

# Chapter 2

## On-Line Coupled Meteorology and Chemistry Models in the US

Yang Zhang

### 2.1 Introduction

The climate–chemistry–aerosol–cloud–radiation feedbacks are important in the context of many areas including climate modelling, air quality (AQ)/atmospheric chemistry modelling, numerical weather prediction (NWP) and AQ forecasting, as well as integrated atmospheric–ocean–land surface modelling at all scales. Some potential impacts of aerosol feedbacks include a reduction of downward solar radiation (direct effect); a decrease in surface temperature and wind speed but an increase in relative humidity and atmospheric stability (semi-direct effect), a decrease in cloud drop size but an increase in drop number via serving as cloud condensation nuclei (first indirect effect), as well as an increase in liquid water content, cloud cover, and lifetime of low level clouds but a suppression of precipitation (the second indirect effect). Aerosol feedbacks are traditionally neglected in meteorology and AQ modelling due largely to historical separation of meteorology, climate, and AQ 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 2003) for those feedbacks may give different results and future climate changes may be affected by improved air quality. Accurately simulating those feedbacks requires fully-coupled models for meteorological, chemical, physical 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 on-line models are reviewed. Several representative models developed in the US are used to illustrate the current status of on-line coupled models. Major challenges and recommendations for future development and improvement of on-line- coupled models are provided.

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Y. Zhang

Department of Marine, Earth and Atmospheric Sciences, North Carolina State University, Raleigh, NC 27695, USA

e-mail: yang\_zhang@ncsu.edu

## 2.2 History of Coupled Chemistry/Air Quality and Climate/Meteorology Models

### 2.2.1 Concepts and History of On-Line Models

Atmospheric chemistry/air quality and climate/meteorology modelling was traditionally separated prior to mid. 1970s. The three-dimensional (3D) atmospheric chemical transport models (ACTMs) until that time were primarily driven by either measured/analyzed meteorological fields or outputs at a time resolution of 1–6 h from a mesoscale meteorological model on urban/regional scale or outputs at a much coarser time resolution (e.g., 6-h or longer) from a general circulation model (GCM) (referred to as off-line coupling). In addition to a large amount of data exchange, this off-line separation does not permit simulation of feedbacks between AQ and climate/meteorology and may result in an incompatible and inconsistent coupling between both meteorological and AQ 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 off-line climate/meteorology models. Such feedbacks, on the other hand, are allowed in the fully-coupled on-line models, without space and time interpolation of meteorological fields but commonly with higher computational costs.

The earliest attempt in coupling global climate/meteorology and chemistry can be traced back to late 1960s, when 3D transport of ozone and simple stratospheric chemistry (e.g., the Chapman reactions, the  $\text{NO}_x$  catalytic cycle, and reactions between hydrogen and atomic oxygen) was first incorporated into a GCM to simulate global ozone ( $\text{O}_3$ ) production and transport (e.g., Hunt 1969; Clark 1970; Cunnold et al. 1975; Schlesinger and Mintz 1979). In such models, atmospheric transport and simple stratospheric  $\text{O}_3$  chemistry are simulated in one model, accounting for the effect of predicted  $\text{O}_3$  on radiation heating and the effect of radiation heating on atmospheric circulation, which in turn affects distribution of  $\text{O}_3$ . Since mid. 1980s, a large number of on-line global climate/chemistry models have been developed to address the Antarctic/stratospheric  $\text{O}_3$  depletion (e.g., Cariolle et al. 1990; Cariolle and Deque 1986; Rose and Brasseur 1989; Austin et al. 1992; Rasch et al. 1995; Jacobson 1995), tropospheric  $\text{O}_3$  and sulfur cycle (e.g., Roelofs and Lelieveld 1995; Feichter et al. 1996; Barth et al. 2000), tropospheric aerosol and its interactions with cloud (e.g., Chuang et al. 1997; Lohmann et al. 2000; Jacobson 2000, 2001a; Easter et al. 2004). The coupling in most on-line models, however, has been enabled only for very limited prognostic gaseous species such as  $\text{O}_3$  and/or bulk aerosol (e.g., Schlesinger and Mintz 1979) or selected processes such as transport and gas-phase chemistry (i.e., incompletely- or partially-coupling). This is mainly because such a coupling largely restricts to gas-phase/heterogeneous chemistry and simple aerosol/cloud chemistry and microphysics and often neglects the feedbacks between prognostic chemical species (e.g.,  $\text{O}_3$  and aerosols) and radiation (e.g., Roelofs and Lelieveld 1995;

Eckman et al. 1996; Barth et al. 2000) and aerosol indirect effects (e.g., Liao et al. 2003), with a few exceptions after mid. 1990s when truly-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).

