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Online coupled meteorology and chemistry models: history, current status, and outlook

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Abstract

The climate-chemistry-aerosol-cloud-radiation feedbacks are important processes occurring in the atmosphere. Accurately simulating those feedbacks requires fully-coupled meteorology, climate, and chemistry models and presents significant challenges in terms of both scientific understanding and computational demand. This paper reviews the history and current status of development and application of online coupled models. Several representative online coupled meteorology and chemistry models developed in the U.S. such as GATOR-GCMOM, WRF/Chem, CAM3, MIRAGE, and Caltech unified GCM are included along with case studies. Major model features, physical/chemical treatments, as well as typical applications are compared with a focus on aerosol microphysics treatments, aerosol feedbacks to planetary boundary layer meteorology, and aerosol-cloud interactions. Recommendations for future development and improvement of online coupled models are provided.

1 Introduction

The climate-chemistry-aerosol-cloud-radiation feedbacks are important in the context of many areas including climate modeling, air quality/atmospheric chemistry modeling, numerical weather and air quality forecasting, as well as integrated atmospheric-ocean-land surface modeling at all scales. Some potential impacts of aerosol feedbacks include a reduction of downward solar radiation (direct effect); a decrease in surface temperature and wind speed but an increase in relative humidity and atmospheric stability (semi-direct effect), a decrease in cloud drop size but an increase in drop number via serving as cloud condensation nuclei (CCN) (first indirect effect), as well as an increase in liquid water content, cloud cover, and lifetime of low level clouds but a suppression or enhancement of precipitation (the second indirect effect). Aerosol feedbacks are traditionally neglected in meteorology and air quality modeling due largely to historical separation of meteorology, climate, and air quality communities as well as

our limited understanding of underlying mechanisms. Those feedbacks, however, are important as models accounting (e.g., Jacobson, 2002; Chung and Seinfeld, 2005) or not accounting (e.g., Penner et al., 2003) for those feedbacks may give different results (Penner, 2003; Feichter et al., 2003; Jacobson, 2003a, b) and future climate changes may be affected by improved air quality (Brasseur and Roeckner, 2005). Increasing evidence from field measurements have shown that such feedbacks ubiquitously exist among multimedia including the atmosphere, hydrosphere, lithosphere, pedosphere, and biosphere. For example, a stratocumulus cloud layer just below the advected pollutant layer observed during the 1993 North Atlantic Regional Experiment (NARE) was found to increase pollutant concentrations through the enhancement of the photolytic rates and oxidant levels (Audiffren et al., 2004). Satellite observations have shown that smoke from rain forest fires in tropical areas such as Amazon and Indonesia (Kaufman and Fraser, 1997; Rosenfeld and Lensky, 1998; Rosenfeld, 1999) and burning of agricultural vegetations (Warner, 1968; Rosenfeld and Woodley, 1999) can inhibit rainfall by shutting off warm rain-forming processes. This effect is due to the fact that large concentrations of small CCN in the smoke from biomass burning can nucleate many small cloud droplets, thus inhibiting cloud droplet coalescence into raindrops and riming on ice precipitation (Rosenfeld, 2000). While suppression of rain and snow by urban and industrial air pollution has been reported (Rosenfeld, 2000; Givati and Rosenfeld, 2004, 2005), enhanced rainfall, on the other hand, was also found downwind of urban areas or large sources such as paper mills (Eagen et al., 1974; Jauregui and Romales, 1996) and over major urban areas (Braham et al., 1981; Cerveny and Bailing, 1998), suggesting that giant CCN can enhance precipitation.

Despite significant progress has been made in modeling climate, meteorology, air pollution in the past several decades (Seaman, 2000; Seinfeld, 2004; Seigneur, 2005), several major deficiencies exist in most current global climate-aerosol models (e.g., Johnson et al., 1999, 2001; Mickley et al., 2004; Langner et al., 2005; Sanderson et al., 2006). First, the coarse spatial resolution (e.g., $4^{\circ}\times5^{\circ}$) used in those models cannot explicitly capture the fine-scale structure that characterizes climatic changes

(e.g., clouds, precipitation, mesoscale circulation, sub-grid convective system, etc.). Second, the coarse time resolution (e.g., 6 h average wind field) used in those models (one exception is GATOR/GCMOM, which typically updates meteorology every 5 minutes) cannot replicate variations at smaller scales (e.g., hourly and diurnal). Third, those models typically use simplified treatments (e.g., simple meteorological schemes and chemistry/aerosol microphysics treatments) that cannot represent intricate relationships among meteorology/climate/air quality variables. Fourth, most models simulate climate and aerosols offline with inconsistencies in transport and no climate-chemistry-aerosol-cloud-radiation feedbacks (e.g., Prather et al., 2003; Sanderson et al., 2006). At present, most global air quality models (GAQMs) are still offline. An empirical sulfate-CCN relation for aerosol indirect effects is typically used in most GAQMs. Some feedbacks are accounted for in some global climate/chemistry models (e.g., Chuang et al., 1997, 2002; Ghan et al., 2001a, b, c; Liao and Seinfeld, 2005) but either with simplified treatments or at a coarse resolution or both. Most air quality models at urban/regional scales, on the other hand, use offline meteorological fields without feedbacks and do not simulate aerosol direct and indirect effects (e.g., the EPA's Community Multiple Air Quality (CMAQ) modeling system, Byun and Ching, 1999; Binkowski and Roselle, 2003). Some air quality models are driven by a global model with inconsistent model physics (e.g., Hogrefe et al., 2004; Sanderson et al., 2006). Most regional climate models use prescribed aerosols or simple modules without detailed chemistry, aerosol microphysics, and aerosol-cloud interactions (e.g., Giorgi et al., 1993a, b; Giorgi and Shields, 1999).

The aforementioned model deficiencies in accurately representing atmospheric processes and feedbacks have led to the largest uncertainties in current estimates of direct and indirect effects of aerosols on climate (IPCC, 2001, 2007) as well as the impact of climate on air quality. Accurately simulating those feedbacks requires fully-coupled models for meteorological, chemical, physical, and biological processes and presents significant challenges in terms of both scientific understanding and computational demand. In this work, the history and current status of development and application of

online coupled models are reviewed. Several representative online coupled meteorology and chemistry models developed in the U.S. are used to illustrate the current status of online coupled models. Major model features, physical/chemical treatments, as well as typical applications of these models are evaluated with a focus on aerosol microphysics treatments, aerosol feedbacks to planetary boundary layer meteorology, and aerosol-cloud interactions. Major challenges and recommendations for future development and improvement will be provided.

2 History of online coupled climate/meteorology and air quality modeling

2.1 Concepts, history, and milestones of online coupled models

Atmospheric chemistry/air quality and climate/meteorology modeling were traditionally separated prior to 1970's. The three-dimensional (3-D) chemical transport models (CTMs) until that time were driven by either measured/analyzed meteorological fields or outputs at a time resolution of 1–6 h from a mesoscale meteorological model on urban/regional scale or outputs at a much coarser time resolution (e.g., 6 h or longer) from a general circulation model (GCM) (referred to as offline coupling). In addition to a large amount of data exchange, this offline separation does not permit simulation of feedbacks between air quality and climate/meteorology and may result in an incompatible and inconsistent coupling between both meteorological and air quality models and a loss of important process information (e.g., cloud formation and precipitation) that occur at a time scale smaller than that of the outputs from the offline climate/meteorology models (Seaman, 2000; Grell et al., 2005; Baklanov and Korsholm, 2007). Such feedbacks, on the other hand, can be simulated in the fully-coupled online models, without space and time interpolation of meteorological fields but commonly with higher computational costs.

Both offline and online models are actively used in current regional and global models. Offline models are frequently used in ensembles and operational forecasting, in-

verse modeling, and sensitivity simulations, whereas online models are increasingly used for applications in which the feedbacks become important (e.g., locations with high frequencies of clouds and large aerosol loadings), the local scale wind and circulation system change quickly, and the coupled meteorology-air quality modeling is essential for accurate real-time operational forecasting. Reported differences in simulation results from online and offline models can be fairly small or quite significant, depending on level of complexities of the model treatments and the simulated variables/fields. For example, Mickley et al. (1999) found that differences in the simulated radiative forcing of anthropogenic ozone (O_3) from their global chemistry-climate model operated online and offline are within 2%. Shindell et al. (2001) found that the tropospheric oxidation capacity in terms of hydroxyl radical (OH) simulated by their online coupled model is lower by ∼10% than that of the same model but running offline. Jacobson (2002) and Chung and Seinfeld (2005) reported a positive forcing of fossil-fuel particulate black carbon and organic matter using their online coupled models, whereas other models that do not account for aerosol feedbacks give a strong negative forcing (e.g., Penner et al., 2003). Nevertheless, there is an increasing recognition from science communities that online coupled model systems represent the true, one atmosphere and are urgently needed, although there remain significant work for such models to be mature and their applications are currently limited by computational constrains.

Regardless of the temporal and spatial scales of applications, online coupled models provide powerful platforms for reproducing the feedbacks among multiple processes and variables in varying degrees in one-atmosphere, depending on the framework and degree of the coupling in those models. Two coupling frameworks are conventionally used in all mesoscale and global online coupled models: one couples a meteorology model with an air quality model in which the two systems operate separately but exchange information every time step through an interface (referred to as separate online coupling), the other integrates an air quality model into a meteorology model as a unified model system in which meteorology and air quality variables are simulated together in one time step without a model-to-model interface (referred to as unified on-

line coupling). Transport of meteorological and chemical variables is typically simulated with separate schemes in separate online models but the same scheme in unified online models. Depending on the objectives of the applications, the degrees of coupling and complexities in coupled atmospheric processes in those models vary, ranging from a simple coupling of meteorology and gas-phase chemistry (e.g., Rasch et al., 1995; Grell et al., 2000) to the most sophistic coupling of meteorology, chemistry, aerosol, radiation, and cloud (e.g., Jacobson, 1994, 2004b, 2006a; Grell et al., 2002, 2005). While online coupled models can in theory enable a full range of feedbacks among major components and processes, the degree of coupling in those models varies substantially from slightly-coupled to moderately- or significantly-, or fully-coupled. In the slightly- or moderately-coupled models, only selected species other than water vapor (e.g., O_3 or aerosols) and/or processes (e.g., transport of chemical species other than water vapor or gas-phase chemistry) are coupled and other processes (e.g., solar absorption of O_3 and total radiation budget) remain decoupled. Feedbacks among processes may or may not be accounted for. In the significantly- or fully-coupled models, major processes are coupled and a full range of atmospheric feedbacks are realistically simulated. At present, very few significantly- or fully-coupled online models exist. Most online models are still under development; they are slightly- or moderately-coupled with little or no feedbacks among major atmospheric processes. Depending on the coupled components/processes, those online models can be generally grouped into four main categories: online meteorology and pollutant transport; online meteorology and pollutant transport and chemistry; online pollutant feedbacks to heating rates to drive meteorology; and online pollutant feedbacks to photolysis to drive photochemistry. Examples of each category are given in Table 1; they represent various degrees of coupled treatments for each category, varying from highly simplified to the most sophistic one.

Figure 1 shows chronology of the development history and major milestones in terms of transport of gaseous and aerosols species, their chemistry, and feedbacks among major atmospheric processes for online coupled models on all scales. The earliest

attempt in coupling global climate/meteorology and chemistry can be traced back to late 1960's, when 3-D transport and O_3 and very simple stratospheric chemistry (e.g., the Chapman reactions) were first incorporated into a GCM to simulate global O_3 production and transport simultaneously (e.g., Hunt, 1969; Clark, 1970). Coupled climate-chemistry GCMs developed in mid-late 1970's included additional reactions (e.g., the nitrogen oxides (NO_x) catalytic cycle, and reactions between hydrogen and atomic oxygen) and accounted for the effects of predicted O_3 (but not other gases) on radiation heating and the effect of O_3's heating on atmospheric circulation, which in turn affected the distributions of O_3 (e.g., Cunnold et al., 1975; Schlesimger and Mintz, 1979). 3-D transport of bulk aerosols and their feedbacks into radiation heating to drive meteorology were also be included in some early coupled GCMs (e.g., Atwater, 1975; Joseph, 1976; Covey et al., 1984; Thompson, 1985; Cess e al., 1985; Malone et al., 1986; Ghan et al., 1988). Since the mid 1980's, a larger number of online coupled global climate-chemistry models with various degrees of coupling to chemistry have been developed to address the Antarctic/stratospheric O_3 depletion (e.g., Cariolle et al., 1986, 1990; Rose and Brasseur, 1989; Granier and Brasseur, 1991; Austin and Butchart, 1992; Austin et al., 1992, 2000; Pitari et al., 1992, 2002; Hack et al., 1993; Rasch et al., 1995; Jacobson, 1995; Eckman et al., 1996; Beagley et al., 1997; Shindell et al., 1998; Dameris et al., 1998, 2005; Takigawa et al., 1999; Rozanov et al., 2001; Nagashima et al., 2002; Schnadt et al., 2002), tropospheric O_3 and sulfur cycle (e.g., Levy et al., 1985; Roelofs and Lelieveld, 1995; Roelofs et al., 1998; Feichter et al., 1996, 1997; de Laat et al., 1999; Mickley et al., 1999; Rasch et al., 2000; Barth et al., 2000; Shindell et al., 2001; Grenfell et al., 2001; Wong et al., 2004), and tropospheric aerosols, their direct radiative forcing and interactions with clouds (e.g., Taylor and Penner, 1994; Chuang et al., 1997, 2002; Lohmann and Feichter, 1997; Koch et al., 1999; Kiehl et al., 2000; Lohmann et al., 2000; Jacobson, 2000, 2001a, 2002; Ghan et al., 2001a, b, c; Boucher and Pham, 2002; Menon et al., 2002; Gong et al., 2002, 2003; Iversen and Seland, 2002; Derwent et al., 2003; Liao et al., 2003; Easter et al., 2004; Hauglustaine et al., 2004; Tulet et al., 2005). Such online coupled models have

also been expanded to study climate-carbon cycle-chemistry feedbacks in the middle atmosphere (e.g., Steil et al., 2003; Manzini et al., 2003), and the interactions among atmosphere, biosphere, ocean and land systems (referred to as earth system modeling) since late 1990's (e.g., Prinn et al., 1999; Gordan et al., 2000; Neelin and Zeng, 2000; Cox, 2001; Johnson et al., 2001; Khodri et al., 2001; Jacobson, 2004b, 2005b, 2006a; Jöckel et al., 2005; Collins et al., 2006b; Chou et al., 2006; Doney et al., 2006; Jungclaus et al., 2006; O'Connor et al., 2006). Some online coupled GCMs for stratospheric chemistry have been reviewed in Austin et al. (2003) and Eyring et al. (2005); those for tropospheric chemistry have been reviewed in Ghan et al. (2001c), Easter et al. (2004), and Ghan and Schwartz (2007), and those for earth system modeling have been reviewed in Friedlingstein et al. (2006).

