 2000, respectively. The modeling results indicate the effect of emissions reduction has greater impact than the effect of climate change.

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List of Abbreviations

AERO4	AEROsol module 4
ANL	Argonne National Laboratory
AQM	Air Quality Modeling
BC/BCON	Boundary Condition
BEIS	Biogenic Emissions Inventory System
BVOC	Biogenic Volatile Organic Compounds
CAMx	Comprehensive Air quality Model
CASTNET	Clean Air Status and Trends NETwork
CB-IV	Carbon Bond IV
CFRACT	Total Cloud FRACTION
CMAQ	Community Multi-scale Air Quality modeling system
CONUS	CONTinental United States
CTM	Chemistry-Transport Modeling system
DAO	Atmospheres Data Assimilation Office
FDDA	Four Dimensional Data Assimilation
FNL	NCEP Final Global Analyses
GCAP	Global Change and Air Pollution
GCM	General Circulation Model
GCTM	Global Chemistry-Transport Model
GEOS	Goddard Earth Observing System
GEOS-Chem	Goddard Earth Observing System-CHEMistry
GISS	Goddard Institute for Space Studies
GMAO	NASA Global Modeling Assimilation Office
IC/ICON	Initial Condition
IMAGE	Model to Assess the Greenhouse Effect
IPCC	Intergovernmental Panel on Climate Change
IPCC AR4	Intergovernmental Panel on Climate Change 4th Assessment Report
JFMA	January, February, March, and April

JPROC	J-values (Photolysis rate) PROCessor
LSM	Land Surface Model
MCIP	Meteorology-Chemistry Interface Processor
MDA8	Maximum Daily Average 8-hour
MEGAN	Model of Emissions of Gases and Aerosols from Nature
MJJAS	May, June, July, August, and September
MM5	5th generation Mesoscale Model system
MOZART	Model for OZone And Related chemical Tracers
NAAQS	National Ambient Air Quality Standards
NCAR	National Center for Atmospheric Research
NCEP	National Centers for Environmental Prediction
NEI	National Emissions Inventories
OA	Organic Aerosols
OND	October, November, and December
ORDY-BC	ORDinarY vertical interpolated GEOS-Chem Boundary Conditions
PBL	Planetary Boundary Layer
PDF	Probability Distribution Function
PPFD	Photosynthetic Photon Flux Density
Profile-BC	PROFILE Boundary Conditions
PSU	Pennsylvania State University
PV	Potential Vorticity
RAQM	Regional Air Quality Model
RH	Relative Humidity
RMSE	Root Mean Square Error
SA	Sulfate Aerosols
SMOKE	Sparse Matrix Operator Kernel Emissions
SOA	Second Organic Aerosol
SRES	Special Report on Emission Scenarios
STE	Stratosphere-Troposphere Exchange

STN	Speciation Trends Network
SW	ShortWave radiation at the surface
TOMS	Total Ozone Mapping Spectrometer
Tropo-BC	TROPOpause vertical interpolated GEOS-Chem Boundary Conditions
VISTAS	Visibility Improvement State and Tribal Association of the Southeast
VOC	Volatile Organic Compound
WDR	Wind DiRection
WRF	Weather Research Forecasting system
WSP	Wind Speed
2000M_2000E	2000 Meteorology condition with 2000 current Emissions
2000M_2050E	2000 Meteorology condition with 2050 future Emissions
2050M_2000E	2050 Meteorology condition with 2000 current Emissions
2050M_2050E	2050 Meteorology condition with 2050 future Emissions

Glossary

ACET	Acetone
ALD2	Acetaldehyde
ALK4	C4,5 alkanes
ANH4J	Accumulation Mode Ammonium Mass
ANO3J	Accumulation Mode Nitrate Mass
ASO4J	Accumulation Mode Sulfate Mass
C2H6	Ethane
C3H8	Propane
CO	Carbon Monoxide
FORM	Formaldehyde
H2O2	Hydrogen Peroxide
HNO3	Nitric Acid
HNO4	Pernitric Acid
ISOP	Isoprene
ISPD	Products of Isoprene Reactions
MACR	Methacrolein
MEK	>C3 Ketones
MGLY	Methylglyoxal
MP	Methylhydroperoxide
MVK	Methylvinylketone
N2O5	Dinitrogen Pentoxide
NH3/NH3	Ammonia
NH4	Ammonium
NIT	Inorganic Sulfur Nitrates
NITs	Inorganic Nitrates on surface of sea-salt aerosol
NO2/NO2	Nitrogen Dioxide
NOx	NO + NO2+ NO3 + HNO2
NTR	Organic Nitrate
O3/O ₃	Ozone

OLE	Olefins (Anthropogenic & Biogenic)
OLE2	Olefins (Biogenic)
Ox	O ₃ + NO ₂ + 2NO ₃
PAN	Peroxyacetylnitrate
PAR	Paraffins
PEC	Primary Elemental Carbon
PM10/PM ₁₀	Particles with a Diameter of 10 Micrometers or Less
PM2.5/PM _{2.5}	Particles with a Diameter of 2.5 Micrometers or Less
PMC	Primary Metallic Carbon
PMFINE	Primary PM-fine "other"
PMN	Peroxymethacryloyl nitrate (MPAN)
PNA	Peroxynitric Acid
POA	Primary Organic Aerosols
PPN	Peroxypropionynitrate
PRPE	Propene
PSO4	Primary PM-fine Sulfate
R4N2	C _{4,5} Alkyl nitrates
RCHO	>C ₂ Aldehydes
SO2/SO ₂	Sulfur Dioxide
SO4	Sulfate

CHAPTER I

1 OVERVIEW

1.1 Objective

The objectives of this study were: (1) to develop simulation and analysis tools that could be used to evaluate the future air quality in the United States for year 2050 using Models-3/Community Multi-scale Air Quality modeling system (hereafter, referred as CMAQ); (2) to predict the consequences of climate change on the future air quality; and (3) to assess the impacts of change of Biogenic Volatile Organic Compound (BVOC) to the future air quality. The major challenge in the study was to couple global and regional climate and chemistry models into a coherent and useful modeling approach to predict the ozone and PM_{2.5} air quality in the future.

1.1.1 Introduction

Human activities from industries, fossil fuel burning, agriculture, and animal husbandry are responsible for increasing concentrations of greenhouse gases in the atmosphere (Samenow, 2010). Observations in developing countries have shown that these activities have grown exponentially over the last century. As a direct consequence of these increased activities, huge amounts of greenhouse gases have made it impossible for “Mother Nature” to maintain a balance through chemical sinking processes (IPCC, 2007). Eventually, it has changed the composition of the Earth’s atmosphere. More and more scientific analyses indicated that the increase of greenhouse gases is the primary anthropogenic contributor for global warming and climate change. According to the Intergovernmental Panel on Climate Change (IPCC), “Most of the observed increase in global average temperatures since the mid-20th century was very likely due to the observed increase in anthropogenic greenhouse gas concentrations (IPCC, 2007)”. The “unequivocal” trends of increasing Earth’s global mean temperature in the last century is observed to be approximately 1.0 to 1.7°F from 1906-2005 (IPCC, 2007). In this century, scientists have projected that the atmospheric concentrations of greenhouse gases will continue to rise, which will cause the average global temperatures and sea

levels to rise (Titus and Narayanan, 1995). It will eventually threaten the stability of environment and social systems on the globe.

Global chemistry and climate models have been considered as one of the most important tools to explore or predict the future living environment for mankind. In recent years, particular attention has been given to issues such as extreme weather patterns, mean sea level rising, global warming, and air quality problems as a consequence of human activities. Studies on impacts of these issues are also believed to be extremely important because they may assist in the prevention of national disasters caused by the extreme weather such as, hurricanes, flooding, heat waves, and dust bowls (Gopalakrishnan et al., 2002; Marshall et al., 2008; Westrick and Mass, 2001). Long-term simulations (from decade to century in length) with various scenarios are required to investigate the effects of these issues, which in turn, leads to a high demand for computational resources. In global simulations, the amount of computational resources required is dictated by the complexity of the models, simulation time-steps, and horizontal and vertical resolutions (Richard, 1999). With these limited resources, researchers are required to determine the most efficient configurations while not jeopardizing the global simulations outputs. Reducing horizontal and vertical resolutions seems to be the most feasible way of controlling the use of available computational resources, which has been practiced throughout the research community (Cahill and Mackay, 2003). The computational and design limitations of the global model have restricted the resolution of the global model simulation to be $1^{\circ} \times 1^{\circ}$ degree (about 111 km x 111 km) at the highest. Unfortunately, the results from this coarse resolution are not sufficient to represent the actual conditions in regional scale. Therefore, its usefulness is limited to only demonstrating future air quality and climate trends. It cannot serve as information for short-term emergency response or for decision-making. On the other hand, regional climate and chemistry models have been designed and developed for mesoscale application with high resolutions up to 1-4 km x 1-4 km (Chen et al., 2004; Fu et al., 2008b; Kim et al., 2008; Kumar et al., 2008; Sokhi et al., 2006). These applications relied on outputs from the global models, which then became the lateral boundary conditions for future climate and air quality simulations. On long-term

air quality forecasting, studies have shown that horizontal resolutions of 36–40 km are sufficient for revealing the regional scale condition within the continental United States (Civerolo et al., 2007; Liao et al., 2008). Although finer resolutions (i.e., less than 36 km) can be achieved through downscaling nesting processes, uncertainties of the outputs would be too large when land use changes are not incorporated in the models. Civerolo (2007) successfully integrated land use changes into his study, however, the study area was only limited to a small domain which covers New York metropolitan areas and surrounding states under 4 km resolution (Civerolo et al., 2007). In conclusion, the approach of integrating global model outputs with the regional models on 36 km resolution should sufficiently give a good understanding on both the long-term trend and the detailed air quality information. The results from those simulations can be used for air quality-related policy making. For this study, 12 km resolution simulation was also performed to investigate the effect of the downscaling.

For air quality projection, most studies in the literature (i.e., projecting for year 2010 or 2020) have been centered on emission projection scenarios where future projected emissions with the current/base year climate conditions are used to simulate the future air quality (Arunachalam et al., 2006; Tesche et al., 2006; Yamaji et al., 2008). These studies assumed that the climate conditions in the future year should be similar to the current/base year and the effects of climate change are small enough to be negligible. In short-term projections, this approach would be considered valid since the climate condition is not expected to change much. However, in long-term forecasting, for which global warming plays an important role in defining the climate condition, this assumption would be unacceptable. Wu, et al., (2008) used GEOS-Chem (one of the global chemistry models), to study air quality conditions in year 2050 (Wu et al., 2008b). Their results demonstrated that the effect on ozone due to climate change was as much as 2 to 5 ppbv. Similar results were also found in Civerolo (2007) and Liao (2008). Hence, it is necessary to incorporate climate change effects into the regional model when long-term forecasting on air quality is performed.

Another important component for simulating future air quality is the background tracer concentrations. Unlike global models, where study domain extends to the whole

globe, regional models are required to define a fixed boundary and to supply an hourly lateral boundary condition as inputs for the model. With the recent observed increase of background pollutant concentrations at the boundary of United States, the conventional profile boundary condition (which assumes relatively clean air) is no longer sufficient to represent the background tracer condition (Byun and Ching, 1999; Song et al., 2008). Satellite observations, as well as global chemistry simulations, have shown that these background concentrations of ozone and $\text{PM}_{2.5}$ have increased as much as 20% in the last ten years (Fiore et al., 2003a). The main contribution of this phenomenon is from the effect of intercontinental transport of air pollutants and the effect of climate changes (Liang et al., 2007; Park et al., 2004; Bertschi et al., 2004). Unlike other pollutants, $\text{PM}_{2.5}$ is able to travel at a great distance without change (Heald et al., 2006; Husar et al., 2001). These particles are able to travel from East Asia, across the Pacific Ocean to the continental United States, ultimately impacting the air quality in the United States. Researchers have demonstrated air quality effects from the intercontinental transport of air pollutants through events such as wild fire or dust storm, which are large enough to capture by satellite imagers (Chin et al., 2007). In those studies, movement of plumes were measured either in terms of the light extinction coefficient or the aerosol optical thickness. It is this impairment of local visibility that has confirmed the importance of understanding the long-range transport phenomenon (i.e., the intercontinental transport) of air pollutants. Chemical analysis results of air samples collected during the local visibility episode also confirm the presence of foreign pollutants that are imported through intercontinental transport. The mixing of local and foreign pollutants has made the study of the intercontinental transport more difficult because chemical interactions are involved (Park et al., 2004; Sudo and Akimoto, 2007; Weiss-Penzias et al., 2006). These chemical interactions can be evaluated using a global chemical model, where the reactions of these chemicals are simulated. For future background pollutant concentrations, the global chemistry model (with future climate condition and projected future emissions) can be used to supply the lateral boundary condition for the regional air quality model (where the effects of intercontinental transport of air pollutants are already taken into account). So, reasonable concentrations can be obtained.

1.2 Study Plan

1.2.1 Study Scope

The main goal of the study was to investigate the future air quality in the United States for year 2050 using the regional air quality model, CMAQ. By utilizing climate and air quality models on both global and regional scales, four model scenarios were proposed, which were: (1) 2000 condition with current emissions; (2) 2000 condition with future emissions; (3) 2050 condition with current emissions; and (4) 2050 condition with future emissions. The results from these simulations were compared with each other to explore the effects of climate change and emission growth from human activities using statistical analysis. It is expected that global warming will increase the average temperature of the future climate condition (IPCC, 2007). However, the dynamic spatial and temporal changes of temperature across major metropolitan areas in the United States are unknown (Bell et al., 2007). It is possible that some metropolitan areas in the United States may have a lower average temperature because of the natural variations of climate. Moreover, the effect of raising average temperature is more likely to increase the probability of exceeding 8-hour ozone concentration standards during the summer (Dawson et al., 2007). Currently, a limited research has been performed using both global and regional models to investigate the future air quality in the United States (Civerolo et al., 2007; Civerolo et al., 2008; Vautard and Hauglustaine, 2007) and had been focused on ground level ozone and its related precursors (i.e., NO_x and VOCs). The pioneer work of linking global and regional models together (for both climate and air quality models to investigate the future air quality conditions) have a great influence on the direction of research topics in the years to come.

1.2.2 Proposed Study

The study (modeling 2050 air quality in the United States) was used to evaluate the effect of climate change, caused by human activities, on the regional air quality in the United States. The study was based on year 2000 to year 2050 transient climate simulations using the Goddard Institute for Space Studies (GISS)/NASA General

Circulation Model (GISS GCM) III and applied future emission scenarios used by the IPCC Special Report on Emission Scenarios (SRES) (Nakicenovic et al., 2001). The GISS GCM simulations serve as the meteorology indicator for trends. The outputs of GISS GCM were also used as the input to global chemistry model, Goddard Earth Observing System-CHEMistry (GEOS-Chem), for simulating tropospheric ozone and PM_{2.5}. Eventually both outputs from GISS GCM and GEOS-Chem were used as dynamic boundary conditions for the regional climate model and air quality model (i.e., MM5 and CMAQ). Figure 1.1 shows the summary of the study scope and time-line of the study. This study involved multiple universities and government agencies including Harvard, California Institute of Technology, University of Tennessee, NASA, University of Houston and Argonne National Laboratory (ANL). The study was funded by the USEPA STAR program. The area of my research focus was on the green and brown areas in the diagram of the study scope, where downscaling from global to regional models was performed.

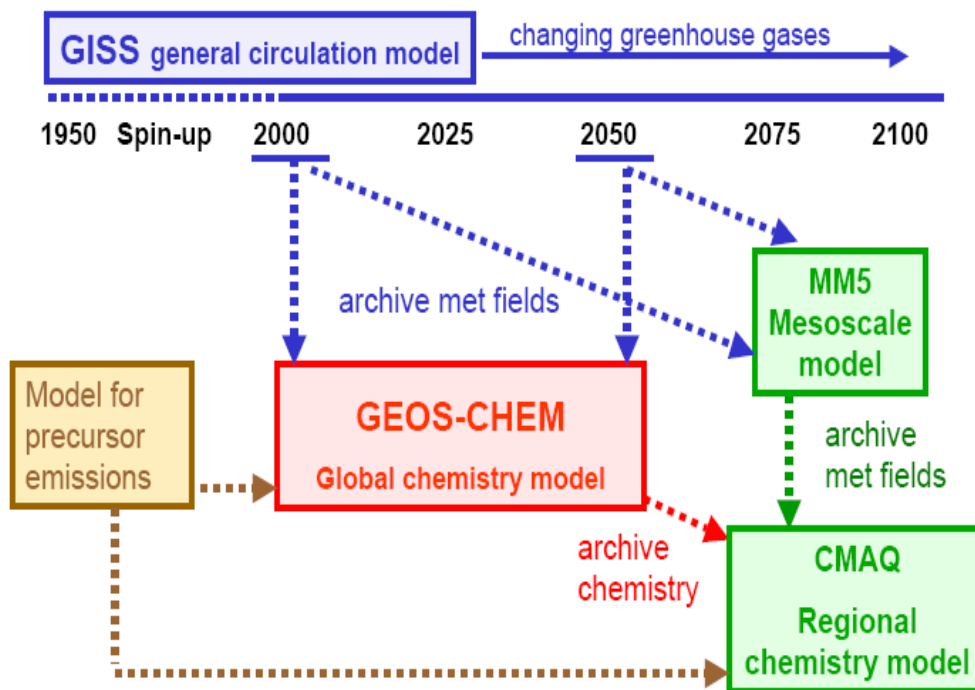


Figure 1.1 Summary of the study scope (Mickley, 2005).

1.2.2.1 Climate Model Downscaling

The climate changes and global warming have a great impact on global and regional air quality. For example, increasing the average temperature by 3.2°C increases the exceedance of National Ambient Air Quality Standards (NAAQS) on ozone by as much as 68% (Bell et al., 2007). Moreover, increasing the deep convection amplifies the chance of lightning, which consequently produces more NO_x and affects the formation of ozone (Wu et al., 2007). The climate variables that have effects on air quality are atmospheric concentrations of carbon dioxide and methane levels; sea surface temperatures; solar radiation; volcanic aerosols; and frequency of lightning. To understand the effects of climate change on regional air quality, analysis of long-term simulations of climate and air quality on both regional and global scales must be performed. By comparing simulations from both current and future climate conditions with various scenarios mentioned early, relative contributions of climate change would be obtained. For current climate condition, simulated climate data in current conditions were used as the inputs for the global climate model. On the other hand, for the future climate condition, projections of atmospheric constituents were needed to incorporate into the climate simulation. The Intergovernmental Panel of Climate Change (IPCC) report categorizes six projection scenarios, which are A1B, A1T, A1FI, A2 B1 and B2 (Nakicenovic et al., 2001). A1B scenario was applied to the United States because this scenario represented the country with a well-balanced progress across all resources and technologies from energy supply to end use (Nakicenovic et al., 2001). For the purposes of simulating regional climate change in the United States, the mesoscale model MM5, with the one-way nested to the GISS GCM, was used to simulate both current and future conditions. The outputs from the regional climate model, MM5, were used as meteorological inputs for regional air quality model, CMAQ. The challenge of scaling down global model data into a regional model is finding a way to maintain correct representations of climate condition. The regional model uses lateral boundary conditions given by the global model to simulate regional climate condition with higher temporal and spatial resolutions. It is important to demonstrate that high-resolution

results have better representations of the local climate condition than those simulated by the global model while preserving similar trends as the global climate condition.

1.2.2.2 Chemistry Model Downscaling

In the last decade, many United States industries have moved their operations to East Asia and South America to cut operational cost. This phenomenon has created rapid industrialization in these third world regions (i.e., Asia and South America). With the lack of any major air quality regulations, these regions have been subjected to severe air quality problems, where high concentrations of PM_{2.5} and O₃ have been observed (Fu et al., 2008a). Recent research has shown that a significant amount of pollution and dust can be transported from Asia to North American across the Pacific Ocean (Chin et al., 2007). The effects of the intercontinental transport to local air quality have alarmed the modeling community, where the traditional approach of using a fixed Initial condition/Boundary condition profile in the regional model such as CMAQ model system has to be changed. The Initial Condition (IC), in here, represents the initial air quality condition within the boundary of the model domain, where as the Boundary Condition (BC) corresponds to the air quality condition in the outer boundary of the model domain. To overcome the shortage of using default IC/BC, the alternative approach of using the global chemistry model to create the IC/BC for the regional air quality model has been suggested. For the purposes of accurately simulating CMAQ for both current and future conditions, a global chemistry model, GEOS-Chem, was applied to provide initial and boundary conditions in CMAQ. For consistency among all models (i.e., GISS GCM, MM5 and CMAQ), GISS GCM model outputs were also used as the meteorological inputs for the GEOS-Chem.

1.2.2.3 Research Tasks

The tasks are completed in this research were:

1. Developed meteorological model interface module between GISS GCM and MM5;
2. Developed air quality model interface module between GEOS-Chem and CMAQ;
3. Simulated 2000 and 2050 meteorological conditions using MM5;

4. Simulated 2000 air quality using CMAQ;
5. Projected emissions changes for 2050;
6. Simulated 2050 air quality with 2050 emissions and 2050 climate using CMAQ;
7. Simulated different scenarios using CMAQ (i.e., 2000 emissions with 2050 climate and 2050 emissions with 2000 climate);
8. Compared global model results with regional model results; and
9. Performed statistical analysis to separate the effect of global warming and emission growth.

This study focused on the changing intensities of the maximum 8-hour ozone concentration caused by climate change and by a change of human activities using CMAQ. The outputs from CMAQ simulations were evaluated and analyzed to provide daily statistical analyses and temporal analyses on both meteorological parameters and atmospheric pollutant concentrations. The variables examined were surface temperature, relative humidity, precipitation, 10m wind speed, 10m wind direction, O₃ and PM_{2.5} species.

CHAPTER II

2 BACKGROUND AND MODELING METHODOLOGY

2.1 Introduction to Global Models

The general circulation model is a global climate model that was developed to simulate the driving processes of the ocean and atmosphere (IPCC, 2008). It is used to make predictions of future climates based on the concentrations of various atmospheric constituents such as carbon dioxide level. Although this model can not necessarily make infallible predictions of the future, it can make reasonably accurate ones (GISS-GCM, 2008). Hence it can be utilized as a source of information for studying the effects of atmospheric changes due to increasing carbon dioxide levels. Conversely, the Global Chemistry-Transport Model (GCTM) was developed to simulate the transport and chemical reactions phenomena of various atmospheric constituents. For predicting the future, GCTM requires reasonably accurate climate conditions (which are often supplied by GCM) to simulate the atmospheric chemical balances.

2.1.1 GISS GCM

General Circulation Model (GISS GCM) of the Goddard Institute for Space Studies is a three-dimensional global atmosphere-ocean model that simulates the earth's climate on a grid-based domain. It numerically solves the physical conservation equations for energy, mass momentum, and moisture in a given domain. The purpose of the model is to predict climate conditions for a decade to century time scales. The model has been under continual development since 1970. The atmosphere-ocean coupling has been incorporated into the model since 1990. The current version of the model, referred as Model III/Model E, has been used to simulate relevant experiments for the Intergovernmental Panel on Climate Change 4th Assessment Report (IPCC AR4) and has been documented in the literature (Joseph and Nigam, 2006; Li et al., 2006; Schmidt et al., 2006; McKibben, 2007).

Model III uses the terrain-following sigma coordinates for the vertical profile. Compared with Model II, the new Model III has improved the model top to above the stratopause with the number of vertical layers increased to 23 as shown in Figure 2.1.

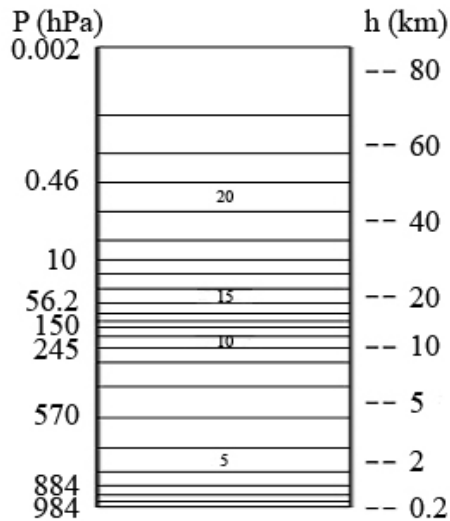


Figure 2.1 Vertical layering for 23-layer model configurations (for graphical convenience, the vertical coordinate shown here is linear in pressure to 150 hPa, logarithmic above) (Schmidt et al., 2006).

For horizontal resolution, Model III uses a Cartesian grid point formulation with either $4^\circ \times 5^\circ$ or $2^\circ \times 2.5^\circ$ latitude by longitude. For special handling of velocity vectors, Model III uses the Arakawa-B grid, where the vectors are assigned to the corner of the grid. Model III allows a 30-minute time-step for all physics calculations; the radiation code is applied on every five physics time-steps, which is 2.5 hours. A new cloud microphysical scheme is implemented in Model III to improve precipitation and surface albedo estimates and effective resolution for pollutant transport has significantly increased for the mean pollutant values in each grid box (McKibben, 2007).

The basic model requires two kinds of input, specified and prognostic variables (GISS, 1997). These specified variables include physical constants, the Earth's orbital parameters, the Earth's atmospheric constituents, the Earth's topography, the Earth's surface distribution of ocean, glacial ice, or vegetation, and many others. The prognostic variables include time-varied fluid mass, horizontal velocity, heat, water vapor, salt, and subsurface mass and energy fields. For outputs, the model produces an un-scaled climate diagnostics file which is separated by each simulated month. Subsequent programs read this file and convert to scaled climate variables. These scaled climate variables can be

effectively used as the initial and boundary conditions in the regional climate models for predicting future regional climate conditions.

Model III can be divided into five separate model modules, which are atmospheric, dynamic ocean, sea ice, land surface, and tracers. The atmospheric model module calculates the solution of the momentum equations based on the dry physics (no water vapor effects in the pressure gradient calculation) and advection variable (such as potential temperature) to determine the surface fluxes in the atmosphere. The ocean model module uses a sub-model, Q-flux/mixed layer model, with the given variables such as total freshwater mass, heat fluxes, surface heat, mass fluxes, and sea surface temperature to determine the actual ocean heat transports. The sea ice model module uses the dynamic oceans model module and the sub-model Q-flux in dynamic ocean model module to determine the ice advection. The ice advection is a function of the atmosphere-ice and ice-ocean momentum stress and the sea surface gradient. The land surface model module defines soil physics and soil characteristics on a six-layers soil scheme. Moreover, the surface and ground hydrology characteristics (i.e., evapotranspiration and runoff) and surface albedo are also determined in this module. The tracer model module tracks four different types of atmospheric tracers, which are atmospheric tracers that follow air mass, tracers that are either purely water-based or have a soluble component, particulate tracers that may interact with the hydrologic cycle, and tracers that are purely oceanic (GISS-GCM, 2008).

2.1.2 GEOS-Chem

The GEOS-Chem model system (hereafter, called GEOS-Chem) is a global three-dimensional model of atmospheric chemistry that is driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling Assimilation Office (GMAO). It was developed by researchers from Harvard, Duke, NASA/GSFC and the University of Washington, as a multipurpose tool for studying a wide range of atmospheric chemistry problems (GEOS-Chem, 2008). For air quality applications, GEOS-Chem is capable of evaluating the impacts of air quality disasters such as volcanic events, wild fire, and dust storm across the globe. Moreover,

air quality issues, such as intercontinental transport of air pollutants, the impact of tropospheric ozone, and visibility degradation, can also be modeled by GEOS-Chem.

GEOS-Chem accepts various meteorological data products including DAO's GEOS, GEOS-STRAT, GEOS-3 and GEOS-4 with either $1^\circ \times 1^\circ$ or $2^\circ \times 2.5^\circ$ global resolution produced by Atmospheres Data Assimilation Office (DAO). These meteorological data have vertical resolutions of 20 to 55 vertical levels. For GEOS-Chem, the highest achievable horizontal resolution is $1^\circ \times 1^\circ$ with 55 vertical levels. Often time, the resolution of GEOS-Chem is limited by the input meteorological data. GEOS-Chem adapts the latitude and longitude coordination system as the default horizontal projection. Detailed inventories of fossil fuel, biomass burning, biofuel burning, biogenic, and aerosol emissions are required for simulating in GEOS-Chem. In the GEOS-Chem model, the state-of-the-art transport (TPCORE) and photolysis (FAST-J) routines, as well as the SMVGEAR II chemistry solver packages are incorporated.