The earliest attempt in coupling meteorology and air pollution in mesoscale models can be traced back to early 1980s (Baklanov et al. 2007 and references therein). Since then, a number of mesoscale on-line coupled meteorology-chemistry models have been developed in North America (e.g., Jacobson 1994, 1997a, b; Mathur et al. 1998; Côté et al. 1998; Grell et al. 2000) and Australia (e.g., Manins 2007) but mostly developed recently by European researchers largely through the COST Action 728 (<http://www.cost728.org>) (e.g., Baklanov et al. 2004, 2007, and references therein). The coupling was enabled between meteorology and tropospheric gas-phase chemistry only in some regional models (e.g., Grell et al. 2000); and 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. 2006; Zhang et al. 2005a, b, 2010a, b; Krosholm et al. 2007; and Misenis and Zhang 2010). Similar to global models, 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).

Two coupling frameworks are conventionally used in all mesoscale and global on-line coupled models: one couples a meteorology model with an AQ model in which the two systems operate separately but exchange information every time step through an interface (referred to as separate on-line coupling), the other integrates an AQ model into a meteorology model as a unified model system in which meteorology and AQ variables are simulated together in one time step without a model-to-model interface (referred to as unified on-line coupling). Transport of meteorological and chemical variables is typically simulated with separate schemes in separate on-line models but the same scheme in unified on-line models. Depending on the objectives of the applications, the degrees of coupling and complexities in coupled atmospheric processes in those models vary, ranging from a simple coupling of meteorology and gas-phase chemistry (e.g., Rasch et al. 1995; Grell et al. 2000) to the most sophisticated coupling of meteorology, chemistry, aerosol, radiation, and cloud (e.g., Jacobson 1994, 2004b, 2006; Grell et al. 2005). While on-line coupled models can in theory enable a full range of feedbacks among major components and processes, the coupling is typically enabled in two modes: partially-coupled where only selected species (e.g.,  $O_3$ ) and/or processes (e.g., transport and gas-phase chemistry) are coupled and other processes (e.g., solar absorption of  $O_3$  and total radiation budget) remain decoupled; fully-coupled where all major processes are coupled and a full range of atmospheric feedbacks can be realistically simulated. At present, very few fully-coupled on-line models exist; and most on-line models are partially-coupled and still under development.

### 2.2.2 History of Representative On-Line Models in the US

In this review, five models on both regional and global scales developed in the US are selected to represent the current status of on-line-coupled models. These include:

- One global-through-urban model, i.e., the Stanford University's Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR/GCMOM) (Jacobson 2001c, 2002, 2004a; Jacobson et al. 2004)
- One mesoscale model, i.e., the National Oceanic and Atmospheric Administration (NOAA)'s Weather Research Forecast model with Chemistry (WRF/Chem) (Grell et al. 2005; Fast et al. 2006; Zhang et al. 2010a)
- Three global models, i.e., the National Center for Atmospheric Chemistry (NCAR)'s Community Atmospheric Model v. 3 (CAM3), the Pacific Northwest National laboratory (PNNL)'s 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; Liao and Seinfeld 2005)

All these models predict gases, aerosols, and clouds with varying degrees of complexities in chemical mechanisms and aerosol/cloud microphysics. The history and current status of these models along with other relevant models are reviewed below.

