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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 the development and application of online-coupled meteorology and chemistry models, with a focus on five representative models developed in the US including GATOR-GCMOM, WRF/Chem, CAM3, MIRAGE, and Caltech unified GCM. These models represent the current status and/or the state-of-the science treatments of online-coupled models worldwide. Their major model features, typical applications, and physical/chemical treatments are compared with a focus on model treatments of aerosol and cloud microphysics and aerosol-cloud interactions. Aerosol feedbacks to planetary boundary layer meteorology and aerosol indirect effects are illustrated with case studies for some of these models. Future research needs for model development, improvement, application, as well as major challenges for online-coupled models are discussed.

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 (RH) 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 and vice versa through various feedback mechanisms (Brasseur and Roeckner, 2005; Jacobson, 2002). Increasing evidence from field measurements have shown that such feedbacks ubiquitously exist among the Earth systems 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

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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 lead to the formation of many small cloud droplets, thus inhibiting cloud droplet coalescence into raindrops and riming on ice precipitation (Rosenfeld, 2000). While the 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.

Although 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) that are developed either based on a general circulation model (GCM) or a global chemical transport model. First, the coarse spatial resolution (e.g., $4^\circ \times 5^\circ$) 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 (except for a few models that use a smaller time step, e.g., GATOR-GCMOM 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., Lohmann and Feichter, 1997; Chuang et al., 1997, 2002; Ghan et al., 2001a, b, c; Nagashima et al., 2002; Steil et al., 2003; Hauglustaine et al., 2004; 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 urban/regional air quality models are driven by a global model with inconsistent model physics (e.g., Langmann et al., 2003; Hogrefe et al., 2004; Tulet et al., 2005; Sanderson et al., 2006). Most regional climate models use prescribed aerosols or simple modules without detailed gas-phase chemistry, aerosol microphysics, and aerosol-cloud interactions (e.g., Giorgi et al., 1993 a, 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 worldwide are reviewed in Sect. 2. Several representative online-coupled meteorology and chemistry models developed in the US are used to illustrate the current status of online-coupled models in Sect. 3. Their major model features, typical applications, and physical/chemical treatments are compared with a focus on aerosol and cloud microphysics treatments and aerosol-cloud interactions. Simulated aerosol feedbacks to planetary boundary layer meteorology and aerosol indirect effects are illustrated with case studies for some of these models in Sect. 4. Major challenges and recommendations for future needs for the development, improvement, and application of online-coupled models are discussed in Sect. 5.

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

2.1 Concepts, history, and milestones of online-coupled models

Atmospheric chemistry or air quality and climate or 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 or chemical fields 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 GCM (referred to as offline coupling). In addition to a large amount of data exchange, this offline separation does not permit simulations 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 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, inverse/adjoint 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 model simulations (e.g., real-time operational forecasting or simulating the impact of future climate change on air quality). Reported differences in simulation results from online and offline models can be fairly small or quite significant, depending on the level of complexities of the model treatments and the simulated variables. 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%. While their online radiation calculation was carried out every 5-hr based on the O_3 fields simulated by a detailed tropospheric O_3-NO_x-hydrocarbon chemistry and did not account for the radiation feedbacks into the climate calculation, their offline radiation calculation was based on the monthly-mean O_3 fields. Shindell et al. (2001) found that the tropospheric oxidation capacity in terms of hydroxyl radical (OH) simulated by their online model is lower by $\sim$10% than that of the same model but running offline. Jacobson (2002) and Chung and Seinfeld (2005) reported a positive forcing of fossil-fuel black carbon (BC) and organic matter using their online-coupled models, whereas other models that do not account for aerosol feedbacks and use a different mixing state treatment for BC 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 constraints.

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 enabled 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 an interface between the two models (referred to as unified online coupling). In models with a unified online coupling, the equations can be solved simultaneously with a nonlinear equation solver or the meteorological and air quality processes can be solved using operator splitting; the latter is more often used at present. The main difference between the two types of coupling is that the 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; Langmann, 2000) sophisticated 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 sophisticated one.

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. Figure 1 shows chronology

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, R95 L00	G91, B88	P92	J94, J95, J96, J97a, J97b	J02, J04a-d	G05	J06 J07	F06	L08	Jö06
Online meteorology and pollutant transport														
O_3	Y	Y					Y							
O_3 and some other gases and families					Y	Y				Y		Y		Y
All photochemically-active gases								Y	Y		Y			
Single bulk or modal aerosol				Y		Y				Y			Y	Y
All discrete, size-resolved aerosol particles								Y	Y		Y	Y		
All chemicals within discrete, size-resolved aerosol particles								Y	Y		Y			
All bulk or modal or size-resolved hydrometeor 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 for others gases					Y	Y								
Time-dependent for all reacting and transported gases								Y	Y	Y	Y	Y	Y	Y
Time-dependent for aerosols with comprehensive dynamics treatments								Y	Y	Y	Y	Y	Y	Y
Online pollutant feedbacks to heating rates to drive meteorology														
No feedback	Y				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		Y		Y
Feedback of all photochemically-active gases to heating rates from spectral radiative transfer								Y	Y		Y			
Feedback of online bulk or modal or size-resolved aerosol to parameterized heating rate				Y		Y				Y		Y		
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			
Online pollutant feedbacks to photolysis to drive photochemistry														
No photolysis				Y										
Photolysis from lookup table or fixed or a preprocessor model, without feedback	Y	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		Y		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		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, M. A. (1975), B88 – Baklanov (1988), C70 – Clark J.H.E. (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), Jö06 – Jöckel et al. (2006), L00 – Langmann (2000), P84 – Penenko et al. (1984), P92 – Pitari et al. (1992), R95 – Rasch et al. (1995), S79 – Schlesinger and Mintz (1979), and T85 – Thompson (1985).