GEOS-Chem has been extensively evaluated with comparisons of various air pollutants (i.e., ozone and $\text{PM}_{2.5}$) and its precursors, and observation data in the United States (Bey et al., 2001; Martin et al., 2002; Sauvage et al., 2007; Fiore et al., 2003a; Fiore et al., 2003b; Hudman et al., 2007). Studies from higher-resolution GEOS-Chem showed that the GEOS-Chem does not induce significant mean bias to the surface PM precursors and ozone (Fiore et al., 2003a; Park et al., 2004; Wu et al., 2008b). The major synoptic forcings of air pollution meteorology are also successfully captured by GEOS-Chem. It should be noted that the GEOS-Chem is not designed to simulate local air quality conditions. Therefore, detailed information, such as diurnal variations and daily maximum value, may not be used. Currently, GEOS-Chem is being used to support regional air quality modeling to provide temporal and spatial initial and boundary conditions.

2.2 Introduction to Regional Models

Regional meteorological modeling is one of the prerequisites for photochemical air quality modeling (AQM), which provides real-time meteorological fields (i.e., moisture and temperature) to the AQM. Two meteorological models, MM5 and WRF were being

considered in this study. MM5 stands for Fifth Generation Mesoscale Model system and WRF stands for Weather Research Forecasting system (Chen et al., 2004). Since MM5 is the recommended model for USEPA in photochemical AQM studies, it has been studied intensively for the continental United States (Morris et al., 2005; Morris et al., 2006a; Streets et al., 2007; Queen et al., 2008). Therefore, MM5 has been selected for this study.

Two of the most common air quality models in the United States were considered in this study, they are CMAQ and CAMx. CMAQ stands for Models-3/Community Multiscale Air Quality model. It is maintained and developed by the USEPA as the preferred model for State Implementation plan (Morris et al., 2005). CAMx stands for Comprehensive Air quality Model. It is also an acceptable model for State Implementation planning and was developed by Environ® (ENVIRON, 2008). Both models have been extensively studied on the continental United States domain for the past decade. From the previous performance studies done on these two models, as reported in the literature, no statistical conclusion can be made on which model is better in model performance (Teschke et al., 2006). CMAQ was arbitrarily selected for the future air quality because I had more extensive experience using CMAQ than CAMx. Moreover, similar air quality studies for future ozone prediction have also been performed on the upper Eastern United States by the University of New York using CMAQ, which is available for model comparisons (Civerolo et al., 2007).

2.2.1 Mesoscale Model (MM5)

The MM5 modeling system is one of the leading three-dimensional meteorological models and is being developed by the National Center for Atmospheric Research (NCAR) and Pennsylvania State University (PSU). MM5 integrates an efficient split, semi-implicit temporal integration scheme and nested-grid capability that allows multiple domains and resolutions to be simulated in a timely fashion (Olerud et al., 2000). With a four-dimensional data assimilation (Newtonian nudging) capability, it greatly improves the quality of model results. For coordinate system, MM5 uses a non-

hydrostatic terrain-following sigma coordinate system in the vertical direction, which is defined as

$$\sigma = (p_0 - p_t) / (p_s - p_t) \quad (2.1)$$

where p_0 is the reference-state pressure, p_t is the pressure at the top of the atmosphere, and p_s is the surface pressure. The reference-state pressure, p_0 , is derived from the terrain height using the following equation

$$Z = -\frac{RA_L}{2g} \left[\ln \frac{p_0}{p_{00}} \right]^2 - \frac{RT_{s0}}{g} \left[\ln \frac{p_0}{p_{00}} \right] \quad (2.2)$$

where p_{00} is sea-level pressure taken to be 10^5 Pa, T_{s0} is the reference temperature at p_{00} , A_L is a measure of lapse rate usually taken to be 50, R is the universal gas constant and g is gravitational acceleration (i.e., 9.81 m/s). The above equation permits a reference-state pressure to be related to the terrain height while it is independent from time. Sigma levels have a value of one at the ground surface and zero at the top of the model domain and number of layers can be adjusted in MM5 based on user configuration. Figure 2.2 shows the vertical structure of MM5.

The center of the grid squares are assigned to the scalars (T, q etc.) and designated by cross points, where the corner points are allocated to the eastward (u) and northward (v) velocity components and referred by dot points. In general, horizontal data with an x and y coordinate system (x, y) are required to interpolate into the model as J and I for assuring consistency with the grid. In MM5, three types of projections are available: Mercator, Lambert conformal, or polar stereographic projection. For the horizontal coordinate system, MM5 uses Arakawa-Lamb B-staggering of the velocity variables with respect to the scalars as shown in Figure 2.3.

The MM5 model system consists of a collection of preprocessors including TERRAIN, PREGRID, REGRIDDER, little_r, and INTERPF. The schematic diagram/flow chart of the modeling system is shown in Figure 2.4.

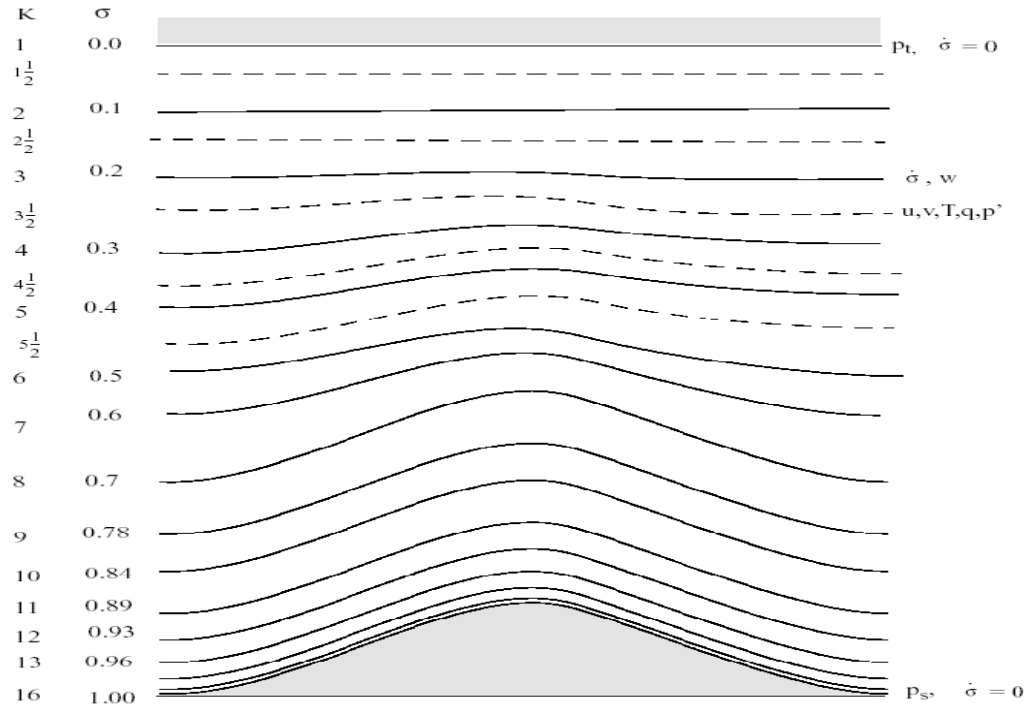


Figure 2.2. Vertical structure of MM5 systems (Dudhia et al., 2005)

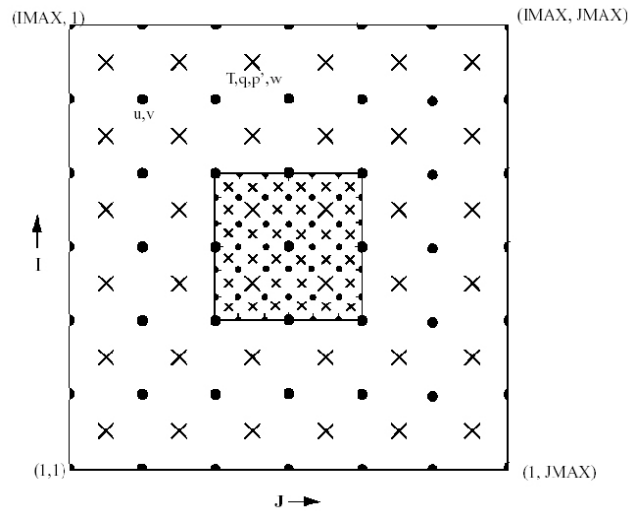


Figure 2.3. Horizontal structure of MM5 system (Dudhia et al., 2005)

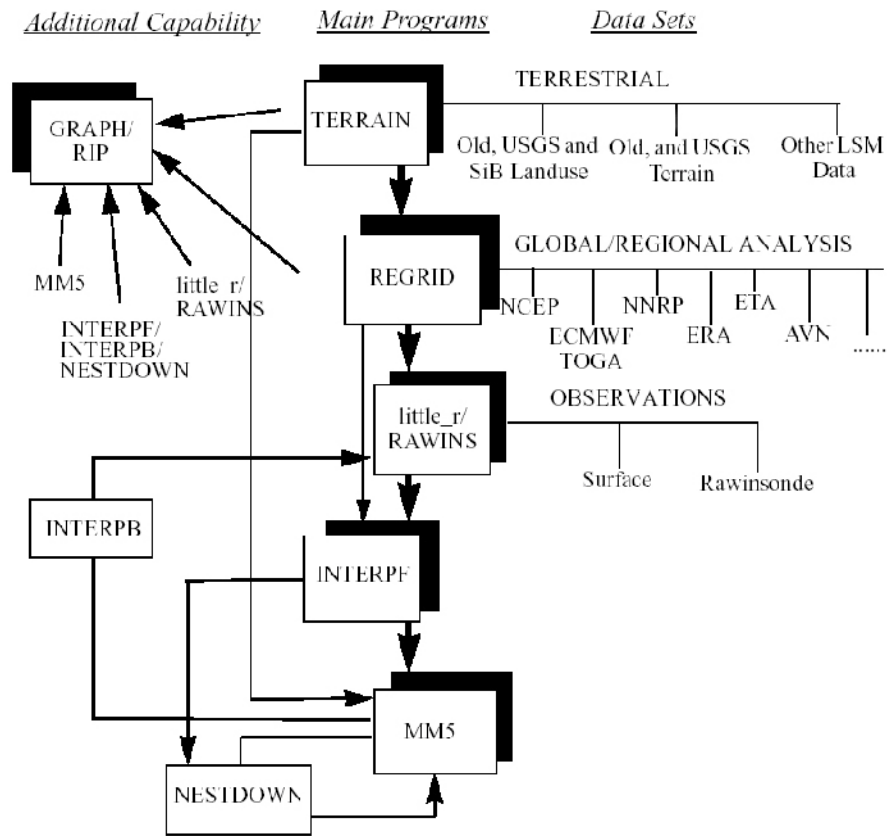


Figure 2.4. Process flow chart of MM5 modeling system (Dudhia et al., 2005)

2.2.1.1 TERRAIN

The TERRAIN preprocessor specifies the horizontal grid of the MM5 model, which allows the user to select model domain sizes, center of the domains, and projection type. TERRAIN interpolates the latitude – longitude, terrain elevation, and vegetation (land use) onto the chosen domain. An additional function of TERRAIN is to generate data (i.e., soil types, vegetation fraction, and annual deep soil temperature) for the land-surface model (LSM).

2.2.1.2 REGRID

The REGRID preprocessor contains two distinct processes, which are PREGRID and REGRIDDER. The PREGRID preprocessor translates grid-based meteorological analyses data into an intermediate data format that the REGRIDDER preprocessor can

process, where as the REGRIDDER preprocessor converts the intermediate data to user specified pressure levels and to user-specified horizontal grid that INTERPF can be processed. The main purpose of the REGRID is to generate the first guess value to an objective analysis (little_r), or to supply the initial and boundary conditions for INTERPF.

2.2.1.3 Little_r/RAWINS

The RAWINS and little_r preprocessors provided objective analysis, Four Dimensional Data Assimilation (FDDA), on the output from REGRIDDER by incorporating the meteorological observation data (FDDA data) from the surface and upper air to improve meteorological analyses (the first guess) on the mesoscale grid. FDDA is the process of incorporating observational data into a forecast model (i.e., MM5) to create an estimate of meteorological conditions. It is a fact that the program functions of RAWINS are the same as those in little_r and the only major difference between RAWINS and LITTER_R is that RAWINS is written in an older Fortran 77 package, where as little_r was written in Fortran 90.

2.2.1.4 INTERPF

The INTERPF pre-processor interpolates the RAWINS/little_r/REGRID output into standard pressure levels and applies it to a user specified vertical grid. This vertical grid is defined in terms of pressure value, (i.e., sigma level) where sigma level of 1.0 is the surface and sigma level of 0 is the top of the model atmosphere, which is explained in an earlier section (Coordinate System). The outputs of INTERPF are a model initial, lateral boundary condition, and a lower boundary condition.

2.2.1.5 MM5

The MM5 model performs the numerical weather prediction using the data generated by the preprocessors (i.e., INTERPF and little_r). MM5 model allows the user to specify initial parameters (i.e., time step) and physics option (i.e., cumulus, PBL, radiation, surface, and microphysics schemes) that match the needs of the user. MM5 can be executed in various computer platforms with multiprocessor capability.

2.2.2 Community Multi-scale Air Quality (CMAQ)

The CMAQ modeling system is a multi-resolution multi-scale air quality model that simulates all atmospheric and land processes that affect the transport, transformation, and deposition of air pollutants and their precursors on both regional and urban scales (Byun and Schere, 2006; Byun and Ching, 1999). It is designed to approach air quality problems as a whole with state-of-science capabilities on the issues of tropospheric ozone, fine particles, toxics, acid deposition, and visibility degradation. The term “one atmosphere” is often used to describe this approach. The results of CMAQ can be used to improve and support (1) the environmental management community's ability to evaluate the impact of air quality management practices for multiple pollutants at multiple scales; and (2) the scientist's ability to better probe, understand, and simulate chemical and physical interactions in the atmosphere.

CMAQ uses the same sigma (pressure level) system as MM5 in the vertical direction. Pressure level number is decided by use and is most commonly 14, 16 or 22 layers. For the horizontal coordinate system, CMAQ uses Arakawa-Lamb C-grid of the velocity variables, where eastward velocities are centered on the eastern and western edges of primary grid cells, and northward velocities are centered on the northern and southern edges. The C-grid yields the best precision when it calculates velocity vectors (Arakawa and Lamb, 1977).

The CMAQ modeling system consists of three main modeling components: (1) a meteorological modeling system, (2) an emission models system, and (3) a Chemistry-Transport Modeling system (CTM). The common meteorological modeling systems for CMAQ are WRF and MM5. For regulatory purposes, MM5 is the only model accepted by USEPA. The Fortran-based, Sparse Matrix Operator Kernel Emissions (SMOKE) is the preferable emissions model. The purpose of the emission model systems is to estimate the amount of pollutants being emitted into the atmosphere by man-made and natural processes and convert them into a grid-based temporal format, which CMAQ accepts. As for CTM, it is an internal module of CMAQ where the chemistry-transport modeling system is used to simulate the chemical transformation and resulting atmospheric changes using predefined gas phase chemical mechanisms (i.e., RADM2 and

CB-IV, CB-V and SAPPRO99). The processes involved in CTM are chemical and energy balances, advection, diffusion, convection, cloud and aerosol formation. These three components are linked together by converting all its outputs to a common grid with same resolutions, same domain sizes, and same projections using the following interface processors: MCIP, JPROC, ICON, and BCON that will be described in the following section. These processors prepare the requisite input information for initial and boundary conditions and photolysis rates for the CCTM. Figure 2.5 shows the logical diagram for CMAQ.

2.2.2.1 Meteorology-Chemistry Interface Processor (MCIP)

The MCIP converts model outputs from the meteorology model (i.e., WRF and MM5) for the CTM. Various processes, such as, coordinate system conversion; cloud parameter computation; and surface and planetary boundary layer (PBL) parameters are executed in MCIP.

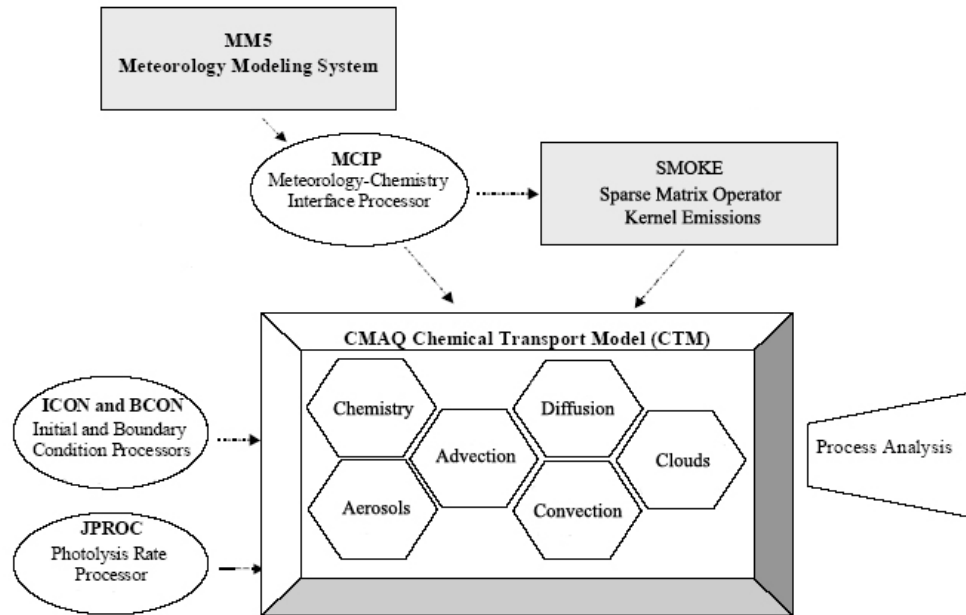


Figure 2.5. CMAQ logical flow chart (Byun and Schere, 2006).

2.2.2.2 *ICON and BCON*

The Initial Conditions and Boundary Conditions (ICON and BCON) supply individual chemical concentrations for the beginning of a simulation and for the grids surrounding the modeling domain, respectively. The ICON and BCON processors accept data from previous CMAQ model simulations, clean troposphere vertical profiles, or the outputs of global chemical models.

2.2.2.3 *J-values (Photolysis rate) PROCessor (JPROC)*

The photolysis processor (JPROC) computes temporal photolysis rates using the given information such as vertical ozone profiles, temperature profiles, a profile of the aerosol number density, the earth's surface albedo and Total Ozone Mapping Spectrometer (TOMS) satellite data for the CTM. The output of the JPROC is tabulated into a lookup table of photo-dissociation reaction rates.

2.3 **Downscaling from Global to Regional Models**

The global climate model, GISS-GCM III was used to simulate meteorological conditions in the United States at year 2000 (1995 to 2002) and year 2050 (2045 to 2052) on a 4° x 5° horizontal resolution. The output of the results in GISS-GCM III was required to interpret and interpolate into a finer resolution, which could be accepted by MM5 as an input of the lateral boundaries conditions. This was one of the major tasks in the dissertation to develop a linkage program between GISS-GCM III and MM5. Since no program was available to the public. During the dissertation, an interface program called GISS2MM5 was developed to facilitate this linkage process. The program provided a great contribution to the research community for future air quality study using GISS-GCM III.

As for the air quality model, the global chemistry model, GEOS-Chem was adopted to provide IC/BC files for CMAQ. To allow the GEOS-Chem output to be used in the CMAQ model, an interface program was needed to be developed to convert all GEOS-Chem species into CMAQ acceptable species.

2.3.1 GCM to MM5

Two MM5 preprocessors, REGRID and little_r, were involved in the linkage between these two models. The function of REGRID preprocessor is to convert and interpolate input data into a file that contains the initial meteorological condition on a specific grid-based domain, whereas, little_r is used to blend meteorological observational data with a “first guess” value from REGRID and to then perform FDDA objective analysis nudging. A module was developed for each of these preprocessors. Both programs were written in Linux/Unix based Fortran 90. Both programs were intended to preserve as much information as possible from GISS-GCM III. Detailed program methodology and general procedure were documented in the following sections.

2.3.1.1 REGRID Linkage

The REGRID preprocessor involves two sequential programs, which are pregrid and regridder. The internal program, pregrid, reads pressure-level meteorological fields from MM5 pre-accepted sources (NCAR or NCEP data) and modifies them to an intermediate format. Since GISS-GCM III data was not one of the pre-accepted sources, no pregrid process was needed. The second program, regridder, takes the intermediate-format data with the TERRAIN file and converts it into a lateral boundaries condition file. The output of the file contains both 3-dimensional meteorological fields (i.e., wind, temperature, relative humidity, and geo-potential height) and 2-dimensional fields (i.e., sea-level pressure, and sea surface temperature).

The intention of the REGRID linkage was to create a program that acted as the function of pregrid which converts GISS-GCM III data into the intermediate format. Specific input format and data types were required by REGRID for all foreign datasets. The GISS-GCM III data contained 23 layers of pressure levels. Both 2-D and 3-D data were stored within the same file. For 2-D data, it was treated as one of the 3-D layer files. A program was developed to convert all the data into specific formats and units and output into an intermediate format. The intermediate format used two-dimensional horizontal (i.e., pressure-level or surface) slabs of data for both 2-D and 3-D data as the input format. For the horizontal slab, each slab defined a single pressure level of all

meteorological variables. The slab file had to be written as unformatted FORTRAN records for the MM5 preprocessor to be recognized. Four records were defined in every horizontal slab as shown in Figure 2.6.

The first record is a format version number. The second record specifies information to all types of grid-based data. The third record specifies information to a particular grid type. The fourth record is the 2-dimensional slab of data. Table 2.1 shows the mapping table between REGRID and GISS-GCM III.

The map projection conversion and domain data selection between these two models was handled by IPROJ in Record 2 and Record 3, respectively, where the regridded internally converts input data into a TERRAIN-defined map projection and selects only the data that is contained within a fixed domain. One of the acceptable map projection types of REGRID is the Latitude and Longitude coordinate system. Since the GISS-GCM III outputs were based on the Lat-Lon coordinate system, no additional conversion was needed.

2.3.1.2 *little_r* Linkage

The primary function of *little_r* is to read in both output from REGRID and observations from *little_r* to perform objective analysis, where it nudges a “first guess” value with observations and outputs it on pressure levels. This objective analysis improves MM5 meteorological analyses on the grid-based domain. The meteorological variables that are accepted by *little_r* are pressure, temperature, u component of wind, v component of wind, and relative humidity. In *little_r*, four objective analysis techniques are available, which include (1) Cressman scheme; (2) Ellipse Scheme; (3) Banana Scheme; and (4) Multiquadric scheme.

Record1: IFV Record 2: HDATE, XFCST, FIELD, UNITS, DESC, XLVL, NX, NY, IPROJ Record 3: STARTLAT, STARTLON, DX, DY, XLONC, TRUELAT1 Record 4: SLAB
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Figure 2.6. Standard slab format for REGRID

Table 2.1. Mapping data for GISS-GCM III downscaling to REGRID

	REGRID	Units	GISS-GCM III	Units
T	* Air temperature	K	Surface Air Temperature	C
U	* Grid-relative u-component of the horizontal wind	m/s	U Surface Wind	m/s
V	* Grid-relative v-component of the horizontal wind	m/s	V Surface Wind	m/s
RH	* Relative humidity	%	Surface Relative Humidity	%
HGT	* Geopotential height	m	Geopotential height	m
PMSL	* Sea-level pressure	Pa	Sea Level Pressure	mb
SST	* Sea-surface Temperature	K	Skin temperature of open water	C
SKINTMP	* Skin Temperature	K	-	-
SOILT010	Ground temperature from 0 to 10 cm	K	Temperature 1st Layer of soil	C
SOILT200	Ground temperature from 10 to 200 cm	K	Interpolate from Temperature 1st, 2nd, and 3rd Layer of soil	C
SOILT400	Ground temperature from 200 to 400 cm	K	Interpolate from Temperature 2nd, 3rd, and 4th Layer of soil	C
SOILM010	Soil moisture from 0 to 10 cm	K	Moisture 1st Layer of soil	Fraction
SOILM200	Soil moisture from 10 to 200 cm	K	Interpolate from Moisture 1st, 2nd, and 3rd Layer of soil	Fraction
SOILM400	Soil moisture from 200 to 400 cm	K	Interpolate from Moisture 2nd, 3rd, and 4th Layer of soil	Fraction
SEAICE	Binary flag for the presence (1.0)/absence (0.0) of sea ice	-	Fraction sea ice or lake ice & Fraction glacial ice or land ice	-
LANDSEA	Binary flag for land(1.0)/water(0.0) masking	-	Fraction bare ground	-
SOILHGT	Terrain elevation	m	-	m
WEASD	Water equivalent of accumulated snow depth	kg/m ²	Snow amount over land & Snow Fraction over land & size of grid	m
SNOWCOVR	Binary flag for the presence (1.0) or absence (0.0) of snow on the ground	-	Snow Fraction over land	-

The first three techniques use several successive scans to nudge a “first-guess” field toward the neighboring observed values. It calculates distance-weighted “radius of influence”, R , from all observation points, and uses it to adjust the “first-guess” field. The last technique uses hyperboloid radial basis functions as the basis of objective analysis.

For the purpose of linkage between GISS-GCM III and MM5, GISS-GCM III data was used for both input of REGRID, as well as the input of little_r. By incorporating GISS-GCM III data as observation data for little_r, better conservation of original GISS-GCM III meteorological conditions was achieved. A little_r linkage program was created to convert GISS-GCM III data into a little_r recognizable report format. A report format contains a single observation or group of observations that has the same latitude /longitude coordinate. Each report consists of four basic records, which are: (1) A report header record; (2) data records (could be multiple records); (3) end data record; and (4) end report record. The report header record is composed of a 600-character-long record and is used to store site information such as location, station ID, station type, and station elevation. The data records store values of pressure, height, temperature, dewpoint, wind speed, and wind direction, where each pressure level is separated into a single record. The end data record contains only dummy data of 777777 and the end report stores record count information for the report. Table 2.2 and 2.3 show the mapping between little_r and GISS-GCM III.

2.3.1.3 *GISS-GCM Gadget*

The output of GISS-GCM III covered the entire world with 4 x 5 resolutions. However, for little_r, only a regional scale domain was needed. For considering the processing time in little_r, a supplementary algorithm was written to pre-select observational data based on the latitude and longitude coordinates. Any observation that is 0.5 degree away from the edge of the domain grid, which defined by TERRAIN, was removed before it converted into little_r input format. This algorithm had greatly reduced the processing time of little_r.

Table 2.2. Mapping data for GISS-GCM III downscaling to little_r – part 1

Little_r	Units	GISS-GCM III	Units
<u>Report header format</u>			
Latitude	degree	Grid_Y	Grid #
Longitude	degree	Grid_X	Grid #
ID	-	GISS_GRID_X&Y	-
Name	-	GISS_GRID_X&Y	-
platform	-	GISS_MODEL_RESULT	-
source	-	BOGUS GISS_OUTPUT	-
elevation	-	0	-
Num_vld_fld	-	no. of fields in data record	-
Num_error	-	0	-
Num_warning	-	0	-
Seq_num	-	Sequential number (I.e. 1,2... etc.)	-
Num_dups	-	0	-
is_sound	-	T	-
bogus	-	T	-
discard	-	F	-
Sut	-	-888888	-
julian	-	-888888	-
Date_char	YYYYMMDDHHmmss	JulianDay	JulianDay
slp, qc	Pa	Sea Level pressure	mb
ref_pres, qc	-	-888888	-
ground_t, qc	-	-888888	-
sst, qc	-	-888888	-
Psfc, qc	-	-888888	-

Table 2.3. Mapping data for GISS-GCM III downscaling to little_r – part 2

Little_r	Units	GISS-GCM III	Units
precip, qc	-	-888888	-
t_max, qc	-	-888888	-
t_min, qc	-	-888888	-
t_min_night, qc	-	-888888	-
p_tend03, qc	-	-888888	-
p_tend24, qc	-	-888888	-
cloud_cvr, qc	-	-888888	-
ceiling, qc	-	-888888	-
<u>Format of data records</u>			
pressure of observation, qc	Pa	Sea Level Pressure	mb
height, qc	m (MSL)	Geopotential height	m (MSL)
temperature, qc	K	Surface Air Temperature	K
Dew_point, qc	-	-888888	-
wind speed, gc	-	-888888	-
direction, qc	-	-888888	-
u, qc	m/s	U Surface Wind	m/s
v, qc	m/s	V Surface Wind	m/s
rh, qc	%	Surface Relative Humidity	%
thickness, qc	-	-888888	-

Another supplementary algorithm, observation distribution algorithm, was written to create a higher resolution observation data matrix. Regular GISS-GCM III output contained only one observation point within a 4 x 5 resolution grid. This coarse resolution may not have sufficient observation points when processing little_r with a finer grid-based resolution. Therefore, this algorithm was used to distribute a single observation point into multiple observation points within the same GISS-GCM III grid. The algorithm permitted us to choose their observation density of the observation data applied to the little_r.