Jacobson (1994, 1997a, b) and Jacobson et al. (1996) developed the first unified fully-coupled on-line model that accounts for major feedbacks among meteorology, chemistry, aerosol, cloud, radiation on urban/regional scales: a gas, aerosol, transport, and radiation AQ model/a mesoscale meteorological and tracer dispersion model (GATOR/MMTD, also called GATORM). Grell et al. (2000) developed a unified on-line coupled meteorology and gas-phase chemistry model: Multiscale Climate Chemistry Model (MCCM, also called MM5/Chem). Built upon MM5/Chem and NCAR's WRF, Grell et al. (2002) developed a unified fully-coupled on-line model, WRF/Chem, to simulate major atmospheric feedbacks among meteorology, chemistry, aerosol, and radiation. This is the first community on-line model in the US. 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. In WRF/Chem, transport of meteorological and chemical variables is treated using the same vertical and horizontal coordinates and the same physics parametrization with no interpolation in space and time. In addition to Regional Acid Deposition Model v.2 (RADM2) in MM5/Chem, WRF/Chem includes 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). Two additional gas-phase mechanisms

and two new aerosol modules have been recently incorporated into WRF/Chem by external developers (Fast et al. 2006; Zhang et al. 2005a, b, 2007, 2010a; Pan et al. 2008). The two new gas-phase mechanisms are the Carbon-Bond Mechanism version Z (CBMZ) (Zaveri and Peters 1999) and the 2005 version of Carbon Bond mechanism (CB05) of Yarwood et al. (2005). 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, 2010c).

On a global scale, a number of climate or AQ models have been developed in the past three decades among which very few of them are on-line models. Since its initial development as a general circulation model without chemistry, CCM0 (Washington 1982), the NCAR's Community Climate Model (CCM) has evolved to be one of the first unified on-line climate/chemistry models, initially with gas-phase chemistry only (e.g., CCM2 (Rasch et al. 1995) and CCM3 (Kiehl et al. 1998; Rasch et al. 2000)) and most recently with additional aerosol treatments (e.g., CAM3 (Collins et al. 2004, 2006a, b; and CAM4 (<http://www.cesm.ucar.edu>)). Jacobson (1995, 2000, 2001a) developed a unified fully-coupled Gas, Aerosol, TranspOrt, Radiation, and General circulation model (GATORG) built upon GATORM and a 1994 version of the University of Los Angeles GCM (UCLA/GCM). Jacobson (2001b, c) linked the regional GATORM and global GATORG and developed the first unified, nested global-through-urban scale Gas, Aerosol, Transport, Radiation, General Circulation, and Mesoscale Meteorological model, GATOR/GCMM. GATOR/GCMM was designed to treat gases, size- and composition-resolved aerosols, radiation, and meteorology for applications from the global to urban (<5 km) scales and accounts for radiative feedbacks from gases, size-resolved aerosols, liquid water and ice particles to meteorology on all scales. GATOR/GCMM was extended to Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model (GATOR/GCMOM) in Jacobson (2004a, 2006) and Jacobson et al. (2004, 2006). Built upon NCAR CCM2 and PNNL Global Chemistry Model (GChM), MIRAGE1 was developed and can be run off-line or fully-coupled on-line (Ghan et al. 2001a, b, c and Easter et al. 2004). In MIRAGE2, the gas/aerosol treatments are an integrated model imbedded in NCAR CAM2 (i.e. unified on-line coupling). Several on-line-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 under development (e.g., Liao et al. 2003). Among all 3D models that have been developed for climate and AQ studies at all scales, GATOR/GCMOM, MIRAGE, and WRF/Chem represent the state of science global and regional coupled models; and GATOR/GCMOM 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 feedback from gases, aerosols, and clouds to meteorology and radiation on all scales in one model simulation.