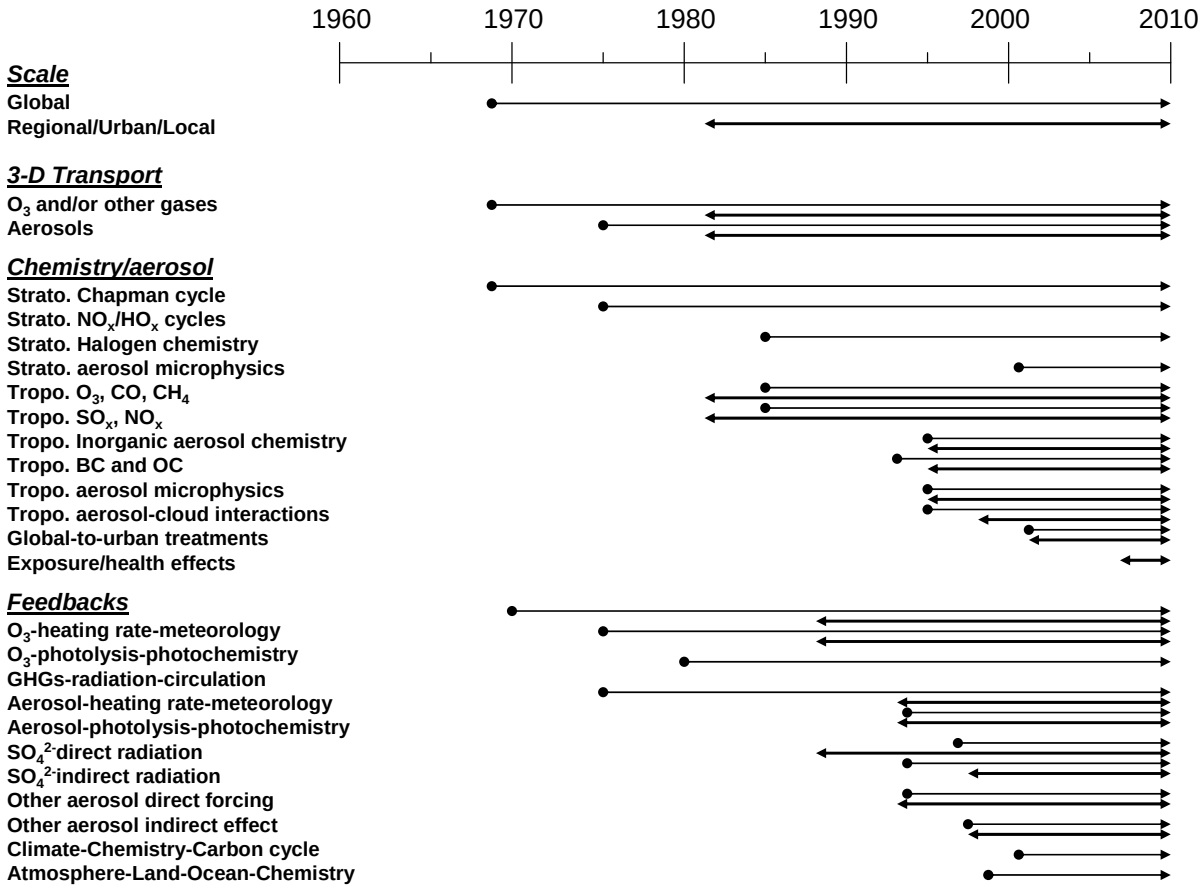

Fig. 1. The development history in chronological order and milestones in terms of chemistry/aerosol and feedback treatments for online-coupled models. ●⟶ and ⟷ indicate the time and treatments in global and regional models, respectively.

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 of 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; Schlesinger and Mintz, 1979). 3-D transport of bulk aerosols and their feedbacks into radiation heating to drive meteorology were also 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). The earliest attempt in coupling meteorology and air pollution in local to regional scale models can be traced back to early 1980s. 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 included highly-simplified aerosol treatments and the direct radiation feedbacks of bulk aerosols to heating/reflection and other atmospheric processes at a local scale.

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; and 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 Jöckel et al., 2006), both tropospheric and stratospheric chemistry (Jöckel et al., 2006 and Teyssèdre et al., 2007), 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; Stier et al., 2005, 2007; and Lohmann et al., 2007). 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 and 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; and O'Connor et al., 2006). The online-coupled meteorology-chemistry models developed at urban/regional scales for studies of tropospheric air pollutants and their interactions with regional climate and meteorology include those 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, 2005; Fast et al., 2006; and Kaminski, 2007), Asia (e.g., Uno et al., 2001; 2003), Australia (e.g., Manins, 2007), and Europe (e.g., Tulet et al., 1999, 2003, 2005, 2006; Langmann, 2000, 2007; Langmann et al., 2008; 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; and Korsholm et al., 2007). Some of European online models were developed through the European Cooperation in Science and Technology (COST) action 728 (http://www.cost728.org). Among these mesoscale models, the work done by Jacobson (1994, 1997a, b) is the one with the highest degree in coupling. In his model, chemistry is solved for all transported gases; all chemically-active gases and size-resolved aerosol components are transported; and feedbacks of all photolyzing gases and aerosols to meteorology through heating rates and to photolysis through actinic fluxes are treated (see Table 1). Some of the mesoscale online meteorology-chemistry models have been coupled with population exposure and health effects (e.g., Jacobson, 2007 and Baklanov et al., 2007b). Several online-coupled regional climate-chemistry/aerosol models have also been developed 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 and 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 and 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 et al., 2004, 2006; Zhang et al., 2005a, b; Hu and Zhang, 2006; Gustafson et al., 2007; Korsholm et al., 2007; Sofiev, 2007; and Knoth and Wolke, 2007). 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), Textor et al. (2006), and Ghan and Schwartz (2007), and those for earth system modeling have been reviewed in Friedlingstein et al. (2006). Some of the mesoscale online-coupled models have been briefly reviewed in Baklanov (1990, 2007) and Baklanov et al. (2007a).

The coupling in most 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; Schlesinger 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; Lamarque et al., 2005) and between aerosols and clouds (e.g., Liao et al., 2003; Lamarque et al., 2005). 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). 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-cloud-radiation feedbacks is treated in very few mesoscale models (e.g., Jacobson, 1994, 1997a, b; Grell et al., 2005).

2.2 History of online-coupled models developed in the US

The current status of a number of online models in Europe has been reviewed in Baklanov and Korsholm (2007) and Baklanov et al. (2007a). Most of the European online models were developed in recent years, and very few of them are fully-coupled models that account for all major feedbacks. In this work, five online models on both regional and global scales developed in the US are selected to represent the current status of online-coupled models worldwide 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 Forecasting/Chemistry model (WRF/Chem) (Grell et al., 2005; Fast et al., 2006), and three global models, i.e., the Community Atmospheric Model v. 3 (CAM3) (Collin et al., 2006a), the Model for Integrated Research on Atmospheric Global Exchanges version 2 (MIRAGE2); Textor et al., 2006; Ghan and Easter, 2006), 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. While GATOR-GCMOM, WRF/Chem, and MIRAGE represent the state-of-the-science online-coupled models in the world with many feedbacks accounted for, CAM3 and Caltech unified GCM represent the current transition of 3-D models from offline to online in which meteorology and chemistry are coupled and feedbacks among various processes are being accounted for. In the following section, history and current status of the five models along with other relevant models developed in the US are reviewed.

Jacobson (1994) developed a unified fully-coupled online meteorology-chemistry-aerosol-radiation model on urban and 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 done in an either slightly- or somewhat incompletely-coupled fashion 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)/the National Center for Atmospheric Research (NCAR) nonhydrostatic mesoscale model (MM5, Grell et al., 1994) was coupled online only with the gas-phase chemical mechanism of the 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 US (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 mesoscale meteorology/chemistry/aerosol/radiation model, WRF/Chem. WRF/Chem represents the first community online-coupled model in the US. Different from other models, a community model refers to a model that is publicly available. This type of model represents synergetic model development efforts by contributors from community and also a major trend of development and application of current models including online-coupled models. 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., Fast-J photolysis algorithm does not account for the feedbacks of all photochemically-active gases to photolysis), the degree of coupling for many atmospheric processes is much more significant as compared with earlier work. In WRF/Chem, transport of meteorological and chemical variables is treated using the same vertical and horizontal coordinates and the same transport scheme with no interpolation in space and time. The meteorological model is 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 is 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 and 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 vapor (H_2O), O_3, carbon dioxide (CO_2), other trace gases such as nitrous oxide (N_2O), methane (CH_4), trichlorofluoromethane (CFC-11), dichlorofluoromethane (CFC-12), chlorofluorocarbon 22 (CFC-22), and carbon tetrachloride ($CC1_4$), 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 recently been incorporated into the latest version of WRF/Chem (version 2.2) by external developers (Fast et al., 2004, 2006; Zhang et al., 2005a, 2007; Hu and Zhang, 2006, 2007; Huang et al., 2006; Pan et al., 2008). The two new gas-phase mechanisms are the Carbon-Bond Mechanism version Z (CBM-Z) (Zaveri and Peters, 1999) and the 2005 version of Carbon Bond mechanism (CB05) of Yarwood et al. (2005) and Sarwar et al. (2005, 2008) (both are variants of Carbon Bond Mechanism IV (CBM-IV) of Gery et al., 1989). The two new aerosol modules are the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008) and the Model of Aerosol Dynamics, Reaction, Ionization, and Dissolution (MADRID) (Zhang et al., 2004). 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, it however does not account for the feedbacks of other radiatively absorbing gases such as NO_2, formaldehyde (HCHO), peroxyacetyl nitrate (PAN), hydroperoxy radical (HO_2), and nitric acid (HNO_3) to the online calculation of photolysis. 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.

On a global scale, a number of climate or air quality models have been developed in the US 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 GCM without chemistry, CCM0 and CCM1 (Washington, 1982; Williamson et al., 1987), the NCAR CCM has evolved to be one of the first-generation unified online climate-chemistry models in the US 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) and online calculations of soil dust and seat salt emissions (Mahowald et al., 2006a, b).