The coupling in most current global online coupled climate-chemistry models, however, is largely incomplete; and has been done only for very limited prognostic gaseous species such as O_3 and/or bulk sulfate aerosol or selected processes such as transport and gas-phase chemistry (i.e., slightly- or moderately-coupling, e.g., Hunt, 1969; Atwater, 1975; Schlesimger and Mintz, 1979; Taylor and Penner, 1994). This is mainly because such a coupling typically restricts to gas-phase or parameterized chemistry (and heterogeneous chemistry in some cases) and simple aerosol/cloud chemistry and microphysics and often neglects the feedbacks between prognostic chemical species (e.g., O_3 and aerosols) and radiation (e.g., Roelofs and Lelieveld, 1995; Eckman et al., 1996; Barth et al., 2000; Wong et al., 2004) and between aerosols and clouds (e.g., Liao et al., 2003). There are, however, a few exceptions after mid 1990's when significantly- or fully-coupled systems were developed to enable a full range of feedbacks between meteorology/climate variables and a myriad of gases and size-resolved aerosols (e.g., Jacobson, 1995, 2000; Ghan et al., 2001a, b, c).

While a large number of online coupled global climate-chemistry GCMs have been developed for simulating global climate change and air quality studies for more than three decades, there exist fewer coupled meteorology- (or climate-) chemistry models at urban and regional scales. This is largely due to the historic fact that mesoscale

meteorology models and air pollution models were developed separately. The development of mesoscale coupled meteorology-chemistry models was driven by the needs for forecasting air quality in real-time and simulating feedbacks between air quality and regional climate as well as responses of air quality to changes in future regional climate, land use, and biogenic emissions. The earliest attempt in coupling meteorology and air pollution in local to regional scale models can be traced back to early 1980's. The one-way coupling of 3-D transport of gases and gas-phase chemistry with meteorology was included at meso-to-regional scales (e.g., Marchuk, 1982; Penenko et al., 1984; Penenko and Aloyan, 1985; and Bazhin et al., 1991) and local-to-meso scale (e.g., Aloyan et al., 1982; Baklanov, 1988). In addition to the one-way coupling of transport and gas-phase chemistry, Baklanov (1988) also simulated highly-simplified aerosol treatments and the direct radiation feedbacks of bulk aerosols to heating/reflection and other atmospheric processes at a local scale (Alexander Baklanov, personal communication, Danish Meteorological Institute, 2007). Some of those early online models were briefly reviewed in Baklanov (1990, 2007). Since then, a number of mesoscale online coupled meteorology-chemistry models have been developed in North America (e.g., Jacobson, 1994, 1997a, b; Mathur et al., 1998; Xiu et al., 1998; Côté et al., 1998; Grell et al., 2000; Kaminski, 2007), Asia (e.g., Uno et al., 2001; 2003), Australia (e.g., Manins, 2007), and Europe through the European Cooperation in Science and Technology (COST) action 728 (http://www.cost728.org) (e.g., Wolke et al., 2003; Chenevez et al., 2004; Baklanov et al., 2004, 2007a, b, and references therein; Vogel et al., 2006; Vogel, 2007; Maurizi, 2007; Korsholm et al., 2007). Among these models, the most representative work was done by Jacobson (1994, 1997a, b), in which chemistry was solved for all transported gases; all chemically-active gases and size-resolved aerosol components were transported; and feedbacks of all photolyzing gases and aerosols to meteorology through heating rates and to photolysis through actinic fluxes were treated (see Table 1). Some of the online meteorology-chemistry models have been coupled with population exposure and health effects (e.g., Jacobson, 2007; Baklanov et al., 2007b). Several online coupled regional climate-chemistry/aerosol models have also been de-

veloped since late 1999, with either a sulfate-like tracer (e.g., Qian and Giorgi, 1999) or highly simplified sulfate chemistry (e.g., Qian et al., 2001; Giorgi et al., 2002) simulated in a regional climate model. The coupling was enabled partially, i.e., only between meteorology and tropospheric gas-phase chemistry in some regional online models (e.g., Grell et al., 2000; Taghavi et al., 2004; Arteta et al., 2006); and significantly to fully, i.e., among more processes/components including meteorology, chemistry, aerosols, clouds, and radiation (e.g., Jacobson, 1994, 1997a, b; Jacobson et al., 1996; Mathur et al., 1998; Grell et al., 2005; Fast, 2005; Fast et al., 2006; Zhang et al., 2005a, b; Hu and Zhang, 2006; Korsholm et al., 2007; Sofiev, 2007; and Knoth and Wolke, 2007). Similar to global models, the feedbacks between meteorology and chemical species are often neglected in many local-to-regional scale online models (e.g., Uno et al., 2001, 2003), and a full range of climate-chemistry-aerosol-cloudradiation feedbacks is treated in very few mesoscale models (e.g., Jacobson, 1994, 1997a, b; Grell et al., 2005).

2.2 History of representative online coupled models in the U.S.

The current status of a number of online models in Europe has been reviewed in Baklanov and Korsholm (2007) and Baklanov et al. (2007a). In this work, five online models on both regional and global scales developed in the U.S. are selected to represent the current status of online coupled models and reviewed in details. These models include one global-through-urban model, i.e., the Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR-GCMOM) (Jacobson, 2001b, 2002, 2004a, b; Jacobson et al., 2004), one mesoscale model, i.e., the Weather Research and Forecast model with Chemistry (WRF/Chem) (Grell et al., 2005; Fast et al., 2006), and three global models, i.e., the Community Atmospheric Model version 3 (CAM3) (Collin et al., 2006a), the Model for Integrated Research on Atmospheric Global Exchanges (MIRAGE) (Ghan et al., 2001a, b; Easter et al., 2004), and the Caltech unified GCM (Liao et al., 2003, 2004, 2006; Liao and Seinfeld, 2005). All these models predict gases, aerosols, and clouds with varying degrees of complexities in

chemical mechanisms and aerosol/cloud microphysics. In the following section, history and current status of the five models along with other relevant models developed in the U.S. will be reviewed.

Jacobson (1994) developed a unified fully-coupled online meteorology/chemistry/aerosol/radiation model on urban/regional scale: a gas, aerosol, transport, and radiation air quality model/a mesoscale meteorological and tracer dispersion model (GATOR/MMTD, also called GATORM) (Jacobson, 1994; 1997a, b; Jacobson et al., 1996). This is the first fully-coupled online model in the history that accounts for all major feedbacks among major atmospheric processes based on first principles (Jacobson, 2006a), since early work on the coupling of meteorology and chemistry were either significantly or somewhat incomplete and the feedbacks among multiple processes in those online models were either omitted or largely simulated with simplified parameterizations. In an early version of GATOR/MMTD, all meteorological and chemical processes were solved simultaneously online but with separate transport schemes for meteorological and chemical variables. The two-way feedbacks between gases/aerosols and meteorology through solar and thermal-IR radiative transfer were accounted for. The same transport scheme was developed for GATOR/MMTD in 1997 to solve transport of water vapor, energy, and column pressure in MMTD and of chemical species in GATOR (Jacobson, 1997c). GATOR/MMTD has been applied to simulate gases and aerosols over Los Angeles (LA) Basin (Jacobson et al., 1996; Jacobson, 1997a, b), the effects of aerosols on vertical photolysis rate and temperature profiles (Jacobson, 1998), nitrated and aromatic aerosols and nitrated aromatic gases as sources of ultraviolet light absorption (Jacobson, 1999a), the effects of soil moisture on temperatures, winds, and pollutant concentrations in LA (Jacobson, 1999b), and the effects of different vehicle fuels on cancer and mortality (Jacobson, 2007). The results from those model applications have been rigorously evaluated with available measurements.

Grell et al. (2000) developed a unified coupled online meteorology and chemistry model: Multiscale Climate Chemistry Model (MCCM, also called Mesoscale

Model (MM5)/Chem). In this model, the Penn State University (PSU) /NCAR nonhydrostatic mesoscale model (MM5, Grell et al., 1994) was coupled online only with the gas-phase chemical mechanism of Regional Acid Deposition Model, version 2 (RADM2, Chang et al., 1989; Stockwell et al., 1990). No aerosol and radiation processes were treated in MM5/Chem. MM5/Chem was applied and evaluated with several testbeds in the U.S. (e.g., McKeen et al., 2003; Eder et al., 2005; Bao et al., 2005; Kang et al., 2005; Kim and Stockwell, 2007). Built upon their work on MM5/Chem, Grell at al. (2002) developed a unified significantly-coupled meteorology/chemistry/aerosol/radiation model, WRF/Chem following the first workshop on Modeling Chemistry in Cloud and Mesoscale Models held at National Center for Atmospheric Research (NCAR) in March 2000. WRF/Chem represents the first community online coupled model in the U.S. Since its first public release in 2002, WRF/Chem has attracted a number of external developers and users from universities, research organizations, and private sectors to continuously and collaboratively develop, improve, apply, and evaluate the model. Although the coupling of all simulated processes in current version of WRF/Chem is not as completed as that of GATOR/MMTD and some couplings are still partially completed and/or largely based on parameterizations (e.g., feedbacks of photochemically-active gases and aerosols to photolysis via Fast-J photolysis algorithm), the degree of coupling for many atmospheric processes is much more significantly as compared with earlier work and will become more complete as more developers from community contribute to its further development. In WRF/Chem, transport of meteorological and chemical variables is treated using the same vertical and horizontal coordinates and the same physics parameterization with no interpolation in space and time. The meteorological model was based on the NCAR's WRF that offers options for hydrostatic and nonhydrostatic, with several dynamic cores (e.g., the Advanced Research WRF with the Eulerian Mass (ARW) and the Nonhydrostatic Mesoscale Model NMM), and many options for physical parameterizations for applications at different scales. The chemistry model of WRF/Chem was largely based on MM5/Chem of Grell et al. (2000), but with an additional gas-phase mechanism:

the Regional Atmospheric Chemistry Mechanism (RACM) of Stockwell et al. (1997) and a new aerosol module: the Modal Aerosol Dynamics Model for Europe (MADE) (Ackermann et al., 1998) with the secondary organic aerosol model (SORGAM) of Schell et al. (2001) (referred to as MADE/SORGAM). The photolytic rates of photochemical reactions are calculated online using the Tropospheric Ultraviolet and Visible radiation model (TUV) algorithm of Madronich (1987), in which the radiative transfer model of Chang et al. (1989) is used to calculate actinic flux due to absorption by two gases (i.e., O_2, O_3), Rayleigh scattering, and scattering and absorption by aerosols and clouds. The feedbacks of gases and aerosols to radiation heating are simulated using atmospheric longwave radiation schemes (e.g., the RRTM of Mlawer et al. 1997) and the shortwave radiation schemes (e.g., the MM5 scheme of Dudia (1989) and the Goddard scheme of Chou and Suarez 1994) (Skamarock et al., 2005). RRTM is a spectral-band scheme based on the correlated-k method and uses precalculated tables to simulate feedbacks to longwave due to water (H_2O), O_3, carbon dioxide (CO_2), other trace gases, and clouds. The MM5 shortwave scheme simulates a simple downward integration of solar flux. It accounts for clear-air scattering and absorption of H_2O only (instead of all photolyzing gases) using parameterizations and cloud albedo and absorption using look-up tables. The Goddard shortwave scheme is used in a two-stream approach that accounts for scattered and reflected components over 11 spectral bands.