2.3.2 GEOS-Chem to CMAQ

The global chemistry model, GEOS-Chem was adopted to provide CMAQ IC/BC files. Figures 2.7 (a) and (b) show an example of NO_x and O_x concentrations from 2001 GEOS-Chem output. To allow the GEOS-Chem output to be used in the CMAQ model, the GEOS2CMAQ IC/BC interface module (GEOS2CMAQ) was developed by the University of Tennessee, Knoxville, Air Quality Modeling Group. Detailed program methodology and a general procedure for generating CMAQ IC/BC files were documented as follows. The GEOS2CMAQ program was written in Linux/Unix based Fortran 90. It was intended to convert existing GEOS-Chem output to a CMAQ IC/BC output. Since the GEOS-Chem and the CMAQ models did not share common temporal, spatial, and chemical schemes, converting the GEOS-Chem output to CMAQ IC/BC involved time-step interpolation, vertical and horizontal interpolations, chemical species conversion, adding additional chemical species, and unit conversion.

2.3.2.1 Time-step Interpolation

The temporal resolution of standard GEOS-Chem output was every 3rd hour, whereas the temporal resolution of CMAQ boundary condition was every hour. To match the temporal resolution, the GEOS2CMAQ program had assumed the 1st hour of the GEOS-Chem data was same as the 2nd and 3rd hours of the data. Hence, hourly profile of CMAQ boundary condition was generated.

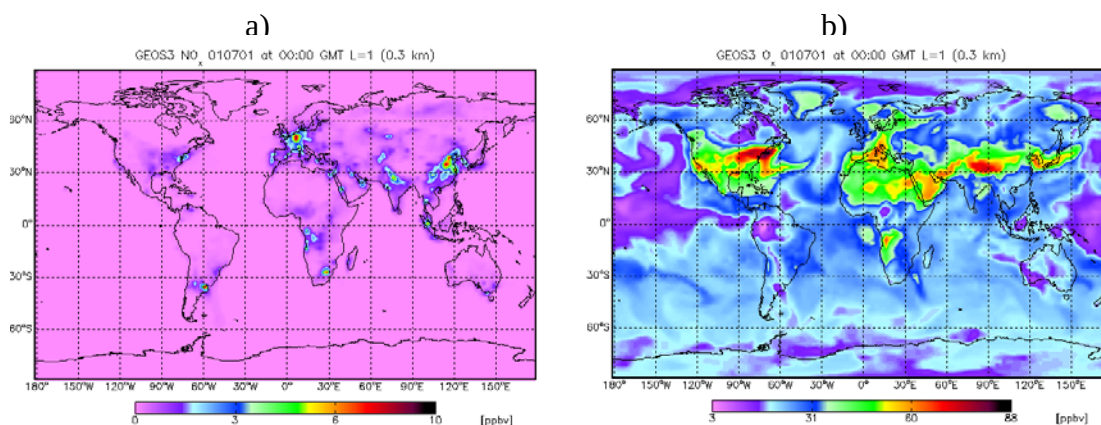


Figure 2.7. Example of GEOS-Chem output: a) NO_x concentration; b) O_3 concentration

For CMAQ initial condition, since only data from the 1st hour were required for each simulation period, no interpolation of temporal data was required.

2.3.2.2 *Spatial Interpolation*

In the matter of spatial interpolation, algorithms were developed in the GEOS-Chem2CMAQ program to execute both vertical and horizontal interpolations simultaneously. The horizontal interpolation algorithm found the column of GEOS-CHEM cells in which the arbitrary CMAQ grid cell was located and assigned species concentration values from the GEOS-Chem output to the CMAQ grid cell. When a CMAQ grid cell fell between two GEOS-Chem grid cells, the average concentration from those two grid cells was used. The vertical interpolation algorithm compared the height of each GEOS-Chem vertical layer with the height of each arbitrary CMAQ vertical layer to determine whether vertical interpolation was required. When a CMAQ layer fell between two GEOS-Chem layers, vertical interpolation was performed.

2.3.2.3 *Horizontal Interpolation Algorithm*

The GEOS-Chem output was organized in a sequential binary format. Each binary record contained an array of data that was associated with all chemical species in different vertical layers within the same horizontal grid cell. The center location of the grid cell, in terms of latitude and longitude coordinates, was calculated based on the grid-based model resolution. Linking the GEOS-Chem output to CMAQ IC/BC, the arbitrary information from the CMAQ model, such as map projection type, domain center, longitude /latitude and domain resolution was required and was available from one of the MCIP outputs, GRIDCRO2D file (Byun and Schere, 2006;Byun and Ching, 1999). With the given horizontal grid information from both models, the algorithm first computed the projected horizontal locations from each CMAQ grid cell and assigned the nearest GEOS-Chem grid cell to the CMAQ grid cell. This latter information was used in the vertical interpolation algorithm step to estimate species concentrations for the CMAQ grid cell.

2.3.2.4 Vertical Interpolation Algorithm

The vertical interpolation algorithm was designed to extract the vertical layers information from the GEOS-Chem output and assigned it to the CMAQ IC/BC. Since the GEOS-Chem model did not have the same vertical layers structure as the CMAQ model, model interpolation, using size-apportioning (i.e., height) interpolation, was required. Two possible scenarios occurred in the interpolation: “containing” and “not containing”. For the “containing” scenario, where the corresponding CMAQ layer was vertically contained within a GEOS-Chem layer (as shown in Figure 2.8 [a]), the concentration values of all species from the GEOS-Chem layer were directly assigned to the CMAQ layer.

On the other hand, for the “not containing” scenario, where the CMAQ grid cell vertically overlapped with one or more GEOS-Chem layers (as shown in Figure 2.8b), the concentration values of all species were calculated according to overlapping percentage and concentration of each related layer using the following equation.

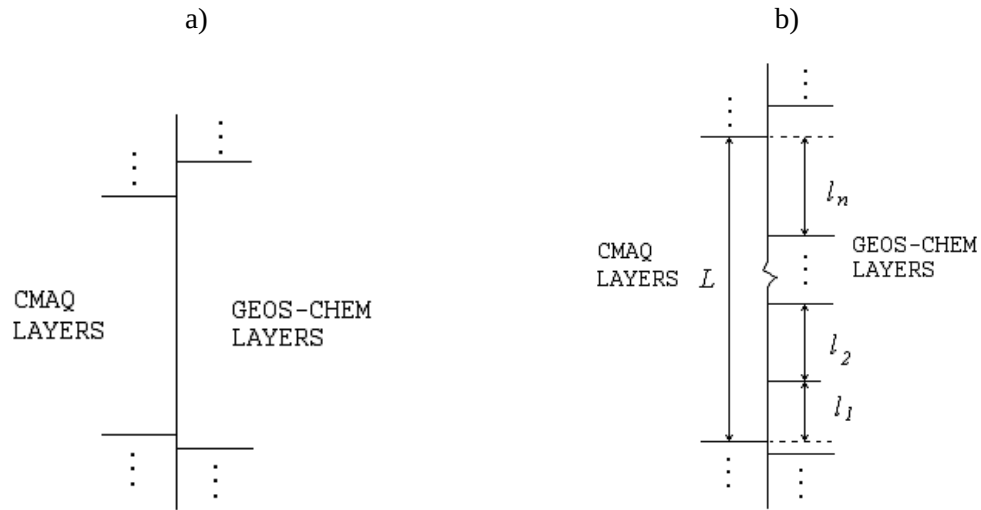


Figure 2.8. Vertical Interpretation: a) CMAQ layer contained within a GEOS-Chem layer; b) CMAQ layer overlaps with more than one GEOS-Chem layer

$$C = \sum_{i=1}^n G_i \frac{l_i}{L} \quad (2.3)$$

where $L = \sum_{i=1}^n l_i$, C is the estimated concentration in a CMAQ unit cell, L is the height of the CMAQ grid cell, n is the number of GEOS-Chem layers overlapping the CMAQ cell vertically, l_i is the height overlapped with i^{th} of the n overlapping GEOS-CHEM layers, and G_i is the interpreted concentration of the GEOS-Chem unit cell in the i^{th} overlapping GEOS-Chem layer. The interpolated C value is an approximation of the corresponding concentration in the CMAQ grid cell.

2.3.2.5 Chemical Species Conversion

The GEOS-Chem output contained a total of 37 species in which 30 species were used for converting GEOS-Chem species to CMAQ IC/BC species. Table 2.4 shows the conversion formulas that were used in the current GEOS2CMAQ program. Using the formulas given in the Table 2.4, the GEOS-Chem species were converted to the CMAQ recognizable chemical species. The GEOS-Chem output contained only 30 species of the CMAQ CB-IV required species. It did not cover all the CMAQ CB-IV species required in IC/BC. When missing species found in the CMAQ IC/BC, the GEOS2CMAQ program was automatically assigning the default IC/BC profile concentrations, which prevented a missing data error from occurring when running the CMAQ model. It should be noted that since not all species in GEOS-Chem were interpolated into CMAQ, it is expected that the species total was not conserved in CMAQ, which might result an underestimated of species concentrations on those missing species in the IC/BC.

2.3.2.6 Unit Conversion

For gas species, the output unit of the CMAQ was the same as the unit in the GEOS-Chem, which was ppm_v . However, for particulate species, the CMAQ required unit was $\mu g/m^3$ and the unit of the GEOS-Chem was given as ppm_v . Therefore, unit conversion was required for all particulate species.

Table 2.4. Chemical mapping formula from GEOS-CHEM to CMAQ CB-IV Scheme

CMAQ CB-IV specie	GEOS-CHEM species
[NO ₂]	[NO _x]
[O ₃]	[O _x]-[NO _x]
[N ₂ O ₅]	[N ₂ O ₅]
[HNO ₃]	[HNO ₃]
[PNA]	[HNO ₄]
[H ₂ O ₂]	[H ₂ O ₂]
[CO]	[CO]
[PAN]	[PAN] + [PMN] + [PPN]
[MGLY]	[MP]
[ISPD]	[MVK] + [MACR]
[NTR]	[R ₄ N ₂]
[FORM]	[CH ₂ O]
[ALD2]	1/2[ALD2] + [RCHO]
[PAR]	[ALK4] + [C ₂ H ₆] + [C ₃ H ₈] + [ACET] + [MEK] + 1/2[PRPE]
[OLE]	1/2 [PRPE]
[ISOP]	1/5 [ISOP]
[SO ₂]	[SO ₂]
[NH ₃]	[NH ₃]
[ASO ₄ J]	[SO ₄]
[ANH ₄ J]	[NH ₄]
[ANO ₃ J]	[NIT] + [NITs]

It should be noted that GEOS-Chem arbitrarily selected the unit of ppm_V as the reporting unit for both gaseous and aerosol pollutants. Using the assumption of homogeneous distribution of species concentration for each simulation grid in GEOS-Chem, the unit for particulate species could be interchangeable between ppm_V and $\mu g/m^3$ using the equation given in equation 2.4. The equation for the unit conversion is given as follows:

$$\frac{Den_{air} \times 1000}{28.97} \times Conc \times Mw \quad (2.4)$$

where Den_{air} is air density provided by MCIP results (kg/m^3), 28.97 is the dry air molecular weight (g/mol), $Conc$ is the concentration value to be transformed (ppm_v), and Mw is the molecular weight of the species(g/mol).

CHAPTER III

3 STUDY OF DOWNSCALING TECHNIQUE FOR THE LINKAGE OF GLOBAL AND REGIONAL AIR QUALITY MODELING

3.1 Declaration

This chapter is a slightly revised version of a paper with the same title published on the Atmospheric Chemistry and Physics by Yun Fat Lam; Joshua Fu. (doi:10.5194/acp-10-4013-2010)

3.2 Introduction

Regional air quality models are designed to simulate the transport, production, and destruction of atmospheric chemicals at the tropospheric level. Particular interest is given at the planetary boundary layer (PBL) where human activities reside (Byun and Schere, 2006). Performance of the regional models depends greatly on the temporal and spatial quality of the inputs (i.e., emission inventories, meteorological model outputs, and boundary conditions). Recently, establishing proper boundary conditions (BCs) has become a crucial process as the effects of intercontinental transport of air pollutants (Heald et al., 2003; Chin et al., 2007; Lin et al., 2008) and enhancement of background pollutant concentrations emerged (Vingarzan, 2004; Ordonez et al., 2007; Fiore et al., 2003a). Various studies suggested that utilizing dynamic global chemical transport model (CTM) outputs as the BCs for the regional air quality model would be the best option for capturing the temporal variation and spatial distributions of the tracer species (Fu et al., 2008a; Byun et al., 2004; Morris et al., 2006a; Tang et al., 2007). For example, Song et al., (2008) applied the interpolated values from a global chemical model, RAQMS, as the lateral BCs for the regional air quality model, CMAQ and evaluated simulated CMAQ results with ozone soundings. Simulations were performed on the standard CMAQ seasonal varied profile BCs and dynamic BCs from RAQMS. The results demonstrated that the scenario with dynamic BCs performed better than the scenario with profile BCs in terms of the prediction of vertical ozone profile.

The quality of BCs depends on the vertical, horizontal, and temporal resolutions of global CTM outputs. The latitudinal location and seasonal variation are also playing an important role, which defines the tropopause height that influences the vertical interpolation

process between global and regional models (Bethan et al., 1996;Stohl et al., 2003). In the MICS-Asia project, high concentrations of ozone (i.e., 500 ppbv) have been observed in CMAQ BCs when the regional model's layers reach above or beyond the tropopause height during the vertical interpolation process (Fu et al., 2008a). This high ozone aloft in BCs has created problems for the regional tropospheric model (such as CMAQ) since it does not have a stratospheric component or stratosphere-troposphere exchange mechanism. As a result, unrealistically high ozone concentrations were observed at the surface layer during the regional CTM simulations. Tang et al. (2008) studied various CTM lateral BCs from MOZART-NCAR, MOZART-GFDL, and RAQMS. They observed that CTM BCs have induced a high concentration of ozone in the upper troposphere in CMAQ; this high ozone aloft quickly mixed down to the surface resulting in an overestimation of surface ozone. Mathur et al. (2008), suggested that the overestimation of O₃ might also be partially contributed by the inadequate representation of free tropospheric mixing due to the selection of a coarse vertical resolution (Mathur et al., 2008;Tang et al., 2008). Since the rate of vertical transport of flux is highly sensitive to temperature and moisture-induced buoyancies, correctly representing deep convection or flux entrainment at the unstable layer in the meteorological model becomes critical to modeling ozone vertical mixing. It should be noted that the single PBL scheme in the meteorological model is not sufficient to simulate the correct vertical layer structure on the broad aspect of environmental conditions (i.e., terrain elevation and PBL height) in the existing domain. As a result, it introduces uncertainties and errors to the process of determining vertical transport of O₃ in the air quality model (Zangl et al., 2008;Perez et al., 2006). For the downscaling problem, Tang et al. (2008) has commented that using outputs of the GCTM as BCs may not necessarily be better than the standard profile-BC, which highly depends on location and time. The quick downward mixing in CMAQ has caused an erroneous prediction of surface ozone when both tropospheric and stratospheric ozone are included in the CTM BCs. (Al-Saadi et al., 2007;Tang et al., 2007;Tang et al., 2008) Therefore, correctly defining tropopause height for separating troposphere and stratosphere becomes crucial to the prevention of stratospheric influence during the vertical interpolation process for CMAQ and other regional CTMs simulation.

The tropopause is defined as the boundary/transitional layer between the troposphere and the stratosphere, which separates by distinct physical regimes in the atmosphere. The height of tropopause ranges from 6 km to 18 km depending on seasons and locations (Stohl et al., 2003). In the United States, the typical tropopause height in summer ranges from 12 km to 16 km, but drops to 8 km to 12 km in winter (Newchurch et al., 2003). Various techniques were developed for identifying the altitude of tropopause, which are based on temperature gradient, potential vorticity (PV), and ozone gradient. In meteorological studies, such as satellite and sonde data analysis, temperature gradient method, also referred to as the thermal tropopause method, is the most commonly used technique, which searches the lowest altitude where the temperature lapse rate decreased to less than 2 °K/km for the next 2 km and defines that as tropopause (WMO, 1986). In climate modeling, PV technique, referred to as dynamic technique, is often applied to define tropopause. PV is a vertical momentum up drift parameter and is expressed by PV unit ($1 \text{ PVU} = 10^{-6} \text{ K} \cdot \text{m}^2/\text{kg} \cdot \text{s}$). The threshold value of the tropopause lies between ± 1.6 to 3.5 PVU depending on the location on the globe (Hoinka, 1997). Recently, in an attempt to improve the regional model (i.e., the pure tropospheric model), CMAQ (to simulate ozone at the lower stratosphere) was performed using Potential Vorticity relationship. Location-independent correlation between PVU with ozone concentrations was applied to correct the near/above-tropopause ozone concentrations in CMAQ. The fundamental disadvantage of using such technique is the implementation of a single correlation profile (i.e., $R^2 = 0.7$) to represent the entire study domain (i.e., the continental United States). It shows that a slight shift of PV value in the profile could result in a big change of ozone concentration, up to 100 ppbv. In addition, this profile may not be applicable for all locations in the domain due to the limited amount of data in the literature (Mathur et al., 2008). In GCTM downscaling, the ozone gradient technique, referred to as chemical tropopause or ozone tropopause, is more appropriate for defining tropopause since we have observed the stratospheric level of ozone (i.e., about 300 ppbv) at the level of thermal and dynamic tropopause (Lam et al., 2008b).

Ozone tropopause is defined by atmospheric ozone concentration, which observes a sharp transition from low concentrations to high concentrations from troposphere to stratosphere. The defined O₃ tropopause is consistently lower than the thermal and dynamic

tropopause (Bethan et al., 1996). The height of tropopause affects both the stratosphere-troposphere exchange (STE) as well as the transport of O₃ at upper troposphere. (Holton et al., 1995;Stohl et al., 2003) In global CTM, well-defined vertical profiles of troposphere, tropopause, and stratosphere are established for simulating STE, upper tropospheric advection, and other atmospheric processes. Collins (2003) estimated that the net O₃ flux from the stratosphere could contribute 10 to 15 *ppbv* of the overall tropospheric ozone. (Collins et al., 2003), where Stohl (2003) has found about 10% to 20% of tropospheric ozone are originated from stratosphere (Collins et al., 2003). The advantage of employing CTM outputs as BCs gives a better representation of upper troposphere and the effect of STE can be taken into account. Although global CTM is capable of simulating tropospheric conditions, the temporal and spatial resolutions may not be sufficient to represent the daily and monthly variability of surface conditions since the monthly chemical profile of budget is used. Several researchers have demonstrated the outputs of global CTM can be used in the area of surface background conditions and trends (Park et al., 2006;Fiore et al., 2003a). However, it also indicated that the global CTM is inadequate to predict the peak magnitude of O₃ at the surface since it is not intended to describe detailed surface flux condition at a high temporal and spatial resolution. Therefore, the regional air quality model remains indispensable for simulating the surface O₃ conditions.

In this study, we have developed a linking tool to provide lateral BCs of the USEPA Community Multiscale Air Quality (CMAQ) model with the outputs from GEOS-Chem (Byun and Schere, 2006;Lam et al., 2008b;Li et al., 2005). One full year of GEOS-Chem data in 2002 are analyzed and summarized to explore the seasonal variations of O₃ vertical profiles and tropopause heights in global CTM with available ozonesonde data in the United States are used to verify the performance of the GEOS-Chem model. Evaluations are conducted to measure the potential impact of changing tropopause height to the performance of the interpolated BCs toward the regional CTM. A new algorithm, “tropopause-determining algorithm”, which is based on chemical (O₃) tropopause definition, is proposed for the vertical interpolation process during downscaling to remove stratospheric effects from the global CTM toward the regional CTM. Verifications of the new algorithm are performed using three sets of CMAQ simulations, which are: (1) the static lateral BCs from predefined

profile is used as an experimental control for GEOS-Chem data inputs; (2) standard dynamic lateral BCs from GEOS-Chem using original vertical interpolation; and (3) the modified dynamic lateral BCs from GEOS-Chem, and is intended to show the improvement of the proposed idea using the observation data from ozonesonde and CASTNET. Moreover, it demonstrates the necessity of filtering the tropospheric portion of global GMC outputs for the inputs in regional air quality modeling.

3.3 Description and Configuration of Models Used

In this study, GEOS-Chem global chemistry model output is used to provide lateral boundary conditions for the regional air quality model CMAQ, where meteorological inputs are driven by the MM5 mesoscale model. The model setups are described as follows.

3.3.1 GEOS-Chem

GEOS-Chem global chemistry model output is one of the most popular global models for generating BCs for the CMAQ regional model. (Morris et al., 2005; Eder and Yu, 2006; Tesche et al., 2006; Streets et al., 2007; Tagaris et al., 2007) Many studies demonstrated that GEO-Chem is capable of capturing the effects from intercontinental transport of air pollutants and increasing background concentrations (Heald et al., 2006; Liang et al., 2007; Park et al., 2003). Please note the above referenced studies may have used different versions of GEOS-Chem. For example, Heald et al. (2006) used version 4.33 of GEOS-Chem, where as Liang et al. (2007) and Park et al. (2003) used version 7.02.

GEOS-Chem is a hybrid (stratospheric and tropospheric) 3-D global chemical transport model with coupled aerosol-oxidant chemistry (Park et al., 2006). It uses 3-hour assimilated meteorological data such as winds, convective mass fluxes, mixed layer depths, temperature, clouds, precipitation, and surface properties from the NASA Goddard Earth Observing System (GEOS-3 or GEOS-4) to simulate atmospheric transports and chemical balances. In this study, all GEOS-Chem simulations were carried out with 2° latitude by 2.5° longitude ($2^{\circ} \times 2.5^{\circ}$) horizontal resolution on 48 sigma vertical layers. The lowest model levels are centered at approximately 50, 200, 500, 1000, and 2000 m above the surface.

Figure 3.1 (a) and (b) show the vertical layer structure of GEOS-Chem. The grey areas indicate the height range of tropopause in summer and winter in literature. A full-year simulation was conducted for year 2002, which was initialized on September 1, 2001 and continued for 16 months. The first four months were used to achieve proper initialization, and the following 12 months were used as the actual simulation results. All simulations were conducted using version 7.02 with GEOS-3 meteorological input. Detailed discussion of GEOS-Chem of version 7.02 is available elsewhere (Park et al., 2004).

For the purpose of developing a new algorithm for the downscaling linkage application, the outputs from GEOS-Chem in 2002 were being analyzed for investigating the variation of tropopause heights. Many published studies have already demonstrated the ability of GEOS-Chem to predict an ozone vertical profile using ozonesonde and satellite observations (Liu et al., 2006; Fusco and Logan, 2003; Martin et al., 2002), therefore, no detailed performance analysis was conducted in this study. Note that GEOS-Chem simulates stratospheric ozone with the Synoz algorithm (McLinden et al., 2000), which gives us the right cross-tropopause ozone flux but no guarantee of correct ozone concentrations in the region. That is because, until recently, cross-tropopause transport of air in the GEOS fields was sometimes too fast. This is discussed for example in Bey et al. 2001, Liu et al., 2001, Fusco and Logan 2003. Nevertheless, for this study, simple model verifications were still conducted on the GEOS-Chem outputs using available ozonesonde data in the United States (Newchurch et al., 2003). Particular interest was given to upper troposphere and tropopause regions (1000 hPa to 50 hPa), where the downscaling process could be influenced by stratospheric ozone. Figure 3.2 shows the yearly variability of GEOS-Chem with ozonesonde data. It is observed that 99.5 % of GEOS-Chem outputs are contained within the statistical range of the observation data, which gives a good indication of reasonable model results. For the Boulder and Huntsville sites, good model performances were found at higher pressure when the pressure fell between 1000 hPa to 300 hPa. Consistent under-predictions were observed at the upper atmosphere when the pressures dropped below 250hPa.

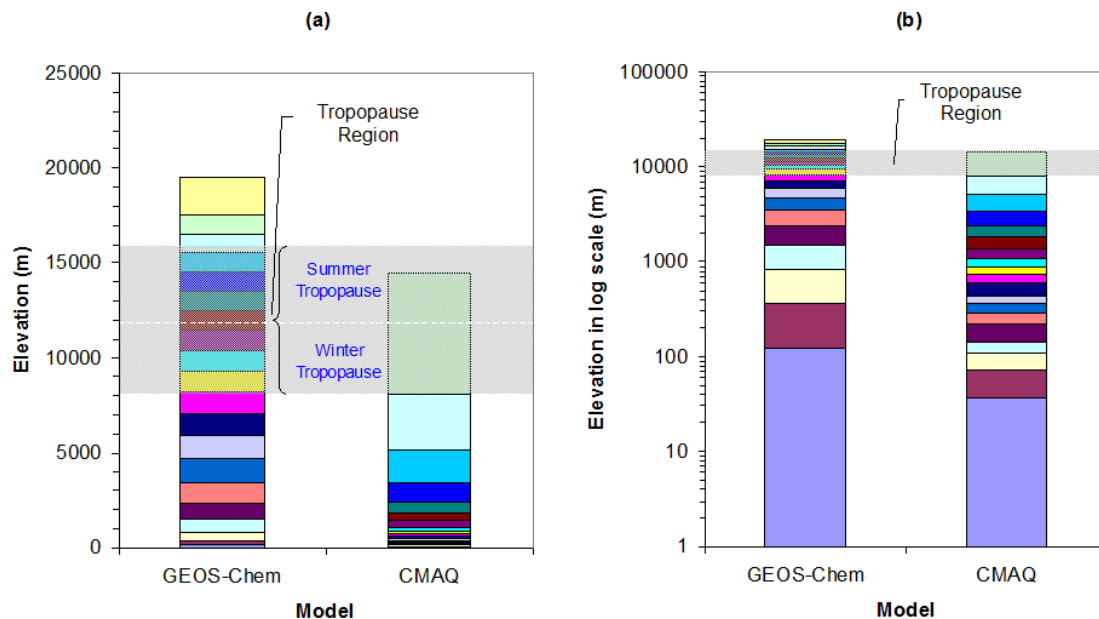


Figure 3.1. Vertical layer structure comparison between GEOS-Chem and CMAQ, (a) arithmetic scale, and (b) log scale.

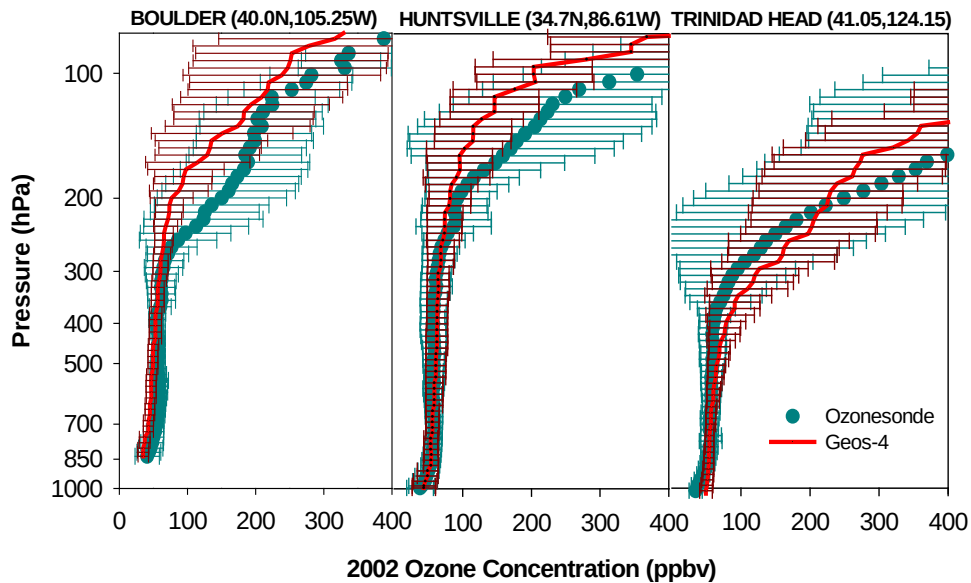


Figure 3.2. Yearly variability of GEOS-Chem outputs verses ozonesonde.