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 is designed to treat gases, size- and composition-resolved aerosols, radiation, and meteorology for applications from the global to urban (<5 km) scales and includes 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 are

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 O_3 in northern and central California and global direct forcing of BC (Jacobson, 2001c, d, 2002). GATOR-GCMM was extended to Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR-GCMOM) in Jacobson (2004a, b, 2005b, 2006b) and Jacobson et al. (2004, 2006b, 2007) by the 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.

MIRAGE2 is an aerosol-climate model built upon the NCAR CAM2 climate model. Most of its treatments of aerosol chemistry and physics are from its predecessor MIRAGE1 (Ghan et al., 2001a, b, c; Easter et al., 2004) which used the same Pacific Northwest National Laboratory (PNNL) Global Chemistry Model (GChM) but a different framework (i.e., the NCAR CCM2 climate model coupled online with a chemical transport model). The NCAR CCM2 climate model and GChM in MIRAGE1 can be run offline or online via an interface (i.e., separate online coupling) (Ghan et al., 2001 a, b, c; Easter et al., 2004). In MIRAGE2, the gas/aerosol treatments are an integrated model imbedded in NCAR CAM2 (i.e., unified online coupling). As a result, the treatment of the cloud processing of gas/aerosols in MIRAGE2 is closer to that in CAM2, as compared to MIRAGE1. Also, the transport/advection treatments in MIRAGE 2 are numerically identical for water and gas/aerosol species. The prescribed CH_4, NO_x, and O_3 but prognostic steady state OH and HO_2 are used in MIRAGE 1 and offline oxidant chemistry (except for prognostic H_2O_2 using offline HO_2) is used in MIRAGE 2. Both MIRAGE 1 and 2 contain identical aerosol treatments. The aqueous chemistry and wet removal are simulated online in MIRAGE 2.

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 worldwide; 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 aforementioned online-coupled meteorology and chemistry models developed in the US 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.

3.1 Chemistry, emissions, and typical model applications

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, CBM-Z, and CB05) with 156–237 chemical reactions among 52–77 chemical species and three aerosol modules (i.e., MADE/SORGAM, MOSAIC, and MADRID). CBM-Z extends the original CBM-IV mechanism to function properly at regional to global spatial scales and longer time periods than the typical urban air-shed simulations. The CBM-Z version implemented in WRF/Chem also includes a condensed dimethylsulfide (DMS) photooxidation mechanism (Zaveri, 1997) to simulate the temperature-dependent formation of SO_2, H_2SO_4, and methanesulfonic acid (MSA) in the marine environment. Compared with CBM-IV, the main changes in CB05 include updates of kinetic data (i.e., rate coefficients) and photolysis data (i.e., absorption cross-sections and quantum yields), extended inorganic reaction set (e.g., reactions involving H_2 and NO_3), explicit acetaldehyde, propionaldehyde and higher aldehydes, alkenes with internal double bonds (internal olefins) (e.g., 2-butenes), oxygenated products and intermediates (e.g., higher organic peroxides and peroxycarboxylic acids), and lumped terpene chemistry. In the latest version of WRF/Chem (v. 2.2) released in March, 2007, a generic chemical kinetic solver, the Kinetic PreProcessor (KPP), has been included to facilitate the users to incorporate any new gas-phase chemical mechanisms into WRF/Chem. 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 4 or 8 sections, but it can be changed to any number of sections) is

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

Model System/Scale	Meteorology Model	Chemical Transport Model (Main features)	Emissions	Typical Applications	Example References
GATOR-GCMOM and 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	Online: all natural gases and particles Offline: anthropogenic and volcanic emissions	Current/future met/chem/rad feedbacks; Direct/indirect effects; AQ/health effect	Jacobson, 1994, 1997a, b, 2001c, 2002, 2004a, b; Jacobson et al., 2004, 2006a, 2007
WRF/Chem (Mesoscale)	WRF	RADM2, RACM, CBM-Z, CB05 (156–237 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	Online: biogenic and sea-salt emissions Offline: anthropogenic emissions and other natural emissions	Forecast/hindcast, Met/chem feedbacks; O_3, $PM_{2.5}$; Aerosol direct and indirect effects	Grell et al. (2005); Fast et al. (2006); McQueen et al. (2005, 2007); Zhang et al. (2005a, b, 2007) ; Tie et al., 2007; Gustafson et al., 2007
CAM3 and Predecessors (Global)	CCM3/ CCM2/ CCM1	Sulfur chemistry (14 reactions), prescribed CH_4, N_2O, CFCs/MOZART4 gas-phase chemistry (167 reactions, 63 species); Bulk aqueous-phase sulfate chemistry of S(IV) (4 equilibria and 2 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Online: soil dust, sea-salt, and biogenic emissions Offline: anthropogenic emissions and other natural emissions	Climate; Direct/indirect effects; Hydrological cycle	Rasch et al., 1995, 2006; Kiehl et al., 1998; Collins et al., 2004, 2006a, b; Lamarque et al., 2005; Heald, 2007
MIRAGE2 and 1 (Global)	CAM2/ CCM2	Gas-phase CO-CH_4-oxidant chem.(MIRAGE 1 only); Bulk aqueous-phase sulfate chemistry (6 equilibria and 3 kinetic reactions); Mode-resolved simple aerosol treatment; Prognostic aerosol/cloud treatments	Online: soil dust and sea- salt emissions Offline: anthropogenic emissions and other natural emissions	CO (MIRAGE 1 only), Aerosol mass/number, Sulfur cycle; Direct/indirect effects	Ghan et al., 2001a, b, Zhang et al., 2002; Easter et al., 2004; Textor et al., 2006; Ghan and Easter, 2006
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	Online: soil dust and sea- salt emissions Offline: anthropogenic emissions and other natural emissions	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

used in MOSAIC and MADRID. RADM2 and RACM have been coupled with MADE/SORGAM and CBM-Z has been coupled with MOSAIC and MADRID; CB05 has been coupled with MOSAIC and MADRID (Zhang et al., 2007a; Pan et al., 2008). While CBM-Z and MOSAIC have been included in the latest released version 2.2 of WRF/Chem, CB05 and MADRID are being tested by the author's group and will be released in the near future. MADE/SORGAM is coupled with the bulk RADM aqueous-phase chemistry that simulates aqueous-phase chemistry of sulfate with 5 kinetic reactions, MOSAIC/MADRID is coupled with the bulk Carnegie Mellon University (CMU) 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 some 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 levels of details, a simple mechanism with prescribed methane (CH_4), nitrous oxide (N_2O), chlorofluorocarbons (CFCs), radicals (e.g., OH, 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 uses offline oxidants for the carbon monoxide (CO)-CH_4-oxidant chemistry (except for prognostic H_2O_2 using offline HO_2) and treats the gas-phase oxidation of SO_2 and DMS by OH. Its aqueous-phase chemistry includes dissolution equilibria of SO_2, H_2O_2, O_3, sulfuric acid (H_2SO_4), and methane sulfonic acid (MSA) and aqueous-phase kinetic reactions of S(IV) with H_2O_2 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.