Two additional gas-phase mechanisms, two new aerosol modules, and one photolytic algorithm have been recently incorporated into WRF/Chem by external developers (Fast, 2005; Zhang et al., 2005a, 2007; Fast et al., 2006; Hu and Zhang, 2006, 2007; Huang et al., 2006). The two new gas-phase mechanisms are the Carbon-Bond Mechanism version Z (CBMZ) (Zaveri and Peters, 1999) and the 2005 version of Carbon Bond mechanism (CB05) of Yarwood et al. (2005) and Sarwar et al. (2005, 2006) (both are variants of Carbon Bond Mechanism IV (CBMIV) of Gery et al., 1989). The two new aerosol modules are the Model for Simulating Aerosol Interactions and

Chemistry (MOSAIC) (Zaveri et al., 2008[1]) and the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (Zhang et al., 2004). RADM2 and RACM have been coupled with MADE/SORGAM and CBMZ has been coupled with MOSAIC and MADRID; CB05 is being coupled with MOSAIC and MADRID (Zhang et al., 2007). While MADE/SORGAM uses a modal approach with three lognormally-distributed modes to represent aerosol size distribution, the sectional approach with a number of size sections (currently with 8 sections, but it can be changed to any number of sections) is used in MOSAIC and MADRID. An alternative photolysis algorithm, the Fast-J scheme of Wild et al. (2000), has been incorporated into WRF/Chem by Fast et al. (2006). Fast-J scheme computes photolysis rates from the predicted O_3, aerosol, and clouds following a Legendre expansion of the exact scattering phase function. CBM-Z can use the photolysis rates from either Fast-J or TUV. The aerosol optical depth, single scattering albedo, and phase function expansion coefficients are calculated as a function of the refractive indices and size distribution based on predicted aerosol mass and composition. WRF/Chem and its variations have been applied for both real-time forecasting (e.g., Grell et al., 2005; Kang et al., 2005; McKeen et al., 2005; 2007; Pagowski et al., 2006) and hindcasting (e.g., Fast, 2005; Fast et al.,2006; Zhang et al., 2005a, 2007; Frost et al., 2006; Hu and Zhang, 2006; Huang et al., 2006; Hu et al., 2007).

On a global scale, a number of climate or air quality models have been developed in the past three decades among which very few of them are online coupled models (e.g., the NCAR Community Climate Model (CCM) (which was renamed later as Community Atmospheric Model CAM); the Pacific Northwest National Laboratory (PNNL)'s MIRAGE; the Stanford University's GATORG (which was later extended as a global-through-urban model, GATOR/GCMOM), and the Caltech unified GCM). Since its initial development as a general circulation model without chemistry, CCM0 and CCM1 (Washington, 1982; Williamson et al., 1987), the NCAR CCM has evolved to be one

[1]Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for simulating aerosol interactions and chemistry, J. Geophys. Res., submitted, 2008.

of the first-generation unified online climate-chemistry models in the U.S. following pioneer work by Hunt (1969) and Clark (1970), initially with gas-phase chemistry only (e.g., CCM2 (Hack et al., 1993; Rasch et al., 1995) and CCM3 (Kiehl et al., 1998; Rasch et al., 2000; Barth et al., 2000)) and most recently with additional aerosol treatments (e.g., CAM3 (Collins et al., 2004, 2006a, b; Rasch et al., 2006a, b) and CAM4 (http://www.ccsm.ucar.edu)).

Jacobson (1995, 2000, 2001a) developed a unified fully-coupled Gas, Aerosol, TranspOrt, Radiation, and General circulation model (GATORG). Similar to GATOR-MMTD on urban/regional scales, this is the first fully-coupled global online model in the history that accounts for all major feedbacks among major atmospheric processes based on first principles. While the gas-aerosol-radiation modules in GATORG are the same as those in GATORM, GATORG uses a 1994 version of the University of Los Angeles General Circulation Model (UCLA-GCM) (Arakawa and Lamb, 1981) to generate meteorology. GATORG was used to study global direct aerosol radiative forcing (Jacobson, 2000, 2001a). Jacobson (2001b, c) linked the regional GATORM and global GATORG and developed the first in the history unified, nested global-through-urban scale Gas, Aerosol, Transport, Radiation, General Circulation, and Mesoscale Meteorological model, GATOR-GCMM. GATOR-GCMM was designed to treat gases, size- and composition-resolved aerosols, radiation, and meteorology for applications from the global to urban (<5 km) scales and has switches to run in global mode, regional mode, nested mode, and with/without gases, aerosols and cloud microphysics, radiation, meteorology, transport, deposition and sedimentation, and surface processes. All processes in all nested domains were exactly the same, except for the horizontal boundary conditions and solutions to the momentum equation that are different on global and regional scales. GATOR-GCMM accounts for radiative feedbacks from gases, size-resolved aerosols, liquid water and ice particles to meteorology on all scales and has been applied to study weather and tropospheric ozone in northern and central California and global direct forcing of black carbon (Jacobson, 2001c, d, 2002). GATOR-GCMM was extended to Gas, Aerosol, TranspOrt, Radiation, General Circu-

lation, Mesoscale, Ocean Model (GATOR-GCMOM) in Jacobson (2004a, b, 2005b, 2006b) and Jacobson et al. (2004, 2006b, 2007) by addition of a 2-D ocean module with 3-D energy diffusion to the deep ocean and treatments of multiple-distribution, size-resolved cloud hydrometeors and interactions between these hydrometeors and size- and distribution-resolved aerosols. GATOR-GCMOM has been applied to study the effect of black carbon (BC) within clouds and precipitation on global climate (Jacobson, 2006b), feedbacks of aerosols to urban climate over California (Jacobson et al., 2007), and effects of ethanol versus gasoline vehicles on cancer and mortality in the U.S. (Jacobson, 2007).

MIRAGE consists of a climate model and a chemical transport model that can be run offline or coupled online (Ghan et al., 2001 a, b, c; Easter et al., 2004). The climate model is the PNNL version of the NCAR CCM2. The chemical transport model is the PNNL Global Chemistry Model (GChM). MIRAGE has been applied to simulate global air quality, aerosol direct and indirect radiative forcing and evaluated rigorously with available gas, aerosol, and cloud measurements (e.g., Ghan et al., 2001a, b, c; Easter et al., 2004). Several other online coupled global climate/aerosol models with full oxidant chemistry have also been developed since early 2000 but most of them do not include all feedbacks, in particular, aerosol indirect effects; and they are still under development (e.g., Liao et al., 2003). Among all 3-D models that have been developed for climate and air quality studies at all scales, GATOR-GCMOM, MIRAGE, and WRF/Chem represent the state of the science global and regional coupled models; and GATOR-GCMOM (Jacobson, 2001 a, b, c, 2004 a, b) appears to be the only model that represents gas, size- and composition-resolved aerosol, cloud, and meteorological processes from the global down to urban scales via nesting, allowing feedbacks from gases, aerosols, and clouds to meteorology and radiation on all scales in one model simulation.

3 Current treatments in online coupled models

In this section, model features and treatments of major aerosol and cloud processes for the five representative online coupled meteorology and chemistry models developed in the U.S. (i.e., GATOR-GCMOM, WRF/Chem, CAM3, MIRAGE2, and Caltech unified GCM) are reviewed and intercompared. The review is presented in terms of model systems and typical applications, aerosol and cloud properties, aerosol and cloud microphysics and aerosol-cloud interactions. As shown in Table 2, the five models consist of a meteorology model (either a GCM or a mesoscale model) and a chemical transport model with different levels of details in gas-phase chemistry and aerosol and cloud treatments ranging from the simplest one in CAM3 to the most complex one in GATOR/GCMOM. GATOR/GCMOM uses an extended Carbon Bond mechanism (CBM-EX) with 247 gas-phase reactions among 115 chemical species. Its aqueous chemical mechanism simulates 64 kinetic aqueous-phase reactions for sulfate, nitrate, organics, chlorine, oxidant, and radical chemistry and offers options for bulk or size-resolved chemistry. Its aerosol and cloud modules provide comprehensive treatments for size-resolved, prognostic aerosol/cloud properties and processes. WRF/Chem offers four options for gas-phase mechanisms (i.e., RADM2, RACM, CBMZ, and CB05) with 156–237 chemical reactions among 52–77 chemical species and three aerosol modules (i.e., MADE/SORGAM, MOSAIC, and MADRID), although not all gas-phase mechanisms are currently coupled with the same aqueous-phase chemical mechanism or the same aerosol module. MADE/SORGAM uses the bulk RADM aqueous-phase chemistry that simulates aqueous-phase chemistry of sulfate with 5 kinetic reactions, MOSAIC/MADRID simulates bulk Carnegie Mellon University aqueous-phase mechanism for chemistry of sulfate, nitrate, and oxidants that includes 147 reactions among 71 species. While all three aerosol modules provide size-resolved (in terms of either mode or section) prognostic aerosol treatments, they differ in many aspects of aerosol treatments for thermodynamics and dynamics. All three aerosol modules simulate aerosol direct radiative forcing, MOSAIC also simulates aerosol indirect forcing. CAM3

offers gas-phase chemistry with different level of details, a simple mechanism with prescribed methane (CH_4), nitrous oxide (N_2O), chlorofluorocarbons (CFCs), radicals (e.g., OH, hydroperoxy radical (HO_2) and nitrate radical (NO_3)), and oxidants (e.g., O_3) and simulated sulfur dioxide (SO_2)/dimethyl sulfide (DMS) chemistry and a more comprehensive mechanism with 167 chemical reactions among 63 species from the Model for Ozone and Related Chemical Tracers version 4 (MOZART4). It simulates bulk sulfate chemistry with dissolution equilibria of SO_2, hydrogen peroxide (H_2O_2), O_3, and sulfurous acid (H_2SO_3) and aqueous-phase kinetic reactions of dissolved sulfur compounds with oxidation state IV (S(IV)) with H_2O_2 and O_3. It includes prognostic aerosol/cloud treatments but with prescribed size distribution for all aerosol components except for dust and sea salt. MIRAGE2 includes carbon monoxide (CO)-CH_4-oxidant chemistry and oxidation of SO_2 and DMS by OH with prescribed CH_4, NO_x, and O_3. Its aqueous-phase chemistry includes dissolution equilibria of SO_2, H_2O_2, methyl hydroperoxide (CH_3O_2H), O_3, sulfuric acid (H_2SO_4), and methane sulfonic acid (MSA) and aqueous-phase kinetic reactions of S(IV) with H_2O_2, CH_3O_2H, and O_3 in cloud water. It provides mode-resolved simple aerosol treatment with prognostic aerosol/cloud properties and processes. Caltech unified GCM uses the Harvard tropospheric O_3-NO_x-hydrocarbon chemistry with 305–346 reactions among 110–225 species. Its bulk aqueous-phase chemistry simulates aqueous-phase oxidation of S(IV) by H_2O_2 and O_3. Among the five models, it has the simplest aerosol treatments and no treatments for aerosol-cloud interactions.

Those models have been developed for different applications. GATOR-GCMOM has been applied for simulation of feedbacks among meteorology, chemistry and radiation on urban-to-global scales for both current and future emission/climate scenarios, estimates of global aerosol direct/indirect effects (e.g., Jacobson, 2002), and the linkage between air quality and health effect (Jacobson, 2007). WRF/Chem was developed and applied for real-time air quality forecasting (e.g., Grell et al., 2005; Kang et al., 2006), although it has also been applied retrospectively for simulating concentrations and distributions of tropospheric O_3 and particles with aerodynamic diameters less

than or equal to 2.5 μm ($PM_{2.5}$) (e.g., Fast, 2005; Zhang et al., 2005a; Fast et al., 2006). The feedbacks between meteorology and chemistry via aerosol radiation are studied; aerosol indirect effect through affecting cloud formation, lifetime, and precipitation is being studied with MOSAIC (Jerome Fast, personal communication, PNNL, 2007; Zhang et al., 2007, 2008a). CAM3 and its predecessors were developed for global climate applications to simulate global aerosol direct/indirect effects (e.g., Kiehl et al., 2000; Collins et al., 2006a), global transport and chemistry of trace gas species (e.g., Rasch et al., 1994, 2000; Barth et al., 2000), global climate dynamic circulation (Hurrell et al., 2006) and global hydrological cycle (Hack et al., 2006). MIRAGE2 and its predecessors were developed to simulate global climate and aerosols. It has been applied to simulate global transport and chemistry of trace gases and aerosols (e.g., Easter et al., 2004) and global cloud radiative forcing (e.g., Ghan et al., 1997a, b) and aerosol direct/indirect effects (e.g., Ghan et al., 2001a, b, c). Caltech unified GCM has been applied to simulate global chemistry-aerosol interactions; aerosol direct radiative forcing; the role of heterogeneous chemistry; impact of future climate change on O_3 and aerosols (Liao et al., 2003, 2004, 2006; Liao and Seinfeld, 2005).

As shown in Table 3, the treatments of aerosol properties in those models are different in terms of composition, size distribution, aerosol mass/number concentrations, mixing state, hygroscopicity, and radiative properties. MIRAGE2 treats the least number of species including sulfate, BC, organic carbon (OC), sea-salt, and dust. CAM3, WRF/Chem, and Caltech unified GCM treat the same species but with nitrate and ammonium. GATOR/GCMOM treats 47 species including sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, crustal species and their salts. Both CAM3 and MIRAGE2 use modal approach with four modes to represent aerosol size distributions. GATOR/GCMOM uses sectional approach with 17–30 size sections for typical applications. WRF/Chem offers both approaches depending on the aerosol module selected (e.g., modal approach with 3 modes for MADE/SORGAM, and sectional approach for MOSAIC and MADRID). Caltech unified GCM prescribes size distribution of sea-salt and dust with the sectional distribution but that of other aerosols with the modal dis-

tribution. Size distribution of all aerosol components are prescribed in Caltech unified GCM and that of all aerosols except sea-salt and dust is prescribed in CAM3; they are predicted in the other three models. Prescribed aerosol size distribution may introduce significant biases in simulated aerosol direct and indirect radiative forcing that highly depends on aerosol size distributions. The mixing state of aerosols affects significantly the predictions of direct/indirect radiative forcing. For example, the direct forcing of BC is 0.27, 0.78, and 0.54 $W\,m^{-2}$ for externally-mixed, well-mixed, and core treatments, respectively (Jacobson, 2000). The core treatment results in values of absorption/scattering coefficients and single scattering albedo that are lower than those with well-mixed treatment but higher than those with the externally-mixed assumption. Most models assume aerosols to be either completely externally- or internally-mixed. The internally-mixed (i.e., well-mixed) hydrophilic treatment for BC is unphysical and reality lies between the externally-mixed, hydrophobic and core treatments. Available measurements indicate that BC particles are coated with a shell containing other soluble species such as sulfate, nitrate, and ammonium (e.g., Katrlnak et al., 1992, 1993; Pósfai et al., 1999). Among the five models, GATOR/GCMOM is the only model treating internal/external aerosol mixtures with a coated BC core. It treats one or more size distributions with the latter representing aerosols with different sources and mixing states (e.g., freshly-emitted BC, internally mixed aerosols, and aerosols with a coated BC core), other four models treat a single aerosol distribution in either external or internal mixtures (e.g., external mixture in CAM3, internal mixture in WRF/Chem, externally mixed modes with internal mixtures within each mode in MIRAGE2, and BC, OC, and mineral dust externally-mixed with internally-mixed other aerosols in Caltech unified GCM).