3.3.2 MM5 and CMAQ

The CMAQ meteorological inputs are driven by NCAR's 5th generation Mesoscale Model version 3.7. (MM5) with hourly temporal resolution, 36 km horizontal resolution, and 34 sigma vertical layers. All MM5 simulations were conducted using the one-way nested approach from 108 km over North America (140 - 40W, 10 - 60 N) down to 36 km continental United States (128 - 55W, 21 - 50 N). For meteorological initial and boundary conditions, the NCEP Final Global Analyses (FNL) data (i.e., ds083.2) with resolution of 1° by 1° from the United States National Centers for Environmental Prediction (NCEP) was used. For MM5 simulations, 4-D analysis nudging technique was employed to reproduce the observed weather conditions using the surface and upper layers observations from DS353.4 and DS464.0, respectively. The new Kain-Fritsch cumulus, Mix-phase micro-physic, RRTM long-wave radiations, planetary boundary layer (PBL) and land surface model (LSM) were configured in the simulations. A detailed summary of MM5 configuration is listed in Table 3.1. For CMAQ, Lambert conformal projection with true latitude limits of 25 and 40 was used on 148 by 112 grid cells with horizontal resolution of 36 km. A total of 19 sigma vertical layers were extracted from MM5. The lowest model levels were centered at approximately 20, 50, 90, 130, 180, 250, 330, and 400 m above the surface as shown in Figure 3.1 (a) and (b). The center of the horizontal domain was set at 100W and 40N. This domain covers the entire continental United States with part of the Mexico and Canada (referred to as CONUS domain), which is shown in Figure 3.3. In CMAQ simulations, three scenarios with different lateral boundary conditions were performed, which included profile boundary conditions (Profile-BC), ordinary vertical interpolated GEOS-Chem boundary conditions (ORDY-BC), and vertical interpolated GEOS-Chem boundary conditions using the new algorithm (Tropo-BC). All of these simulations were configured with Carbon Bond IV (CB-IV) chemical mechanism with aerosol module (AERO3). The detailed configuration is also shown in Table 3.1.

Table 3.1. MM5 and CMAQ model configurations for 2002 simulations.

MM5 Configuration	
Model version	3.7
Number of sigma level	34
Number of grid	156 x 120
Horizontal resolution	36 km
Map projection	Lambert conformal
FDDA	Analysis nudging
Cumulus	Kain-Fritsch 2
Microphysics	Mix-phase
Radiation	RRTM
PBL	Pleim-Xiu
LSM	Pleim-Xiu LSM
LULC	USGS 25-Category
CMAQ Configuration	
Model version	4.5
Number of Layer	19
Number of grid	148 x 112
Horizontal resolution	36 km
Horizontal advection	PPM
Vertical advection	PPM
Aerosol module	AERO3
Aqueous module	CB-IV
Emission	VISTAS emissions (NEI 2002 G)
Boundary condition I	CMAQ Predefined Vertical Profile
Boundary condition II	2002 GEOS-Chem

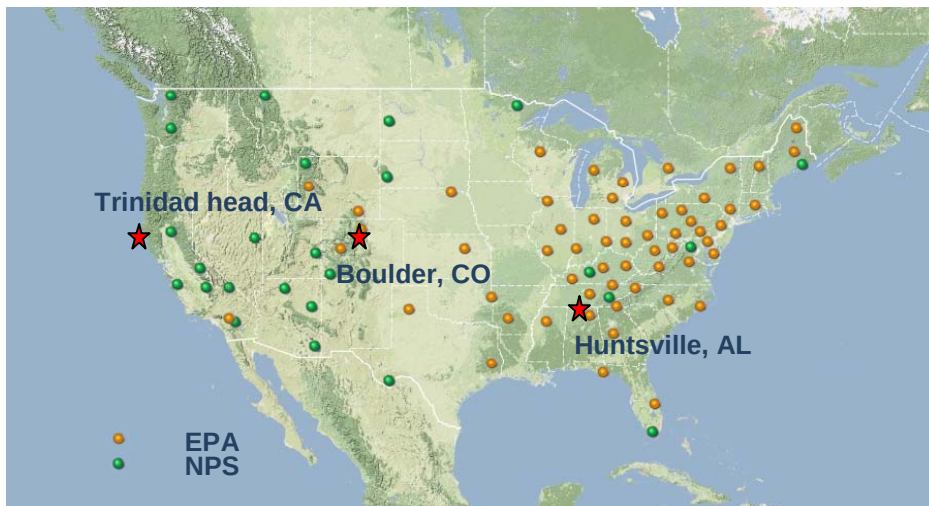


Figure 3.3. The CONUS domain with observation sites marked in green or orange from CASTNET and ozonesondes in red star.

3.3.3 Linkage Methodology between GEOS-Chem and CMAQ

The GEOS-Chem outputs were extracted as CMAQ lateral boundary conditions using GEOS2CMAQ linkage tool, which involved grid structure association, horizontal/vertical interpolation, and chemical mapping processes. A summary of the systematic flowchart of the linkage methodology is shown in Figure 3.4. It should be noted that most of the regional models including CMAQ do not utilize top boundary condition as input. As a result, in this study, no top boundary condition is generated. In the linkage process, GEOS2CMAQ applied the ‘nearest neighbor’ method to associating the latitude/longitude formatted GEOS-Chem outputs with the CMAQ Lambert Conformal gridded format. Horizontal interpolating process then utilized the results to interpolate the GEOS-Chem outputs into CMAQ gridded format for each vertical layer column. For Tropo-BC, a newly developed tropopause-determining algorithm, which is based on chemical (O_3) tropopause definition, was implemented in the vertical interpolating process to identify the tropopause height. Moreover, it separated the troposphere from the stratosphere for each horizontal grid. Different interpolating processes were employed in the tropospheric and the stratospheric regions. A detailed discussion may be found in the latter section of this document.

For the chemical mapping process, 37 GEOS-Chem species were transformed into 30 CB-IV mechanism species of CMAQ according to the chemical definitions given in Table 2.4. The GEOS-Chem species with the same definitions as CB-IV species were mapped directly into CMAQ; where as other species were mapped by partitioning and/or regrouping processes. For example, total oxidants O_x species in GEOS-Chem were defined as the combination of O_3 and NO_x . Therefore, to obtain O_3 concentrations, O_x was subtracted by NO_x species in the GEOS-Chem. Other species, such as paraffin carbon bond (PAR), were composed of multiple species in GEOS-Chem. Regrouping was required to reconstruct the CB-IV corresponding species, which is shown as follows:

$$PAR = ALK4 + C_2H_6 + C_3H_8 + ACET + MEK + \frac{1}{2}PRPE \quad (3.1)$$

For chemicals that were not supported by GEOS-Chem, CMAQ predefined boundary conditions were used to maintain the full list of CMAQ CB-IV species.

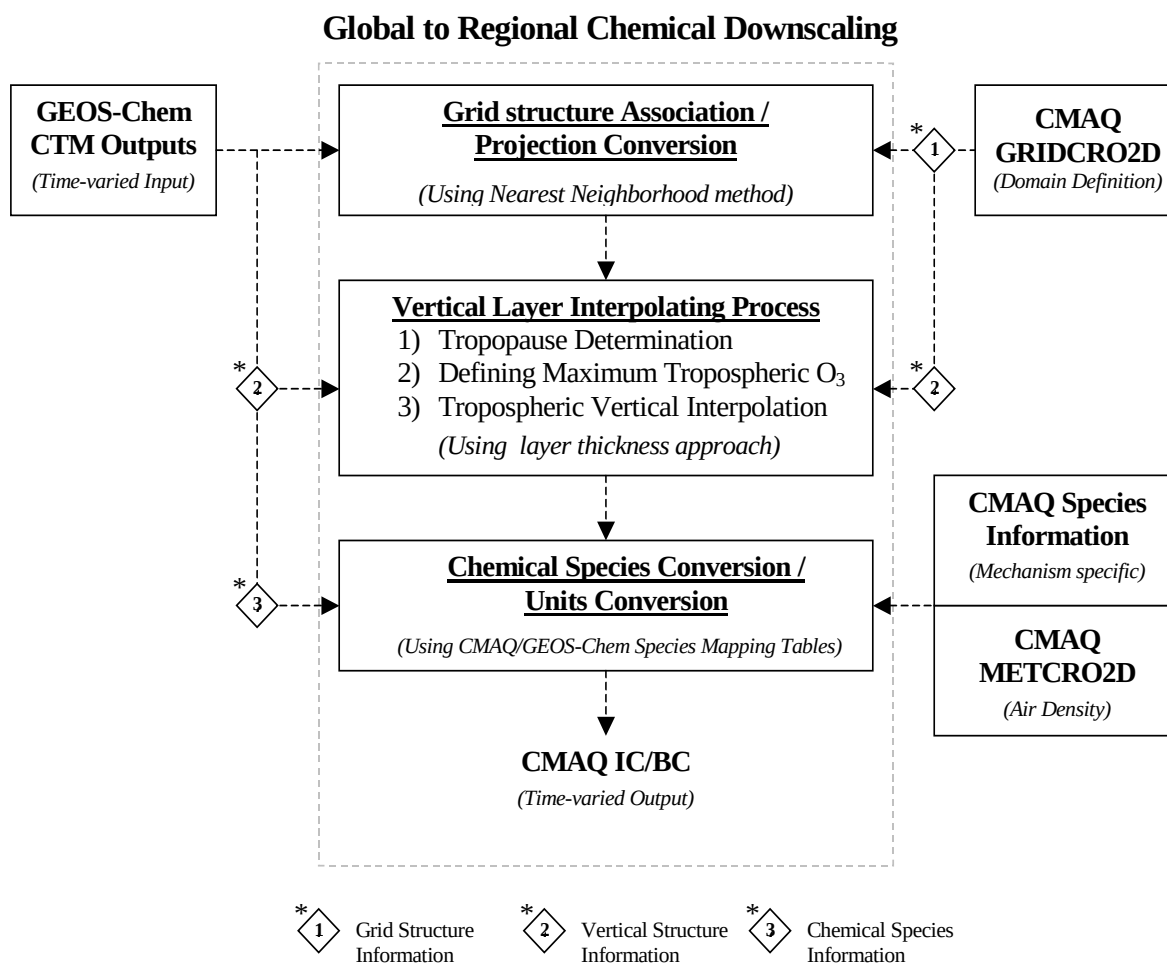


Figure 3.4. Systematic flowchart of global to regional chemical downscaling.

3.3.4 Tropopause Determining Algorithm

The newly developed tropopause-determining algorithm was added to the ordinary interpolating process (i.e., uses pressure level as the only criteria in the interpolating process) for handling the near tropopause and stratosphere interpolating processes, which is essential to correct and represent the global model outputs in the regional model. We have utilized the chemical/ozone tropopause definition described in Bethan (1996), instead of thermal and dynamic tropopause definitions, as the basis for separating the stratosphere and the troposphere. Although thermal and dynamic tropopauses are more commonly used in determining the tropopause, we have identified that these tropopauses are inappropriate for this application because of the observed stratospheric ozone effect at the troposphere. Since the purpose of determining tropopause is to exclude stratospheric pollutants concentrations from the global model during the interpolating process, ozone tropopause is better suited for this application. Ozone tropopause is defined as the location at which an abrupt change of ozone concentration occurred. Our algorithm finds the ozone tropopause by finding the largest negative rate of change of slope (i.e., could be negative) from the plot of elevation verses ozone concentration. In other words, we have taken the second derivative of elevation with respect to ozone concentration and found the lowest value.

$$H_{Tropo(C_i)} = \max \left| \frac{C_{i+1} - C_i}{H_{i+1} - H_i} - \frac{C_i - C_{i-1}}{H_i - H_{i-1}} \right| \quad \text{where } 8km < H < 19km \quad (3.2)$$

Each rate of change of slope requires 3 data points or 2 line segments, upon which two line slopes were calculated. In the tropopause level, which is indicated by the largest negative rate of change of slope, a combination of a small concentration change in the first segment with a large concentration change in the second segment were obtained. Occasionally, a false tropopause was identified when an extremely small change of ozone concentration in the first segment or negative change of ozone concentrations in the second segment occurred. To ensure the tropopause found by this method is a reasonable tropopause height with no stratospheric effect, we have cross checked the tropopause results with thermal tropopause heights (i.e., ozone tropopause should be lower than thermal tropopause), as well as the

maximum concentrations of ozone should exceed 300 ppbv as found in the literature (McPeters et al., 2007).

For the vertical interpolating process in GEOS2CMAQ, stratospheric ozone is excluded by limiting the maximum ozone concentration at the tropopause level while generating CMAQ lateral boundary conditions. Unlike some of the studies, without enforcing any upper bound limit or using predefined maximum ozone concentration, we have dynamically determined the altitude of the tropopause for each grid and time-step in GEOS-Chem outputs for use in the vertical interpolating process (Morris et al., 2006b; Song et al., 2008; Tang et al., 2007).

3.4 Results and Discussion

3.4.1 CMAQ Lateral Boundary Conditions

We have generated CMAQ lateral boundary conditions from every third hour GEOS-Chem output for VISTAS CMAQ simulation using GEOS2CMAQ linkage tool. Figure 3.5 shows the vertical ozone profiles from GEOS-Chem with CMAQ vertical layers for both summer and winter. It should be noted that the tropopause in summer is much higher than the tropopause in winter. As a result, less stratospheric ozone is included in summer than winter when the vertical interpolating process is performed. In Figure 3.6, comparisons of Profile-BC, ORDY-BC, and Tropo-BC for June 22, 2002 is shown on the CONUS domain. The top row represents the 1st CMAQ layer (~ 1000 millibars) and the bottom shows the top CMAQ layer (i.e., 19th layer~ 140 millibars). These plots are intended to demonstrate the horizontal distribution of ozone concentrations across the CONUS domain. The Profile-BC was designed to represent the relatively clean air conditions for the CONUS boundaries. It enforces a pre-defined vertical profile with no temporal and spatial dependencies. In general, the surface ozone concentrations (i.e., 1st layer) range between 30 to 35 ppbv and they progressively increase and reach a peak ozone concentration of 70 ppbv at the top (i.e., 19th layer). The ORDY-BC and Tropo-BC were both generated using the linkage methodology described earlier. This methodology intends to incorporate the effects of intercontinental transport of air pollutants and the rise in background ozone concentrations into CMAQ by utilizing GEOS-Chem outputs (Bertschi et al., 2004; Fiore et al., 2003a; Park et al., 2004).

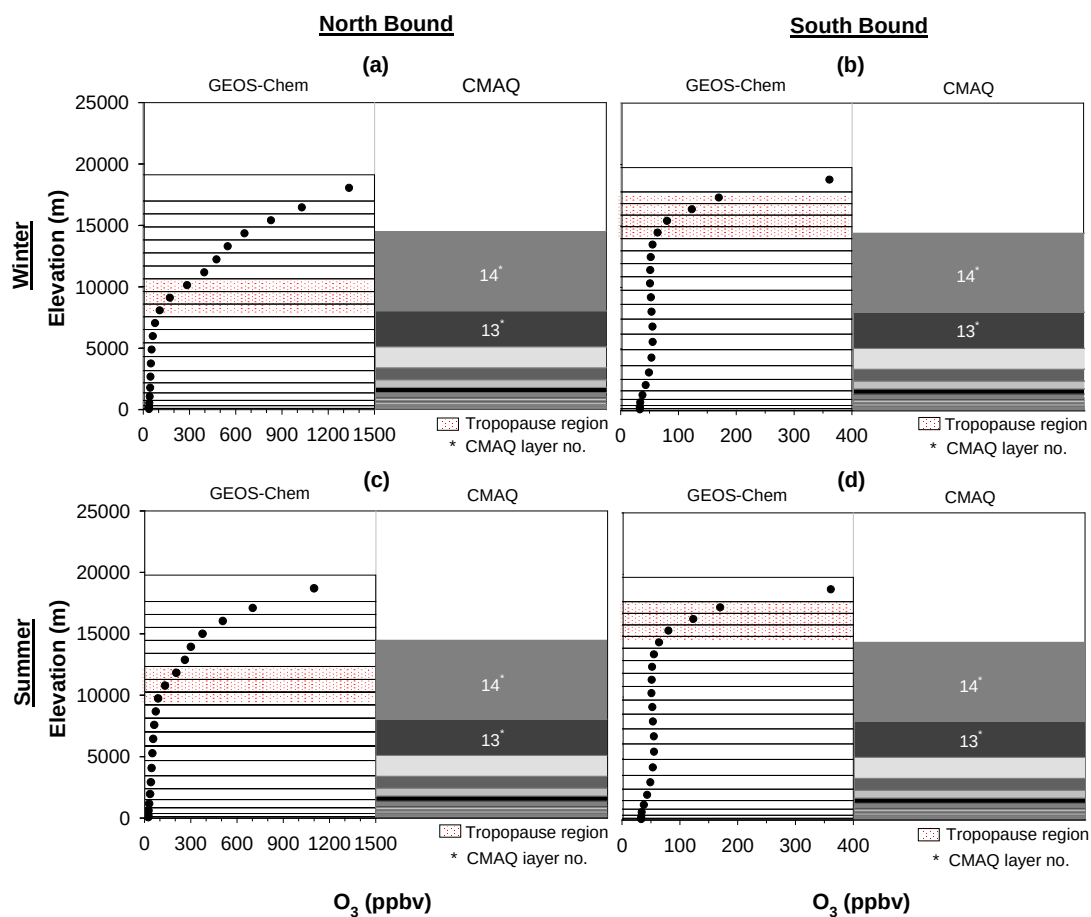


Figure 3.5. Vertical ozone profiles from GEOS-Chem with CMAQ layers for both summer and winter, (a) north bound in winter, (b) south bound in winter, (c) north bound in summer, and (d) south bound in summer.

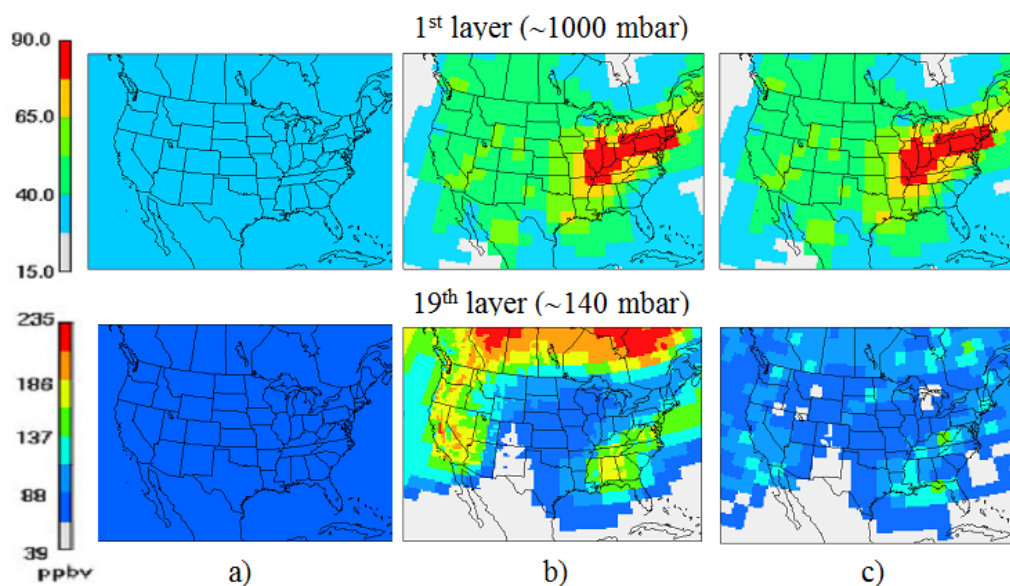


Figure 3.6. Comparison of different lateral boundary conditions in 1st and 19th layers, a) Profile-BC, b) ORDY-BC, and c) Tropo-BC.

The temporal and horizontal variations in GEOS-Chem were captured into CMAQ to reflect daily diurnal differences in concentrations. In ORDY-BC and Tropo-BC, the difference was the vertical interpolating process. ORDY-BC uses the ordinary vertical interpolating process, where as Tropo-BC uses the ordinary vertical interpolating process with the tropopause-determining algorithm that excludes pollutants in the stratosphere from the interpolating process. In the surface level (1st layer), both ORDY-BC and Tropo-BC perform identically; ozone concentrations ranged from 19 *ppbv* to 90 *ppbv* depending on location and time on June 22nd. For other days in 2002 (i.e., January, June, and July), ozone concentration could reach up to 130 *ppbv* at the surface. In the top level (19th layer), the ORDY-BC ozone reaches as much as 235 *ppbv* and the Tropo-BC ozone achieves up to 160 *ppbv* in the CONUS domain on June 22nd. For other days in 2002, the ORDY-BC and Tropo-BC ozone reaches up to 714 *ppbv* and 205 *ppbv*, respectively. In Considine (2008), the reported maximum mean tropopause ozone concentration from observations in North America is about 235 *ppbv* based on the thermal tropopause definition. We would have expected that if Considine's analyses used the ozone tropopause as its definition, the maximum tropopause ozone concentrations should be lower since the ozone tropopause is constantly lower than the

thermal tropopause at the upper troposphere. So, the maximum ORDY-BC ozone of 714 *ppbv* would be too high in the troposphere and would impractically bring high ozone to surface level, where as the maximum Tropo-BC ozone of 205 *ppbv* has fallen within a reasonable value in the United States. It should be noted that the Considine's data is concentration at higher latitudinal locations. With the direct proportional relationship between latitudinal location and tropopause ozone concentration, we would expect that the reported 235 *ppbv* should be a high end of the ozone concentration at the tropopause in the United States.

As tests of the lateral boundary conditions' responses to the GEOS2CMAQ linkage tool, we have extracted the vertical profiles of various CMAQ boundary conditions for selected months to investigate the seasonal effects of the data. Figure 3.7 shows average monthly ozone vertical distribution from all four boundaries of the CONUS domain: East, West, South, and North are shown in various colors with average vertical temperature profiles for January, June, and July. January represents the winter condition where tropopause is relatively low as a consequence of cold temperatures; July characterizes the summer condition with possible high surface ozone concentration. The additional month of June is selected because we have occasionally observed high effects of stratospheric ozone to the surface ozone from the MISC-ASIA study (Fu et al., 2008a). As expected, Profile-BC on the left has shown no seasonal variation. In contrast, the ORDY-BC and Tropo-BC are both showing a seasonal dependence. The ORDY-BC in the middle panel has shown a strong seasonal difference at the top CMAQ layer (i.e., blue line). This dependence directly relates to the seasonal difference in ozone tropopause heights as a result of temperature differences. In the ORDY-BC interpolating process, the amount of stratospheric ozone included in boundary conditions is governed by the altitude of ozone tropopause. It is highly sensitive with elevation because ozone is exponentially increased with altitude beyond tropopause or at stratosphere. The vertical structures of CMAQ and GEOS-Chem are also playing an important role. With the constant elevations in CMAQ layers, the higher the tropopause is located, the less stratospheric effect will result. As shown in Figure 3.7, the monthly average ozone concentrations for ORDY-BC on North bound at the top CMAQ layer for January, June, and July are 362 *ppbv*, 207 *ppbv*, and 172 *ppbv*, respectively.

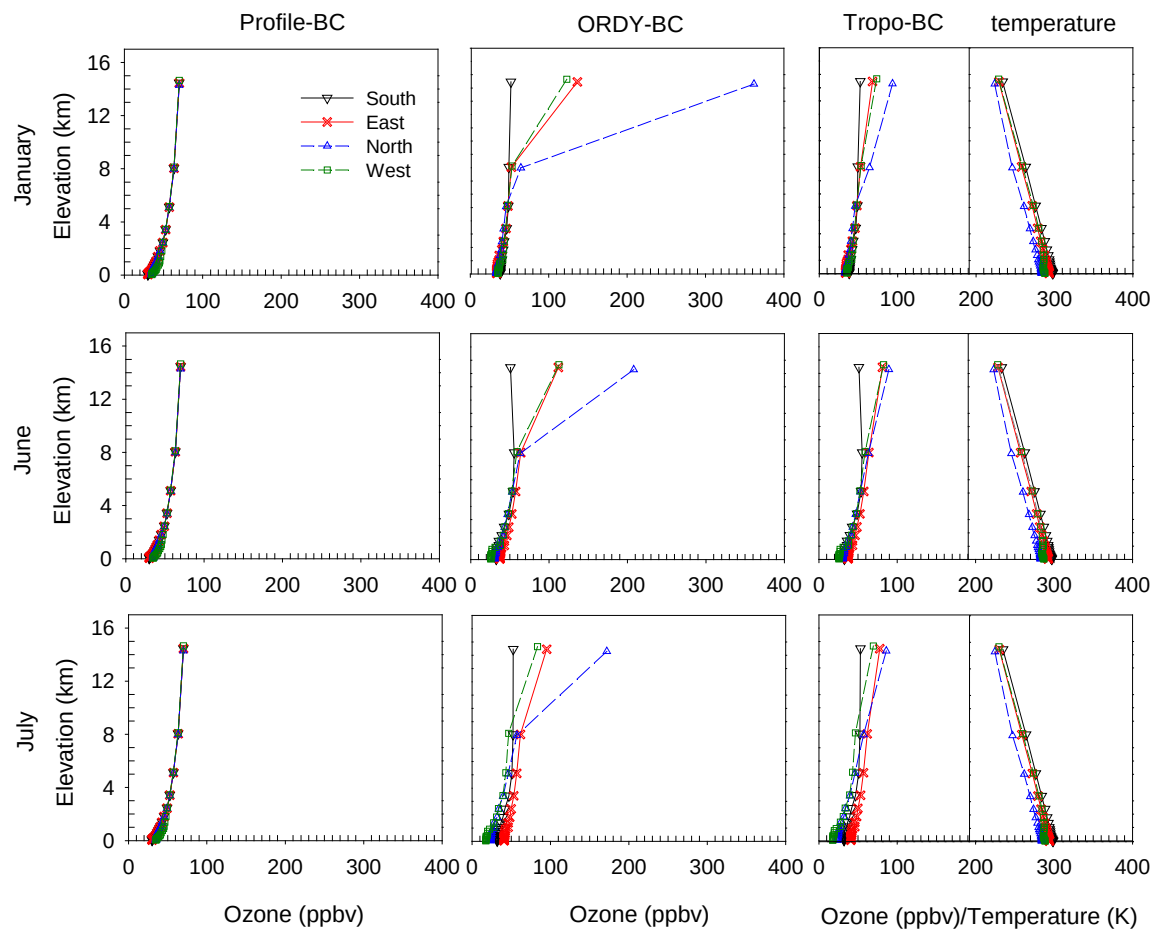


Figure 3.7. Monthly vertical distribution of ozone from CMAQ BCs: South (black line), East (Red line), North (blue line), and West (green line) of CONUS domain in January, June and July with temperature profiles for Profile-BC (left), ORDY-BC (middle) and Tropo-BC (right).

As recalled from early comparisons with Considine (2008), this average concentration in January is too high. For Tropo-BC, shown on the right panel, little seasonal variation is observed at the top CMAQ layer. The average monthly ozone concentrations of 94 ppbv, 90 ppbv, and 86 ppbv are found on the North bound for January, June, and July, respectively. These results demonstrate the effects of tropopause-determining algorithm, which have limited the stratospheric effects from the BCs.

In addition to the seasonal effect, latitudinal effect is also observed in Figure 3.7, where South bound (i.e., downward triangle in black) has the lowest concentration and the North bound (i.e., upward triangle in blue) exhibits the highest concentration at the upper CMAQ layers (top two layers) on both ORDY-BC and Tropo-BC. The latitudinal effect is mainly induced by the temperature differences at troposphere on different boundaries. The vertical temperature profile in CMAQ on the right shows a decrease in temperature with increase in elevation; no temperature inversion is observed. This indicates all CMAQ layers have fallen within the troposphere because it illustrates the pattern of tropospheric lapse rate.

3.4.2 CMAQ Outputs

The CMAQ model was used to simulate the surface ozone concentrations in 36 km CONUS domain using Profile-BC, ORDY-BC and Tropo-BC with VISTAS emissions inventories (Morris et al., 2006b). Figure 3.8 shows the CMAQ simulated vertical distribution of monthly ozone in Boulder, CO, Huntsville, AL, and Trinidad head, CA with available ozonesonde for the months of January, June, and July. In the plot, the elevation is taken from the mid-point of each CMAQ layer. It should be noted that CMAQ is a tropospheric model. Therefore, the maximum concentration of ozone should not exceed the reported maximum tropopause concentration of 235 ppbv. It is observed that ORDY-BC (i.e., in the black triangle) overestimates the January ozone concentrations for all locations in all altitudes (i.e., top panels). Moreover, overestimations are also observed in June at Boulder (i.e., middle left panel) and Trinidad Head at upper altitude (i.e., middle right panel). The overestimations in ORDY-BC mainly resulted from bad lateral boundary conditions (i.e., unreasonable ozone concentration at the troposphere) propagated through the downscaling process.