Emissions used in these models include both natural and anthropogenic emissions. Emissions of some sources and species are a strong function of temperature (e.g., biogenic VOC emissions from vegetation, evaporative emissions for anthropogenic VOCs), solar radiation (e.g., isoprene emissions), precipitation (e.g., mercury emissions from soils), and wind speed (e.g., dust emissions from soil erosion and sea salt emissions). In order to accurately simulate the effect of climate and meteorological changes on air quality in a truly integrated manner, meteorologically-dependent emissions should be treated online. Currently, emissions are, however, treated offline in most models and very few models include online emissions for all meteorologically-dependent species. In GATOR-GCMOM, emissions of all natural gases and particles (e.g., sea spray and its chemicals, soil dust and its chemicals, lightning chemicals, pollen, spores, bacteria, biogenic gases, soil NO_x, and DMS from the ocean) are treated online and are affected by simulated meteorological conditions. The effect of meteorology on the height of emissions from biomass-burning and volcanos is accounted for. WRF/Chem contains online calculation of emissions of biogenic isoprene, monoterpenes, other biogenic VOCs, and nitrogen emissions by the soil based on the US EPA Biogenic Emissions Inventory System (BEIS) (www.epa.gov/asmdnerl/biogen.html) and sea-salt (Grell et al., 2005; Fast et al., 2006). While MIRAGE 1 and MIRAGE 2 simulate sea-salt emissions online, dust emissions can be simulated either online (e.g., Easter et al., 2004) or offline (e.g., Textor et al., 2006). Caltech unified GCM simulates the emissions of soil dust and sea-salt online. CAM3 simulates the emissions of soil dust and sea-salt (Tie et al., 2005; Mahowald et al., 2006 a, b) as well as biogenic species online based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) of Guenther et al. (2006) that has been incorporated into CAM3 along with the Model for Ozone and Related Chemical Tracers version 4 (MOZART-4) (Lamarque et al., 2005; Heald, 2007) (http://www.essl.ucar.edu/LAR/2006/catalog/ACD/hess.htm), although offline biogenic emissions can also be used in some CAM3 simulations.

Those models have been developed for different applications. GATOR-GCMOM has been applied for studying the effect of BC within clouds and precipitation on global climate (Jacobson, 2006b), the simulation of feedbacks among meteorology, chemistry and radiation on urban-to-global scales for both current and future emission/climate scenarios, the estimates of global aerosol direct/indirect effects (e.g., Jacobson, 2002; Jacobson et al., 2007), and the effects of ethanol versus gasoline vehicles on cancer and mortality in the US (Jacobson, 2007). WRF/Chem and its variations were developed and applied for real-time air quality forecasting (e.g., Grell et al., 2005; Kang et al., 2005; McKeen et al., 2005; 2007; Pagowski 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 et al., 2004, 2006; Zhang et al., 2005a, Frost et al., 2006; Hu and Zhang, 2006; Huang et al., 2006; Hu et al., 2007; Xie et al., 2007; Gustafson et al., 2007). 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 (Gustafson et al., 2007; Zhang et al., 2007a, 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 the 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 CO, sulfur cycle, 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). These results have been evaluated rigorously using available gas, aerosol, and cloud measurements (Ghan et al., 2001a, b, c; Easter et al., 2004; Kinne et al., 2004, 2005). 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).

3.2 Aerosol properties

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. GATOR-GCMOM treats 47 species including sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water (H_2O), carbonate

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 (Global-through-urban)	47 species (sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water, carbonate, crustal species such as Ca^{2+}, K^{+}, and Mg^{2+})	Sectional (17–30)[a]: variable, multiple size distributions	A coated core, internal/external mixtures	Predicted/Predicted	Simulated hydrophobic-to-hydrophilic conversion for all aerosol components	Simulated volume-average refractive indices and optical properties based on core-shell MIE theory
WRF/Chem (Mesoscale)	Sulfate, nitrate, ammonium, BC, OC, and water in all three aerosol modules, sea-salt and carbonate in MOSAIC/MADRID, and menthansulfonate in MOSAIC	Modal (3): variable (MADE/SORGAM) Sectional (8): variable (MOSAIC/MADRID) single size distribution	Internal	Predicted/ Predicted	Similar to MIRAGE2	Similar to MIRAGE2
CAM3 (Global)	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water	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 conversion rate	Prescribed RI and optical properties for each aero. type, size, and wavelength, for external mixtures
MIRAGE2 (Global)	Sulfate, BC, OC, sea-salt, dust, water	Modal (4): variable; single size distribution	Externally mixed modes with internal mixtures within each mode	Predicted/Diagnosed or predicted	Simulated (volume averaged) with prescribed hygroscopities for OC and dust	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, water, Ca^{2+}	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-}, NH_4^{+}, NO_3^{-}, sea-salt, and H_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

[a] The number in the parentheses indicates the total of aerosol size sections or modes used in typical applications of the models.

(CO_3^{2-}), and crustal species (e.g., calcium (Ca^{2+}), potassium (K^{+}), and magnesium (Mg^{2+})) and their salts. MIRAGE2 treats the least number of species including sulfate, BC, organic carbon (OC), sea-salt, dust, and water (H_2O). Nitrate and ammonium are treated in CAM3, WRF/Chem, and Caltech unified GCM. Additional species such as calcium (Ca^{2+}) and carbonate (CO_3^{2-}) are treated in WRF/Chem-MOSAIC/MADRID. Both CAM3 and MIRAGE2 use modal approaches with four modes to represent aerosol size distributions. GATOR-GCMOM uses a 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 with 8 sections for MOSAIC and MADRID for typical applications). MOSAIC and MADRID can be applied for any number of size sections. Caltech unified GCM prescribes size distribution of sea-salt and dust with the sectional distribution but that of other aerosols with the modal distribution. 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 errors 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 for externally-mixed (i.e., distinct from other aerosol particles), 0.78 for well-mixed (i.e., incorporated within other aerosol particles), and $0.54\,W\,m^{-2}$ for core treatments (i.e., a black-carbon core could be surrounded by a well mixed shell), according

to 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 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 the transition of externally-mixed aerosols into internally-mixed aerosols with a coated BC core. It treats one or more size distributions of aerosols. The multiple aerosol size distributions represent aerosols with different sources and mixing states (e.g., freshly-emitted BC, internally-mixed aerosols, and aerosols with a coated BC core). The 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 aerosol modes with internal mixtures of aerosol components within each mode in MIRAGE2, and BC, OC, and mineral dust externally-mixed with internally-mixed other aerosols in Caltech unified GCM).

All five models predict aerosol mass concentration. CAM3 can also use offline aerosol mass concentrations. Aerosol number concentration is diagnosed (e.g., CAM3) or predicted (e.g., GATOR-GCMOM, WRF/Chem) or both (e.g., MIRAGE 2), but it is not included in the Caltech unified GCM. It is noted that a fixed standard deviation is used for both Aitken and accumulation modes in MADE/SORGAM in WRF/Chem, which introduces errors in simulated aerosol number and mass size distributions (Zhang et al., 1999). The simulated aerosol direct and indirect forcing depend on particle size and hygroscopicity, 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 RIs of wet aerosol in each mode. Volume mixing is assumed for all components, including insoluble components. The main difference between Caltech unified GCM and both MIRAGE2 and WRF/Chem is that Caltech unified GCM prescribes size distribution (e.g., a sectional distribution for seasalt and dust and a standard gamma distribution for 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.

3.3 Model treatments of cloud properties

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 sophisticated 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 simulates prognostically a bulk cloud condensate that includes cloud water and cloud ice with water/ice fractions determined diagnostically, and precipitation is treated diagnostically. WRF/Chem includes several bulk microphysical schemes such as the Kessler scheme (Kessler 1969), the Purdue Lin scheme (Lin et al., 1983; Chen and Sun, 2002), and WRF Single-Moment (WSM) 6-class graupel scheme (Hong et al., 2006). The Purdue Lin scheme used with MOSAIC has 6 prognostic variables: water vapor, 2 bulk cloud categories (cloud water and ice), and 3 bulk precipitation categories (rain, graupel, snow/aggregates). The cloud droplet number was added by PNNL as a prognostic variable in the expanded Lin scheme that is used with MOSAIC in WRF/Chem. Both MIRAGE2 and WRF/Chem predict cloud size distribution as a single, modal distribution (Barrie et al., 2001). CAM3 treats bulk liquid and ice with