All the five models predict aerosol mass concentration, which can also be prescribed in CAM3 and MIRAGE2. Aerosol number concentration is diagnosed from mass and size distribution in CAM3, predicted in GATOR/GCMOM, either diagnosed or predicted in WRF/Chem and MIRAGE2, but it is not included in the Caltech unified GCM. The simulated aerosol direct and indirect forcing depend on particle size and hygroscop-

icity, which should be included in atmospheric models for an accurate prediction. GATOR simulates hydrophobic-to-hydrophilic conversion for all aerosol components, MIRAGE2, WRF/Chem, and Caltech unified GCM simulate this conversion but with prescribed hygroscopicities. For example, MIRAGE assumes a hydroscopicity of 0.14 for OC, which is one-fourth of the value for ammonium sulfate (0.51). For BC, a very small nonzero value (10^{-10}) is assumed to avoid computational difficulties (Ghan et al., 2001a). In Caltech unified GCM, this conversion is simulated by assuming an exponential decay lifetime of 1.15 days (Liao et al., 2003). CAM3 treats hydrophobic and hydrophilic BC/OC but with a fixed conversion rate. It also prescribes the hygroscopicity of individual aerosol components. One difference between MIRAGE and CAM3 is that MIRAGE treats BC and OC from boreal fires, but CAM3 does not. For aerosol radiative properties, refractive indices (RIs) vary as a function of particle size and composition for both aerosols and cloud droplets (as well as precipitation). GATOR/GCMOM assumes a BC core surrounded by a shell where the RIs of the dissolved aerosol components are determined from partial molar refraction theory and those of the remaining aerosol components are calculated to be volume-averaged based on core-shell Mie theory. MIRAGE2, WRF/Chem, and Caltech unified GCM predict RIs and optical properties using Mie parameterizations that are function of wet surface mode radius and wet RI of each mode. Volume mixing is assumed for all components, including insoluble components. The main difference between Caltech unified GCM and MIRAGE2 (and WRF/Chem) is that Caltech unified GCM prescribes size distribution (e.g., sectional distribution for sea-salt and dust and standard gamma distribution other aerosols), but MIRAGE2 predicts it. Caltech unified GCM assumes that dust is externally-mixed with internal mixtures of other aerosols (which is different from the aerosol mixing state assumption used in the aerosol thermodynamics simulation). In CAM3, RIs and optical properties are prescribed for each aerosol type, size, and wavelength of the external mixtures.

Table 4 summarizes model treatments of cloud properties, reflecting the levels of details in cloud microphysics treatments from the simplest in Caltech unified GCM to the

most sophistic in GATOR-GCMOM. Hydrometeor types in clouds in GATOR/GCMOM include size-resolved liquid, ice, graupel, and aerosol core components. Liquid drops are assumed to be spherical. Ice crystals and graupel are assumed to be non-spherical. Their non-sphericity is modeled as a collection of spheres of the same total volume-to-area ratio and total volume as the nonspherical particles. GATOR/GCMOM uses prognostic, multiple size distributions (typically three, for liquid, ice, and graupel), each with 30 size sections. MIRAGE2 and WRF/Chem simulate bulk single condensate in single size distribution, with either a prescribed modal distribution (MIRAGE2) or a predicted sectional distribution (WRF/Chem-MOSAIC). CAM3 treats bulk liquid and ice with the same prognostic droplet size treatment as MIRAGE2. Droplet size distribution in both models has a prescribed dispersion so that liquid water content is proportional to number times effective radius cubed. Caltech unified GCM treats bulk liquid and ice with their distributions diagnosed from predicted cloud water content. Among the five models, Caltech unified GCM is the only model that prescribes cloud droplet number, which is predicted in all other four models. It assumes a cloud droplet number of 60 and 170 cm^{-3} respectively, for liquid phase clouds over ocean and land, and 0.06 cm^{-3} for all ice clouds based on observations (Del Genio et al., 1996). CAM3, MIRAGE2, and WRF/Chem use the same treatment for droplet number, with droplet nucleation parameterized by Abdul-Razzak and Ghan (2000). GATOR treats prognostic, size- and composition-dependent cloud droplet number from multiple aerosol size distributions.

While an empirical relationship between sulfate aerosols and CCN is commonly used in most atmospheric models, CCN is calculated from Köhler theory using the aerosol size distribution and hygroscopicity in all models but Caltech unified GCM. MIRAGE and WRF/Chem treat the same CCN composition, except with different size representations. Other than Caltech unified GCM that does not treat CCN and Ice Deposition Nuclei (IDN), all other four models treat the competition among different aerosol species but the hydrophobic species are not activated in CAM3 since it assumes external-mixture. Among the five models, GATOR/GCMOM is the only model that simulates composition of IDN. For CCN spectrum, MIRAGE and WRF/Chem sim-

ulate it as a function of aerosol size and hygroscopicity based on Köhler theory. CAM uses prescribed CCN spectrum. GATOR predicts spectra of both CCN and IDN with 13–17 sections and 1–16 size distributions for typical applications. MIRAGE and CAM use a prognostic parameterization in terms of cloud water, ice mass, and number to predict cloud radiative properties. WRF/Chem also uses the same method but with sectional approach. Caltech unified GCM simulates cloud optical properties based on MIE theory and prescribed Gamma distribution for liquid clouds and phase functions of Mishchenko et al. (1996) (Liao et al., 2003). GATOR/GCMOM simulates volume-average cloud refractive indices (RIs) and optical properties based on MIE theory and an iterative dynamic effective medium approximation (IDEMA) to account for multiple BC inclusions within clouds. The IDEMA is superior to classic effective-medium approximation that is used by several mixing rules such as the volume-average RI mixing rule, the volume average dielectric constant mixing rule, the Maxwell-Garnett mixing rule, and the Bruggeman mixing rule in two aspects (Jacobson, 2006a). First, the IDEMA accounts for polydispersion of spherical absorbing inclusions within the medium and gives different efficiencies at a given wavelength for a given volume fraction but with different size distributions of absorbing material, as occurs in reality. Second, the IDEMA also accounts for light interactions as a function of size of the material included.

Table 5 shows model treatments of aerosol chemistry and microphysics that differ in many aspects. Caltech unified GCM treats aerosol thermodynamics only, the rest of models treat both aerosol thermodynamics and dynamics such as coagulation and new particle formation via homogeneous nucleation. It uses a thermodynamic module, ISORROPIA (equilibrium in Greek) of Nenes et al. (1998), for inorganic aerosols with regime equilibrium among sulfate, nitrate, ammonium, sea-salt, and water. Similar to many global models, MIRAGE2 does not treat nitrate; it simulates a simple inorganic aerosol equilibrium involving ammonium sulfate ($(NH_4)_2SO_4$) and precursor gases. MOZART4 aerosol module in CAM3 uses regime equilibrium for sulfate, ammonium, and nitrate that accounts for cases with sulfate neutral, rich, and very rich. GATOR/GCMOM uses EQUISOLV II that simulates equilibria of all major inor-

ganic salts and crustal species and that provides the most comprehensive treatments among inorganic aerosol thermodynamic modules used in 3-D models (Zhang et al., 2000). In WRF/Chem, different equilibrium modules are used in different aerosol modules. The inorganic aerosol equilibrium modules are the Model for an Aerosol Reacting System (MARS) – version A (MARS-A) in MADE/SORGAM, the Multicomponent Equilibrium Solver for Aerosols (MESA) with a new activity coefficient module Multicomponent Taylor Expansion Method (MTEM) (MESA-MTEM) in MOSAIC, and ISORROPIA in MADRID. Both MARS-A and ISORROPIA use regime equilibrium, whereas MESA-MTEM does not. Sodium chloride is not treated in MARS-A but treated in ISORROPIA and MESA-MTEM. Zhang et al. (2000) evaluated five inorganic aerosol modules used in major 3-D air quality models including MARS-A of Binkowski and Shankar (1995), SEQUILIB of Pilinis and Seinfeld (1987), the model Simulating the Composition of Atmospheric Particles at Equilibrium 2 (SCAPE2) of Kim et al. (1993a, b) and Kim and Seinfeld (1995), the EQUIlibrium SOLVer Version 2 EQUISOLV II of Jacobson (1999c), and the Aerosol Inorganics Model version 2 (AIM2) of Clegg et al. (1998a, b). Among the five modules, MARS-A treats the simplest chemistry with the highest computational speed; it may not be applicable to dry areas with low relative humidities (RHs) and coastal areas, although its results are comparable to those from other modules under high RH conditions (Zhang et al., 2000). The results of ISORROPIA were compared with those of EQUISOLV II in both box model with 11200 test cases and 3-D model over continental U.S. (Zhang and Jacobson, 2005a). While results of ISORROPIA are consistent with those of benchmark and EQUISOLV II, larger bias may occur for relative humidity (RH) $\leq$40 or $\geq$99 for most species. An improved version of ISORROPIA has been developed and implemented in WRF/Chem-MADRID. MESA (Zaveri et al., 2005a) is designed to efficiently solve the complex solid-liquid partitioning within each aerosol size bin using a pseudo-transient continuation technique. In this approach the equilibrium reactions are formulated as pseudo-transient precipitation and dissolution reactions, and the resulting set of stiff non-linear ordinary differential equations (ODEs) are integrated until the system reaches a steady-state to obtain the equilibrium solu-

tion. The mean activity coefficient of an electrolyte in a multi-component solution used in equilibrium simulation is calculated by the MTEM mixing rule (Zaveri et al., 2005b) on the basis of its values in binary solution of all the electrolytes present in the mixture at the solution water activity, assuming that the water activity is equal to the ambient relative humidity. MTEM has been evaluated for several multi-component systems representing various continental and marine aerosols and was found to be significantly more accurate than the approaches employed by Bromley (1973), Kusik and Meissner (1978), and Metzger et al. (2002). The activity coefficients used in MESA, however, are limited for 298 K, which may not be applicable for mid-upper tropospheric conditions. In addition, the equilibrium is solved for a limited number of species and expanding the system of equations may be difficult.

Several major approaches have been used in 3-D models to simulate secondary organic aerosol (SOA) including saturation or fixed aerosol yield (e.g., Pandis et al., 1992), absorption/adsorption (Pankow, 1994 a, b), dissolution (Jacobson, 1997a), dynamic condensation (Jacobson, 1997a), and combination of absorption and dissolution (Pun et al., 2002; Griffin et al., 2002). Both CAM3 and MIRAGE2 use prescribed aerosol yields for a few condensable semi-volatile organic compounds (SVOCs), which is the simplest, computationally most efficient approach but it does not provide a mechanistic understanding of SOA formation. GATOR/GCMOM simulates SOA formation from 10–40 classes SVOCs via condensation and dissolution based on Henry's law. Caltech unified GCM simulates formation of SOA based on a reversible absorption of 5 classes of biogenic SVOCs and neglect that from anthropogenic SVOCs. In MADE/SORGAM and MOSAIC in WRF/Chem, SOA formation via reversible absorption of 8 classes SVOCs is simulated based on smog-chamber data of Odum et al. (1997) and Griffin et al. (1999). Two approaches are used to simulate SOA formation in WRF/Chem-MADRID (Zhang et al., 2004). MADRID 1 uses absorptive approach for 14 parent VOCs (2 anthropogenic, and 12 biogenic) and 38 SOA species (4 anthropogenic, and 34 biogenic). MADRID 2 combines absorption and dissolution approaches to simulate an external mixture of 42 hydrophilic and hydrophobic VOCs,

which are grouped into 10 surrogate compounds based on their affinity for water, origin, number of carbon, volatility, and dissociation properties (Pun et al., 2002). MADRID 2 is currently inactivated in WRF/Chem since its gas-phase chemical mechanism, the California Atmospheric Chemical Mechanism (CACM), has not yet been implemented into WRF/Chem.