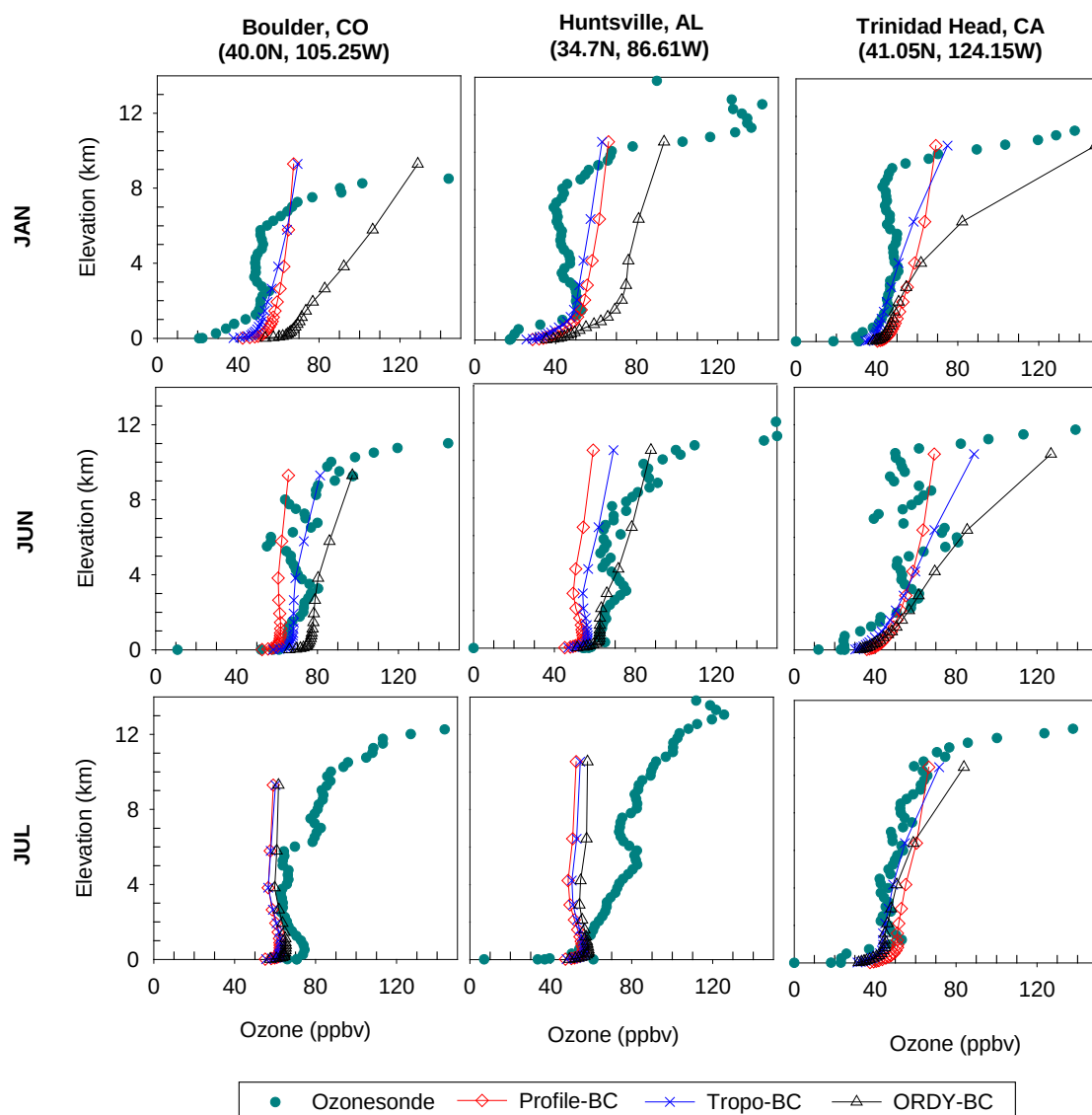


Figure 3.8. CMAQ simulated monthly vertical distribution of ozone for Profile-BC (red line), Tropo-BC (blue line) and ORDY-BC (black line) with ozonesonde.

By removing stratospheric ozone from ORDY-BC, which is demonstrated by Tropo-BC, CMAQ outputs have shown a much better result when compared with ozonesonde. For Profile-BC, similar results as Tropo-BC are observed; slight extra overestimations are found in January and slight extra underestimations are found in June and July when compared with Tropo-BC. Overall, Tropo-BC shows the best agreement with ozonesonde data. It should be noted that the underestimations in Huntsville in July are unrelated to the selection of lateral BCs since very little differences are observed among different lateral BCs. The underestimations in here demonstrate once again that the CMAQ model is incapable of simulating the upper ozone concentration in the area where a large change of upper ozone concentration occurred. We believe that this may be resolved if CMAQ can implement the STE mechanism along with supplementary upper boundary condition from GCM.

Figure 3.9 and 3.10, respectively, show the outputs of the average monthly surface ozone concentrations and the maximum monthly surface ozone concentrations for January (top frames), June (middle frames), and July (bottom frames). The maximum ozone concentrations within the domain are also listed at the corner and denoted in blue or white. In Figure 3.9, the output results show that similar ozone concentration patterns are found across the CONUS domain among all three BCs with some exceptional high ozone being observed in the ORDY-BC. It is believed that these high ozone concentrations occurring in the western United States in ORDY-BC are the consequence of high ozone observed at the top layer of CMAQ boundaries discussed earlier. The undesirable boundary conditions (i.e., ORDY-BC) produce abnormal surface ozone concentrations for both January and June. Since ozone is a photochemical pollutant driven by NO_x, VOCs, and temperature, we would expect higher monthly average ozone should be observed in July rather than in January. In the top frames, the reported maximum average ozone concentrations in January for Profile-BC, ORDY-BC, and Tropo-BC are 55 ppbv, 69 ppbv, and 50 ppbv, respectively. A similar trend is observed for June. For July (bottom frames), the effects of stratospheric ozone in ORDY-BC become minimal due to the fact that the tropopause is much higher than other months at the top layer. As a result, fewer differences are found among these three scenarios.

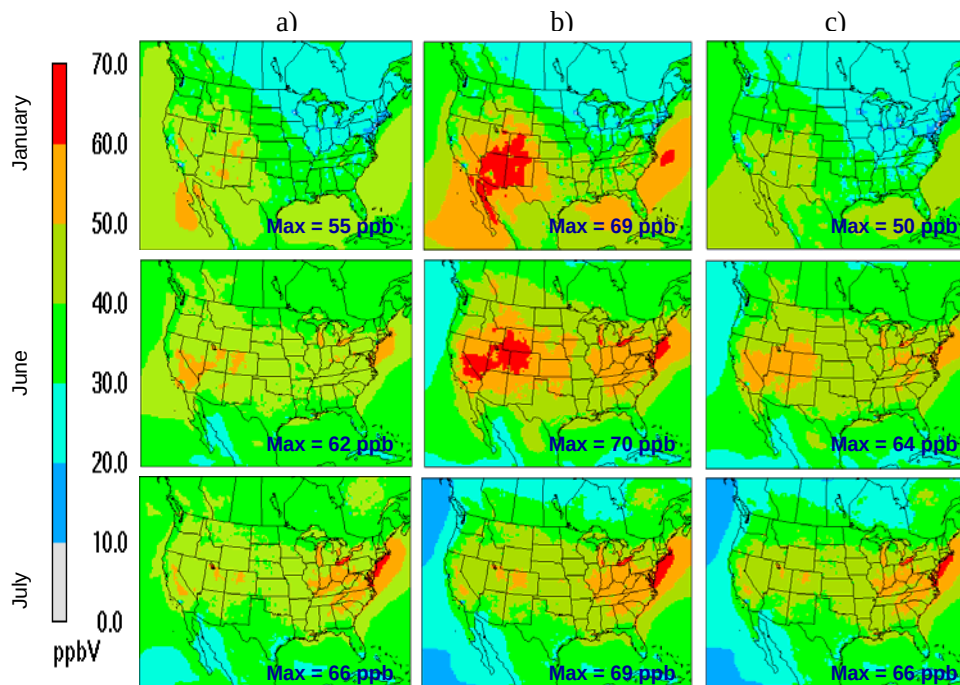


Figure 3.9. Comparisons of monthly average ozone concentrations in January, June and July from CMAQ outputs; a) Profile-BC (left), b) ORDY-BC (middle), and c) Tropo-BC (right). The maximum concentration within the domain is shown at the bottom of right hand corner.

Figure 3.10 shows that the monthly maximum 8-hour ozone concentration in January on ORDY-BC is in excess of 150 *ppbv* over the western United States. The result indicates that the effect of stratospheric ozone in lateral boundary conditions has a significant impact on surface ozone concentrations, as a result of the high ozone aloft mixing downward quickly. The large differences observed between ORDY-BC and Profile-BC/Tropo-BC reveal an important message, which is “excluding stratospheric ozone on tropospheric model during the downscaling process is extremely important. We have found the concentration differences between these scenarios could be as much as 87 *ppbv* in January. These differences gradually decrease with temperature increasing through June and July. The effects of lateral BCs in ORDY-BC have contributed to the high surface concentrations observed in the western United States in January and June. Since both ORDY-BC and Tropo-BC utilize a dynamic algorithm to interpolate the vertical ozone profile for each horizontal grid for lateral BCs, the variations in the western boundary are observed primarily due to the treatments of stratospheric ozone.

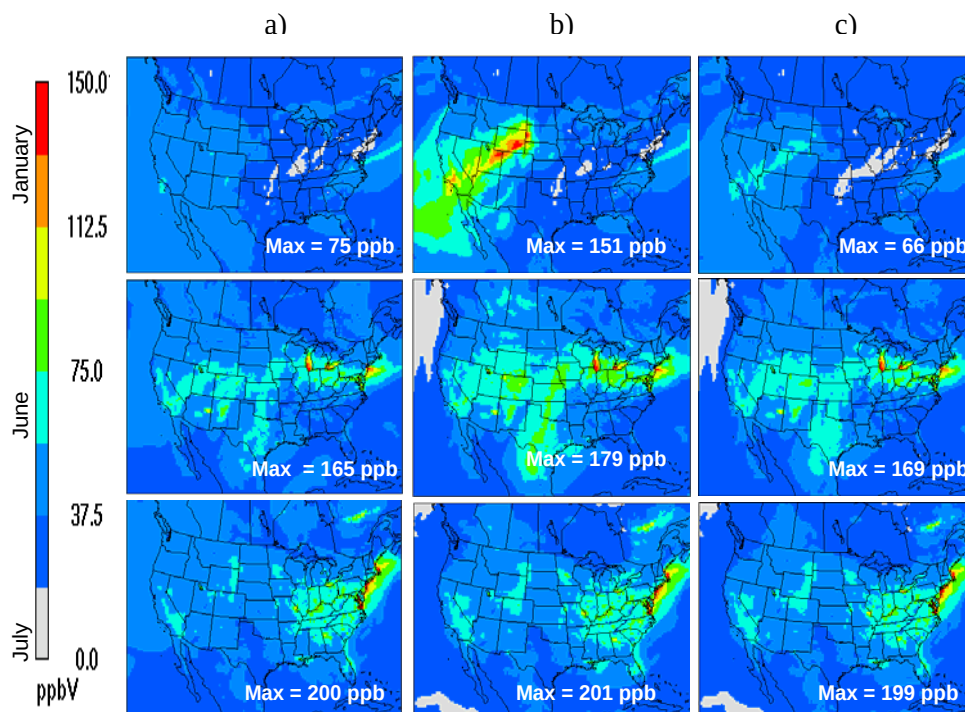


Figure 3.10. Comparisons of monthly maximum 8-hour surface ozone concentrations in January, June and July from CMAQ outputs; a) Profile-BC (left), b) ORDY-BC (middle), and c) Tropo-BC (right). The maximum concentration within the domain is shown at the bottom of right hand corner.

Note that the Tropo-BC is intended to demonstrate the effectiveness of the tropopause-determining algorithm of separating the stratospheric and tropospheric ozone for the lateral boundary condition.

3.4.3 CMAQ Performance Analyses

Model performance analyses on all three cases have been performed using the entire Clean Air Status and Trends Network (CASTNET) dataset, in which 70+ observation sites across the CONUS domain from both EPA and the National Park Service (NPS) are included. It should be noted that our study only simulates the 36 km domain and it is intended to demonstrate the effects of different BCs. Hence, the results in root mean square error in this research may be higher than the one in a finer resolution CMAQ. Figure 3.11 shows the simulated and measured surface ozone for the months of January, June, and July at the nearest locations of the ozonesonde sites found in CASTNET network (See Figure 3.3

denoted in red star). In the plot, blue, purple, green, and red colors correspond to observation, Profile-BC, ORDY-BC, and Tropo-BC, respectively. And the top, middle, and bottom panels show the first 15 day's outputs for January, June, and July, respectively. It should be noted that, due to limitation of the size of the plot, we have only documented the first 15 days of data in Figure 3.11. However, our analyses are based on a full month of data. The quoted number below each point represents root mean square error (RMSE) for each case, with the same color scheme used on the plot.

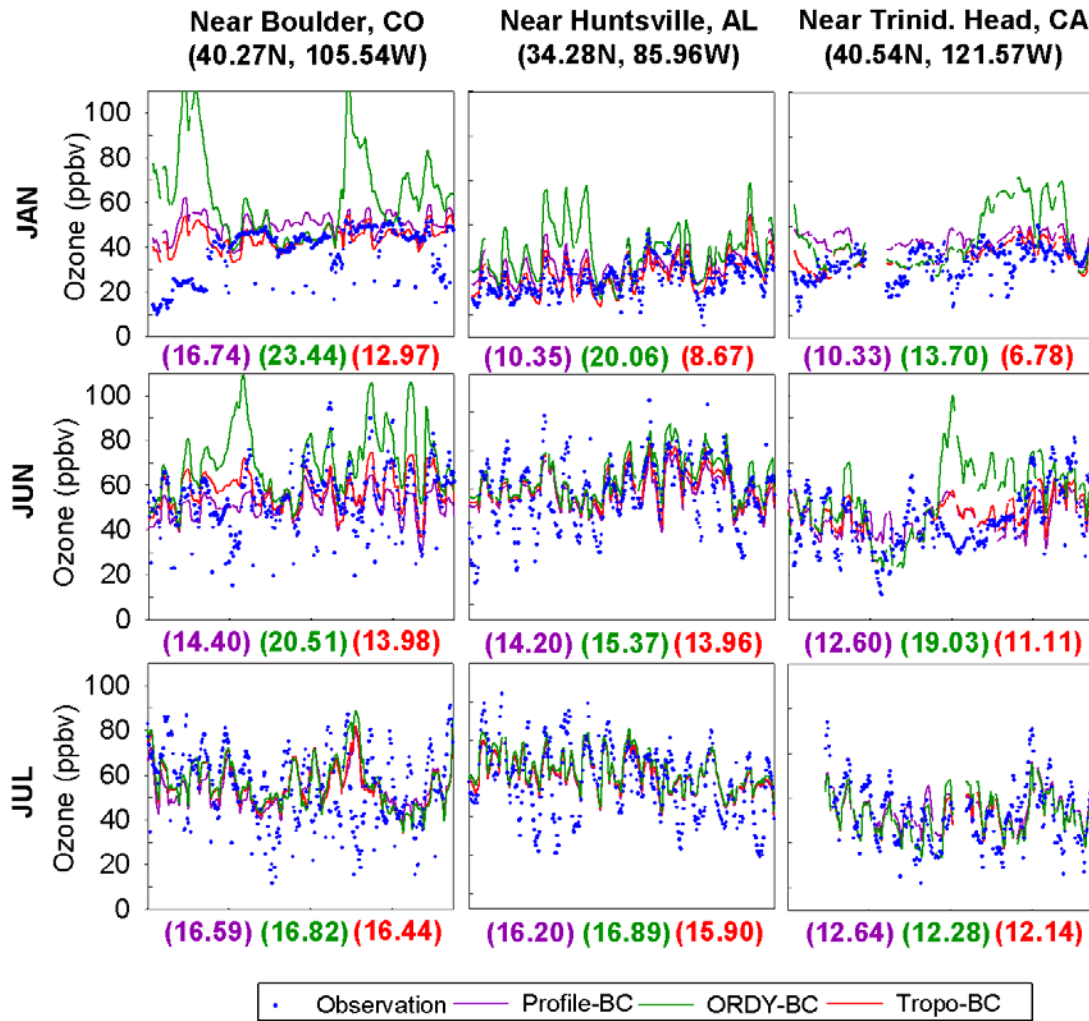


Figure 3.11. Comparison of simulated and measured surface ozone concentration for month of January, June and July from the selected sites. The quoted value at the bottom of each plot reveals the root mean square error of each case.

3.4.4 ORDY-BC

In these time series plots, model results indicate the surface ozone in ORDY-BC has over predicted in January and June (i.e., top and middle panels) and it is in agreement with our results early in Figure 3.10. In comparisons of RMSE, ORDY-BC has shown the worst prediction of surface ozone comparing with others. The RMSE reaches as much as 23.0 ppbv. The highest RMSE occurs at the conditions where the tropopause is low in January and at “near Boulder” site (top left panel). This large RMSE strongly ties to the parameters such as air temperature, altitudinal, and latitudinal locations. Since “near Boulder” is located much higher in altitude (i.e., Boulder at about 1650 m above mean sea level) than Huntsville and Trinidad head, the larger amount and quicker downshift of uncontrolled stratospheric ozone is expected at the surface of ORDY-BC. This did not happen in Profile-BC and Tropo-BC since both of them do not contain any stratospheric ozone. For air temperature, January has much lower air temperature than June and July. With the relationship of air temperature, it is directly proportional to tropopause height; lower air temperature means a lower tropopause height. Therefore, a larger amount of aloft ozone is included in the lateral boundary condition of ORDY-BC and results from a huge over prediction of surface ozone in “near Boulder”. This low temperature effect has also contributed to the high RMSE found in “near Huntsville” and “near Trinidad head” sites in January.

Another high RMSE(s) is found in “near Boulder” and “near Trinidad head” in June. These high RMSE(s) most likely relate to the low tropopause height resulting from low air temperature. We believe latitudinal location might explain why “near Boulder” and “near Trinidad head” observed high RMSE, where as “near Huntsville” did not. In general, the higher latitudinal location is, the lower temperature will be when it is further away from the equator. The low temperature condition affects the downscaling process by changing the tropopause height and resulting in more stratospheric ozone in the lateral boundary conditions in ORDY-BC. To demonstrate the effect of tropopause due to air temperature and latitudinal location, we calculated the RMSE in all CASTNET sites for each boundary condition. Moreover, we subtracted the RSME in ORDY-BC to the RSME in Tropo-BC to yield a net RSME to account for stratospheric ozone effect, denoted as NET-RSME. Note

that the difference between ORDY-BC and Tropo-BC is the extra stratospheric concentrations from GEOS-Chem. Therefore, we use the differences in RMSE as an indicator for stratospheric effects on surface ozone performance. Multivariate statistical fitting is performed on NET-RMSE with monthly average column temperature and latitudinal location. Figure 3.12 shows the results from statistical analyses: (a) multivariate fitting for NET-RMSE on each month, (b) sensitivity analysis on multivariate fitting for the month of June. Note that the equations on top of Figure 3.12 (a) are the best-fit equations for temperature and latitude. These equations are used to generate the NET-RMSE predicted in Figure 3.12 (a) and they do not represent the best-fit equations for the straight lines shown in Figure 3.12 (a). For January, NET-RMSE is highly correlated with latitudinal location and air temperature with R^2 of 0.73 and RMSE of 2.73. For June, only air temperature is correlated to NET-RMSE with R^2 of 0.3. And for July, no correlation is found on either latitudinal location or air temperature. Since NET-RMSE is an indicator of the stratospheric effect from the lateral BCs, we believed that no correlation observed in July implies the average column air temperature has reached a certain level at which tropopause height is higher than the upper boundary of CMAQ. Thus, no stratospheric ozone is included in the lateral BCs. To determine the temperature at which there is no stratospheric effect, we have performed sensitive fittings on June's data because it contains both stratospheric effect sites and non-stratospheric effect sites. Figure 3.12b shows the results of the sensitive test and the observed break point temperature is about 252K, at which the lowest RMSE and the highest R^2 are obtained. These results are consistent with our early explanations of why bad predictions of ORDY-BC occurred in January and June and similar predictions as Tropo-BC are found in July. Table 3.2 shows the monthly average column temperature along with NET-RMSE in all three ozonesonde sites for all months. For January, all three sites have the average temperature lower than 252K. Therefore, a large NET-RMSE caused from stratospheric ozone is expected. For June, Boulder and Trinidad head are equal or below 252K, where as Huntsville is above 252K. Hence, a large NET-RMSE(s) is observed in those two sites and a small NET-RMSE is found in Huntsville. These results are in agreement with our conclusions made earlier on the time-series plots in Figure 3.11.

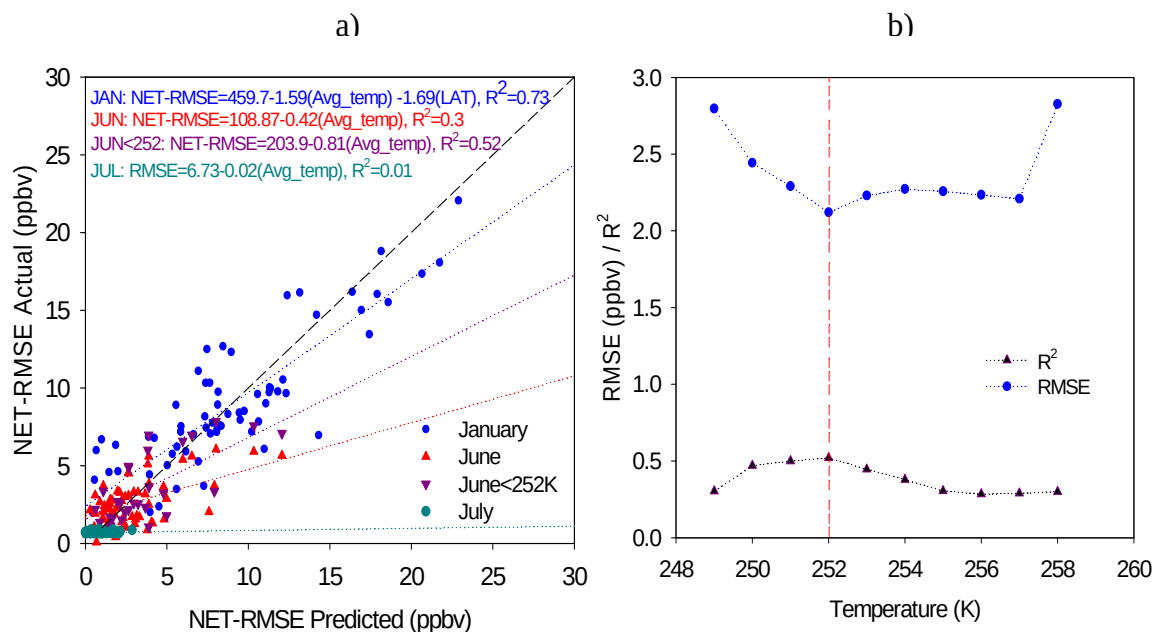


Figure 3.12. Statistical analysis outputs from CASTNET sites: a) NET-RMSE actual Vs. NET-RMSE predicted, b) sensitivity analysis on best-fit equation for June data.

Table 3.2. Summary of NET-RMSE and average column temperatures for the sonde sites.

	Boulder, CO	Huntsville, AL	Trinidad head, CA
JANUARY	Tc = 236 K NET-RMSE = 10.5 ppbv	Tc = 246 K NET-RMSE = 11.4 ppbv	Tc = 242 K NET-RMSE = 6.9 ppbv
JUNE	Tc = 247 K NET-RMSE = 6.5 ppbv	Tc = 254 K NET-RMSE = 1.4 ppbv	Tc = 252 K NET-RMSE = 7.9 ppbv
JULY	Tc = 253 K NET-RMSE = 0.39 ppbv	Tc = 257 K NET-RMSE = 1.0 ppbv	Tc = 255 K NET-RMSE = 0.1 ppbv

Tc is average vertical column temperature; NET-RMSE is the RMSE differences between Profile-BC and Tropo-BC

Overall, these results stress the important relationship of temperature and seasonal changes in the GCM downscaling process.

3.4.5 Profile-BC

For Profile-BC versus lateral boundary conditions from GCTM, Tang et al. (2007), have found that the performance of boundary conditions from GCTM may not necessarily be better than Profile-BC. Moreover, different GCTM outputs also yield different results. The performance of lateral boundary conditions from GCTM (GCTM-LBC) highly depends on locations and scenarios of the GCTM-LBC, also the type of GCTM used. Al-Saadi et al. (2007), suggested that this phenomenon might relate to the ozone aloft in GCTM-LBC, where rapid transports of stratospheric ozone into the surface level are observed. In addition, they have found that GCTM-LBC enhances the model errors of ozone concentration at the surface in the range of 6 to 20 ppbv in Trinidad Head in August.

Since these studies have selected the summer ozone season (i.e., August) as their study period, we expected that the effect of stratospheric ozone would be minimal based on the relationship we developed earlier. However, this did not happen. In this case, we suspect their average column temperature in August for Trinidad head may not be hot enough to exclude the stratospheric ozone from the GCTM-LBC interpolating process, or it may be affected by the quality of GCTM-LBC as inputs where strong boundary influx of ozone affects the simulation results. Nevertheless, these studies have indicated that GCTM-LBC preprocessing may be required. In our study, we have implemented the tropopause-determining algorithm, which is based on chemical tropopause definition, as the preprocessor for generating ORDY-BC and denoted as Tropo-BC. Note that ORDY-BC is one kind of GCTM-LBC. The intention of the tropopause algorithm is an attempt to improve the ozone simulation at the surface.

Figure 3.11 shows the RMSE for both Profile-BC and Tropo-BC. The results show that the RMSE in Profile-BC is always higher than the RMSE in Tropo-BC, whereas the ORDY-BC have either greater or less than Profile BC depending on the locations. Although the differences between Profile-BC and Tropo-BC in RMSE was found to be within 1 to 2 ppbv in June and July, and 3 to 4 ppbv in January, the results have demonstrated the

tropopause-determining algorithm has successfully prevented the high surface ozone estimates, which Tang et al. (2007) and Al-Saadi et. al. (2007) mentioned in their study.

3.4.6 Tropo-BC

For overall performance of Tropo-BC, we have included additional statistical analyses using all CASTNET data. Table 3.3 shows the summary of RMSE and mean bias (MB) for all three BCs. In the table, the entire United States have been broken down into three regions, which are West Coast (West), Central United States (Central), and East Coast (East). The average RMSE for all three months in all stations is calculated to be 14.2 ppbv, 13.3 ppbv, and 17.6 ppbv for Profile-BC, Tropo-BC, and ORDY-BC, respectively. We observed that the RMSE in Tropo-BC is always lower than both Profile-BC and ORDY-BC for every region and every month.

Table 3.3. Summary of NET-RMSE and average column temperatures for the sonde sites.