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 (Global-through-urban)	Size-resolved liquid, ice, graupel, aerosol core components in stratiform subgrid convective clouds	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 (Mesoscale)	bulk water vapor, rain, snow, cloud ice, cloud water, and graupel or a subset of them, depending on microphsics schemes used in both stratiform and subgrid convective clouds	Prognostic, modal, single size distribution (MOSAIC)	Similar to MIRAGE2 (MOSAIC)	Similar to MIRAGE2 but sectional; CCN only	Similar to MIRAGE2 but sectional, CCN only	Similar to MIRAGE2 but sectional (MOSAIC)
CAM3 (Global)	Bulk liquid and ice in both stratiform and subgrid convective clouds	Prognostic in microphysics calculation but prescribed in sedimentation and radiation calculation as a function of temperature by phase and location	Prescribed or prognostic (similar to MIRAGE2)	All treated species except hydrophobic species; CCN only	Prescribed; CCN only	Similar to MIRAGE2
MIRAGE2 (Global)	Bulk liquid and ice in both stratiform and subgrid convective clouds	Prognostic, 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 in both stratiform and subgrid convective clouds	Diagnosed from predicted cloud water content; single size distribution	constant cloud droplet number based on observations	None	None	Simulated based on MIE theory with different parameterizations for liquid and ice clouds

the same prognostic droplet size treatment as MIRAGE2 in microphysics calculation, but the droplet size treatment is prescribed in sedimentation and radiation calculation as a function of temperature by phase and location (Boville et al., 2006). All five models distinguish large-scale stratiform and subgrid convective clouds but with some differences in their treatments. For example, in GATOR-GCMOM and MIRAGE2, large-scale stratiform clouds can cover a fraction of a grid cell. In WRF/Chem, stratiform clouds have a cloud fraction of 0 or 1 and the aerosols are not affected by sub-grid convective clouds (e.g., Kain-Fritsch option). Neglecting sub-grid cloud treatments may introduce large errors for the horizontal grid resolution greater than 15-km. For both resolved and convective clouds in GATOR-GCMOM, microphysics is explicit and involves growth of water vapor onto discrete size-resolved aerosol particles to form discrete, size-resolved clouds and precipitation (liquid, ice, and graupel), and aerosol inclusions are tracked in each size of each hydrometeor distribution (Jacobson and Kaufman, 2006), whereas other models do not contain such detailed treatments.

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, although the prescribed cloud droplet number can also be used in the cloud miscrophysics parameterization of

Rasch and Kristjánsson (1998) in CAM3. Caltech unified GCM 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 similar treatments for droplet number, with droplet nucleation parameterized by Abdul-Razzak and Ghan (2000). WRF/Chem-MOSAIC diagnoses the total number activated from the sectional size distribution of the CCN, which is then used to predict the droplet number that has a modal distribution. 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 2 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 2 and WRF/Chem simulate it as a function of aerosol size and hygroscopicity based on Köhler theory. CAM3 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 2 and CAM3 use a prognostic parameterization in terms of cloud water and 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 distributions 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.

3.4 Aerosol thermodynamics and dynamics

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 the EQUIlibrium SOLVer Version 2 (EQUISOLV II) of Jacobson (1999c) that simulates equilibria of all major inorganic 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). EQUISOLV II has been extended to the Predictor of Nonequilibrium Growth (PNG)-EQUISOLV II to overcome the oscillatory problem in solving the equilibrium and growth at a long time step (150–300 s) (Jacobson, 2005a). 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) of Binkowski and Shankar (1995) 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 and EQUISOLV II. They found that MARS-A has the fastest computational speed but it may not be applicable to dry areas with low relative humidities (RHs) and coastal areas. Zhang and Jacobson (2005a) evaluated ISORROPIA and EQUISOLV II in both a box model with 11 200 test cases and a 3-D model over continental US. While they found that ISORROPIA gives results that are consistent with those of benchmark and EQUISOLV II under most conditions, larger bias may occur for RHs $\leq$40 or $\geq$99 for most species, mainly because of the use of an approximate treatment for water content and solid-liquid equilibrium in the mutual deliquescence region at moderate and low RHs (Ansari and Pandis, 1999; Zaveri et al., 2008) and errors in activity coefficients used at very high RHs. An improved ISORROPIA (version 1.7) has been developed and implemented in WRF/Chem-

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
GATOR-GCMOM (Global-through-urban)	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) (PNG-EQUISOLV II) for all treated species
WRF/Chem (Mesoscale)	MARS-A (SORGAM) MESA-MTEM (MOSAIC) ISORROPIA (MADRID)	Reversible absorption (8 classes VOCs) based on smog-chamber data (SORGAM) Absorption (MADRID1) and combined absorption and dissolution (MADRID2). No SOA treatment in MOSAIC	Binary homogeneous nucleation of H_2SO_4 and H_2O of Kulmala et al. (1998 b) (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 modes only	1. Full equilibrium for HNO_3 and NH_3 in MADE/SORGAM and all species in MADRID 2. Dynamic for H_2SO_4 in MADE/SORGAM; Dynamic for all species in MOSAIC and MADRID 3. Hybrid in MADRID
CAM3 (Global)	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 $(NH_4)_2SO_4$ and NH_4NO_3
MIRAGE2 (Global)	Sulfate assumed to be $(NH_4)_2SO_4$, no nitrate	Prescribed SOA yield for monoterpenes	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 modes only; Brownian diffusion	Dynamic approach for H_2SO_4 and MSA
Caltech unified GCM (Global)	ISORROPIA with regime equili. for sulfate, nitrate, ammonium, sea-salt, and water	Reversible Absorption for 5 biogenic VOC classes	None	None	None	Full equilibrium involving $(NH_4)_2SO_4$ and NH_4NO_3

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. MESA and EQUISOLV II are evaluated against the AIM Model III and they give overall similar results in terms of both mass growth factors and performance statistics relative to the AIM Model III for the 16 cases tested in Zaveri et al. (2005a). A major factor contributing to the differences in simulated results from various aerosol thermodynamic modules is the activity coefficients used in these modules. For example, EQUISOLV II and ISORROPIA account for temperature-dependence for all activity coefficients when such information are available, the activity coefficients used in MESA, however, are limited for 298 K, which may introduce errors for their applications for upper tropospheric and stratospheric conditions (e.g., the tropical tropopause

where the temperature may fall below 200 K and activity coefficients of species may deviate largely from their values at 298 K). While EQUISOLV II provides a generic code for aerosol thermodynamic calculation, most other modules (e.g., ISORROPIA and MESA) require non-trivial efforts to expand the system of equations for more species and/or other temperatures and/or the re-development of some parameterizations used.

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 volatile organic compounds (VOCs), 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 VOCs via condensation and dissolution based on Henry's law. Caltech unified GCM simulates the formation of SOA based on a reversible absorption of 5 classes of biogenic VOCs and neglect that from anthropogenic VOCs. In MADE/SORGAM in WRF/Chem, SOA formation via the reversible absorption of 8 classes of VOCs is simulated based on smog-chamber data of Odum et al. (1997) and Griffin et al. (1999). The same approach for SOA modeling has been used in an offline version of MOSAIC, which, however, has not been incorporated into WRF/Chem for 3-D applications. Two approaches are used to simulate SOA formation in WRF/Chem-MADRID (Zhang et al., 2004). MADRID 1 uses an 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 1 has been upgraded to Sesqui-MADRID (MADRID 1.5) and now treats phase separation (i.e., a relatively hydrophilic phase and a relatively hydrophobic phase) within the organic particulate phase when thermodynamically favorable (Pun et al., 2008). MADRID 2 has been modified to be compatible with any gas-phase mechanism (Pun et al., 2006). A variation of MADRID 2 that is computationally efficient has been incorporated into the Mesoscale Nonhydrostatic Chemistry (Meso-NH-C) model of Tulet et al. (2003) that couples meteorology and chemistry online (Tulet et al., 2005, 2006). Simulated SOA concentrations by most 3-D models are, however, lower than observations for several reasons. For example, these models use the yields for aromatics and monoterpene oxidation under high NO_x conditions (e.g., Odum et al., 1997; Griffin et al., 1999; Ng et al., 2007 a, b). Some SOA precursors in these models may be missing (e.g., isoprene SOA is not simulated in MADE-SORGAM), which have been shown to be important at both global and regional scales (e.g., Henze and Seinfeld, 2006; Zhang et al., 2007b).