## 2.3 Current Treatments in On-Line Coupled Models in the US

In this section, model features and treatments for the five representative on-line coupled meteorology and chemistry models developed in the US are reviewed in terms of model systems and typical applications, aerosol and cloud properties, aerosol and cloud microphysics and aerosol–cloud interactions. As shown in Table 2.1, four out of the five models are unified on-line models (i.e., GATOR/GCMOM; WRF/Chem, CAM3, and Caltech unified GCM) and one (i.e., MIRAGE) is a separate on-line model, all 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. Those models have been developed for different applications. As shown in Table 2.2, 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. For example, MIRAGE2 treats the least number of species, and GATOR/GCMOM treats the most. Size distribution of all aerosol components are prescribed in Caltech unified GCM and that of all aerosols except sea-salt and dust is prescribed in CAM3; they are predicted in the other three models. Prescribed aerosol size distribution may introduce significant biases in simulated aerosol direct and indirect radiative forcing that highly depends on aerosol size distributions. The mixing state of aerosols affects significantly the predictions of direct/indirect radiative forcing. The internally-mixed (i.e., well-mixed) hydrophilic treatment for BC is unphysical and reality lies between the externally-mixed, hydrophobic and core treatments. Among the five models, GATOR/GCMOM is the only model treating internal/external aerosol mixtures with a coated BC core. All the five models predict aerosol mass concentration, but only some of them can predict aerosol number concentration (e.g., GATOR/GCMOM, WRF/Chem, and MIRAGE2). For aerosol radiative properties, GATOR/GCMOM assumes a BC core surrounded by a shell where the refractive indices (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 parametrizations that are function of wet surface mode radius and wet RI of each mode. Volume mixing is assumed for all components, including insoluble components. The main difference between Caltech unified GCM and MIRAGE2 (and WRF/Chem) is that Caltech unified GCM prescribes size distribution, but MIRAGE2 predicts it. In CAM3, RIs and optical properties are prescribed for each aerosol type, size, and wavelength of the external mixtures.

Table 2.3 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. GATOR/GCMOM uses prognostic, multiple size distributions (typically three, for liquid, ice, and graupel), each with 30 size sections. MIRAGE2 and WRF/Chem simulate bulk condensate in single size distribution, with either a modal distribution (MIRAGE2) or a sectional

Table 2.1 Model systems and typical applications of on-line models

Model system/scale	Met. model	Chemical transport model (main features)	Typical applications	Example References
GATOR/GCMOM & Predecessors (Global-through-urban)	MMTD GCMOM GCMOM	Gas-phase chemistry: CBM-EX: (247 reactions, 115 species); bulk or size-resolved aqueous-phase sulfate, nitrate, organics, chlorine, oxidant, radical chemistry (64 kinetic reactions); size-resolved, prognostic aerosol/cloud with complex processes	Current/future met/chem/rad feedbacks; Direct/indirect effects; AQ/health effect	Jacobson (1994, 1997a, b, 2001c, 2002, 2004a, b), Jacobson et al. (2004, 2007), Jacobson (2006)
WRF/Chem (Mesoscale)	WRF	RADM2, RACM, CBMZ, 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	Forecast/hindcast, Met/chem. feedbacks; O <sub>3</sub> , PM <sub>2.5</sub> ; aerosol direct and indirect effects	Grell et al. (2005), Fast et al. (2006), Zhang et al. (2005a, b, 2007, 2010a, b), Tie et al. (2007), Misernis and Zhang (2010)
CAM3 & Predecessors (Global)	CCM3/CCM2/ CCM1	Sulfur chemistry (14 reactions) prescribed CH <sub>4</sub> , N <sub>2</sub> O, CFCs/ MOZART4 gas-phase chemistry (167 reactions, 63 species); Sulphur chemistry; Bulk aqueous-phase sulfate chemistry of S(IV) (4 equilibria and 2 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Climate; direct/indirect effects; hydrological cycle	Rasch et al. (1995), Kiehl et al. (1998), Collins et al. (2004, 2006a, b)

(continued)

Table 2.1 (continued)

Model system/scale	Met. model	Chemical transport model (main features)	Typical applications	Example References
MIRAGE2 & 1 (Global)	CAM2/CCM2	Gas-phase CO-CH4-oxidant chem. (MIRAGE1 only); bulk aqueous-phase sulfate chemistry (6 equilibria and 3 kinetic reactions); Mode-resolved simple aerosol treatment; Prognostic aerosol/cloud treatments	Trace gases and PM; Sulphur cycle; Direct/indirect effects	Ghan et al. (2001a, b, c), 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 <sub>3</sub> -NO <sub>x</sub> -hydrocarbon chemistry (305–346 reactions, 110–225 species); bulk aqueous-phase chemistry of S(IV) (5 equilibria and 3 kinetic reactions); prognostic aerosol/cloud treatments with prescribed size distribution	Global chemistry–aerosol interactions; aerosol direct radiative forcing; the role of heterogeneous chemistry; impact of future climate change on O <sub>3</sub> and aerosols	Liao et al. (2003), Liao and Seinfeld (2005)