		Profile-BC	Tropo-BC	ORDY-BC
JANUARY	ALL	RMSE = 11.9 ppbv MB = 7.3 ppbv	RMSE = 10.3 ppbv MB = 3.9 ppbv	RMSE = 19.8 ppbv MB = 13.2 ppbv
	WEST	RMSE = 16.8 ppbv MB = 14.6 ppbv	RMSE = 13.0 ppbv MB = 9.8 ppbv	RMSE = 23.5 ppbv MB = 18.3 ppbv
	CENTRAL	RMSE = 10.1 ppbv MB = 6.6 ppbv	RMSE = 8.2 ppbv MB = 2.4 ppbv	RMSE = 23.6 ppbv MB = 16.1 ppbv
	EAST	RMSE = 11.2 ppbv MB = 6.3 ppbv	RMSE = 10.1 ppbv MB = 3.2 ppbv	RMSE = 18.0 ppbv MB = 11.5 ppbv
JUNE	ALL	RMSE = 14.3 ppbv MB = 0.3 ppbv	RMSE = 13.8 ppbv MB = 1.9 ppbv	RMSE = 16.4 ppbv MB = 7.2 ppbv
	WEST	RMSE = 18.3 ppbv MB = 4.3 ppbv	RMSE = 15.2 ppbv MB = 2.0 ppbv	RMSE = 19.9 ppbv MB = 7.2 ppbv
	CENTRAL	RMSE = 12.5 ppbv MB = -4.5 ppbv	RMSE = 11.3 ppbv MB = -1.3 ppbv	RMSE = 16.0 ppbv MB = 6.1 ppbv
	EAST	RMSE = 14.1 ppbv MB = 1.1 ppbv	RMSE = 14.1 ppbv MB = 2.9 ppbv	RMSE = 15.9 ppbv MB = 7.6 ppbv
JULY	ALL	RMSE = 16.3 ppbv MB = 4.2 ppbv	RMSE = 15.8 ppbv MB = 3.4 ppbv	RMSE = 16.6 ppbv MB = 5.3 ppbv
	WEST	RMSE = 19.8 ppbv MB = 4.3 ppbv	RMSE = 16.9 ppbv MB = 4.1 ppbv	RMSE = 16.9 ppbv MB = 6.0 ppbv
	CENTRAL	RMSE = 13.7 ppbv MB = -2.4 ppbv	RMSE = 13.3 ppbv MB = -3.1 ppbv	RMSE = 13.7 ppbv MB = -1.4 ppbv
	EAST	RMSE = 16.4 ppbv MB = 6.2 ppbv	RMSE = 16.3 ppbv MB = 6.1 ppbv	RMSE = 17.3 ppbv MB = 8.1 ppbv

ALL - All stations; WEST - West of 115W; CENTRAL - Between 115W and 94W; EAST - East of 94W

RMSE is root mean square error; MB is mean bias

This demonstrates the Tropo-BC is the best method of generating lateral boundary condition for CMAQ. In the table, large differences (i.e., average in 3 ppbv) between Tropo-BC and Profile-BC are observed in the “West”. It should be noted that this large RMSE improvement in the “West” was mainly contributed by the sites that are located in Washington. The magnitude of changing RMSE in Washington ranges from 4 to 12 ppbv. The poor performance of Profile-BC in RMSE in the “West” has shown that Profile-BC has failed to estimate the impact from intercontinental transport of air pollutants from East Asia across the Pacific Ocean. Moreover, it fails to represent the actual geospatial variations of lateral boundary in the United States.

For the performance of Tropo-BC in all other regions, minor improvement is observed when compared with Profile-BC. Large improvement is found in month of January. Since Profile-BC uses a fixed BC concentration and this fixed BC concentration is usually higher than the actual background ozone in winter, as a result, overestimation of surface ozone in Profile-BC is observed. This demonstrates the importance of using dynamic BCs instead of the static BCs. Figure 3.13 shows the distributions of RMSE differences among these three scenarios for each of the CASTNET sites. If we consider ± 1 ppbv as model variability, then we conclude that only 5% or less of the sites in Tropo-BC have poorer performance compared with Profile-BC. In these 5% of the sites, we have observed the Tropo-BC overestimated the nighttime ozone concentration in June. In comparison with ORDY-BC, Tropo-BC is outperformed for every observation site in January. Strong improvement in Tropo-BC is found in both January and June. In the plot, we have observed 10% or less of the sites in Tropo-BC have poorer performance than in ORDY-BC (i.e., right side panel). We believed that this 10% is contributed by the nature of underestimation of ozone in 36 km resolution. Since the surface ozone in ORDY-BC is always higher than in Tropo-BC, the improvement may not actually be counted. For the overall performance, Tropo-BC has outperformed ORDY-BC in every month for all regions. These results, once again, demonstrate that the removal of stratospheric ozone using our tropopause-determining algorithm strongly improves the performance of surface ozone simulations in CMAQ.

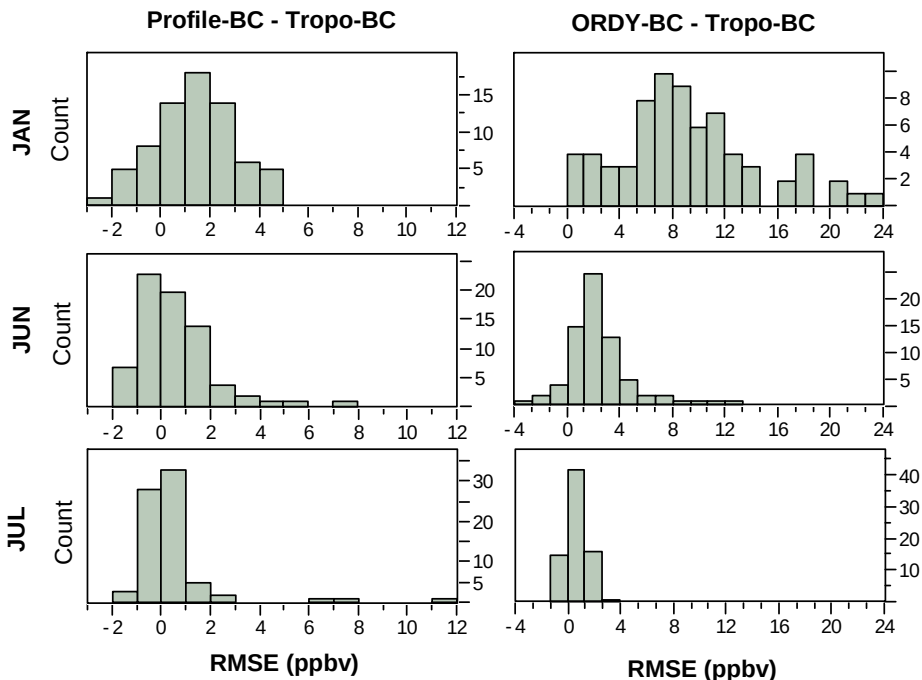


Figure 3.13. Summary of the RMSE distributions for the differences among these three scenarios for each CASTNET sites.

3.5 Conclusion

In this study, we have successfully integrated our newly developed tropopause-determining algorithm, based on chemical tropopause definition, into the methodology of downscaling from the global chemical model (i.e., GEOS-Chem) into the regional air quality model (i.e., CMAQ). The purpose of the algorithm is to resolve the inconsistency of vertical structures between GEOS-Chem (i.e., containing both the tropospheric and stratospheric components) and CMAQ (containing only the tropospheric component). It identifies the height of tropopause from GCTM outputs and applies tropopause ozone concentration as the maximum ozone concentration at the CMAQ lateral boundary condition. As a result, it excludes any stratospheric ozone from being included in the regional air quality model. Since CMAQ is only designed for tropospheric application with no top boundary input, any stratospheric ozone or stratospheric intrusion should be considered inapplicable in CMAQ. In our results, we have found that the GCTM output (i.e., GEOS-Chem) with the tropopause-determining algorithm (i.e., Tropo-BC) always yields a better result than that with the fixed BCs (i.e., Profile-BC). Moreover, Tropo-BC also yields better results than that with the

GCM BCs (i.e., ORDY-BC). For Profile-BC, we have observed the fixed BCs tend to overestimate surface ozone concentration during wintertime and underestimate in summertime. For ORDY-BC, strong over prediction of surface ozone is observed as a result of stratospheric ozone from the upper atmosphere. These results are similar to the findings in Tang et al., where a large overestimation is observed in CMAQ surface ozone when applying GCTM-BC. Fortunately, using our new tropopause algorithm technique (i.e., Tropo-BC) with the global model input (i.e., GEOS-Chem), we have resolved the high surface ozone issue observed in GCTM-BC, while maintaining good vertical ozone prediction in the upper air. For further improving the model simulations, we recommended that all vertical layers from MM5 (i.e., 34 layers) should be used in CMAQ, instead of 19 layers created from vertical collapsing. This way, it will break down the original CMAQ top layer into 5 separated layers with a thickness of 1.0 to 1.5 km for vertical transport. It is believed that the top CMAQ layer (i.e., 6 km deep) is relatively too thick; it may give a wrong representation of transport of flux in the upper troposphere.

In statistical analysis, we have performed a correlation study on the average tropospheric column temperature and stratospheric effect using the RMSE differences between ORDY-BC and Tropo-BC. The results show that a break point temperature, which separates the temperature region between stratospheric effect and non-stratospheric effect in the chemical downscaling process, is about 252 K. This value can be used as a quick check to see whether or not a particular region or day in the regional model is having a stratospheric effect from GCTM-BC. Nevertheless, this temperature is based on statistical analysis and may contain certain statistical errors. Therefore, we recommend only using this value as a screening tool.

In conclusion, we have demonstrated the advantage of using the tropopause-determining algorithm along with time-varying GCTM lateral BC for air quality predictions of the tropospheric ozone. We have advanced the existing technique on how GCTM data can be incorporated into CMAQ lateral BC. This methodology can be applied on different GCTM data for downscaling purposes to yield a better surface ozone prediction in a regional CTM.

CHAPTER IV

4 IMPACTS OF FUTURE CLIMATE CHANGE AND EFFECTS OF BIOGENIC EMISSIONS ON SURFACE OZONE AND PARTICULATE MATTER CONCENTRATIONS IN THE UNITED STATES

4.1 Declaration

This chapter is a slightly revised version of a paper with the same title, which will be submitted to Atmospheric Chemistry and Physics by Yun F. Lam, Joshua S. Fu, Shiliang Wu, Loretta J. Mickley, and Daniel J. Jacob.

4.2 Introduction

Properly representing the transport and chemical transformation of air pollutants has always been one of the greatest challenges of simulating regional air quality in global climate/chemistry models. The accuracy of the results strongly depends on the selection of grid resolution (i.e., usually $\approx 1^\circ \times 1^\circ$ or large), land use information, emissions input and temporal resolution (i.e., 3-hr) (Chin et al., 2007; Civerolo et al., 2007; Ito et al., 2009; Knutti et al., 2008; Wu et al., 2007; Fiore et al., 2005). It has been observed that the coarse resolution used by global models may not be sufficient to represent appropriate meteorological characteristics of some regions (i.e., complex terrain regions) because of over simplifying the vertical grid structure and land use information in the models. This issue has been reported in the regional model, as well when a relatively coarse resolution (i.e., 36 km) is used (Arunachalam et al., 2006; Kim et al., 2010). Nevertheless, global models have been widely recognized as a practical tool for predicting long-term climate and air quality trends, evaluating intercontinental long-range transport of air pollutants and large-scale climate and air quality impact studies, such as those on dust storms and the stratospheric ozone hole (Wu et al., 2008a; Chin et al., 2007; Vingarzan, 2004). To integrate useful information from global models into regional-scale models, downscaled global climate and chemistry outputs have been developed in recent years for resolving the issue of insufficient temporal and spatial resolutions. Various air quality studies have implemented the downscaling methodology for evaluating the influence of climate change, land-use modification, and different emissions

projection scenarios on both anthropogenic and biogenic emissions on the regional-scale in the United States (Civerolo et al., 2007; Xiaoyan et al., 2008; Jacobson and Streets, 2009; Zhang et al., 2008).

In the area of regional climate change and air quality, a wide range of temperature and ozone concentration changes have been reported from both global and regional model studies in the United States. Depending on the type of model used and emissions projection scenarios (i.e., the IPCC A1B) selected, the projected future ozone concentrations vary greatly. In the global model perspective, Wu et al., have predicted a 1–3°C increase of temperature in 2050, which would result in an extra 2–5 ppbv of surface ozone in the Northeast and the Mid-north of the United States and a reduction of ozone in the Southeast United States on a non-emission change scenario using GISS-GCM/GEOS-Chem coupling models (Lam et al., 2008a; Wu et al., 2008a). They also found that the anticipated emissions reductions (40% for NO_x) in the IPCC A1B scenario would have a greater effect (i.e., -2 to -15 ppbv) than the climate change (i.e., +2 to +5 ppbv) on the daily maximum 8-hr ozone. Huang et al., simulated the future air quality in 2048–2052 (summer) using the Model for Ozone And Related chemical Tracers (MOZART) under the IPCC A1Fi (i.e., dirtiness) and the B1 (clearness) scenarios (Ho-Chun et al., 2008). They found that the Southeast United States would have the largest sensitivity of surface ozone in response to the emission changes with +25% to -24% for the A1Fi and B1 scenarios in 2048–2052, while less sensitivity of surface ozone would be shown on the Midwest and the Northeast of the United States and Texas. They suggested that the future United States air quality projected by MOZART is less sensitive to the emissions scenarios simulated by the Regional Air Quality Model (RAQM) in those locations. It is doubtful that they have found the trend of surface ozone mixing ratio from MOZART is consistently higher than the RAQM, which is unlikely to occur at a coarse grid resolution. They commented that the overestimation of ozone in MOZART is caused by over-estimation of anthropogenic emissions. Their study revives the erroneous notion that consistent emissions input between the global and the regional models should be used when model comparisons are performed.

In the regional model perspective, the downscaled results on the impacts of climate change have also varied largely across the geospatial regions. Some studies have found

climate change has large adverse effects on the future air quality. The large increase of temperature (i.e., 1–2° K) has encouraged the formation of ozone and resulted in an extra 5–10 ppbv compared to the current air quality condition. (Bell et al., 2007; Nolte et al., 2008) Dawson et al., found that the sensitivity of temperature change in the regional O₃ averages 0.34 ppbv K⁻¹ (i.e., 1 – 3 ppbv for a 2.5° K increase) for the eastern United States (Dawson et al., 2008). They have also suggested that a 2.5° K increase of temperature leads to a 30% increase of exceedance on the maximum daily average 8-hr (i.e., 80 ppbv) standard. Bell et al., found that the climate change alone contributed an increase of 4.8 ppbv on average ozone across the United States, with the largest increase at 9.6 ppbv, which corresponds to an additional 68% of exceedances in the 8-hr standard in 2050 (Bell et al., 2007). Although the effect of climate change on temperature and stagnant air flow would encourage the formation of ozone, most researchers have found the anticipated emissions reduction from IPCC cases (A1 B1, A1B) in the United States tends to compensate for the effect of climate change on ozone formation with or without considering the positive feedback from Biogenic Volatile Organic Compounds (BVOC) and yields an overall ozone reduction of –4 to –15% in 2050. It is suggested that effects of anthropogenic emissions account for more overall change of ozone formation than the climate change (Tagaris et al., 2007; Jacob and Winner, 2009; Nolte et al., 2008; Zhang et al., 2008).

The accuracy of these studies has been tied strongly to the methodology used for downscaling, the choice of resolution, and selection of projection emission scenarios. It is observed that most of the climate studies mentioned above have used the Biogenic Emissions Inventory System (BEIS) for estimating BVOC emissions (Weaver et al., 2009). While recent studies showing that BEIS may have underestimated isoprene emission compared to the Model of Emissions of Gases and Aerosols from Nature (MEGAN), these studies may also lead to underestimating the effect of climate change in the VOC-limited region, such as in the Northeast region of the United States (Sakulyanontvittaya et al., 2008; Guenther et al., 2006; Fiore et al., 2005; Kunkel et al., 2008). The estimates of BVOC on isoprene emission from MEGAN are about 50% more than the estimates from BEIS (Pouliot, 2008; Pouliot and Pierce, 2009). Therefore, it is possible that the additional BVOC from MEGAN may lead to a large increase of ozone and further alter the relationship found between the effect of

climate change and the effect of change of anthropogenic emissions found in previous studies. Furthermore, the majority of these climate change studies have used grid resolutions of 30 km or larger (i.e., except for Hogrefe et al.,’s group), with recent studies suggesting 12 km resolution may be the better choice for studying regional air quality, the grid resolutions of 30 km or larger may produce an additional bias to climate change studies and may result in underestimation of ozone formation in regional-scale studies (Hogrefe et al., 2007b;Hogrefe et al., 2007a;Kim et al., 2010). Since the sensitivities of scalability and the effects of BVOC on the climate change scenarios have not been studied, revisiting the future air quality in 2050 with the implementation of those concepts is important to further investigate the effect of climate change on a regional scale.

In this study, the global GISS General Circulation Model (GISS GCM III) and GEOS-Chem model outputs were downscaled into regional models under the framework of Global Change and Air Pollution (GCAP) to incorporate the future climate information and chemical boundary conditions (Jacob et al., 2009). We simulated the entire year of 2000 (i.e., present) and 2050 (i.e., future) climate and air quality conditions at a fine resolution (i.e., 12 km) to examine the effect of climate change on regional air quality under various emissions scenarios (i.e., IPCC A1B future projection scenarios with MEGAN and BEIS biogenic emissions scenarios) using the Community Multiscale Air Quality Modeling System (CMAQ). We concentrated on three aspects of climate change/air quality studies: (1) The assessment of air quality impacts on global climate change (i.e., Southeastern United States) using fine resolution CMAQ; Because only a few studies have documented fine resolution results in climate change/air quality studies, it provides additional insight into the effect of model resolution selection to the future climate and air quality predictions; (2) the comparison of global model results (i.e., 4° x 5° resolution) with regional outputs results (i.e., 36 km and 12 km resolution) to identify the discrepancy in the prediction of future regional air quality trends between the global model and the regional model; and (3) the impacts of using different biogenic emissions inventories (i.e., BIES and MEGAN) on the future air quality studies and the role of BVOC in the future climate studies were examined. In the study, we focused on ozone (O₃) and fine particulate (PM_{2.5}) because it possesses significant human health impacts. Eight scenarios are performed in the study, which include various

combinations of emissions scenarios within current and future meteorology. We do not include the impacts of future land cover changes or biomass burning, which is highly uncertain in 2050. It is expected that this study will, overall, give a broader understanding of the discrepancy between global and regional outputs for air quality application in the area of future climate change scenarios.

4.3 Methodology - GCAP Modeling System

The GCAP modeling system consists of four models, spanning from global to regional scale. In the global perspective, the GEOS-Chem modeling system driven by the GISS III GCM/CTM was used to provide global air quality conditions in a coarse resolution. Details of the global chemical and meteorological models implemented in the current study can be found in Schmidt et al. (2006), and Wu et al. (2008). In the regional perspective, the outputs of the GEOS-Chem were downscaled to provide chemical initial and boundary conditions for CMAQ, while the outputs of the GISS GCM/CTM III were used as the inputs for the Penn State/NCAR Mesoscale Model (MM5). Descriptions of the models' setup and emission scenarios are discussed below.

4.3.1 Global Models

The GEOS-Chem CTM driven by meteorological fields from the GISS GCM III (an updated version of the model described by Rind et al., (Rind et al., 1999)) was used to simulate the present and future air quality in the United States. The GISS GCM III simulation was initialized on 1 June 1950 and continuously simulated to the end of 2050. The resolution of $4^{\circ} \times 5^{\circ}$ with 23 vertical layers extending from the surface to 0.002 hPa (up to 85 km in altitude) was used to simulate this transient climate event (Rind et al., 2007). For meteorological consistency, the same temporal resolution was used in the GEOS-Chem CTM with 3-hour meteorological data of mixing depths and surface variables (i.e., temperature; winds; precipitation and albedo; and solar radiation) and 6-hour meteorological data (i.e., winds, convective mass fluxes, temperature, humidity, cloud optical depths, and cloud fractions) generated from the GISS GCM III. In this study, GEOS-Chem (version 7.03.06) was used, which includes a fully coupled treatment of tropospheric ozone-NO_x-VOC

chemistry and aerosols to simulate both O₃ and PM_{2.5} concentrations in the United States (Park et al., 2004). Four scenarios were evaluated: (1) present meteorology with current emissions, (2) present meteorology with future emissions, (3) future meteorology with current emissions, and (4) future meteorology with future emissions. The descriptions of the forecasted future emissions were described in the emissions section. Full-year simulations were conducted for year 2000 and year 2050, which were initialized on 1 September, 1999 and 2049 and continued for 16 months for each year. The first four months served for proper initialization, and the following 12 months were used as the actual simulation results. Details of the global models' setup were described by Wu et al. (2007).

4.3.2 Regional Models

The initial and boundaries conditions of the regional models (both MM5 and CMAQ) were downscaled from the outputs of the global models' simulations (GISS GCM III and GEOS-Chem). Details of the downscaling methodology were described in Lam and Fu (2010). The CMAQ was driven by NCAR's 5th generation Mesoscale Model, version 3.7. (MM5) with hourly resolution. The horizontal resolutions of 36 km and 12 km with 34 sigma vertical layers were used. All MM5 simulations were conducted using the one-way nested approach from 108 km over North America (140 - 40W, 10 - 60 N) down to 36 km continental United States (128 - 55W, 21 - 50 N) and eventually down to 12 km VISTAS domain (96 - 71W, 23 - 45 N), as shown in Figure 4.1.



Figure 4.1. The CONUS domain with the selected study areas boxed in red.

For meteorological initial and boundary conditions, the GISS GCM III data with resolution of 4° by 5° was used with the 4-D analysis nudging technique to reproduce the weather conditions similar to the GISS GCM III outputs. The Kain-Fritsch cumulus, Mix-phase micro-physic, RRTM long-wave radiations, Eta planetary boundary layer (PBL) and NOAH land surface model (LSM) were configured in the simulations. A detailed summary of the MM5 configuration is shown in Table 4.1.

For CMAQ, the Lambert conformal projection with true latitude limits of 25 and 40 was used on 148 by 112 grid cells and on 177 by 168 grid cells with horizontal resolution of 36 km and 12 km, respectively. The center of the horizontal domain was set at 100W and 40N. The 36 km domain covers the entire continental United States, and part of Mexico and Canada (referred to as CONUS domain) and the 12 km domain covers all the southeastern states. A total of 14 sigma vertical layers were extracted from MM5 with the lowest model levels centered at approximately 18, 52, 105, 215, 360, 545 m above the surface. For CMAQ simulations, the same scenarios described in the GEOS-Chem, with additional simulations on biogenic emissions (BEIS and MEGAN emissions scenarios), were performed to investigate the sensitivities of climate change from biogenic emissions on a regional scale. All of these simulations were configured with the Carbon Bond IV (CB-IV) chemical mechanism with aerosol module (AERO4) with boundary conditions generated from downscaling GEOS-Chem outputs. The detailed configuration of CMAQ setting is also listed in the Table 4.1.

4.4 Emissions and Simulation Scenarios

4.4.1 Anthropogenic Emissions

The base year for the present-day anthropogenic emission inventories is 2000. These emission files are based on the 1999 EPA's National Emissions Inventories (NEI 1999), 1995 Canadian point sources for Eastern Canada and 2000 Environment Canada (EC) area and mobile inventories (<http://www.ec.gc.ca/inrp-npri>), and the 1999 BRAVO Mexican emission inventory. The emissions through the Sparse Matrix Operator Kernel Emission system (SMOKE 1.4) was processed to generate CMAQ-ready emission files for both 36 and 12 km domains.

Table 4.1. MM5 and CMAQ model configurations for 2000 and 2050 Simulations.

MM5 Configuration	
Model version	3.7
Number of sigma level	34
Number of grid	169 x 133 / 181 x 190
Horizontal resolution	36 km/ 12 km
Map projection	Lambert conformal
FDDA	Analysis nudging
Cumulus	Kain-Fritsch 2
Microphysics	Mix-phase
Radiation	RRTM
PBL	Mellor-Yamada-Janjic (Eta)
LSM	Noah LSM
LULC	USGS 25-Category
CMAQ Configuration	
Model version	4.6
Number of Layer	14
Number of grid	148 x 112 / 177 x 168
Horizontal resolution	36 km / 12 km
Horizontal advection	PPM
Vertical advection	PPM
Aerosol module	AERO4
Aqueous module	CB-IV
Emission	EPA's NEI 1999
Boundary condition	GEOS-Chem*

* corresponding year of GEOS-Chem outputs are used

For future-year anthropogenic emission estimates (i.e., forecasting from 2000 to 2050), we first calculated the future (monthly) projection rates/growth factors based on the IPCC A1B scenario for ozone and aerosol precursors emissions using the integrated Model to Assess the Greenhouse Effect (IMAGE socioeconomic model (Streets et al., 2004)). Afterward, we applied these monthly growth factors to different categories (fossil fuel, biofuel, and biomass burning) of present-day emissions files to generate future-year emissions for each day. It should be noted that the same methodology described above has been applied in both GEOS-Chem and CMAQ to maintain emissions consistency between the global and the regional models. We have estimated the differences in total anthropogenic NO_x and VOC emissions between those two models at the present-year (2000) were at the levels of 10% or less for the continental United States. The calculated total NO_x and VOC emissions are 19.4 and 15.7 Tg/year for the GEOS-Chem and are 21.0 and 17.2 Tg/year for the CMAQ, respectively. Table 4.2 lists the summary of annual anthropogenic emissions growth rate used in this

study. For the ease on model comparisons, the same sub-domain definitions described in Wu et al. (2008) were implemented (see the red box shown in Figure 4.1) (Lam et al., 2008a). The sub-domains include three areas: 1) Northeast, 2) Southeast, and 3) Midwest. The Northeast domain covers all the eastern states from Indiana to the Atlantic coast in east-west direction and from Kentucky to Michigan in south-north direction (87.5 – 67.7 W, 37.2 – 45.7 N). The Southeast domain includes the majorities of the Visibility Improvement State and Tribal Association of the Southeast (VISTAS) states and with a half of Kentucky and West Virginia (97.6 – 73.3W, 29.8 - 37.2N) and the Midwest domain contains all the mid-northern states and up to the middle of Wyoming. (107.4 – 87.5 W, 38.6 – 49.8 N). As shown in Table 4.2, a large increase of acetaldehyde (ALD2) and formaldehyde (FORM) are projected as a result of the increase of the biomass burning in the future. The projection values could be up to 2.5 times higher than the present-day emissions. In contrast, a huge reduction of sulfur dioxide (SO₂) is proposed due to the anticipated future fuel emissions controls. For nitrogen oxide (NO₂), an overall reduction of 60% is forecasted across the Eastern United States, with the largest reductions of 60 to 70% reductions in the fossil fuel combustion sector. It should be noted that these NO₂ reductions in the United States, have been compensated by the increase emissions in Mexico and yield a smaller reduction factor (0.64) for the continental United States domain.

4.4.2 Biogenic Emissions

For biogenic emissions, two emission factor-based models, the Biogenic Emissions Inventory System (BEIS) v3.12 and the Model of Emissions of Gases and Aerosols from Nature. (MEGAN) v2.02 (<http://bai.acd.ucar.edu/Megan/index.shtml>), were used to generate the hourly biogenic emissions inventories for both current and future climate scenarios. Corresponding years (i.e., 2000 or 2050) of temperature and solar radiation data generated from the Meteorology-Chemistry Interface Processor (MCIP) (i.e., a meteorological preprocessor of CMAQ using MM5 outputs) were used to take into account the change of biogenic emissions from natural sources caused by the change in meteorological conditions. We did not consider lightning as a source of NO_x in the upper troposphere for either the present or future climate simulations.

Table 4.2. The annual projection rates of anthropogenic emissions from 2000 to 2050.

CMAQ species	By Category*			By Region			
	FOSSIL FUELS	BIOFUEL	BIOMASS BURNING	Cont. U.S.	North east	South east	Mid west
NO2	--	-	+	0.64	0.55	0.54	0.54
CO	-	-	+	0.83	0.64	0.69	0.69
ALD2	-	-	++	1.40	1.91	2.24	1.83
FORM	-	-	++	1.41	1.91	2.24	1.84
OLE2	-	-	+	0.74	0.52	0.50	0.53
PAR	-	-	+	0.77	0.56	0.53	0.54
NH3	-	-	=	1.21	1.25	1.27	1.35
PMC	-	-	=	0.62	0.50	0.62	0.63
PM10	-	-	=	0.62	0.50	0.62	0.63
PMFINE	-	-	=	0.62	0.50	0.62	0.83
PEC	-	-	=	0.73	0.59	0.57	0.48
POA	-	-	=	0.63	0.54	0.65	0.67
PSO4	--	-	++	0.61	0.44	0.36	0.24
SO2	--	-	++	0.68	0.46	0.37	0.28

* “=” is the value within $\pm 10\%$, “-“ is 10-50% of reduction, “- -“ is more than 50% of reduction

“+” is 10-50% of increase and “+ +” is more than 50% of increase

In this study, 1 km spatial resolution land use and vegetation database (i.e., leaf area index and plant functional type) was employed in the 36 and 12 km domains on both biogenic models. The same land use and vegetation patterns as 2000 was assumed in all scenarios. The main differences between MEGAN and BEIS are the method of estimating isoprene emission and the emission factors used in the models (Sakulyanontvittaya et al., 2008). In BEIS, the isoprene emission is calculated by empirical algorithms described in Guenther et al., which follows a mathematical function that depends on temperature and solar radiation (Guenther et al., 1993). An increase in temperature causes an initial rise in isoprene emission trailed by a slow decline when the temperature reaches about 38 °C (Zhang et al., 2008). In MEGAN, isoprene is characterized by two separate emissions processes, the light-and-temperature-dependent direct emissions from chloroplasts without storage and purely temperature-dependent emissions from storage pools. Each process utilizes an individual dependence factor to adjust the total isoprene emission. MEGAN calculates the plant-specific isoprene emission by multiplying all those dependence factors with the base/standard emission factor for each type of plant. Equation 1 shows the factor-based emission formula used in MEGAN.

$$EM = \varepsilon * \gamma_{LAI} * \gamma_P * \gamma_T * \gamma_{CE} \quad (4.1)$$

where ε is the base emission factor, γ_{LAI} is the Leaf Area Index Factor, γ_P is the PPFD Emission Activity Factor (light-dependence) and is a function of solar angle and above canopy Photosynthetic Photon Flux Density (PPFD), γ_T is the Temperature Response Factor and γ_{age} is the Leaf Age Factor. Sakulyanontvittaya et al. (2008) estimated the average hourly isoprene emissions in MEGAN are about 61% and 47% higher than the emissions generated by BEIS for July 2001 and January 2002, respectively. It should be noted that the isoprene emission factor used by BEIS is significantly lower than the factors used by other models, which may lead to underestimate the total isoprene emission (Arneth et al., 2007). In this study, the annual isoprene emission in MEGAN is about 53% higher than in BEIS in year 2000. Similar geospatial distributions of isoprene emissions are observed between the two models (not shown). Figures 4.2a and 4.2b show the daily average isoprene and total BVOC emissions in BEIS and MEGA. The major isoprene emission differences occurred in the months of May, June, July, August, and September (MJJAS) when abundances of heat and solar radiation are present. For total BVOC, similar magnitudes of emissions (7% difference in the CONUS domain) are found between the two models. However, significant geospatial disagreements are observed (not shown).