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 the 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 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) in WRF/Chem-MADRID uses an approach that 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 (version 2.2) uses the Adaptive Step Time-split Explicit Euler Method (ASTEEM) method, which has recently been updated to the Adaptive Step Time-split Euler Method (ASTEM) method to reduce the stiffness more effectively using several methods and to allow the use of longer time step ($\sim$100 s) in an offline version of MOSAIC (Zaveri et al., 2008). Coagulation is currently not treated in CAM3 but simulated with a modal approach with several lognormally-distributed modes in MIRAGE2, a sectional approach with a number of size sections in GATOR-GCMOM, and both approaches in WRF/Chem (e.g., MADE/SORGAM uses the modal approach; MOSIAC and MADRID use the sectional approach). 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.

Three approaches are typically used to simulate gas/particle mass transfer in 3-D air quality models: full equilibrium, dynamic, and hybrid. No condensation equation is explicitly solved in the full equilibrium approach (although a weighting factor based on condensational growth law may be used to distribute the transferred mass material from gas to particulate phase over the particle size distribution), whereas condensation is explicitly solved for all particles in the dynamic approach and for particles with diameter greater than a threshold (a typical threshold value of 1 to 2.5 μm is assumed) in the hybrid approach. In such cases, condensation is a sub-process of gas/particle mass transfer. For gas/particle mass transfer, CAM3 and Caltech unified GCM use the simplest full equilibrium approach. MIRAGE2 uses a dynamic approach for H_2SO_4 and MSA. 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 is used for HNO_3 and NH_3 in MADE/SORGAM. A dynamic approach is used for H_2SO_4 in MADE/SORGAM and all species 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 and improve 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. (2008). The box MADRID tests of Hu et al. (2008) 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.

3.5 Aerosol-cloud interactions and cloud processes

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 2 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.

Activation of aerosol particles that can behave as CCN to produce cloud droplets 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 2 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. Important 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-

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	Sedimentation of aerosols and cloud droplets
GATOR-GCMOM (Global-through-urban)	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 on aerosol size and composition	Size-resolved aerosol-hydrometeor coag. (washout), calculated precip. rate dependent on aerosol size and composition	Size-resolved coagulation between hydrometeors and between all aerosols and all hydrometeors	Two-moment discrete size-dependent sedimentation for all aerosol particles and hydrometeors (mass and number) that vary with altitude; sedimentation below cloud leads to shrinkage as a function of drop size
WRF/Chem (Mesoscale)	The same as MIRAGE2 but sectional (MOSAIC)	The same as MIRAGE2 but sectional (MOSAIC); bulk CCN only	The same as MIRAGE2 but sectional	Similar to MIRAGE2 but sectional	Similar to MIRAGE2 but sectional	Two-moment sedimentation for aerosol particles (mass and number) at surface; sedimentation for all hydrometeors or a subset of them, depending on microphysics schemes
CAM3 (Global)	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 on aerosols	Prescribed bulk scav. efficiency, no-size dependence	None	Bulk cloud/ice sedimentation; sedimentation below cloud leads to complete evaporation/sublimation
MIRAGE2 (Global)	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 (nucleation) scavenging, Brownian diffusion (for activated particles), autoconversion and collection for bulk cloud droplets, precip. rate independent of aerosols	Calculated modal scavenging coeff. using a parameterization of the collection efficiency of aerosol particles by rain drops, with size dependence	Modal coagulation between cloud droplets, between cloud droplets and precipitating particles, and between aerosol and precipitating particles	Two-moment sedimentation for aerosol particles (mass and number) at the surface; no sedimentation for cloud droplets/ices, cloud-borne and ice-borne aerosol particles.
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

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 effects are accounted for in GATOR-GCMOM. Aerosol-cloud interaction is currently not treated in Caltech unified GCM.

Aerosols are removed through dry deposition in the absence of hydrometerors and through wet deposition following in- and below- cloud scavenging, in addition to be activated as CCN to form cloud droplets. After activation, cloud droplets (and cloudborne aerosol particles) are removed via autoconversion (i.e., the collision/coalescence of cloud drops to become rain drops and get into precipitation) and via collection/accretion by (existing) precipitation (rain, snow, graupel). CAM3 assumes that in-cloud scavenging occurs via prescribed activation and autoconversion. 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 (nucleation) scavenging, Brownian diffusion (for activated particles), autoconversion, and collection. 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 is treated somewhat differently in GATOR-GCMOM as in other models because of differences in the 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 dependence of precipitation rates on discretely size resolved aerosols via nucleation scavenging and impact scavenging is taken into account in GATOR-GCMOM, but are neglected in other models. The main differences between treatments of cloud-processing of aerosols in GATOR-GCMOM and other models are (1) other models treat removal of aerosols as an empirical function of the rainfall rate without physical interactions of size-resolved aerosols with size-resolved rainfall, and (2) other models do not always track all the aerosol components that the clouds formed on within size resolved cloud drops.

For below-cloud scavenging, CAM3 prescribes scavenging efficiencies. MIRAGE2 and WRF/Chem calculate a scavenging coefficient (=scavenging rate/precipitation rate) using a parameterization of the collection efficiency of aerosol particles by rain drops (due to convective Brownian diffusion and gravitational/inertial capture). 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.

Sedimentation refers to the layer-by-layer sinking of aerosol particles, hydrometeor particles (e.g., cloud drops/ice, cloud-borne and ice-borne particles, and precipitating particles) as a function of their sizes. Sedimentation of cloud droplets and precipitating particles 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 aerosol particles, liquid, ice, graupel particles, and their chemical inclusions with fall speeds for both mass and number concentrations (i.e., so-called two-moment methods) 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 mass and number concentrations of 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). MIRAGE2 also treats two-moment sedimentation for aerosol particles, but it does not treat sedimentation of cloud droplet/ice and cloud-born and ice-born particles. Depending on schemes for cloud microphysics used in WRF/Chem, the sedimentation process is treated for all the hydrometeor categories treated or a subset of them for both mass and number concentrations (Skamarock et

al., 2005). 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 treatment in CAM3 is not physical and prevents an accurate simulation of the physical feedbacks of aerosol particles to climate.

4 Case studies

To illustrate the importance of the feedbacks discussed previously, several case studies on some of the feedbacks using some of the aforementioned models are provided below. These include the feedbacks of aerosols to PBL meteorology by WRF/Chem-MADRID, the feedbacks of aerosols to wind fields and precipitation by GATOR-GCMOM, and the feedbacks of aerosol/cloud to indirect aerosol radiative forcing by MIRAGE2 and CAM3. These studies represent the current status of model capability in simulating such feedbacks with the state-of-the-science treatments.

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 US. The TexAQS-2000 was carried out around the Houston area where exceedance of the National Ambient Air Quality Standard (NAAQS) of 120 ppb O_3 occurs most frequently and VOC reactivities are typically much higher than in other urban areas in the US WRF/Chem uses mass (hydrostatic pressure) coordinates. The horizontal grid spacing is 12-km and the vertical resolution is 57 layers from surface to tropopause with vertical intervals varying from 15 m in the surface layer to 600–680 m near/at the domain top (∼16km). 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. The cloud microphysical scheme is thus turned off. 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).

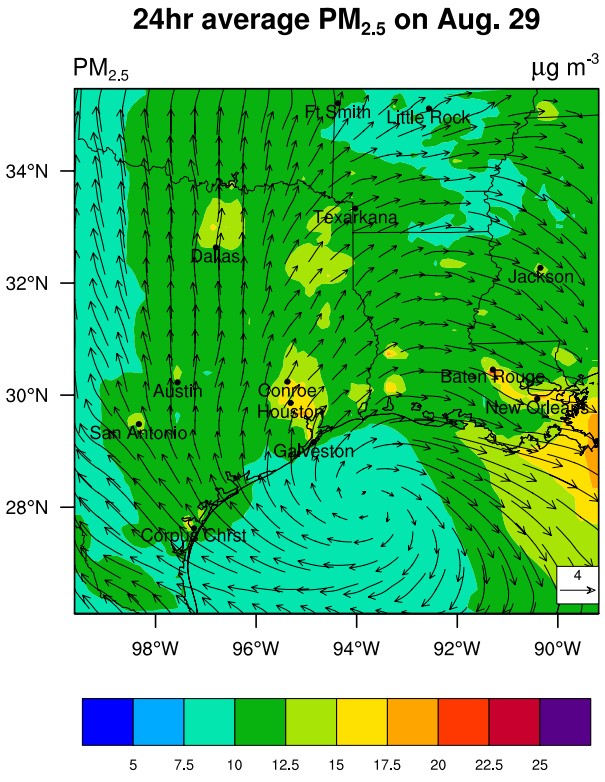

Fig. 2. The spatial distribution of the 24-h average $PM_{2.5}$ concentrations and the 24-h average wind fields predicted by WRF/Chem-MADRID on 29 August 2000 (Zhang et al., 2005a).