New particle formation via binary homogeneous nucleation is simulated in all models except for CAM3, and that via ternary nucleation based on Napari et al. (2002) is only simulated in GATOR-GCMOM. Different models use different equations that account for dependence of new particle formation rates in different ways on number concentration or critical vapor pressure of H_2SO_4, critical new particle formation rate, temperature, and RH. The binary parameterization of Harrington and Kreidenweis (1998) used in MIRAGE2 is based on the calculations of nucleation rates performed by Jaecker-Voirol and Mirabel (1989), which calculates the absolute nucleation rates based on heteromolecular homogeneous nucleation theory of the H_2SO_4–H_2O_2 system. The parameterizations of Kulmala et al. (1998) used in MADE/SORGRAM, Wexler et al. (1994) used in MOSAIC in WRF/Chem, and Vehkamäki et al. (2002) used in GATOR/GCMOM are derived based on the classical binary homogeneous nucleation model that simulates nucleation kinetics and accounts for hydration. The parameterization of Kulmala et al. (1998) predicts binary nucleation rates up to 2–3 orders of magnitude lower than those predicted by most other binary nucleation parameterizations due to the fact that its derivation contains mistakes in the kinetic treatment for hydrate formation (Vehkamäki et al., 2002; Noppel et al., 2002; Zhang and Jacobson, 2005b). The parameterization of McMurry and Friedlander (1979) used in WRF/Chem-MADRID simulates gas-to-particle conversion between nucleation of new particles and condensation on existing particles, which is a more realistic approach than that based on the absolute prediction of a nucleation rate.

While CAM3 assumes instantaneous condensation of inorganic species, other models simulate dynamic condensation of condensable species based on similar growth laws but with different numerical condensational algorithms. For example,

GATOR/GCMOM and WRF/Chem-MADRID use the Analytical Predictor of Condensation (APC) with the moving center scheme, WRF/Chem-MADE/SORGAM uses the modal approach of Binkowski and Shankar (1995), WRF/Chem-MOSAIC uses the Adaptive Step Time-split Explicit Euler Method (ASTEEM) method. Zhang et al. (1999) evaluated performance of several condensational algorithms including APC and the modal approach of Binkowski and Shankar (1995). Coagulation is currently not treated in CAM3 but simulated with a modal approach in MIRAGE2, a sectional approach in GATOR/GCMOM, and both in WRF/Chem-MADE/SORGAM and MOSIAC. Different from other model treatments, GATOR accounts for van der Waals and viscous forces, and fractal geometry in simulating coagulation among particles from multiple size distributions (Jacobson and Seinfeld, 2004). While van der Waals and fractal geometry may enhance coagulation, viscous forces tend to retard the rate of van der Waals force enhancement in the continuum regime. For gas/particle mass transfer, CAM3, MIRAGE2, and Caltech unified GCM use the simplest full equilibrium approach. GATOR/GCMOM uses a computationally efficient dynamic approach with a long time step (150–300 s) (PNG-EQUISOLV II) for all treated species (Jacobson, 2005a). In WRF/Chem, a full equilibrium approach is used in MADE/SORGAM, a dynamic approach is used in MOSAIC. In the dynamic approach of MOSAIC, ASTEEM is coupled with MESA to solve the dynamic gas-aerosol partitioning over multiple size bins. Characteristic times for semi-volatile trace gases to reach equilibrium can vary significantly (by up to several orders of magnitude) among particles with different sizes, making the coupled system of ordinary differential equations for gas-aerosol mass transfer extremely stiff. ASTEEM is developed to reduce the stiffness of the system by first computing the condensation of H_2SO_4 and NH_3 only for all the aerosol size bins, then computing condensation of HNO_3, HCl, and NH_3 gases for one size bin in a time-split fashion over a time-splitting interval. The value of the time splitting interval is determined such that the maximum overall change in the gas-phase concentrations over the interval is restricted to less than ∼10%. This approach improves computational efficiency by allowing the solver to take longer time steps with only a relatively small loss in accuracy. MADRID offers

three approaches: full equilibrium, dynamic, and hybrid; their performance has been evaluated in Zhang et al. (1999) and Hu et al. (2007b). The box MADRID tests of Hu et al. (2007b) have shown that the bulk equilibrium approach is computationally-efficient but fails to predict the distribution of semi-volatile species (e.g., ammonium, chloride, and nitrate) because of the equilibrium and internal mixture assumptions. The hybrid approach exhibits the same problem for some cases as the bulk equilibrium approach since it assumes bulk equilibrium for fine particles. The kinetic approach predicts the most accurate solutions with variable computational efficiencies depending on whether a small time step is required.

Table 6 summarizes the treatments of aerosol-cloud interactions and cloud processes used in the five models. Water uptake is a very important process affecting calculations of both direct and indirect forcing. CAM3 simulates bulk equilibrium with RH for external mixtures only. MIRAGE and WRF/Chem-MOSAIC simulate hygroscopic growth in equilibrium with RH based on Köhler theory. Water uptake is calculated as a function of RH, the mean dry radius, the relative contributions of each aerosol component to the total particle hygroscopicity, and the aerosol water content from previous time step. Aerosol water content in GATOR/GCMOM is calculated based on discrete size-resolved equilibrium using the Zdanovskii-Stokes-Robinson (ZSR) method (Zdanovskii, 1948; Stokes and Robinson, 1966); it simulates the mutual deliquescent RH (MDRH). The ZSR method is also used to simulate aerosol water uptake in Caltech unified GCM. No hysteresis effect is accounted for in CAM3 and Caltech unified GCM, but it is treated in other models.

Aerosol activation by cloud droplets to form CCN is an important process affecting simulations of aerosol-cloud interactions, and aerosol direct and indirect forcing. CAM3 uses empirical, prescribed activated mass fraction for bulk CCN. MIRAGE and WRF/Chem use a mechanistic, parameterized activation module that is based on Köhler theory to simulate bulk CCN. In Köhler theory, the number of particles activated is expressed in terms of supersaturation S, which is primarily determined by aerosol properties (i.e., number, size, and hygroscopicity) and updraft velocity. Impor-

tant parameters for activation such as the peak supersaturation, S_{max}, mass of activated aerosols, and the size of the smallest aerosol activated are calculated using the parameterizations of Abdul-Razzak et al. (1998) and Abdul-Razzak and Ghan (2000) that relate the aerosol number activated directly to fundamental aerosol properties. The effects of Kelvin and Rault's law for liquid activation are partially taken into account in those parameterizations. GATOR-GCMOM also simulates a mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory. At high-resolution regional scales, the saturation ratios at equilibrium (S) are determined from Köhler theory as a function of aerosol particle composition and size, accounting for the Kelvin effect and Raoult's law for liquid activation and the Kelvin effect for ice activation. Aerosol composition of a given size affects the Kelvin term through the surface tension and Raoult's law through the molality term (Jacobson et al., 2007). On the global scale and coarse regional scales, the water vapor available for condensation is determined from cumulus and stratus parameterizations. The cumulus parameterization treats subgrid clouds, and aerosol particles are convected within each of these clouds. Liquid and ice from the cumulus/stratus parameterization are evaporated/sublimated and regrown onto size- and composition-resolved aerosol particles (Jacobson, 2003c). One difference between the treatments in GATOR/GCMOM and MIRAGE2 is that the MIRAGE activation parameterization neglects size-dependence of the water vapor diffusivity coefficient and mass transfer coefficient, which may lead to an underestimation of cloud droplet number concentration. In addition, it does not treat the kinetic effect (i.e., mass transfer limitation) for larger particles for which the equilibrium Köhler theory may be inappropriate. Such size-dependence and kinetic effect are accounted for in GATOR/GCMOM. Aerosol-cloud interaction is currently not treated in Caltech unified GCM but it is being implemented (Hong Liao, Chinese Academy of Sciences, China, personal communications, 2007).

Aerosols are removed through dry deposition in the absence of hydrometerors and wet deposition following scavenged in- and below- cloud. CAM3 assumes that in-cloud scavenging occur via prescribed activation and autoconversion (i.e., the colli-

sion/coalescence of cloud drop to become rain drops and get into precipitation). Caltech unified GCM treats autoconversion and nucleation scavenging with prescribed scavenging coefficient for sea-salt and dust and a first-order precipitation-dependent parameterization for other aerosols. The in-cloud scavenging processes in MIRAGE2 and WRF/Chem include activation, Brownian diffusion (for both interstitial and activated particles), autoconversion, and nucleation scavenging. The dependence of autoconversion on droplet number is neglected in both models. All those processes are included for discrete size-resolved clouds in GATOR/GCMOM. Note that autoconversion in GATOR/GCMOM is somewhat different from that in other models because of their differences in cloud treatments. Cloud droplets are treated to be size-resolved in GATOR/GCMOM but bulk in other models. Consequently, other models treat autoconversion for bulk cloud droplets whereas GATOR/GCMOM treats coagulation for discrete size-resolved cloud droplets into rain drops/ice crystals (which is analogous to autoconversion for bulk clouds). The effects of aerosols on precipitation rates are taken into account in GATOR/GCMOM, but are neglected in other models.

For below-cloud scavenging, CAM3, MIRAGE2, and WRF/Chem-MOSAIC prescribe scavenging efficiencies and Caltech unified GCM assumes the first-order precipitation-dependent scavenging parameterization, whereas GATOR/GCMOM simulates discrete size-resolved aerosol-hydrometeor coagulation (washout). The dependence of below-cloud scavenging and precipitation rates on aerosol size and composition is accounted for in GATOR/GCMOM but either partially (e.g., Caltech unified GCM calculates size-dependent scavenging efficiency) or completely neglected in other models. Among the five models, GATOR/GCMOM is the only model that treats coagulation between different size sections from different size distributions for various hydrometeors (e.g., liquid-liquid, liquid-ice, liquid-graupel, ice-ice, ice-graupel, graupel-graupel) and that between aerosols and hydrometeors. MIRAGE2 and WRF/Chem simulate coagulation between cloud droplets, between cloud droplets and precipitating particles, and between aerosol and precipitating particles for one size distribution of each type of hydrometeors.

Droplet sedimentation refers to the layer-by-layer sinking of drops as a function of droplet size. Sedimentation to the ground in the bottom layer is precipitation if a model treats sedimentation layer by layer. GATOR-GCMOM treats layer by layer sedimentation of discrete size-resolved discrete liquid, ice, and graupel particles with fall speeds as a function of their sizes. As droplets fall below clouds, they shrink as a function of size. Some may completely evaporate, releasing their aerosol cores back to the air. Some may hit the ground as precipitation. CAM3 treats sedimentation of bulk liquid and ice particles, each with a single fall speed that is calculated as a function of a mass-weighted effective radius of ice particles (Boville et al., 2006). For bulk ice, the effective radius is calculated for a size distribution that is assumed to be a function of temperature only. For bulk liquid, no size distribution is assumed; the effective radius is determined from the bulk liquid water mass and the total number concentration of particles. Liquid and ice particles falling from one layer to the next within a cloud do not coagulate as a function of size. All hydrometeors falling below a cloud are evaporated/sublimated completely without releasing aerosol cores. No precipitation resulted from sedimentation unless the cloud exists in the bottom layer (Note that precipitation is calculated as a separate autoconversion in CAM3). Droplet sedimentation is neglected in MIRAGE and WRF/Chem. Droplet sedimentation is not explicitly treated in Caltech unified GCM because it does not resolve the scales of vertical motion relevant to sedimentation; it is however implicitly accounted for by parameterizing the limiting autoconversion rate as a decreasing function of the large scale vertical velocity (Del Genio et al., 1996). A discrete cloud size distribution as it is used in GATOR-GCMOM is necessary to realistically simulate all cloud microphysical processes (e.g., condensation/evaporation, deposition/sublimation, collision-coalescence, contact freezing, rainout, washout, sedimentation) from first principles rather than parameterizations. The droplet sedimentation treatments in CAM3 is not physical and prevents an accurate simulation of the physical feedbacks of aerosol particles to climate.

4 Case studies

4.1 WRF/Chem-MADRID

WRF/Chem-MADRID has been applied to simulate a 5-day episode (12:00 UTC 28 August through 12:00 UTC 2 September of 2000) from the Texas Air Quality Study (TexAQS-2000) in the southern U.S. The TexAQS-2000 was carried out around the Houston area where the exceedance of the National Ambient Air Quality Standard (NAAQS) 120 ppb O_3 standard occurs most frequently and VOC reactivities are typically much higher than other urban areas in the U.S. WRF/Chem uses the mass (hydrostatic pressure) coordinates. The horizontal grid spacing used is 12 km and the vertical resolution is 57 layers from surface to tropopause with vertical intervals varying from 15 m in layer 1 to 600–680 m near/at the domain top. The initial conditions, boundary conditions, and emissions are the same ones as the WRF/Chem simulations with MOSAIC described in Fast et al. (2006). Cloud barely occurred during this episode, cloud microphysic scheme is thus turned off, and no aerosol-cloud interaction and aerosol indirect effects were simulated. The simulation results have been evaluated against in situ observations for gas-phase species (e.g., O_3, SO_2, nitrogen dioxide (NO_2), and nitric oxide (NO), $PM_{2.5}$, and its composition and remote sensing measurements (e.g., aerosol optical depths) (Zhang et al., 2005c, 2007; Hu et al., 2006).