Table 2.2 Treatments of aerosol properties of on-line models

Model system	Composition	Size distribution	Aerosol mixing state	Aerosol mass/number	Aerosol hygroscopicity	Aerosol radiative properties
GATOR/GCMOM	47 species (sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water, crustal)	Sectional (17–30); 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 similar to MIRAGE2
WRF/Chem	Sulfate, nitrate, ammonium, BC, OC, water in all 3 aerosol modules, sea-salt, and carbonate in MOSAIC/MADRID	Modal (3): variable (MADE/SORGAM); Sectional (8); variable (MOSAIC/MADRID); single size distribution	Internal	Predicted/Predicted	similar to MIRAGE2	
CAM3	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water	Modal (4): predicted dust and sea-salt, prescribed other aerosols; single size distribution	External	Predicted/Predicted 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	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 BC/OC with prescribed hygroscopicities for OC and dust	Parameterized RI and optical properties based on wet radius and RI of each mode

(continued)

Table 2.2 (continued)

Model system	Composition	Size distribution	Aerosol mixing state	Aerosol mass/number	Aerosol hygroscopicity	Aerosol radiative properties
Caltech unified GCM (Global)	Sulfate, nitrate, ammonium, BC, OC, sea-salt, dust, water, $\text{Ca}^{2+}$	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 $\text{SO}_4^{2-}$ , $\text{NH}_4^+$ , $\text{NO}_3^-$ , sea-salt, and $\text{H}_2\text{O}$ ; different aerosol mixing states for chemistry and radiative forcing calculation	Predicted aerosol mass; aerosol number not included	Simulated BC/OC with prescribed hygroscopicities	Simulated optical properties based on Mie theory with size- and wavelength-dependent refractive indices

Table 2.3 Treatments of cloud properties of on-line models

Model system	Hydrometeor types in clouds	Cloud droplet distribution	Cloud droplet number	CCN/IDN composition	CCN/IDN spectrum	Cloud radiative properties
GATOR/GCMOM	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 similar to MIRAGE2 but sectional (MOSAIC)
WRF/Chem	Bulk water vapour, rain, snow, cloud ice, cloud water, graupel or a subset of them depending on microphysics schemes used in both stratiform and subgrid convective clouds	Prognostic, sectional, single size distribution (MOSAIC)	similar to MIRAGE2 (MOSAIC)	similar to MIRAGE2 but sectional; CCN only	similar to MIRAGE2 but sectional, CCN only	
CAM3	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	similar to MIRAGE2	All treated species except hydrophobic species; CCN only	Prescribed; CCN only	similar to MIRAGE2

(continued)

Table 2.3 (continued)

Model system	Hydrometeor types in clouds	Cloud droplet size distribution	Cloud droplet number	CCN/IDN composition	CCN/IDN spectrum	Cloud radiative properties
MIRAGE2	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 parametrizations for liquid and ice clouds

distribution (WRF/Chem/MOSAIC). CAM3 treats bulk liquid and ice with the same prognostic droplet size treatment as MIRAGE2. 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 the other four models. CAM3, MIRAGE2, and WRF/Chem use the same treatment for droplet number, with droplet nucleation parameterized by Abdul-Razzak and Ghan (2000). GATOR treats prognostic, size- and composition-dependent cloud droplet number from multiple aerosol size distributions. While an empirical relationship between sulfate aerosols and CCN is commonly used in most atmospheric models, CCN is calculated from Köhler theory using the aerosol size distribution and hygroscopicity in all models but Caltech unified GCM. 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 for CCN but the hydrophobic species are not activated in CAM3 since it assumes an external-mixture. Among the five models, GATOR/GCMOM is the only model that simulates composition of IDN. MIRAGE and CAM use a prognostic parametrization in terms of cloud water, ice mass, and number to predict cloud radiative properties. WRF/Chem also uses the same method but with sectional approach. Caltech unified GCM simulates cloud optical properties based on MIE theory and prescribed Gamma distribution for liquid clouds. GATOR/GCMOM simulates volume-average cloud RIs and optical properties based on MIE theory and an iterative dynamic effective medium approximation (DEMA) to account for multiple BC inclusions within clouds. The DEMA is superior to classic effective-medium approximation that is used by several mixing rules such as the volume-average RI mixing rule (Jacobson 2006).