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 patterns 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. 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 overestimation of primary BC and organic matter (OM) emissions and high aerosol boundary conditions. Figure 3 shows the vertical profiles of $PM_{2.5}$ concentrations and the differences in vertical temperature (T) and water vapor (Q_v) mixing ratio between simulations with and without PM at five different times on August 29 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 PBL vary significantly from time to time during a day, depending on magnitudes and timing of precursor emissions and related meteorological

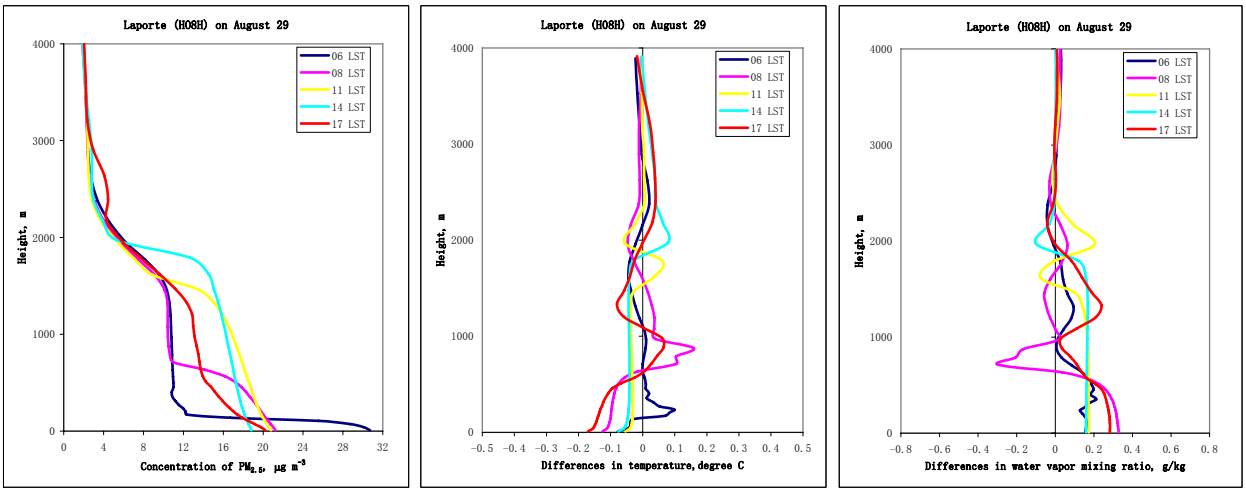

Fig. 3. The vertical distributions of the hourly $PM_{2.5}$ concentrations and differences in vertical distributions of temperatures and water vapor mixing ratios between simulation with and without aerosols by WRF/Chem-MADRID at La Porte, TX at five times (6 a.m., 8 a.m., 11 a.m., 2 p.m., and 5 p.m.) on 29 August 2000 (Zhang et al., 2005c).

conditions such as atmospheric stability, the depth of mixing height, and temperature. The surface $PM_{2.5}$ reaches the highest at 6 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 2 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. The relatively high $PM_{2.5}$ concentrations at/near surface reduce net downward solar/thermal-IR radiation, which in turn causes a decrease in T and an increase in Q_v at/near surface. 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: a so-called California (CA) Grid at a resolution of 0.2°×0.15°(∼21.5 km×14.0 km) and a so-called 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 levels 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 heating 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 for the South coast, CA and the CA grids. AAPPG decreases precipitation in the LA basin and the mountains beyond the basin in February. In August, when precipitation is low, most reductions occur offshore and in the foothills of the San Bernardino Mountains. Some precipitation increases are found on the downslope sides of the San Bernardino and San Gabriel Mountains. Those results are consistent with the findings of Givati and Rosenfeld (2004, 2005).

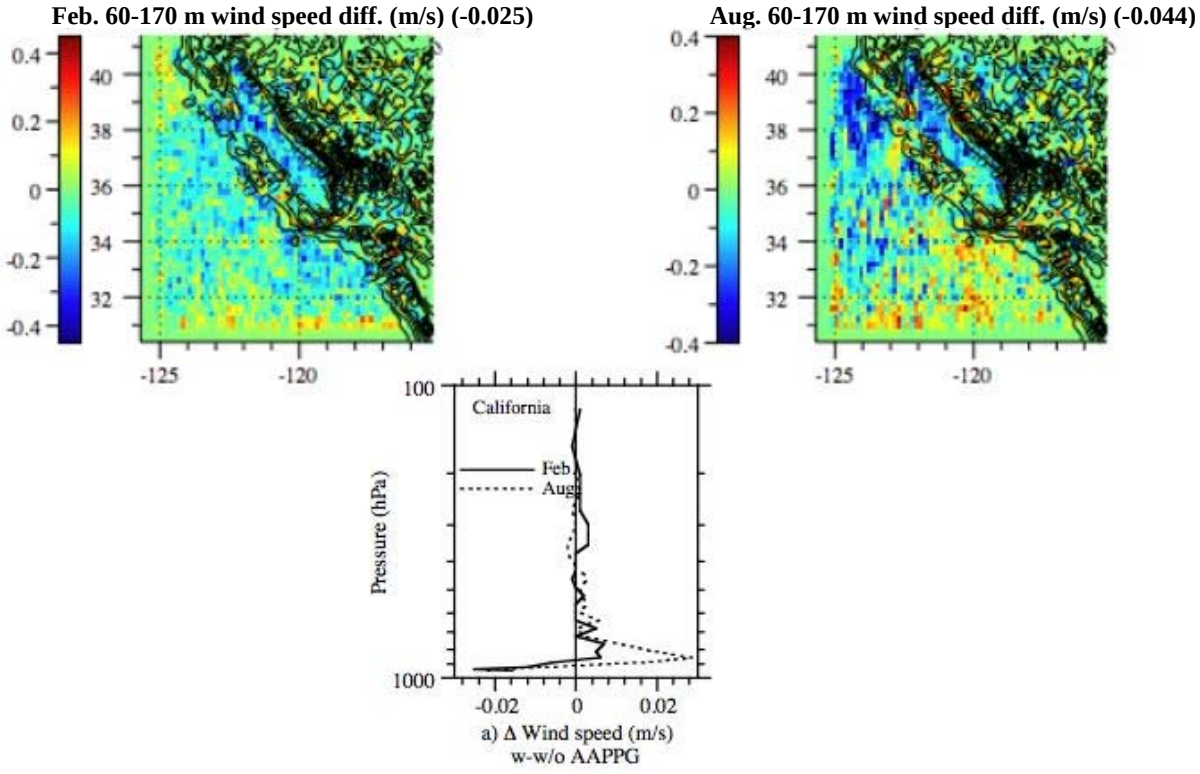

Fig. 4. Differences in the spatial distributions of near-surface wind speeds over California grid and in the domainwide-average vertical distributions of wind speeds between simulation with and without AAPPG by GATOR-GCMOM in February and August 1999. The contours in black lines in the spatial distribution plots indicate topography in meters (provided by M. Z. Jacobson, Stanford University, 2007).