Figure 2 shows the spatial distribution of the predicted 24 h average $PM_{2.5}$ concentrations and the 24 h average wind field on 29 August (central daylight time CDT), 2000. The predicted $PM_{2.5}$ distribution is consistent with the pattern of emissions and wind field. The emissions of primary $PM_{2.5}$ species such as BC and other unknown inorganic $PM_{2.5}$ are high in Houston, the emissions of SO_2 are high in Baton Rouge and the emissions of CO and NO_x are relatively high in Dallas, resulting in relatively high $PM_{2.5}$ concentrations in those cities and their vicinity areas. The normalized mean biases (NMBs) of the hourly O_3 and $PM_{2.5}$ predictions are 19.8% and 41.7%, indicating a moderate overprediction that can be attributed to several factors including overestimate of primary BC and organic matter (OM) emissions and high aerosol boundary condi-

tions. Figure 3 shows the vertical profile of $PM_{2.5}$ concentrations and the differences in vertical temperature (T) and water vapor (Qv) mixing ratio between simulations with and without PM at five different times on 29 August at LaPorte that is located in the east of Houston at the coastal area of the Galveston Bay. As shown, $PM_{2.5}$ concentrations at surface and in the planetary boundary layer (PBL) vary significantly from time to time during a day, depending on magnitudes/timing of precursor emissions and related meteorological conditions such as atmospheric stability, the depth of mixing height, and temperature. The surface $PM_{2.5}$ reaches the highest at 06:00 a.m. due to high emissions of primary $PM_{2.5}$ and precursors of secondary $PM_{2.5}$ from motor vehicles and relatively-shallow mixing height. The $PM_{2.5}$ concentration in the PBL reaches the highest at 02:00 p.m. due likely to the effect of bay breeze. As expected, T and Q_v respond strongly to changes in $PM_{2.5}$concentrations, with maximum changes coincide with maximum gradients in $PM_{2.5}$ concentrations in the PBL. T reduces by up to 0.18°C at/near surface but increases by 0.16°C in PBL. Water vapor mixing ratio increases by 3.2% at/near surface but decreases by 3% in the PBL. While the decrease in T and increase in Qv at/near surface are directly caused by reduced net downward solar/thermal-IR radiation in the absence of $PM_{2.5}$, the opposite changes in the PBL may be caused by radiation absorption of particles and advection of long- or moderately-lived greenhouse gases that absorb thermal-IR radiation emitted by particles aloft.

4.2 GATOR/GCMOM

GATOR/GCMOM has been applied to simulate the effect of aerosol feedbacks into regional climate changes over a global domain at a horizontal resolution of 4°′ SN×5°′ WE and two nested domains: California Grid at a resolution of 0.2°×0.15° (~21.5 km×14.0 km) and the South Coast Air Basin Grid: at a resolution of 0.045°×0.05° (~4.7 km×5 km) (Jacobson et al., 2007). The vertical resolutions are 39 sigma layers up to 0.425 hPa for the global domain and 26 layers up to 103.5 hPa, each matching the bottom 26 global layers (with five layers in the bottom 1 km for all domains). The baseline simulations were conducted for two 1-month periods in 1999:

February and August. In sensitivity simulations, emissions of anthropogenic aerosol particles and their precursor gases (AAPPG) such as BC, OC, sulfate, nitrate, fugitive dust, SO_x, NO_x, NH_3, and reactive organic gases (ROGs) were turned off. Over the LA basin, AAPPG is found to reduce net downward surface total solar irradiance, near-surface temperatures, and surface wind speeds; increase RHs, aerosol and cloud optical depths, cloud fractions, cloud liquid water; and either increase or decrease precipitation depending on location and magnitude of precipitation intensity.

Figure 4 shows the effect of AAPPG on near-surface wind speeds and vertical profiles of wind speeds over California grid simulated by GATOR/GCMOM in February and August 1999. Aerosols decrease surface wind speed but increase boundary-layer wind speed. The decease is driven primarily by two factors: the cooling at the surface due to the reduction in surface solar radiation and the warming in the upper boundary-layer due to the heat caused by the absorbing aerosols. Both factors stabilize the air, reducing turbulence which in turn reduces vertical flux of horizontal momentum, thus slowing transfer of fast winds aloft to the surface (Jacobson et al., 2007). Figure 5 shows the effect of AAPPG on precipitation in the South coast, CA and the CA grids. AAPPG decreased precipitation in the LA basin and the mountains beyond the basin in February. In August, when precipitation was low, most reductions occurred offshore and in the foothills of the San Bernardino Mountains. Some precipitation increases were found on the downslope sides of the San Bernardino and San Gabriel Mountains. Those results are consistent with findings of Givati and Rosenfeld (2004, 2005).

4.3 CAM3 and MIRAGE2

3-year global simulations after 4-month spin-up were conducted with CAM3 and MIRAGE2 to understand the differences in simulated aerosol direct and indirect forcing due to different aerosol and cloud microphysical treatments. No nudging was used in those simulations. The horizontal resolution is 4°latitude×5°longitude and the vertical resolution is 26 layers from surface to 3.5 hPa. Baseline simulations (CAM3_B and MIRAGE2_B) were conducted with default aerosol modules (MOZART4 in CAM3

and PNNL's aerosol module in MIRAGE2, see major differences in Tables 1–5). Four sensitivity simulations were conducted: a CAM3 simulation with constant droplet sedimentation (CAM3_S1), a CAM3 simulation with the same configurations as CAM3_S1 but offline coupling (CAM3_S2), a MIRAGE2 simulation with the same configurations as MIRAGE2_B but with offline coupling (MIRAGE2_S1), and a CAM3 simulation with the same configurations as CAM3_S2 but with PNNL's aerosol module in replacing MOZART4 (CAM3_S3).

Figure 6 shows results from those simulations. The first aerosol indirect effect (FAIE) from CAM_B is much larger than that from MIRAGE_B (3.2 vs. 0.38 $W m^{-2}$), the prediction of MIRAGE_B is much closer to the total aerosol indirect forcing of 0.75 $W m^{-2}$ estimated by IPCC (2007). MIRAGE has no droplet sedimentation. Compared with results using bulk sedimentation that is calculated based on mass-weight effective radius of liquid and ice particles, the magnitude of FAIE in CAM3 decreases by ~30% with a constant sedimentation velocity because sedimentation is reduced. While this result demonstrates the sensitivity of simulated FAIE to droplet sedimentation treatments, neither treatments (i.e., bulk or constant) are realistic because of the use of empirical parameterizations instead of the first principles that treat the sedimentation velocity of particles of individual size. Both online and offline simulations use the same monthly mean aerosol concentrations. But on shorter time scales the online has variability so that less aerosol is present under cloudy conditions, due to enhanced scavenging in clouds. As expected, using an offline aerosol calculation increases magnitude of FAIE in both CAM3 and MIRAGE2 because of presence of increased aerosol under cloudy conditions. The use of MIRAGE aerosol module in offline CAM3 significantly reduces FAIE in CAM3, suggesting that addition of an aerosol treatment that allows aerosol size distribution to shift with increasing emissions is likely to produce a smaller indirect effect, particularly when it is interactive (Ghan, 2007).

5 Major challenges and future directions

Significant progress has been made in the past two decades in the development of online coupled climate- (or meteorology-) chemistry and their application for modeling global/regional climate, meteorology, and air quality, as well as the entire earth system. Several major challenges exist. First, accurately representing climate-aerosol-chemistry-cloud-radiation feedbacks in 3-D air quality/climate models will remain a major scientific challenge in developing a future generation of coupled models for the years to come. There is a critical need for advancing the scientific understanding of key processes include real-time emissions that contribute the model uncertainties to a large extent; the two-way/chain effects among climate, meteorology, chemistry, aerosol, cloud and radiation; the size-/composition-resolved aerosol/cloud microphysics for multiple size distributions (e.g., new particle formation, SOA, and aerosol/cloud interactions); and subgrid variability. Second, representing scientific complexity within the computational constraint will continue to be a technical challenge. Key issues include (1) the development of benchmark model and simulation and use of available measurements to characterize model biases, uncertainties, and sensitivity and to develop bias-correction techniques (e.g., chemical data assimilation); (2) the optimization/parameterization of model algorithms with acceptable accuracy. Third, integrated model evaluation and improvement, laboratory/field studies for an improved understanding of major properties/processes will also post significant challenges, as they involve researchers from multiple disciplinaries and requires a multidisciplinary and or interdisciplinary approach. Key issues include (1) continuously operation of monitoring networks and remote sensing instrument to provide real-time data (e.g., the AirNow surface monitoring network and Satellite) for data assimilation/model evaluation, and (2) the development of process-oriented models to isolate complex feedbacks. Fourth, a unified modeling system that allows a single platform to operate over the full scale will represent a substantial advancement in both the science and the computational efficiency. Major challenges include globalization/downscaling with consistent model

physics and two-way nesting with mass conservation and consistency. The only such model that exists is the GATOR-GCMOM, although other global-through-urban fully-coupled models such as global-to-urban WRF/Chem model are being developed (e.g., O'Connor et al., 2006; Zhang et al., 2008b). Such an unified global-to-urban scale modeling system allows a single platform to operate over the full scale. It represents a substantial advancement in both the science and the computational efficiency, with a new scientific capability for studying important problems that require a consideration of multi-scale feedbacks. For example, locally-emitted air pollutants can affect human health at a neighborhood-scale and air quality and climate at all scales and the changes in climate in turn affect further emissions of biogenic species; locally lifted dust particles can affect local and global circulations, which in turn affects their further lifting. Finally, integrated earth system modeling for multi-media (e.g., atmosphere, biosphere, ocean, land surface, etc.) will represent models of next generation that can best replicate human's environment. Most current earth system models for atmosphere-land surface-ocean do not include detailed chemistry, aerosol, and cloud treatments and biogeochemical cycles, integration of such complexities into the earth system models will pose unprecedented challenges for the entire science communities.

Appendix A

List of Acronyms and Symbols

Acronym	Definition
3-D	three-dimensional
AAPPG	the anthropogenic aerosol particles and their precursor gases
AIM2	the Aerosol Inorganics Model version 2
APC	the Analytical Predictor of Condensation
ASTEEM	the Adaptive Step Time-split Explicit Euler Method
ARW	the Advanced Research WRF with the Eulerian Mass
BC	black carbon
CACM	the California Atmospheric Chemical Mechanism
CAM3	the Community Atmospheric Model version 3
CB05	the 2005 version of Carbon Bond mechanism
CBM-EX	The Stanford University's extended Carbon Bond mechanism
CBMZ	the Carbon-Bond Mechanism version Z
CCM	the NCAR Community Climate Model
CCN	cloud condensation nuclei
CDT	central daylight time
CFCs	chlorofluorocarbons
CH_4	methane
CH_3O_2H	methyl hydroperoxide
CMAQ	the EPA's Community Multiple Air Quality
CO	carbon monoxide
CO_2	carbon dioxide
CTMs	chemical transport models
DEMA	the iterative dynamic effective medium approximation
DMS	dimethyl sulfide
EQUISOLV II	the EQUIlibrium SOLVer version 2

Acronym	Definition
EPA	the U.S. Environmental Protection Agency
GCM	general circulation model
GAQMs	global air quality models
GATORG	the Gas, Aerosol, TranspOrt, Radiation, and General circulation model
GATOR-GCMOM	the Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model
GATOR/MMTD (or GATORM)	the gas, aerosol, transport, and radiation air quality model/a mesoscale meteorological and tracer dispersion model
GChM	the PNNL Global Chemistry Model
H_2O	water
H_2O_2	hydrogen peroxide
HO_2	hydroperoxy radical
H_2SO_3	sulfurous acid
H_2SO_4	sulfuric acid
IDN	Ice Deposition Nuclei
IPCC	Intergovernmental Panel on Climate Change
ISORROPIA	"equilibrium" in Greek, refers to The ISORROPIA thermodynamic module
LA	Los Angeles
MADE/SORGAM	the Modal Aerosol Dynamics Model for Europe (MADE) with the secondary organic aerosol model (SORGAM)
MADRID	the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution
MARS-A	the Model for an Aerosol Reacting System (MARS) –version A
MCCM (or MM5/Chem)	The Multiscale Climate Chemistry Model
MESA	the Multicomponent Equilibrium Solver for Aerosols
MM5	the Penn State University (PSU)/NCAR mesoscale model
MIRAGE	the Model for Integrated Research on Atmospheric Global Exchanges
MOSAIC	the Model for Simulating Aerosol Interactions and Chemistry
MOZART4	the Model for Ozone and Related Chemical Tracers version 4
MSA	methane sulfonic acid
MTEM	The Multicomponent Taylor Expansion Method
NAAQS	the National Ambient Air Quality Standard
NCAR	the National Center for Atmospheric Research
NARE	the North Atlantic Regional Experiment

Acronym	Definition
$(NH_4)_2SO_4$	ammonium sulfate
NMBs	normalized mean biases
NMM	the Nonhydrostatic Mesoscale Model
NO_3	nitrate radical
NO	nitric oxide
NO_2	nitrogen dioxide
NO_x	nitrogen oxides
N_2O	nitrous oxide
NOAA	the National Oceanic and Atmospheric Administration
O_3	ozone
OC	organic carbon
ODEs	ordinary differential equations
OH	hydroxyl radical
OM	organic matter
PBL	the planetary boundary layer
$PM_{2.5}$	particles with aerodynamic diameters less than or equal to 2.5 μm
PNNL	the Pacific Northwest National Laboratory
Qv	water vapor
RACM	the Regional Atmospheric Chemistry Mechanism
RADM2	the gas-phase chemical mechanism of Regional Acid Deposition Model, version 2
RHs	relative humidities
RIs	refractive indices
ROGs	reactive organic gases
RRTM	the Rapid Radiative Transfer Model
S(IV)	dissolved sulfur compounds with oxidation state IV
SCAPE2	the model Simulating the Composition of Atmospheric Particles at Equilibrium 2
SO_2	sulfur dioxide
SOA	secondary organic aerosol
STAR	the U.S. EPA-Science to Achieve Results program
SVOCs	semi-volatile organic compounds
T	temperature
TUV	the Tropospheric Ultraviolet and Visible radiation model
UCLA-GCM	the University of Los Angeles General Circulation Model
WRF/Chem	the Weather Research Forecast model with Chemistry

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Table 1. Examples of treatments of online coupling of gas, aerosol, radiative, transport, and meteorological processes.