Table 2.4 shows model treatments of aerosol chemistry and microphysics that differ in many aspects. For example, 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. The degree of complexity varies in terms of number of species and reactions treated and assumptions made in the inorganic aerosol thermodynamic modules used in those models. The simplest module, MARS-A, is used in WRF/Chem/MADE/SORGAM, and the most comprehensive module, EQUISOLV II, is used in GATOR/GCMOM. For secondary organic aerosol (SOA) formation, 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 to 40 classes VOCs via condensation and dissolution based on Henry's law. Caltech unified GCM simulates SOA formation based on a reversible absorption of five classes of biogenic VOCs and neglects that from anthropogenic VOCs. In MADE/SORGAM in WRF/Chem, SOA formation via reversible absorption of eight classes VOCs is simulated based on Caltech smog-chamber data. 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 and 38 SOA species. MADRID 2 combines

**Table 2.4** Treatments of aerosol chemistry and microphysics of on-line models

Model system	Inorganic aero, thermodynamic equilibrium	Secondary organic aerosol formation	New particle formation	Condensation of gases on aerosols	Coagulation	Gas/particle mass transfer
GATOR/GCMOM	EQUISOLV II, major inorganic salts and crustal species	Condensation; dissolution based on Henry's law (10–40 classes VOCs)	Binary homogeneous nucleation of H <sub>2</sub> SO <sub>4</sub> and H <sub>2</sub> O, Ternary nucleation, T- and RH-dependent	Dynamic condensation of all condensible species based on growth law (e.g., H <sub>2</sub> SO <sub>4</sub> , VOCs) using the Analytical Predictor of Condensation (APC) with the moving center scheme	Sectional, multiple size distributions, 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	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 <sub>2</sub> SO <sub>4</sub> and H <sub>2</sub> O; T- and RH-dependent; sectional; different eqs. in different aero modules	Dynamic condensation of H <sub>2</sub> SO <sub>4</sub> and VOCs using the modal approach (SORGAM), of H <sub>2</sub> SO <sub>4</sub> , MSA, and NH <sub>3</sub> using the Adaptive Step Time-split Explicit Euler Method (ASTEEM) method (MOSAIC), and of volatile inorganic species using the	Modal/Sectional (MADE/SORGAM, MOSAIC), single size distribution, fine-mode only	1. Full equilibrium for HNO <sub>3</sub> and NH <sub>3</sub> in MADE/SORGAM and all species in MADRID 2. Dynamic for H <sub>2</sub> SO <sub>4</sub> in MADE/SORGAM; Dynamic for all species in MOSAIC and MADRID 3. Hybrid in MADRID

CAM3	MOZART4 with regime equili. for sulfate, nitrate, and ammonium	Prescribed SOA yield for $\alpha$ -pinene, n-butane, and toluene	None	Instantaneous condensation of inorganic species (MADRID)	APC with the moving center scheme	Full equilibrium involving $(\text{NH}_4)_2\text{SO}_4$ and $\text{NH}_4\text{NO}_3$
	MIRAGE2	Prescribed SOA yield	Binary homogeneous nucleation of $\text{H}_2\text{SO}_4$ and $\text{H}_2\text{O}$ ; T- and RH-dependent	Dynamic condensation of $\text{H}_2\text{SO}_4$ and MSA based on Fuchs and Sutugin growth law	Modal, single size distribution, fine-mode only; Brownian diffusion	Dynamical approach for $\text{H}_2\text{SO}_4$ and MSA
Caltech unified GCM (Global)	ISORROPIA with regime equili. for sulfate, nitrate, ammonium, sea-salt, and water	Reversible absorption for 5 biogenic SVOC classes	None	None	None	Full equilibrium involving $(\text{NH}_4)_2\text{SO}_4$ and $\text{NH}_4\text{NO}_3$