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 $\times$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 2–6). 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 CAM3_B is much larger than that from MIRAGE2_B (3.2 vs. 0.38 $\mathrm{W\,m^{-2}}$), the prediction of MIRAGE2_B is much closer to the total aerosol indirect forcing of 0.75 $\mathrm{W\,m^{-2}}$ estimated by IPCC (2007). MIRAGE2 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 simulation 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 increased aerosol presence under cloudy conditions. The use of MIRAGE2 aerosol module in offline CAM3 significantly reduces FAIE in CAM3, suggesting that the 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).

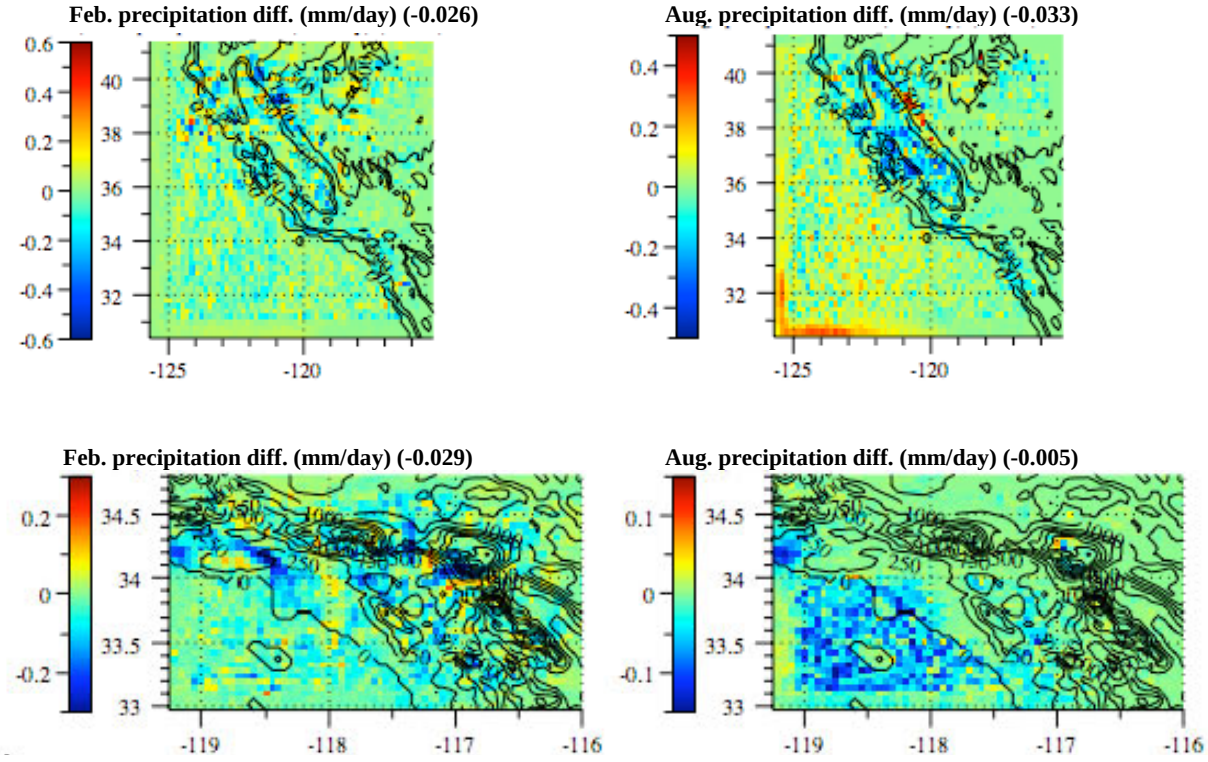

Fig. 5. Differences in the spatial distributions of precipitation over **(a)** California grid, and **(b)** the South Coast grids between simulation with and without AAPPG by GATOR-GCMOM in February and August 1999. The contours in black lines indicate topography in meters (provided by M. Z. Jacobson, Stanford University, 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 models and their application for simulating global/regional climate, meteorology, and air quality, as well as the entire earth system. However, several major challenges exist for further model development, improvement, and application.

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 online-coupled models for the years to come, as many online-coupled models are currently not significantly- or fully-coupled, in particular, such feedbacks are not fully represented in many online-coupled models. There are several key issues associated with such needs. For example, performing an online calculation of all meteorologically-dependent emissions is necessary in all online-coupled models. There is a critical need for further improvement of model treatments of key processes such as the size-/composition-resolved aerosol/cloud microphysics for multiple size distributions (e.g., new particle formation, SOA, and aerosol/cloud interactions) and aerosol-cloud interactions, as well as subgrid variability associated with these processes. In addition, the scientific understanding of the two-way/chain effects among climate, meteorology, chemistry, aerosol, cloud and radiation will continue to be needed for their accurate representations in online-coupled models. Incomplete and/or inaccurate treatments of model inputs (e.g., emissions) and physics treatments (e.g., aerosol/cloud microphysics and feedbacks) will contribute to the model uncertainties to a large extent.

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 the 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 an 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 require a multidisciplinary and or interdisciplinary approach. Key issues include (1) continuous 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, (2)

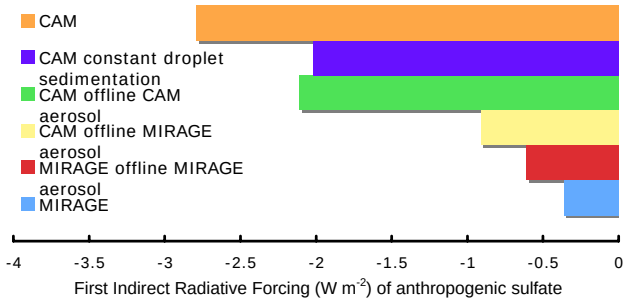

Fig. 6. Global first indirect effect of anthropogenic sulfate simulated by baseline and sensitivity simulations of CAM3 and MIRAGE2 (Ghan, 2007, inclusion with permission of S. J. Ghan, Pacific Northwest National Laboratory, 2007).

the development of process-oriented models to isolate complex feedbacks among various modules/processes in online-coupled models, (3) carefully-designed module/model intercomparison to understand mechanistic differences in various modules embedded in online-coupled models and the resultant differences in simulated feedbacks by the 3-D models. Such comparisons should be conducted using both 0-D (i.e., conducting box-model comparisons for different gas-phase chemical mechanisms and aerosol modules that are used in WRF/Chem), 1-D, and 3-D models (e.g., comparing model performance of several online models against observational data for the same episode) when possible.

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 GATOR-GCMOM, although other global-through-urban fully-coupled models such as the global-to-urban WRF/Chem (GU-WRF/Chem) are being developed (e.g., O'Connor et al., 2006; Zhang et al., 2008b). Such an unified global-through-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. The integration of such complexities into the earth system models will pose unprecedented challenges for the entire scientific 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
ASTEM	the Adaptive Step Time-split Euler Method
ARW	the Advanced Research WRF with the Eulerian Mass
BC	black carbon
CACM	the California Atmospheric Chemical Mechanism
CAM3 (Global)	the Community Atmospheric Model v. 3
CB05	the 2005 version of Carbon Bond mechanism
CBM-EX	The Stanford University's extended Carbon Bond mechanism
CBM-Z	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
CMAQ	the EPA's Community Multiple Air Quality
CMU	Carnegie Mellon University
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
EPA	the US Environmental Protection Agency
GCM	general circulation model
GAQMs	global air quality models
GATORG	the Gas, Aerosol, TranspOrt, Radiation, and General circulation model
GATOR-GCMOM (Global-through-urban)	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

Acronym	Definition
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
NH_4NO_3	ammonium nitrate
$(NH_4)_2SO_4$	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
PAN	peroxyacetyl nitrate
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
Q_v	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
SO_2	sulfur dioxide
SOA	secondary organic aerosol
STAR	the US EPA-Science to Achieve Results program
T	temperature
TUV	the Tropospheric Ultraviolet and Visible radiation model
UCLA-GCM	the University of Los Angeles General Circulation Model
VOC	volatile organic compound
WRF/Chem	the Weather Research Forecast model with Chemistry
ZSR	Zdanovskii-Stokes-Robinson

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