	H69	C70, S79	C75	A75, Jo76 T85, C85, M86	P84, G91, R95	B88	P92	J94, J95, J96, J97a,b	J02, J04a–d,	J06 J07	G05 F06
Online meteorology and pollutant transport											
O_3	Y	Y					Y				
O_3 and some other gases and families					Y	Y					
All photochemically-active gases								Y	Y	Y	
Single bulk or modal aerosol				Y		Y					Y
All discrete, size-resolved aerosol particles								Y	Y	Y	
All chemicals within discrete, size-resolved aerosol particles								Y	Y	Y	Y
All discrete, size-resolved hydrometeor particles and their aerosol inclusions										Y	
Online meteorology and pollutant transport/chemistry/microphysics											
None				Y							
Time-dependent for O_3 only	Y	Y	Y				Y				
Time-dependent for O_3 and some gases; steady-state or family chemistry					Y	Y					
Time-dependent for all reacting and transported gases for others gases								Y	Y	Y	Y
Time-dependent for aerosols with comprehensive dynamics treatments								Y	Y	Y	Y
No feedback	Y				Y						
Feedback of online O_3 to lookup-table heating rate			Y								
Feedback of online O_3 to online parameterized heating rate		Y					Y				
Feedback of a few gases to heating rates from spectral radiative transfer											Y
Feedback of all photochemically-active gases to heating rates from spectral radiative transfer								Y	Y	Y	
Feedback of online bulk or modal aerosol to parameterized heating rate				Y		Y					Y

Table 1. Continued.

	H69	C70, S79	C75	A75, Jo76 T85, C85, M86	P84, G91, R95	B88	P92	J94, J95, J96, J97a,b	J02, J04a–d,	J06 J07	G05 F06
Feedback of all discrete size-resolved aerosols to heating rates from spectral solar and thermal-IR radiative transfer								Y	Y	Y	
Feedback of all discrete size-resolved hydrometeors to heating rates from spectral solar and thermal-IR radiative transfer									Y	Y	
No photolysis				Y							
Photolysis from lookup table or fixed, without feedback	Y	Y			Y	Y					
Feedback of online O_3 only to lookup-table photolysis			Y				Y				
Feedback of a few gases to online photolysis from spectral radiative transfer											Y
Feedback of all gases to online photolysis from spectral radiative transfer								Y	Y	Y	
Feedback of online bulk or modal or size-resolved aerosol to parameterized photolysis schemes											Y
Feedback of all discrete size-resolved aerosols to photolysis from spectral radiative transfer								Y	Y	Y	
Feedback of all discrete size-resolved hydrometeors to photolysis from spectral radiative transfer									Y	Y	

A75-Atwater (1975); B88-Baklanov (1988); C70-Clark, (1970); C75-Cunnold et al. (1975); C85-Cess et al. (1985); F06-Fast et al. (2006); G91-Granier and Brasseur (1991); G05-Grell et al. (2005); H69-Hunt (1969); J94-Jacobson(1994); J95-Jacobson (1995); J96-Jacobson et al. (1996); J97a-Jacobson (1997a); J97b-Jacobson (1997b); J02-Jacobson (2002); J04a-Jacobson et al. (2004); J04b-Jacobson and Seinfeld (2004); J04c-Jacobson (2004a); J04d-Jacobson (2004b); J06-Jacobson and Kaufmann (2006); J07-Jacobson et al. (2007); Jo76-Joseph (1976); P84-Penenko et al. (1984); P92-Pitari et al. (1992); R95-Rasch et al. (1995); S79-Schlesinger and Mintz (1979); and T85-Thompson (1985).

Table 2. Model Systems and Typical Applications of Online Models developed in the U.S.

Model System/Scale	Meteorology Model	Chemical Transport Model (Main features)	Typical Applications	Example References
GATOR-GCMOM & Predecessors (Global-through-urban)	MMTD GCMM GCMOM	Gas-phase chemistry: CBM-EX: (247 reactions, 115 species); Bulk or size-resolved aqueous-phase sulfate, nitrate, organics,chlorine, oxidant, radical chemistry (64 kinetic reactions); size-resolved, prognostic aerosol/cloud with complex processes	Current/future met/ chem/rad feedbacks; Direct/indirect effects; AQ/health effect 2006a, 2007	Jacobson, 1994, 1997a, b, 2001c, 2002, 2004a, b; Jacobson et al.,2004,
WRF/Chem (Mesoscale)	WRF	RADM2, RACM, CBMZ,CB05 (156237 reactions, 52–77 species); bulk aqueous-phase RADM chemistry (MADE/SORGAM) or CMU mechanism (MOSAIC/MADRID; Three aerosol modules (MADE/SORGAM, MOSAIC,and MADRID) with size/mode-resolved, prognostic aerosol/cloud treatments	Forecast/hindcast, Met/chem feedbacks; O_3, $PM_{2.5}$; Aerosol direct effect	Grell et al. (2005); Fast et al. (2006); McQueen et al. (2005, 2007); Zhang et al. (2005a, b, 2007)
CAM3 & Predecessors (Global)	CCM3/	Prescribed CH_4, N_2O, CFCs/MOZART4 gas-phase chemistry (167 reactions, 63 species);	Climate; Direct/indirect effects; Hydrological cycle	Rasch et al. 1995, 2006; Kiehl et al. 1998; Collins et al. 2004, 2006a, b
	CCM2/ CCM1	Bulk aqueous-phase sulfate chemistry of S(IV) (4 equilibria and 2 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution		
MIRAGE2 & 1 (Global)	CAM2/ CCM2	Gas-phase CO-CH_4-oxidant chem.; Bulk aqueous-phase sulfate chemistry (6 equilibria and 3 kinetic reactions); Mode-resolved simple aerosol treatment; Prognostic aerosol/cloud treatments	Trace gases and PM; Direct/indirect effects	Ghan et al. 2001a, b, Zhang et al. 2002; Easter et al. 2004;
Caltech unified GCM (Global)	GISS GCM II'	Harvard tropospheric O_3-NO_x hydrocarbon chemistry (305–346 reactions, 110–225 species); bulk aqueous-phase chemistry of S (IV) (5 equilibria and 3 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Global chemistry-aerosol interactions; aerosol direct radiative forcing; the role of heterogeneous chemistry; impact of future climate change on O_3 and aerosols	Liao et al. 2003, 2004, 2006; Liao and Seinfeld, 2005

Table 3. Treatments of Aerosol Properties of Online Models.

Model System	Composition	Size Distribution	Aerosol Mixing State	Aerosol Mass/Number	Aerosol Hygroscopicity	Aerosol radiative properties
GATOR-GCMOM	47 species (sulfate, nitrate, ammonium,BC, OC, sea-salt, dust, crustal)	Sectional (17–30): variable, multiple size distributions	A coated core, internal/external mixtures	Predicted/Predicted	Simulated hydrophobic-to-hydrophilic conversation for all aerosol components	Simulated volume-average refractive indices and optical properties based on core-shell MIE theory
WRF/ Chem	Sulfate, nitrate, sea-salt ammonium, BC, OC,	Modal (3): variable (MADE/SORGAM) Sectional (8): variable (MOSAIC/MADRID) single size distribution	Internal	Predicted/Diagnosed from mass or predicted	The same as MIRAGE2	The same as MIRAGE2
CAM3	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust	Modal (4): predicted dust and sea-salt, prescribed other aerosols; single size distribution	External	Prescribed or predicted/Diagnosed from mass	hydrophobic and hydrophilic BC/OC with a fixed conversation rate	Prescribed RI and optical properties for each aero. type, size, and wavelength, for external mixtures
MIRAGE2	Sulfate, BC, OC, sea-salt, dust	Modal (4): variable; single size distribution	Externally mixed modes with internal mixtures within each mode	Prescribed or predicted/Diagnosed or predicted	Simulated BC/OC with prescribed hygroscopicities	Parameterized RI and optical properties based on wet radius and RI of each mode
Caltech unified GCM (Global)	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust	Sectional (11) prescribed for sea-salt; Sectional (6) prescribed for mineral dust; Modal (1): prescribed size distribution for other aerosols; single size distribution for all aerosols	BC, OC, and mineral dust externally mixed with internally-mixed SO_4 2-, NH4 + , NO_3 -, sea-salt, and H_2OH_2O; different aerosol mixing states for chemistry and radiative forcing calculation	Predicted aerosol mass; aerosol number not included	Simulated BC/OC with prescribed hygroscopicities	Simulated optical properties based on Mie theory with size- and wavelength-dependent refractive indices

Table 4. Treatments of Cloud Properties of Online Models.

Model System	Hydrometeor types in clouds	Cloud droplet size distribution	Cloud droplet number	CCN/IDN composition	CCN/IDN spectrum	Cloud radiative properties
GATOR-GCMOM	Size-resolved liquid, ice, graupel, aerosol core components	Prognostic, sectional (30), multiple size distributions (3)	Prognostic, size- and composition-dependent from multiple aerosol size distributions	All types of aerosols treated for both CCN/IDN	Predicted with Köhler theory; sectional (13–17); multiple size distributions (1–16) for both CCN/IDN	Simulated volume-average refractive indices and optical properties based on MIE theory and a dynamic effective medium approximation
WRF/ Chem	Bulk single condensate	Prognostic, sectional, single size distribution (MOSAIC)	The same as MIRAGE2 (MOSAIC)	The same as MIRAGE2 but sectional; CCN only	The same as MIRAGE2 but sectional, CCN only	The same as MIRAGE2 but sectional (MOSAIC)
CAM3	Bulk liquid and ice	The same as MIRAGE2	The same as MIRAGE2	All treated species except hydrophobic species; CCN only	Prescribed; CCN only	The same as MIRAGE2
MIRAGE2	Bulk single condensate	Prescribed, modal, single size distribution	Prognostic, aerosol size- and composition-dependent, parameterized	All treated species; CCN only	Function of aerosol size and hygroscopicity based on; Köhler theory; CCN only	Prognostic, parameterized in terms of cloud water, ice mass, and number
Caltech unified GCM (Global)	Bulk liquid and ice	Diagnosed from predicted cloud water content; single size distribution	constant droplet number cloud based on observations	None	None	Simulated based on MIE theory with different parameterizations for liquid and ice clouds

Table 5. Treatments of Aerosol Chemistry and Microphysics of Online Models.

Model System	Inorganic aero. thermodynamic equilibrim	Secondary organic aerosol formation	New particle Formation	Condensation of gases on aerosols	Coagulation	Gas/particle mass transfer
GATORGCMOM	EQUISOLV II, major inorganic salts and crustal species	Condensation; Dissolution based on Henry's law (10–40 classes VOCs)	Binary homogeneous nucleation of H_2SO_4 and H_2O of Vehkamäki et al. (2002), T- and RH-dependent; Ternary nucleation from Napari et al. (2002)	Dynamic condensation of all condensible species based on growth law (e.g., H_2SO_4, VOCs) using the Analytical Predictor of Condensation (APC) with the moving center scheme	Sectional, multiple size distributions, Brownian diffusion, turbulent shear, turbulent inertial motion, gravitational settling, diffusiophoresis, thermophoresis, electric charge, also accounts for van der Waals and viscous forces, and fractal geometry	Dynamic approach with a long time step (150–300 s) (PNGEQUISOLV II) for all treated species
WRF/ Chem	MARS-A (SORGAM) MESA-MTEM (MOSAIC) ISORROPIA (MADRID)	Reversible absorption (8 classes VOCs) based on smog-chamber data (SORGAM and MOSAIC)Absorption (MADRID1)and combined absorption and dissolution (MADRID2)	Binary homogeneous nucleation of H_2SO_4 and H_2O of Kulmala et al. (1998b) (SORGAM) and of McMurry, and Friedlander, (1979) (MADRID); T- and RH-dependent; sectional; different equations in different aero modules	Dynamic condensation of H_2SO_4 and VOCs using the modal approach of Binkowski and Shankar, (1995) (SORGAM), of H_2SO_4, MSA, and NH_3 using the Adaptive Step Time-split Explicit Euler Method (ASTEEM) method (MOSAIC), and of volatile inorganic species using the APC with moving center scheme (MADRID)	Modal/Sectional (MADE/SORGAM, MOSAIC), single size distribution, fine-mode only	1. Full equili. In all aerosol modules 2. Dynamic in MOSAIC and MADRID 3. Hybrid in MADRID
CAM3	MOZART4 with regime equili. for sulfate, nitrate, and ammonium	Prescribed SOA yield for α-pinene, n-butane, and toluene	None	Instantaneous condensation of inorganic species	None	Full equilibrium involving (NH4) $2SO_4$ and $NH4NO_3$
MIRAGE2	Simple equilibrium involving (NH4) $2SO_4$ and precursor gases	Prescribed SOA yield	Binary homogeneous nucleation of H_2SO_4 and H_2O of Harrington and Kreidenweis (1998); T- and RH-dependent	Dynamic condensation of H_2SO_4 and MSA based on Fuchs and Sutugin growth law	Modal, single size distribution, fine-mode only; Brownian diffusion	Simple equilibrium involving $(NH4)2SO_4$ and precursor gases
Caltech unified GCM (Global)	ISORROPIA with regime equili. for sulfate, nitrate, ammonium, sea-salt, and water	Reversible Absorption for 5 biogenic SVOC classes	None	None	None	Full equilibrium involving (NH4) $2SO_4$ and $NH4NO_3$

Table 6. Treatments of Aerosol-Cloud Interactions and Cloud Processes of Online Models.