absorption and dissolution approaches to simulate an external mixture of 42 hydrophilic and hydrophobic VOCs. SOA formation is not treated in MOSAIC. Coagulation is currently not treated in CAM3 but simulated with a modal approach in MIRAGE2, sectional approach in GATOR/GCMOM, and both in WRF/Chem/MADE/SORGAM and MOSIAC. Different from other model treatments, GATOR accounts for van der Waals, viscous forces, and fractal geometry in simulating coagulation among particles from multiple size distributions (Jacobson and Seinfeld 2004). For gas/particle mass transfer, CAM3 and Caltech unified GCM use the simplest full equilibrium approach. MIRAGE2 uses a dynamic approach for  $\text{H}_2\text{SO}_4$  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 2005). In WRF/Chem, a full equilibrium approach is used for  $\text{HNO}_3$  and  $\text{NH}_3$  in MADE/SORGAM, a dynamic approach is used in MOSAIC. MADRID offers three approaches: full equilibrium, dynamic, and hybrid; their performance has been evaluated in Zhang et al. (1999, 2010a) and Hu et al. (2008). Hu et al. (2008) have shown that the bulk equilibrium approach is computationally-efficient but less accurate, whereas the kinetic approach predicts the most accurate solutions but typically with higher CPUs.

Table 2.5 summarizes the treatments of aerosol–cloud interactions and cloud processes. Aerosol activation by cloud droplets to form CCN is an important process affecting simulations of aerosol–cloud interactions, and aerosol direct and indirect forcing. CAM uses empirical, prescribed activated mass fraction for bulk CCN. MIRAGE and WRF/Chem use a mechanistic, parameterized activation module that is based on Köhler theory to simulate bulk CCN. 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 a parametrization of Abdul-Razzak et al. (1998) and Abdul-Razzak and Ghan (2000) that relate the aerosol number activated directly to fundamental aerosol properties. GATOR/GCMOM also simulates a mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory. One difference between the treatments in GATOR/GCMOM and MIRAGE is that the MIRAGE activation parametrization 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, the equilibrium Köhler theory may be inappropriate for larger particles due to the kinetic effect (i.e., mass transfer limitation). Such size-dependence and kinetic effect are accounted for in GATOR/GCMOM. A more detailed description of US integrated models along with example case studies can be found in Zhang (2008).

## 2.4 Major Challenges and Future Directions

Significant progress has been made in the past two decades in the development of on-line coupled climate- (or meteorology-) chemistry and their applications for modelling global/regional climate, meteorology, and air quality, as well as the



**Table 2.5** Treatments of aerosol–cloud interactions and cloud processes of on-line models

Model system	Aerosol water uptake	Aerosol activation aero-CCN/IDN	In-cloud scavenging	Below-cloud scavenging	Sedimentation of aerosols and cloud droplets
GATOR/GCMOM	Size-resolved equilibrium with RH; ZSR equation; simulated MDRH; Hysteresis is treated	Mechanistic, size- and composition-resolved CCN/IDN based on Köhler theory	Size-resolved aerosol activation, Nucl. scavenging (rainout), autoconversion for size-resolved cloud droplets or precip. rate dependent of aerosol size and composition	Size-resolved aerosol-hydrometeor coag. (washout), calculated precip. rate dependent of aerosol size and composition	Two-moment size-dependent sedimentation for all aerosols and hydrometeors
WRF/Chem	The same as MIRAGE2 but sectional (MOSAIC)	The same as MIRAGE2 but sectional (MOSAIC); bulk CNN only	Size-resolved aerosol activation, Nucl. scavenging (rainout), autoconversion for size-resolved cloud droplets or precip. rate dependent of aerosol size and composition similar to MIRAGE2 but sectional	similar to MIRAGE2 but sectional	The same as MIRAGE2
CAM3	For external mixtures only, equilibrium with RH, no hysteresis	Empirical, prescribed activated mass fraction; bulk CCN only	Prescribed bulk activation, autoconversion, precip. rate independent of aerosols	Prescribed bulk scav. efficiency, no-size dependence	Bulk cloud/ice sedimentation
MIRAGE2	Bulk equilibrium with RH based on Köhler theory, Hysteresis is treated	Mechanistic, parameterized activation based on Köhler theory; bulk CCN only	Modal activation, Brownian diffusion, autoconversion, precip. rate independent of aerosols	Calculated modal scaveng. coeff. using a parameterization of the collective efficiency of aerosol particles by rain drops with size dependence	Two-moment sedimentation for aerosols, nosedimentation for cloud droplets/ices

*(continued)*

Table 2.5 (continued)