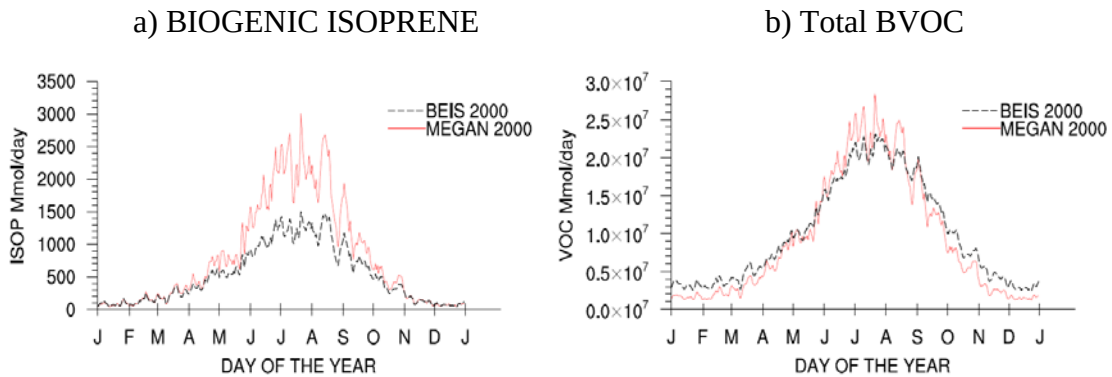


Figure 4.2. Daily average biogenic emissions in the CONUS domain: a) biogenic isoprene; and b) total biogenic VOC.

These discrepancies of emission distributions potentially create differences in the predictions of air quality results between the two models. Table 4.3 shows the emissions breakdown of the present-year biogenic emissions from BEIS and MEGAN. In the Northeast domain, MEGAN shows much larger emission values than BEIS on ALD2, ETH, ISOP, and PAR. The total biogenic VOC emissions in MEGAN is about 60% higher than in BEIS. It is expected that the additional VOC emission in MEGAN may strongly affect the ozone production in the Northeast domain since the Northeast domain is considered as a VOC-limited area and is sensitive to an increase of BVOC. For the Southeast and Midwest domains, -8% and +20% of total VOC differences (MEGAN - BEIS) are found, respectively. To investigate the effect of climate change, the differences in VOC emissions between 2050 and 2000 were also calculated and shown in the right side of the Table 4.3. As resulting from surface warming (i.e., 1.0-2.5°C) and enhancement of solar radiation, both MEGAN and BEIS showed an increase of total biogenic emission by 30% and 21%, respectively. These results (+40% increase in isoprene in MEGAN and +23% in BEIS) are comparable to the values reported in the literature, where BEIS's VOC emissions are increased by 10–90% in the future-year (Zhang et al., 2008; Hogrefe et al., 2004a). It is observed that the emissions estimated by MEGAN is much more climate sensitive than by BEIS, with additional 3-5% increase for most of VOC species and 10% increase for total biogenic VOC.

Table 4.3. The breakdown of species concentrations (in mole) of the present-year biogenic emissions from BEIS and MEGAN, with comparisons of the present-year and future-year biogenic emissions.

Species	Factor (mole)	Present Climate (2000)								% Growth Rate (2050 - 2000)	
		CONUS		Midwest		Northeast		Southeast		CONUS	
		MEGAN	BEIS	MEGAN	BEIS	MEGAN	BEIS	MEGAN	BEIS	MEGAN	BEIS
ALD2	$\times 10^9$	162	149	17	13	11	6	36	28	+24	+19
ETH	$\times 10^9$	75	67	11	9	6	3	14	9	+24	+20
FORM	$\times 10^9$	16	59	2	8	1	3	3	8	+24	+20
ISOP	$\times 10^9$	294	192	26	21	30	14	76	47	+40	+23
NO	$\times 10^9$	71	112	7	8	5	4	17	21	+21	+19
OLE	$\times 10^9$	96	263	11	31	6	10	18	37	+25	+21
PAR	$\times 10^9$	1230	1515	160	167	92	57	245	228	+23	+21
TERPB	$\times 10^9$	71	112	7	8	5	4	17	21	+21	+19
TOL	$\times 10^9$	0.65	20.88	0.10	2.81	0.05	0.99	0.12	2.80	+24	+20
XYL	$\times 10^9$	0.92	0.62	0.07	0.10	0.07	0.07	0.21	0.16	+21	+18
VOC	$\times 10^{12}$	3397	3644	370	404	287	174	767	637	+30	+21

The increase of VOC emission strengthens the impact of climate change on ozone air quality in the United States.

4.4.3 Emissions Scenarios

Overall, eight simulation scenarios were selected and summarized in Table 4.4. The first four scenarios (marked inside of the red dash line) were intended to investigate the effects of downscaling. These scenarios are (1) present meteorology with current emissions, (2) present meteorology with future emissions, (3) future meteorology with current emissions and (4) future meteorology with future emissions, which are identical to the scenarios used in Wu et al. (2008), for GEOS-Chem (Lam et al., 2008a). While the other four scenarios (filled with green) were used to study the impacts of change of biogenic emissions (MEGAN vs. BEIS) in the future climate scenarios.

4.5 Discussion and Results

4.5.1 Comparison of Current and Future Climate

Model performance of meteorological outputs was evaluated by comparing the global GCM outputs to the downscaled MM5 outputs. We have chosen the global GCM outputs as the bases of comparison (instead of observational data) for three reasons: (1) the primary focus of the study was to investigate the effects of downscaling, this type of comparison permitted one to quantify the relative air quality impacts from meteorological downscaling;

Table 4.4. Summary of CMAQ simulations conducted in this study.

Scenario			Model			Scenario Index
Meteorology	Anthrop. Emission	Bio. Emission	GEOS-Chem	CMAQ		
2000	2000	MEGAN (2000)	4° x 5°	36km x 36km	12km x 12km	2000M_2000E_M
2000	2050	MEGAN (2050)	4° x 5°	36km x 36km	12km x 12km	2000M_2050E_M
2050	2000	MEGAN (2000)	4° x 5°	36km x 36km	12km x 12km	2050M_2000E_M
2050	2050	MEGAN (2050)	4° x 5°	36km x 36km	12km x 12km	2050M_2050E_M
2000	2000	BEIS (2000)	-	36km x 36km	-	2000M_2000E_B
2000	2050	BEIS (2050)	-	36km x 36km	-	2000M_2050E_B
2050	2000	BEIS (2000)	-	36km x 36km	-	2050M_2000E_B
2050	2050	BEIS (2050)	-	36km x 36km	-	2050M_2050E_B

(2) the global GCM outputs used for the comparisons have been extensively evaluated with observational data (Rind et al., 2007; Schmidt et al., 2006). This type of comparison should give sufficient understanding of MM5 performance to the current climate condition; and (3) since no observed boundary conditions (i.e., FDDA technique or observation nudging in the GISS GCM) were used to constrain GCM simulations, the characteristics of MM5 outputs is unlikely to follow the hour/daily trends with the observational data. If model performance was done on monthly averaging, comparing MM5 outputs to the GISS outputs would give sufficient meteorological validation—just as if the outputs had been compared with observational data.

For the purpose of air quality evaluation, we have selected several climatic variables that affect air pollution to facilitate our discussion, which included ground temperature (T), relative humidity (RH), precipitation (RAIN), shortwave radiation at the surface (SW), total cloud fraction (CFRACT), wind speed (WSP), wind direction (WDR) and Planetary Boundary Layer (PBL) height. The main focus was placed on temperature since the rise in temperature is expected to worsen the regional air quality in the future by enhancing both biogenic emissions and photochemical reaction rates of gaseous precursors of O₃ and low-volatile secondary PM_{2.5}. In addition to temperature, the change of wind speed and PBL height are also expected to be important to the regional air quality since they had substantial effects on the rates of horizontal and vertical dispersions. Thus, they are expected to have a significant impact on surface O₃ and PM_{2.5} concentrations if significant changes from these variables are observed (Lamsal et al., 2004; Gaza, 1998).

In response to the change of greenhouse gases, surface temperatures in 2050 were predicted to increase 1.0-3.0°K when compared to the base year (2000), as shown in Table 4.5. The major changes of temperature are observed in MJJAS for all three domains. The change of annual average ground temperature is about 2.0 °K for the entire continental United States domain, with the maximum hourly temperature difference of 5–6 °K within the 36 km² grid. These values are similar in magnitudes to previous studies reported in the literature (Bell et al., 2007; Nolte et al., 2008; Zhang et al., 2008). A larger increase in temperature is predicted for the Midwest and Southeast, with a smaller increase is expected for the Northeast (i.e., average +1 °K). The increase in temperature across the United States

is expected to increase the rates of radical production and photochemical reaction and worsen the ozone and PM_{2.5} air quality in the future. The comparison of the GISS outputs to the MM5-36km have shown that the Root Mean Square Error (RMSE) is about +0.25 °K or less for the CONUS domain, with ± 0.6 °K of winter bias and ± 0.2 °K summer bias on the defined domains. Based on the value reported by Dawson et al. (2008), the difference of 0.2 °K would translate into about 0.1–0.2 ppbv increase of surface ozone. With a 0.3 °K difference found between the GISS and MM5 outputs, the effect of downscaling would contribute an additional 0.2-0.3 ppbv of surface ozone in the Northeast domain for the future scenario. For downscaling from the MM5-36km to the MM5-12km, no significant change is found on the average domain-wide temperature for the Southeast domain.

On one hand, higher future temperature promotes higher ozone and PM_{2.5} concentrations. However, the possible increase in PBL height and horizontal wind speed may counteract the effect of temperature by enhancing regional air circulations and thus cause reductions of ozone and PM_{2.5}. Figure 4.3a shows the current and future average wind speeds for the three domains. It is noticed that the average surface wind speeds between 2050 and 2000 (dash-line vs. solid-line on the same color) are similar for all domains, with the maximum difference of 0.4 m/s occurring in the Southeast during MJJAS. The overall annual RMSE is about 0.2 m/s across the CONUS domain.

Table 4.5. Average zonal temperatures of GISS and MM5 outputs in 2000 and 2050.

Year	Type	Midwest			Northeast			Southeast		
		*JFMA	*MJJAS	*OND	*JFMA	*MJJAS	*OND	*JFMA	*MJJAS	*OND
2000	GISS	276.6	295.8	279.8	283.4	296.9	286.9	290.1	299.3	291.8
	US36	277.6	296.4	280.6	281.8	296.2	285.8	290.8	300.0	292.5
	US12	-	-	-	-	-	-	290.8	299.8	292.5
2050	GISS	276.8	298.3	281.2	284.2	297.9	287.0	291.0	301.4	292.1
	US36	277.9	298.7	282.0	282.7	297.4	286.0	291.6	302.0	292.7
	US12	-	-	-	-	-	-	291.6	301.8	292.8
2050	GISS	0.2	2.4	1.4	0.7	1.0	0.1	0.9	2.0	0.3
-	US36	0.4	2.4	1.4	0.8	1.3	0.2	0.8	2.0	0.3
2000	US12	-	-	-	-	-	-	0.8	2.0	0.3

* JFMA is average value from Jan., Feb, Mar., and Apr.;

MJJAS is average value from May, Jun., Jul., Aug., and Sep.;

OND is average value from Oct., Nov., and Dec.

For the Northeast, the average differences between 2050 and 2000 are nearly the same. Figure 4.3b shows the probability distribution function (PDF) of wind speed in the Northeast during MJJAS. It is observed that although the average wind speed is nearly the same, the distribution of wind speed could be quite different. As indicated in Figure 4.3b, the wind speed distributions in 2050 are actually higher than the ones in 2000 when the wind speed reaches about 4.0 m/s in the Northeast. This PDF gives a clear indication of horizontal dispersion, where higher wind speed is found in the Northeast domain for the future. In the Midwest, the average wind speed between 2050 and 2000 is about 0.2 m/s. From the probability distribution curve of the Midwest domain (not shown), an increase in wind speed is also predicted in 2050. Overall, the change of wind speed between 2050 and 2000 is relatively small in all three domains. For the downscaling perspective, a large difference (i.e., 0.7 m/s) was found between the GISS and MM5-36km outputs in the Midwest. The difference is caused by the manner in which topographical variables are implemented by the GISS GCM and MM5 model in the Rocky Mountain area. This difference in wind speed may introduce significant biases to the future air quality in the Midwest when comparing GEOS-Chem outputs to CMAQ outputs.

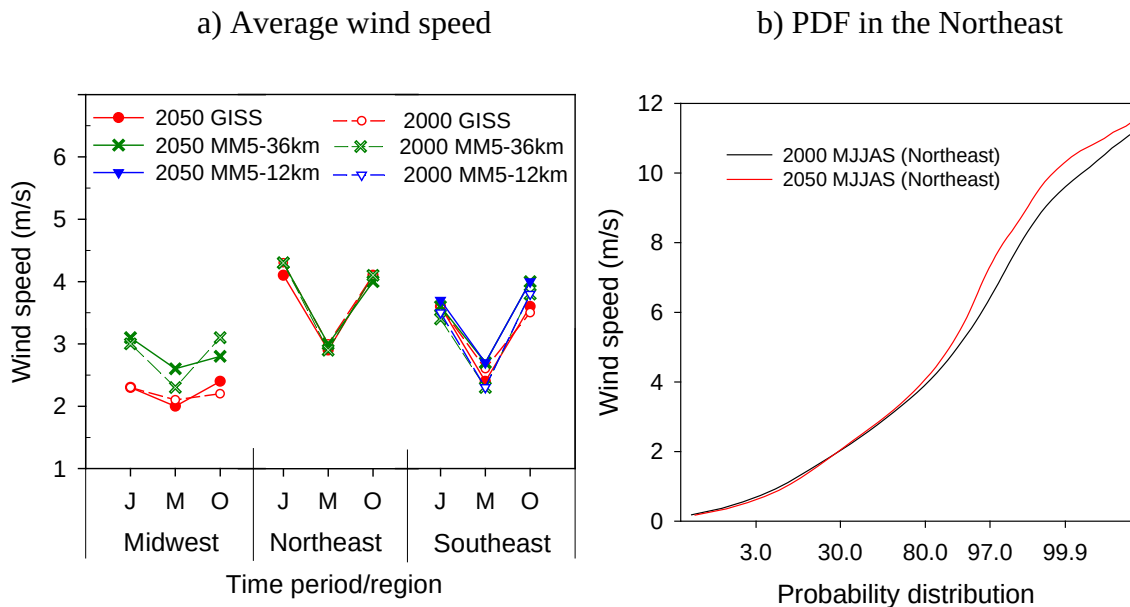


Figure 4.3. Summary of wind speed from GISS and MM5 outputs: a) average wind speed in the study domains and b) the probability distribution function of wind speed in the Northeast.

Figure 4.4 (a - c) show the average difference in PBL heights between 2050 and 2000 during January, February, March, and April (JFMA), MJJAS, and October, November, and December (OND), respectively. It was observed that most of the places in the United States show a minor change of PBL height, except for the Western United States during the months of MJJAS. The maximum difference of PBL height between 2050 and 2000 ranges from – 190 m to 305 m. In the study domains, no significant change of PBL height ($\pm 5\%$ difference) was found to be attributable to climate change. These findings are consistent with the GISS's results, where only $\pm 10\%$ PBL changed for the future year scenario (Mickley et al., 2006). In addition to wind speed and PBL height, other meteorological parameters such as solar radiation, humidity, and precipitation may also affect the ozone and $PM_{2.5}$ air qualities. Early findings show that future temperatures will increase by at least 2 °K. This increase in temperature may be linked to the increase of solar radiation at the surface, a direct proportional relationship between temperature and solar radiation is generally expected. In the Midwest, more solar radiation is predicted reaching the surface due to the decrease of cloud cover. The increase in temperature with less cloud cover in the Midwest may potentially result in a significant increase of ozone under the future climate condition. In contrast, in the Northeast and Southeast, the solar radiation will decrease by 10% due to an increase of cloud cover in the future. Although the average temperatures are increased by 1.0 to 2.0 °K, the increase of cloud cover may limit the amount of solar radiation reaching the surface and possibly result in less ozone formation in these two regions.

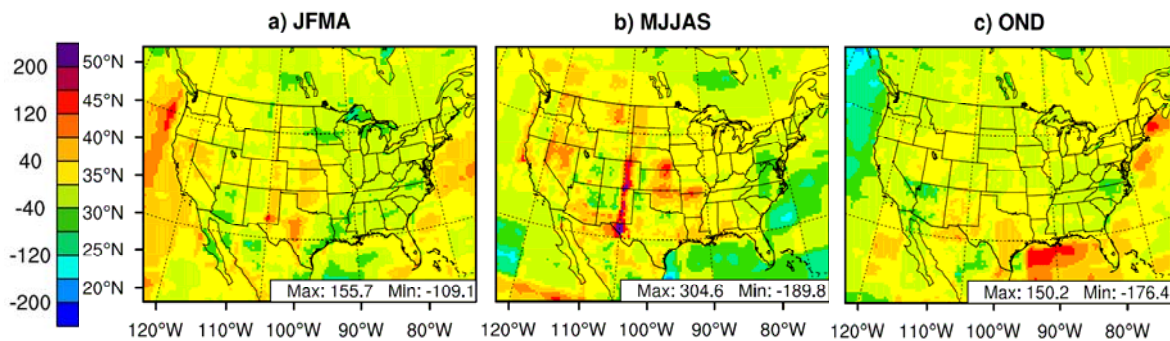


Figure 4.4. Average PBL height difference between 2050 and 2000 from MM5 outputs: a) JFMA, b) MJJAS, and c) OND.

For precipitation, slight change was found for the three regions. The seasonal averages of precipitation (JFMA, MJJAS, and OND) are within a 0.01 cm rainfall difference (Gustafson Jr and Leung, 2007). From geospatial plots (not shown), a slight increase in precipitation was observed in both Northeast and Southeast regions and a slight decrease in precipitation was observed in the Midwest. For relative humidity (RH), the overall changes are about $\pm 5\%$ from 2000 to 2050 among different time periods and domains. A slight increase of moisture was observed in the Northeast, while slight decreases were found in the Midwest and Southeast. As expected, the average relative humidity in the Southeast is much higher than in the Midwest and Northeast. On average, it is about 10 and 15% higher. The average RH in 2050 during MJJAS is about 80%, 75%, and 55% for the Southeast, Northeast, and Midwest, respectively. The high RH possibly enhances chemical deposition rates of SO₂ and also promotes precipitation (Sakamoto et al., 2004). For the downscaling perspective, a large difference of RH is observed between the GISS and MM5-36km outputs. An average of 10% increase of moisture was estimated across those three sub-domains while downscaling the global model into the regional model. For the MM5-36km to MM5-12km outputs, only less than a 1.0% in difference of RH is observed. It is believed that the large difference in RH observed between the GISS and MM5-36km outputs was introduced by the inconsistency of advection schemes and vertical layer structures used by those two meteorological models. The additional RH in MM5 may help the formation of clouds by causing air to increase in elevation and promote more precipitation (Gustafson Jr and Leung, 2007; Gilliam et al., 2006; Queen et al., 2008).

4.5.2 Comparisons of Present Climate Air Quality Using MEGAN Emissions

The qualitative evaluation of CMAQ chemical predictions for the current climate condition was conducted by comparing the average observed quantities of O₃ and PM_{2.5} from the Clean Air Status and Trends Network (CASTNET) <http://www.epa.gov/CASTNET/> and the Speciation Trends Network (STN). The results provided some level of agreement between the observed and our simulated values to justify the use of CMAQ results in representing the future air quality. Due to the difference in meteorological inputs, the hourly comparison was not performed, instead average monthly values was used. Figure 4.5. (a –

b) shows the statistical distributions of maximum 1-hr and 8-hr average O_3 concentrations simulated for the months of MJJAS in 2000. The black and red colors correspond to the 2000 CMAQ simulated value and the CASTNET observed value from 1998 to 2002, respectively. The dashed lines at the top and bottom of each box plot show the maximum and minimum values for the data. The square box specifies the ozone values of 75%, 50%, and 25% tiles and the cross mark shows the monthly mean value. The maximum 1-hr and 8-hr O_3 concentrations are well reproduced with the ozone values slightly under prediction. The average monthly value of ozone is about 50 to 60 ppbv. The good agreement of O_3 suggests that the CO and NO_x emissions used in this study were relatively close to the actual measured emissions from those five years. However, the underestimate also suggests the uncertainty of MM5 meteorological and emissions in CMAQ were derived from our current climate conditions. The O_3 result is about 5-10% lower than the observed values, which is comparable with the results reported by Tagaris et al and Zhange et al., where 2-15% different of O_3 in June, July, and August (JJA) for 2000-2002 (Zhang et al., 2008; Tagaris et al., 2007). Figure 4.6 shows the comparison of monthly average PM_{2.5} from CMAQ outputs and the STN observational network. Identical labeling conventions were used in Figure 4.6, as was used in Figure 4.5.

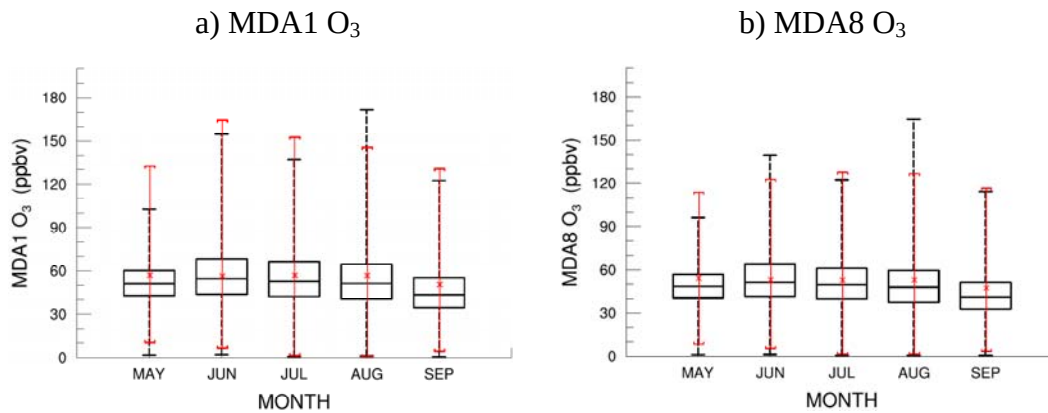


Figure 4.5. Statistical distributions of O_3 concentrations for the months of MJJAS in 2000: a) maximum 1-hr (MDA1), and b) 8-hr average O_3 (MDA1). The black color corresponds to the 2000 CMAQ simulated value, where as the red color represents the CASTNET observed values from 1998 to 2002.

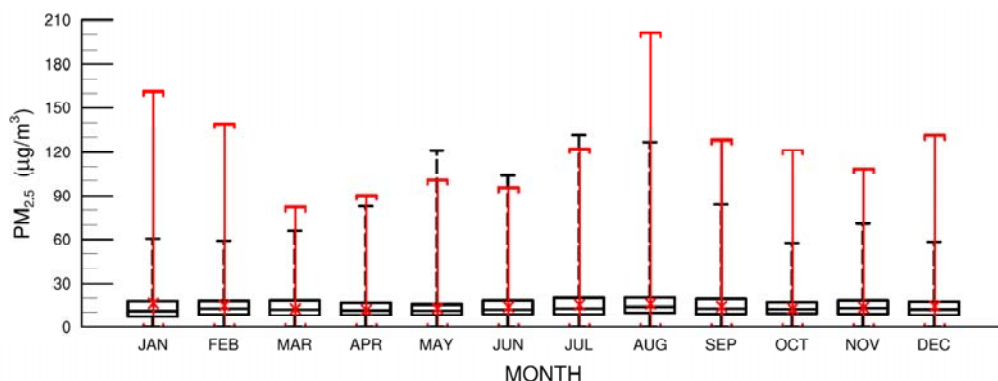


Figure 4.6. Monthly statistical distributions of PM_{2.5} concentrations for year 2000. The black color corresponds to the 2000 CMAQ simulated values, where as the red color represents the CASTNET observed values from 1998 to 2002.

Once again, the average concentration in CMAQ is slightly under predicted for most of the months. As expected, the peak values of PM_{2.5} between simulated and observed values do not match well due to the fact that conservative emissions have been used in the simulation where no special event (such as, a extreme fire event or volcanic eruption) was included in the current emissions. Since the maximum PM_{2.5} value is either lower or close to the maximum observed value, the CMAQ outputs reproduce reasonably well on the current PM_{2.5} level. For the underestimate of PM_{2.5}, Tagaris et al. (2007) suggests that the under-prediction is caused by low aerosol yields, higher vapor pressures, and a lack of isoprene second organic aerosol (SOA) treatment in CMAQ, in which the isoprene SOA accounts for about 0.01–1.52 µg/m³ of PM_{2.5} (Zhang et al., 2008).

4.5.3 Comparisons of CMAQ Simulated Outputs (MEGAN Versus BEIS Inventories)

As mentioned earlier, the MEGAN biogenic model estimated about 50% higher isoprene emission than the BEIS biogenic model. However, due to the fact that the BEIS model estimated higher emissions on other biogenic emissions (terpene and formaldehyde), the resulting difference of total annual biogenic emissions between those two models has turned into about 5%. In summer, isoprene emission contributed a large portion of overall biogenic emissions and resulted in higher overall biogenic emissions in MEGAN. Conversely, the influence of isoprene emission was diminishing when winter approached and resulted in lower overall emissions in MEGAN. To investigate the impacts of using different

biogenic models in the climate change study, both MEGAN and BEIS biogenic emissions were used to simulate both current and future climate conditions using CMAQ. Figures 4.7 a through c show the Maximum Daily Average 8-hour (MDA8) ozone of CMAQ-MEGAN versus CMAQ-BEIS for the months of MJJAS in the year 2000 on the Midwest, Northeast, and Southeast, respectively. The CMAQ-MEGAN represents the CMAQ simulation using MEGAN biogenic emissions, where as the CMAQ-BEIS denotes the CMAQ simulation using BEIS biogenic emissions. Both Midwest and Southeast show only a minor difference in MDA8 ozone between MEGAN and BEIS. The slopes of those two best-fit curves are close to one, which signifies the Midwest and Southeast are insensitive to the increase of VOC emissions. Table 4.6 shows the percentage change of MDA8 ozone and PM_{2.5} in the current and future climate conditions. For ozone, the differences between 2000 and 2050 on the Midwest and Southeast are less than 1.0% (e.g., $|(-1.4)-(-0.6)|=-0.8$), which implies that the impacts of change of biogenic emissions are independent from the selection of simulation year since both years of CMAQ simulations responded similarly. In the Northeast, 10% increase (based on the best-fit line) of MDA8 ozone is observed (shown in Figure 4.7b). The increase of MDA8 ozone implies the Northeast region is perhaps made up from multiple VOC-limited sub-regions. As discussed by (Duncan et al., 2009), the Northeastern region of the United States, such as New York and other metropolitan areas, was a typical radical-limited/VOC-limited region (Kleinman et al., 2000). There is no doubt that the majority of places in the Northeast are more radical-limited/VOC-limited conditions due to a large portion of urban land (Milford et al., 1994; Milford et al., 1989). As with the increase of biogenic emissions in the Northeast domain, the VOC-limited sub-regions within the domain has lead to the increase of MDA8 ozone. For ozone, it was observed that the average changes of MDA8 ozone on the Northeast domain in 2000 and 2050 are 5.4% and 6.0%, respectively, as shown in Table 4.6. For PM_{2.5}, Figure 4.8 shows the chemical breakdown of annual average PM_{2.5}. The left side of the Figure shows the constituent of PM_{2.5} and the right side of the Figure shows the organic carbon (OC) portion of PM_{2.5}. The suffix of “-B” and “-M” indicate the BEIS and MEGAN inventories were used in the CMAQ simulations, respectively. As expected, the change of PM_{2.5} concentration is limited to only organic aerosol from the biogenic VOC.