Model System	Aerosol Water uptake	Aerosol activation aero-CCN/IDN	In-cloud Scavenging	Below-cloud Scavenging	Coagulation involving Hydrometeor	Droplet Sedimentation
GATOR-GCMOM	Size-resolved Equilibrium with RH; ZSR equation; simulated MDRH; Hysteresis is treated	Mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory; accounting for the Kelvin effect and Raoult's law for liquid activation and the Kelvin effect for ice activation	Size-resolved aerosol activation; nucl. scavenging (rainout), autoconversion for size-resolved cloud droplets; precip. rate dependent of aerosol size and composition	Size-resolved aerosol-hydrometeor coag. (washout), calculated precip. rate dependent of aerosol size and composition	Size-resolved coagulation between hydrometeors and between all aerosols and all hydrometeors	Discrete size-dependent sedimentation that varies with altitude; sedimentation below cloud leads to shrinkage as a function of drop size
WRF/ Chem	The same as MIRAGE2 but sectional (MOSAIC)	The same as MIRAGE2 but sectional (MOSAIC); bulk CCN only	The same as MIRAGE2 but sectional	The same as MIRAGE2 but sectional	The same as MIRAGE2 but sectional	The same as MIRAGE2 but sectional
CAM3	For external mixtures only, bulk equilibrium with RH, no hysteresis	Empirical, prescribed activated mass fraction; bulk CCN only	Prescribed , bulk activation autoconversion, precip. rate independent of aerosols	Prescribed bulk scav. efficiency, no-size dependence	None	Bulk sedimentation; sedimentation below cloud leads to complete evaporation/ sublimation
MIRAGE2	Bulk equilibrium with RH based on Köhler theory, Hysteresis is treated	Mechanistic, parameterized modal activation based on Köhler theory; bulk CCN only; partially accounting for the Kelvin effect and Raoult's law for liquid activation	Modal activation, Brownian diffusion (inters./activated), autoconversion for bulk cloud droplets, nucleation scavenging, precip. rate independent of aerosols	Prescribed modal scavenging efficiency with size dependence	Modal coagulation between cloud droplets, between cloud droplets and precipitating particles, and between aerosol and precipitating particles	no droplet sedimentation
Caltech unified GCM (Global)	Bulk equilibrium, ZSR equation, no hysteresis	None	Bulk autoconversion; nucl. scavenging with prescribed scavenging coefficient for sea-salt and dust and a first-order precipitation-dependent parameterization for other aerosols; precip. rate independent of aerosols	First-order precipitation-dependent bulk parameterization; calculated scavenging efficiency with size dependence	None	implicitly accounted for in a parameterization of the limiting autoconversion rate

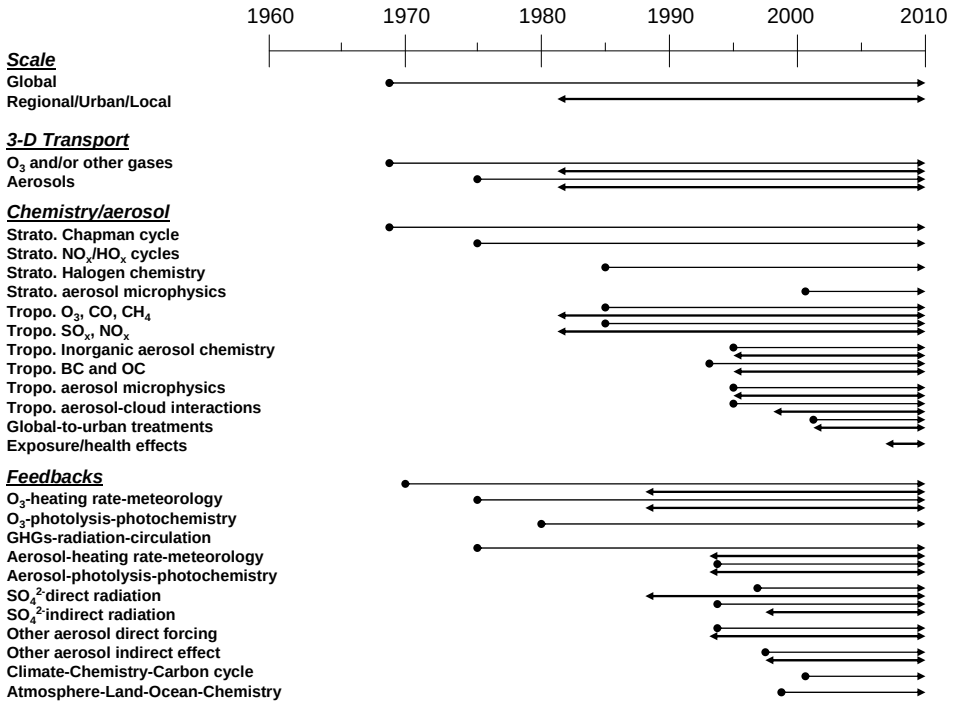

Fig. 1. The development history in chronological order and milestones in terms of chemistry/aerosol and feedback treatments for online coupled models. ●⟶ and ⟷indicate the time and treatments in global and regional models, respectively.

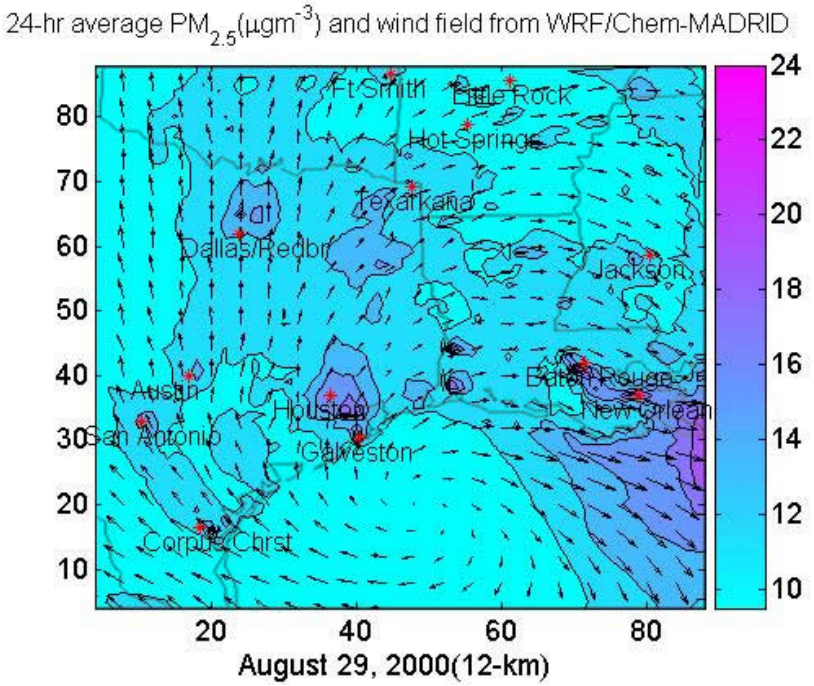

Fig. 2. The spatial distribution of the 24 h average $PM_{2.5}$ concentrations and the 24 h average wind field predicted by WRF/Chem-MADRID on 29 August 2000 (Zhang et al., 2005a).

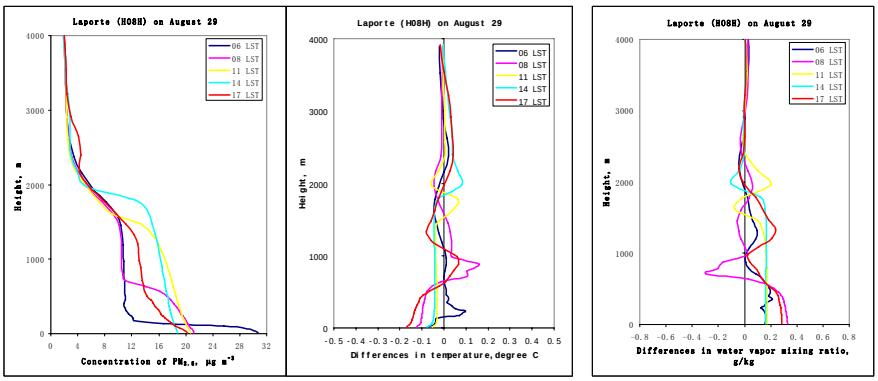

Fig. 3. The vertical distributions of the hourly $PM_{2.5}$ concentrations and differences in vertical distributions of temperatures and water vapor mixing ratios between simulation with and without aerosols by WRF/Chem-MADRID at La Porte, TX at five times (06:00 a.m., 08:00 a.m., 11:00 a.m., 02:00 p.m., and 05:00 p.m.) on 29 August 2000 (Zhang et al., 2005c).

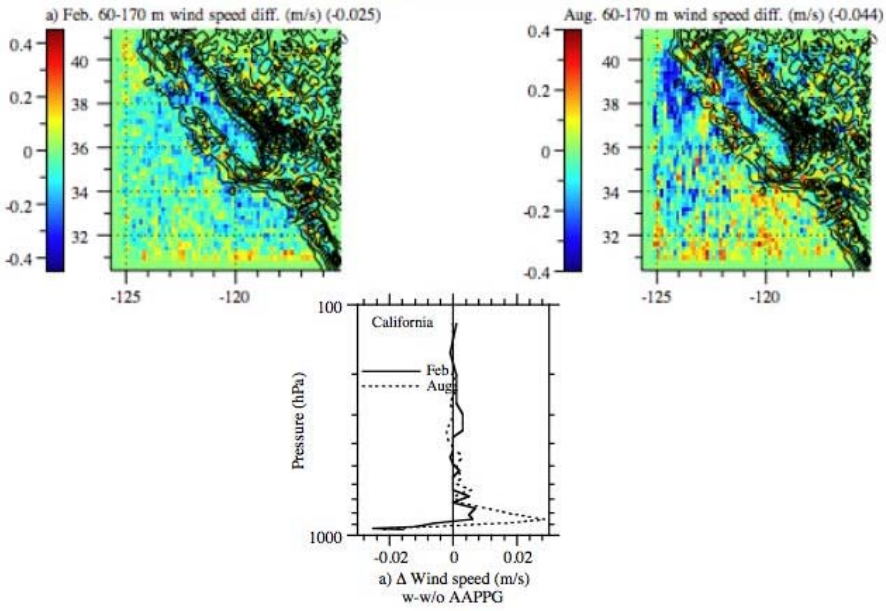

Fig. 4. Differences in the spatial distributions of near-surface wind speeds over California grid and in the domainwide-average vertical distributions of wind speeds between simulation with and without AAPPG by GATOR/GCMOM in February and August 1999 (provided by M. Z. Jacobsen, Stanford University, 2007).

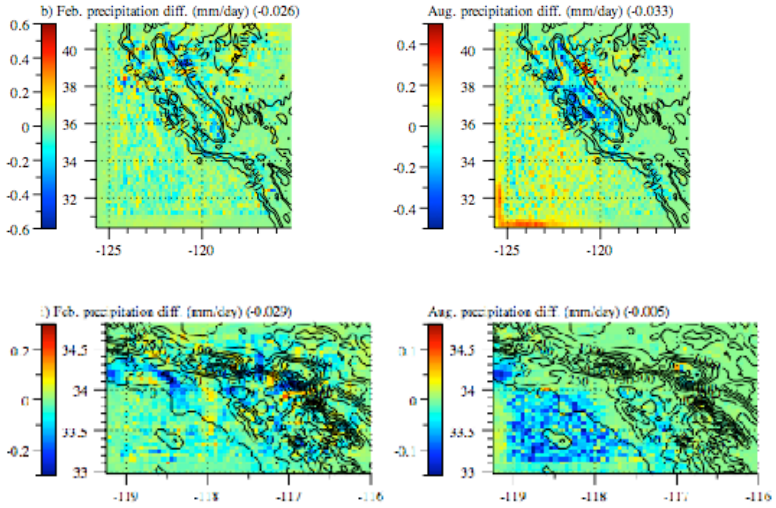

Fig. 5. Differences in the spatial distributions of precipitation over **(a)** California grid, and **(b)** the South Coast grids between simulation with and without AAPPG by GATOR/GCMOM in February and August 1999 (provided by M. Z. Jacobsen, Stanford University, 2007).

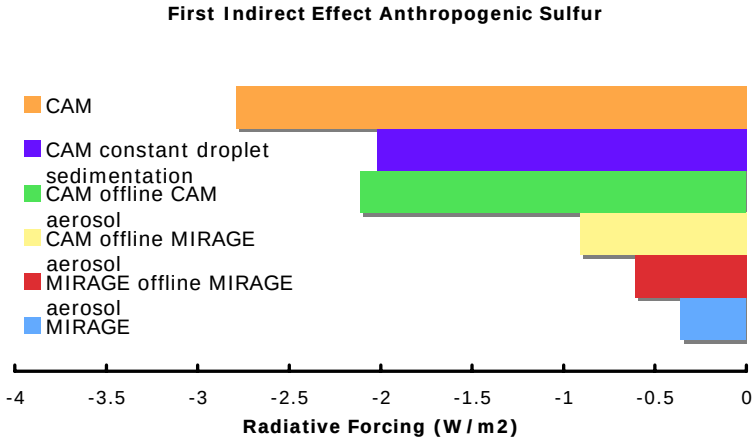

Fig. 6. Global first indirect effect of anthropogenic sulfate simulated by baseline and sensitivity simulations of CAM3 and MIRAGE2 (Ghan, 2007, inclusion with permission of S. J. Ghan, Pacific Northwest National Laboratory, 2007).