Model system	Aerosol water uptake	Aerosol activation aero-CCN/IDN	In-cloud scavenging	Below-cloud scavenging	Sedimentation of aerosols and cloud droplets
Caltech unified GCM (Global)	Bulk equilibrium, ZSR equation, no hysteresis	None	Autoconversion nucl. scavenging with prescribed scavenging coefficient for sea-salt and dust and a first-order precipitation-dependent parametrization for other aerosols; precip. rate independent of aerosols	First-order precipitation-dependent bulk parametrization; calculated scavenging efficiency with size dependence	Implicitly accounted for in a parametrization of the limiting autoconversion rate

entire earth system. Several major challenges exist. First, accurately representing climate–aerosol–chemistry–cloud–radiation feedbacks in 3D climate- or meteorology-chemistry models at all scales will remain a major scientific challenge in developing a future generation of coupled models. There is a critical need for advancing the scientific understanding of key processes. 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/parametrization of model algorithms with an acceptable accuracy. Third, integrated model evaluation and improvement and laboratory/field studies for an improved understanding of major properties/processes will also pose significant challenges, as they involve researchers from multiple disciplines 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., AirNow and Satellite) for data assimilation/model evaluation and (2) the development of process-oriented models to isolate complex feedbacks among various modules/processes in on-line-coupled models. Finally, a unified modelling 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. Such a unified global-to-urban scale modelling system will provide a new scientific capability for studying important problems that require a consideration of multi-scale feedbacks.

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## Appendix – List of Acronyms and Symbols

Acronym	Definition
3D	Three-dimensional
APC	The analytical predictor of condensation
ASTEEM	The adaptive step time-split explicit Euler method
BC	Black carbon
CAM3	The community atmospheric model v. 3

(continued)

Acronym	Definition
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
CFCs	Chlorofluorocarbons
CH <sub>4</sub>	Methane
CMAQ	The EPA's community multiple air quality
CMU	Carnegie Mellon University
CO	Carbon monoxide
CO <sub>2</sub>	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
GATORG	The Gas, Aerosol, TranspOrt, Radiation, and General circulation model
GATOR/GCMOM	The Gas, Aerosol, TranspOrt, Radiation, General Circulation, Mesoscale, Ocean Model
GATOR/MMTD (or GATORM)	The gas, aerosol, transport, and radiation air quality model/a mesoscale meteorological and tracer dispersion model
GChM	The PNNL global chemistry model
H <sub>2</sub> O	Water
H <sub>2</sub> SO <sub>4</sub>	Sulfuric acid
IDN	Ice deposition nuclei
ISORROPIA	"Equilibrium" in Greek, refers to The ISORROPIA thermodynamic module
MADE/SORGAM	The Modal Aerosol Dynamics Model for Europe (MADE) with the secondary organic aerosol model (SORGAM)
MADRID	The model of aerosol dynamics, reaction, ionization, and dissolution
MARS-A	The model for an aerosol reacting system (MARS) –version A
MCCM (or MM5/Chem)	The multiscale climate chemistry model
MESA	The multicomponent equilibrium solver for aerosols
MM5	The Penn State University (PSU)/NCAR mesoscale model
MIRAGE	The model for integrated research on atmospheric global exchanges
MOSAIC	The model for simulating aerosol interactions and chemistry
MOZART4	The model for ozone and related chemical tracers version 4
MSA	Methane sulfonic acid
MTEM	The multicomponent Taylor expansion method
NCAR	The National Center for Atmospheric Research
NH <sub>4</sub> NO <sub>3</sub>	Ammonium nitrate
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	Ammonium sulfate
NO <sub>3</sub>	Nitrate radical
NO <sub>x</sub>	Nitrogen oxides
N <sub>2</sub> O	Nitrous oxide
NOAA	The national oceanic and atmospheric administration
O <sub>3</sub>	Ozone
OC	Organic carbon
PM <sub>2.5</sub>	Particles with aerodynamic diameters less than or equal to 2.5 μm

(continued)

Acronym	Definition
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
RI <sub>s</sub>	Refractive indices
S(IV)	Dissolved sulfur compounds with oxidation state IV
SOA	Secondary organic aerosol
STAR	The US EPA-science to achieve results program
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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