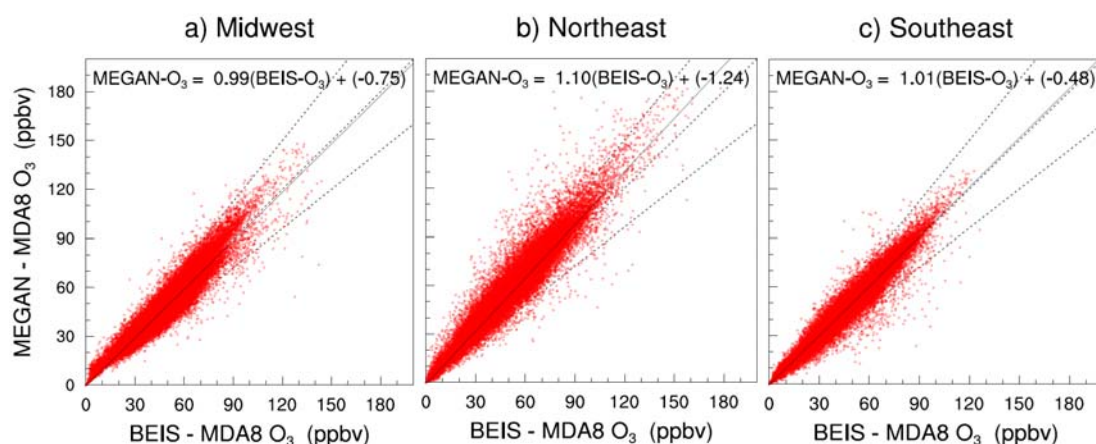


Figure 4.7. Scatter plots of MEGAN-MDA8 ozone versus BEIS-MDA8 ozone for the months of MJJAS in the year 2000 on: a) Midwest, b) Northeast, and c) Southeast.

Table 4.6. The percentage change of MDA8 ozone (in ppbv) and PM_{2.5} (in $\mu\text{g}/\text{m}^3$) for 2000 and 2050.

			Slope		Intercept		(MEGAN- BEIS)/ MEGAN	(MEGAN- BEIS)/ MEGAN
Year			2000	2050	2000	2050	2000	2050
MDA8 ozone	*MJJAS	Midwest	0.99	1.02	-0.1	1.4	-1.4%	-0.6%
		Northeast	1.1	1.11	-1.3	-1.5	5.4%	6.0%
		Southeast	1	1	-0.1	-0.1	0.1%	-0.6%
	*JFMA	Midwest	0.97	0.97	-0.2	-0.2	-3.5%	-4.0%
		Northeast	0.98	0.97	0.0	0.0	-3.3%	-2.6%
		Southeast	0.92	0.92	-0.1	0.0	-4.3%	-5.5%
PM2.5	*MJJAS	Midwest	1.01	1	-0.4	-0.3	-1.7%	-2.1%
		Northeast	1	1	0.0	-0.1	-0.3%	-0.3%
		Southeast	0.97	0.96	-0.2	-0.2	-1.6%	-2.2%
	*OND	Midwest	0.98	0.97	-0.3	-0.3	-6.3%	-6.0%
		Northeast	0.98	0.98	0.0	0.0	-4.2%	-4.3%
		Southeast	0.93	0.9	-0.1	0.0	-6.6%	-7.4%

* JFMA is average value from Jan., Feb, Mar., and Apr.;
MJJAS is average value from May, Jun., Jul., Aug., and Sep.;
OND is average value from Oct., Nov., and Dec.

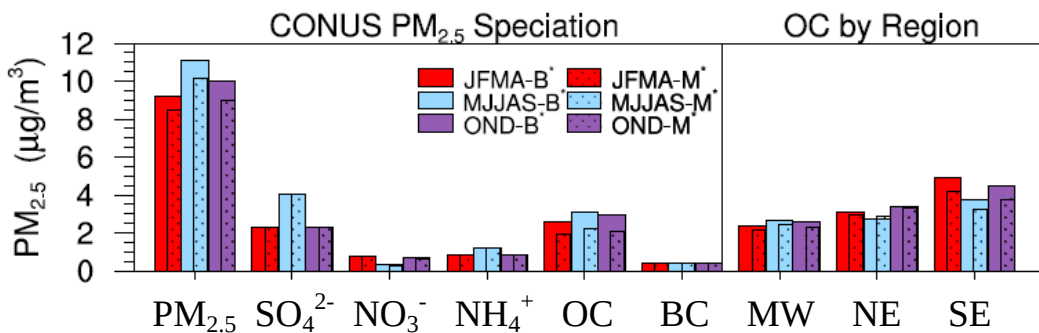


Figure 4.8. The annual average of PM_{2.5} in the CONUS domain (left), and the annual average organic carbon of PM_{2.5} for the Midwest, Northeast, and Southeast.

The overall changes of PM_{2.5} between the MEGAN and BEIS emissions were estimated to be about -5%, -3%, and -6% for the Midwest, Northeast, and Southeast, respectively (shown in the right side of the Figure). The impact of PM_{2.5} in the Southeast domain is much larger than the Midwest and Northeast due to a large difference in isoprene emission between those two biogenic models in the Southeast. To investigate the seasonal impacts of PM_{2.5}, the annual CMAQ outputs have been divided into JFMA, MJJAS, and OND. As shown in Table 4.6 (last two columns), large changes of PM_{2.5} were observed in the months of JFMA and OND (both spring and winter) for all three domains, while insignificant change of PM_{2.5} was found in MJJAS (summer). The largest change of slope, with the value of 0.93, was observed in the Southeast in the months of OND. This value indicates the PM_{2.5} estimates of CMAQ-MEGAN are about 7% lower than the estimates of CMAQ-BEIS. This -7% difference translates into about -2 µg/m³ on average. It is believed that the lower PM_{2.5} in CMAQ-MEGAN was mainly contributed by the lower terpene emission from MEGAN inventories; since terpene emission undergoes oxidation to form condensable gases and eventually converts into second organic aerosols (SOAs).

In the CMAQ simulations, we are aware of the fact that parts of the SOAs pathway for isoprene was missing in the current CMAQ configuration, which might result in lower isoprene SOAs on both CMAQ-MEGAN and CMAQ-BEIS. It is expected that the difference of isoprene emissions between CMAQ-MEGAN and CMAQ-BEIS in the months of JFMA and OND is fairly small, so the impacts of isoprene pathway can be neglected.

However, for MJJAS, a larger impact from the missing isoprene pathway was expected due to the fact that isoprene emission is the dominant species in the biogenic VOC and large differences of VOC emissions were observed between CMAQ-MEGAN and CMAQ-BEIS. (Zhang et al., 2008) and (Boylan, 2005) suggested the maximum impact of lacking of isoprene pathway in CMAQ was about $1.52 \mu\text{g}/\text{m}^3$ and $2.2 \mu\text{g}/\text{m}^3$ of SOAs, respectively. Nevertheless, since the focus of the $\text{PM}_{2.5}$ discussion has been placed on the months of JFMA and OND, the impact of SOAs from isoprene may be ignored because the production of isoprene is low. For different climate conditions (2000 versus 2050), very minor differences (i.e., less than 1.0%) were observed between 2000 and 2050 as shown in the last two columns of Table 4.6. Once again, this indicates that the impact of change of biogenic emissions is independent from climate conditions since the CMAQ simulation results responded in the same way on both years.

4.5.4 Future Ozone Air Quality and Comparison of CMAQ Versus GEOS-Chem

To better understand future air quality, both GEOS-Chem and CMAQ outputs were analyzed to investigate the future air quality trends. As mentioned earlier, both models used the same United States NEI inventories, future emissions projection factors, and meteorological fields from GISS'III GCM. Although the inputs from those models were kept consistently throughout implementations, it is expected that the model outputs might still give an inconsistent prediction of future air quality due to the differences in resolutions, chemical mechanisms, and model sensitivity to climate change. Table 4.7 shows the summary of MDA8 ozone outputs from GEOS-Chem and CMAQ. The four scenarios used in the simulations are: (1) 2000 meteorology condition with 2000 current emissions (2000M_2000E), (2) 2000 meteorology condition with 2050 future emissions (2000M_2050E), (3) 2050 meteorology condition with 2000 current emissions (2050M_2000E), and (4) 2050 meteorology condition with 2050 future emissions (2050M_2050E). It should be noted that all simulations presented in this section are the simulations using MEGAN biogenic emissions and the same notation will be used throughout the paper. As reported by (Lam et al., 2008a), the GEOS-Chem predicted a 2-5 ppbv increase of MDA8 ozone over the Midwest and Northeast domains, while a 3 ppbv

decrease over the Southeast domain from climate change. For CMAQ, it predicted an increase of MDA8 ozone by about 2-4 ppbv (i.e., calculated by taking the average difference between 2050M and 2000M) from climate change for all domains. These results are similar to the findings reported by Zhang et al. (2008), Hogrefe et al. (2004b), and Racherla and Adams (2008), where the MDA8 ozone increased by 20% (1 to 5 ppbv), 1.5-7.5 ppbv, and 5 ppbv (in the 95th percentile), respectively. It is expected that our results may predict less increase of MDA8 ozone than the others since the selected IPCC scenario (i.e., A1B) predicts less future warming than the A2 scenario from Hogrefe et al. (2004b), and Racherla and Adams (2008). For the comparison between 36km and 12km CMAQ simulations, no difference was found on the future climate trends. The largest discrepancy of MDA8 ozone between GEOS-Chem and CMAQ was observed in the Southeast domain, where GEOS-Chem and CMAQ predicted an opposite result on the impact of MDA8 ozone from climate change (e.g., future ozone subtracted by current ozone). We observed that GEOS-Chem predicted higher minimum ozone than CMAQ. Conversely, it also predicted lower maximum ozone than CMAQ due to the restriction of grid resolution (4°x5°). The minimum and maximum ozone concentrations in GEOS-Chem are 15-20 ppbv and 75-85 ppbv, respectively, whereas CMAQ has the maximum and minimum values at 3-5 ppbv and 130-135 ppbv. It is unclear what was causing this discrepancy. However, we believe that this discrepancy may be partially related to the difference in the sensitivity of ozone under the coarse grid resolution at the complex terrain regions.

Table 4.7. Summary of MDA8 ozone (in ppbv) from GEOS-Chem and CMAQ outputs.

MDA8 type	Domain	2000M_2000E		2000M_2050E		2050M_2000E		2050M_2050E	
		GEOS- Chem	CMAQ	GEOS- Chem	CMAQ	GEOS- Chem	CMAQ	GEOS- Chem	CMAQ
Max	MW	106	149	89	125	112	165	94	139
	NE	114	182	99	168	126	186	107	164
	SE-36km	103	132	82	126	100	135	80	126
	SE-12km	-	154	-	145	-	178	-	163
Avg	MW	52	54	49	48	55	55	51	50
	NE	66	57	60	50	69	59	62	52
	SE-36km	66	56	55	51	65	57	53	52
	SE-12km	-	51	-	46	-	50	-	48

Increasing the grid resolution in GEOS-Chem may reduce the discrepancy of MDA8 ozone between GEOS-Chem and CMAQ. Nevertheless, slightly different results were found between GEOS-Chem and CMAQ in the Southeast; both models consistently predicted an increasing response of surface O_3 from climate change on the Northeast and Midwest domains. The average effects on climate change are about +2.0-2.5 ppbv and +1.5-2.0 ppbv for GEOS-Chem and CMAQ, respectively, whereas the average effects from reducing emissions are +4.0-7.0 ppbv and +5.0-7.0 ppbv. It is clear that emissions have stronger impacts than the climate change in the regional air quality.

To further investigate the difference between GEOS-Chem and CMAQ on the MDA8 ozone, the PDF were constructed for all simulated scenarios and are shown in Figure 4.9. The black, green, red, and blue colors represent the scenarios of 2000M_2000E, 2000M_2050E, 2050M_2000E, and 2050M_2050E, respectively. It is observed CMAQ and GEOS-Chem performed quite similarly in the Northeast and Midwest domains, where the order of the color lines are identical. Distinct separation between the colored lines found in a), b), d), and e) demonstrates a discrete relationship was developed among the scenarios. The non-linearity (skew) distribution (e.g., Figure 4.9d) indicates a small value of high ozone concentration was found in the PDF. It should be noted when a line is far more up and left, it implies a higher MDA8 ozone distribution has been found. As expected, the order of lines are red, black, blue, and green and the worst MDA8 ozone air quality occurs in the red line, which corresponds to the scenario where higher overall temperature with no emissions control in the future (2050 meteorological conditions with 2000 current emissions). Since the reduction of emissions is a stronger factor than the increase of temperature from climate change, we will expect that the red and black lines should be in the more up and left position. In the Southeast domain, shown in Figure 4.9c) and f), the order of the colored lines is somewhat different between GEOS-Chem and CMAQ. In GEOS-Chem (9c), the colored lines (red vs. black and green vs. blue) are sticking close together and consequently not able to demonstrate the effect of climate change. Conversely, in CMAQ (9f), clear separations were found among those lines and the effect of climate change was observed.

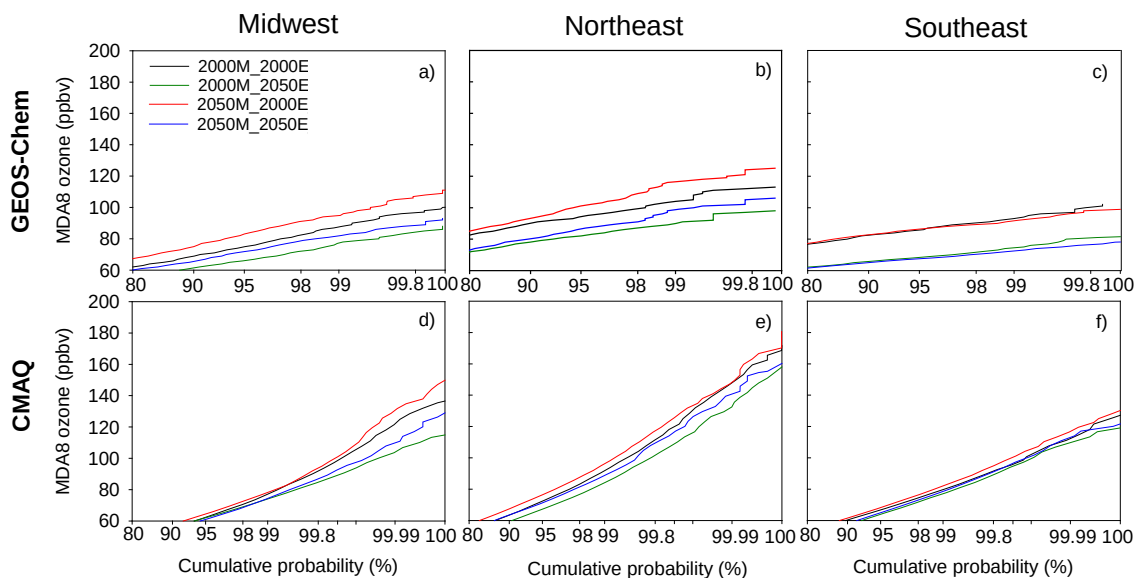


Figure 4.9. The cumulative probability function of MDA8 ozone for GEOS-Chem and CMAQ: a) Midwest for GEOS-Chem, b) Northeast for GEOS-Chem, c) Southeast for GEOS-Chem, d) Midwest for CMAQ, e) Northeast for CMAQ, and f) Southeast for CMAQ.

As mentioned earlier, the inconsistency of the results between GEOS-Chem and CMAQ may relate to meteorological (inconsistent relative humidity and wind speed) and resolution differences ($4^{\circ} \times 5^{\circ}$ vs. $36\text{km} \times 36\text{km}$). Nevertheless, the future climate and air quality predictions in the Southeast have been controversial.

4.5.5 Future PM_{2.5} Air Quality

For PM_{2.5}, since the GEOS-Chem version 7.03.06 did not incorporate sufficient PM_{2.5} species at the moment, no PM_{2.5} comparison between GEOS-Chem and CMAQ will be presented. Table 4.8 shows the summary of PM_{2.5} outputs in CMAQ. The maximum and average PM_{2.5} ranged from 96.1 to 127 $\mu\text{g}/\text{m}^3$ and 5.0 to 11.7 $\mu\text{g}/\text{m}^3$, respectively. It is observed that the effect of climate change had only a minor impact on the future PM_{2.5} concentration, whereas the reduction of emissions contributed a significant reduction of PM_{2.5}. The total PM_{2.5} reductions from current to future was estimated to be about 40 to 50%. For the comparison between 36km and 12km CMAQ simulations, no difference on PM_{2.5} was found on the future climate trends.

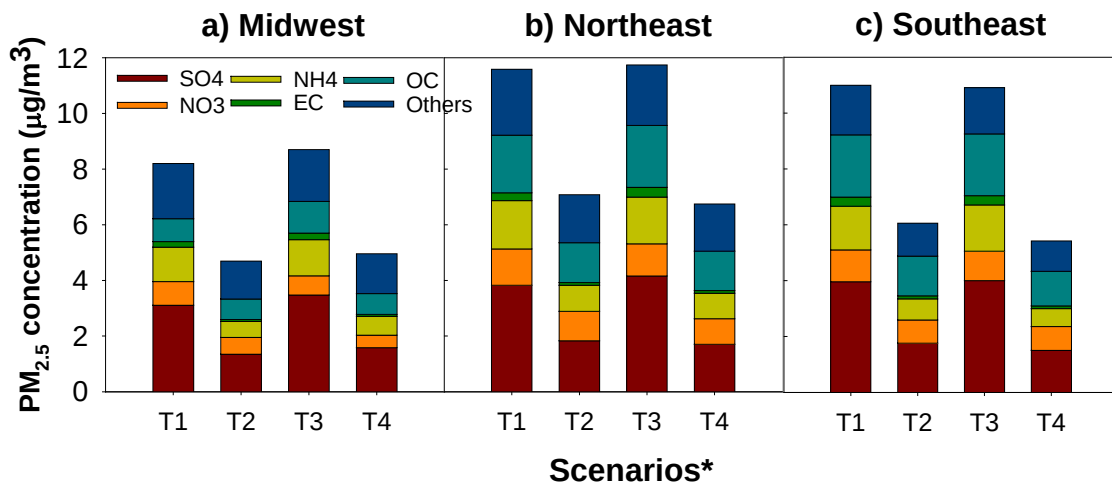
Table 4.8. Summary of PM_{2.5} (in µg/m³) in CMAQ outputs.

PM _{2.5} type	Domain	2000M_2000E	2000M_2050E	2050M_2000E	2050M_2050E
Max	MW	127	130.2	96.1	102
	NE	119.3	116.8	116.6	117.7
	SE-36km	131.4	133.9	107.5	108.5
	SE-12km	115.7	104	105.3	102.5
Avg	MW	8.2	4.7	8.7	5
	NE	11.6	7.1	11.7	6.7
	SE-36km	11	6.1	10.9	5.4
	SE-12km	10.4	5.7	10.5	5.1

Figure 4.10 shows the chemical breakdown of PM_{2.5}. It is expected that a large portion of PM_{2.5} in CMAQ simulations were coming from sulfate aerosols (SA) and organic aerosols (OA). For organic aerosols, no significant change of OA in the future scenario (T4 or 2050M_2050E) was observed. Although researchers have suggested that the increase of temperature might discourage the formation of aerosols by increasing the rate of vaporization (Zhang et al., 2008), in fact, a 5% (1 µg/m³) increase of OA was observed. For sulfate aerosols, a significant reduction of SO₄²⁻ was observed in the future scenario due to the large anticipated reduction of SO₂/SO₄ emissions. The total reduction of sulfate aerosols reaches above 50%. It is believed that the reduction of SO₄²⁻ aerosols allows more radicals to be used for the formation of NO₃⁻ aerosols. However, since the emission of nitrogen oxides was also reduced significantly, the effect from extra radicals have been diminished by the reduction of NO_x and yielded an overall 5% reduction of nitrate aerosols. As a result, the overall change of PM_{2.5} from current condition (T1, 2000M_2000E) to future condition (T4, 2050M_2050E) maintained at about -40% to -50%, as shown in Table 4.8, where the effect of climate change contributed about 10% change of PM_{2.5}, and the emissions accounted for about 90% change of overall PM_{2.5}.

4.6 Conclusion

The CMAQ simulation comparisons of using MEGAN and BEIS biogenic emissions on the climate change scenarios were performed in this study. There was a general increase of MDA8 by about 10 to 12% in the Northeast domain when using MEGAN biogenic emissions. No significant effect was found in the Midwest and Southeast domains.



*T1 - 2000M_2000E, T2 - 2000M_2050E, T3 - 2050M_2000E, and T4 - 2050M_2050E

Figure 4.10. The chemical breakdown of annual average $PM_{2.5}$ in CMAQ for: a) Midwest, b) Northeast, and c) Southeast.

The change of MDA8 ozone in the Northeast domain is mainly triggered by the nature of the VOC-limited region of the domain. For $PM_{2.5}$, as expected, all three domains showed a decrease of organic aerosols by 15% from using MEGAN biogenic emissions. Since the CMAQ version used in this study did not include the pathway of isoprene aerosols, it is expected that the $PM_{2.5}$ results may have been 1-2 $\mu g/m^3$ lower than if the isoprene chemistry had been present. Moreover, since a strong increase of isoprene emission was observed in MEGAN in the future climate condition while it was absent from BEIS, it might have also underestimated the impact of $PM_{2.5}$ when comparing the difference between MEGAN and BEIS simulations. Nevertheless, the change of biogenic emissions was not strong enough to alter the relationship among different climate scenarios on both ozone and $PM_{2.5}$. Therefore, it is concluded that the relationships among different climate change scenarios do not change regardless of which biogenic emissions were used.

For the downscaling, we have found that both GEOS-Chem and CMAQ were in agreement with the MDA8 ozone results on the Midwest and Northeast domains. However, disagreement of the ozone results was found in the Southeast domain, where the GEOS-Chem results were inconclusive. It is believed that the coarse resolution used in GEOS-Chem on the study was insufficient to represent the geospatial relationship in the complex

terrain region. For CMAQ 36km and CMAQ 12km study, no significant difference of output results (i.e., MDA8 ozone and $PM_{2.5}$) in the regional average was observed between those two resolutions. For the future climate condition, MDA8 ozone and average $PM_{2.5}$ were strongly affected by both climate and emissions. The emissions reduction had stronger effects on MDA8 ozone and average $PM_{2.5}$ than the effects from climate change for all three domains. For ozone, the effect from climate change increased the MDA8 ozone by about +1.5-2.0 ppbv, while the emissions reduction decreased the MDA8 ozone by about +5.0-7.0 ppbv. For $PM_{2.5}$, a 90% change in the future climate condition contributed by the emission reduction, where the climate change only contributed by about 10%.

CHAPTER V

5 SUMMARY

5.1 Declaration

This chapter is revised based on papers from: 1) Atmospheric Chemistry and Physics by Yun Fat Lam; Joshua Fu listed in chapter 4, and 2) Atmospheric Environment by Yun F. Lam, Joshua S. Fu, Shiliang Wu, Loretta J. Mickley, and Daniel J. Jacob (anticipated).

5.2 Summary from “**DOWNSCALING TECHNIQUE FOR THE LINKAGE OF GLOBAL AND REGIONAL AIR QUALITY MODELING**”

Recently, downscaling global atmospheric model outputs (GCTM) for the USEPA Community Multiscale Air Quality (CMAQ) Initial (IC) and Boundary Conditions (BC) have become practical because of the rapid growth of computational technologies that allow global simulations to be completed within a reasonable time. The traditional method of generating IC/BC by profile data has lost its advocates due to the weakness of the limited horizontal and vertical variations found on the gridded boundary layers. Theoretically, high quality GCTM IC/BC should yield a better result in CMAQ. Unfortunately, several researchers have found that the outputs from GCTM IC/BC are not necessarily better than profile IC/BC due to the excessive transport of O₃ aloft in GCTM IC/BC. In this paper, we intend to investigate the effects of using profile IC/BC and global atmospheric model data. In addition, we are suggesting a novel approach to resolve the existing issue in downscaling.

In the study, we utilized the GEOS-Chem model outputs to generate time-varied and layer-varied IC/BC for year 2002 with the implementation of tropopause determining algorithm in the downscaling process (i.e., based on chemical (O₃) tropopause definition). The comparison between the implemented tropopause approach and the profile IC/BC approach is performed to demonstrate improvement of considering tropopause. It is observed that without using tropopause information in the downscaling process, unrealistic O₃ concentrations are created at the upper layers of IC/BC. This phenomenon has caused over-prediction of surface O₃ in CMAQ. In addition, the amount of over-prediction is greatly affected by temperature and latitudinal location of the study domain. With the

implementation of the algorithm, we have successfully resolved the incompatibility issues in the vertical layer structure between global and regional chemistry models to yield better surface O₃ predictions than profile IC/BC for both summer and winter conditions. At the same time, it improved the vertical O₃ distribution of CMAQ outputs. It is strongly recommended that the tropopause information should be incorporated into any two-way coupled global and regional models, where the tropospheric regional model is used, to solve the vertical incompatibility that exists between global and regional models.

5.3 Summary from “IMPACTS OF FUTURE CLIMATE CHANGE AND EFFECTS OF BIOGENIC EMISSIONS ON SURFACE OZONE AND PARTICULATE MATTER CONCENTRATIONS IN UNITED STATES”

Simulations of current and future (i.e., 2000 and 2050) average regional ozone and PM_{2.5} concentrations over the United States were performed to investigate the potential impacts of global climate change and emissions on regional air quality using CMAQ. Various emissions and climate conditions with different biogenic emissions and domain resolutions were implemented to study the sensitivity of future air quality trends from the impacts of changing biogenic emissions (i.e., from MEGAN and BEIS) and domain resolutions (i.e., 4° x 5°, 36 km x 36 km, and 12 km x 12 km). A comparison of GEOS-Chem and CMAQ was performed to investigate the effect of downscaling on the prediction of future air quality trends. Meteorological inputs were obtained from dynamically downscaling outputs from GISS GCM into the MM5 regional climate model. Current-year emissions were based on United States EPA 2000 NEI inventories, while future-year emissions were projected from the IMAGE model under the IPCC A1B scenario. Over 50% of NO_x and SO₂ reductions were estimated in the future-year. For ozone, the impacts of global climate change are relatively smaller when compared to the impacts of anticipated future emissions reduction, except for Northeast area where increasing biogenic emissions due to climate change have stronger positive effects (increases) to the regional ozone air quality. The overall effect from both climate change and emission reductions leads to about 10% or 5 ppbv decrease of the maximum daily average 8-hr (MDA8) ozone over the eastern United States. For PM_{2.5}, the impacts of global climate change have shown insignificant

effect, where as the impacts of anticipated future emissions reduction account for the majority of overall PM_{2.5} reductions. The annual average 24-hr PM_{2.5} of the future-year condition was found to be about 40% lower than the one from the current-year condition, of which 60% of its overall reductions are contributed to by the decrease of SO₄ and NO₃ particulate matter. Changing the biogenic emissions model increases the MDA8 ozone by about 5-10% or 3-5 ppbv in the Northeast area. Conversely, it reduces the annual average PM_{2.5} by 5% or 1.0 µg/m³ in the Southeast area. Minor impacts were observed on the relationship between current-year and future-year conditions. For the sensitivity of resolution selection, the overall impact on a domain-wide average is relatively small. The comparison of GEOS-Chem and CMAQ shows GEOS-Chem predicted higher daily maximum 8-hr ozone than CMAQ by 10% and a significant disagreement of ozone trends in the Southeast region.

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Vita

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