FINAL REPORT:
NARSTO WORKSHOP ON AEROSOL MODELING AND PROCESS EVALUATION

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EXECUTIVE SUMMARY

In June 2006, NARSTO conducted a workshop to discuss the representation of aerosol properties and processes in current climate and aerosol models and the prospects for improving the treatment of aerosols in future models. The workshop emphasized representation of the physical and chemical processes involved in the formation and transformation of atmospheric particles and in estimating the effects of these particles on clouds and on radiative energy transfer. However, the problem of modeling the aerosol lifecycle is also important to those developing process-based (as opposed to statistical) air quality forecast models. Thus, the workshop also addressed the problems of developing and evaluating aerosol process modules for air quality forecasting.

The purpose of the workshop was to capture the current status of aerosol treatment in models and recommend improvements. It did not, however, attempt an exhaustive review of the state of aerosol science. The workshop featured four keynote presentations: two discussed current practice and expected improvements in treating aerosol processes in climate and air quality forecast models, and two summarized recent research on performance evaluation by comparing model simulations with atmospheric observations. The presentations provided a foundation for subsequent breakout discussion sessions; participants in these sessions reported back specific recommendations for improving and evaluating aerosol process modules.

Keynote Presentations

Representing Aerosol Processes in Climate Models

Current climate models treat the majority of important aerosol species. Sulfate chemistry is handled within these models; however, the concentrations of most other species are simulated offline using global aerosol models. This compromise, which is made in order to reduce model runtime, means that important feedback mechanisms such as the effects of aerosols on cloudiness, cloud albedo, and precipitation processes cannot be simulated explicitly. In current models, aerosol size distribution is described separately for each aerosol species. Thus, only changes in aerosol mass concentration are simulated. In addition, each aerosol type is assumed to be composed of a single chemical species (i.e., externally mixed). This assumption simplifies the calculation of aerosol optical properties, but it is not a realistic representation of aerosol composition. Actual aerosols are usually aggregates of multiple chemical species (i.e., internally mixed). Thus, the ability of current models to realistically simulate a number of important aerosol effects is compromised. To address some of the more important process deficiencies, the next generation of climate models is expected to include the following:

Aerosol properties

- Nitrate and ammonia will be added to the current suite of aerosol types (i.e., sulfate, dust, sea salt, organic carbon, black carbon)
- Size distribution will be predicted rather than prescribed
- Both internally and externally mixed aerosols will be included
- Cloud-phase (water, ice, or mixed) will be predicted rather than prescribed
• Refractive index will be computed for both internally and externally mixed aerosols
• Hygroscopicity will be calculated for both internally and externally mixed aerosols
• CCN (Cloud Condensation Nuclei) activity will be determined for external mixtures of internally-mixed inorganic and organic salts

**Aerosol processes**
- New particle formation will be included via empirical parameterization
- Formation of organic carbon will be simulated via oxidation of organic aerosol precursors considering multiple hydrocarbon groups and reaction yields dependent upon total organic aerosol
- Aging of black and organic carbon will be incorporated via parameterized treatment of coagulation, condensation, and oxidation effects
- Water uptake will be modeled for both internal and external mixtures, and it will include both hysteresis and kinetic effects
- Aerosol particle activation will be diagnosed rather than prescribed
- Aqueous phase chemistry will be aerosol-size dependent
- Convective transport and removal will be represented via cumulus parameterization with realistic cloud fraction
- In-cloud scavenging will depend upon precipitation rate and aerosol composition. Parameterization will also include the influence of collision/coalescence on aerosol number and subgrid variability in autoconversion.

**Status and Progress in the NOAA/EPA PM Forecasting Program**
The National Oceanic and Atmospheric Administration and the U.S. Environmental Protection Agency are currently developing a deterministic modeling system for providing daily forecasts of ozone and particulate matter (PM) for the United States. A similar activity is underway in Canada. Although there are differences in the demands placed on forecast models, a number of the problems faced in simulating aerosol (or PM) processes are similar to those faced in climate, and especially, global or regional aerosol modeling.

These systems employ a state-of-the-art mesoscale meteorological forecast model to provide the necessary meteorological fields to an appropriate air quality model. The air quality model, which is also state-of-the-art, includes a sophisticated atmospheric chemistry system, and it treats cloud processes (e.g., aqueous-phase chemistry) as well as wet and dry deposition. The air quality model also includes some sort of size-resolving aerosol process module, and it represents both primary and secondary organic aerosols.

Analysis of developmental PM forecast simulations in the United States have illustrated several issues and challenges that will need to be addressed in order to improve short-term (i.e., next-day) PM forecasts:
• Early applications have shown distinct seasonal biases with under-prediction during the warm season and over-prediction during the cool season. These problems are thought to arise, at least partially, from under-prediction of secondary organic aerosol production (during the warm season) and overestimation of primary unspciated PM mass and particulate NO$_3^-$ (during the cool season).
• Representation of real-time, lateral boundary conditions is important for dealing with the effects of large, remote wildfires or dust events.
• Chemical data assimilation using near real-time remote sensing data (e.g., aerosol optical depth) can improve forecast skill during episodic, long-range transport events.
• Real-time emissions forecasting is needed for both large episodic events such as wildfires and local emissions that may exhibit significant day-to-day variability.
• Improvements are needed in the representation of processes related to gas-particle partitioning of both organic and inorganic constituents.

To address this list of problems, the following specific processes require immediate improvement:
1. Representation of heterogeneous NO$_3^-$ formation
2. Aqueous SO$_4^{2-}$ formation and understanding discrepancies between modeled and measured total sulfur
3. Representation of carbonaceous mass, relative contributions of primary and secondary components to carbonaceous mass, and their seasonal variability.
4. Representation of mass-transfer between fine and coarse modes.
5. Improvements in speciation and temporal allocation of primary PM emissions and gaseous precursors.

Lessons from AeroCom
AeroCom is an international collaboration among scientists interested in improving simulations of aerosol effects on climate. AeroCom documents differences in the aerosol modules of global models and assembles global-scale data sets for model evaluation. The objective of the program is to identify and remove weaknesses in aerosol process modeling, including the treatment of aerosol-cloud interactions.

Comparisons made to date in AeroCom indicate several key process issues that need improvement, including:
• Wet Deposition and its partitioning into stratiform and convective fractions.
• Vertical and horizontal transport.
• Formation of secondary organics formation and their contributions to aerosol mass.
• Representation of aerosol aging (e.g. ability for water uptake, aerosol mixing).

Achieving these process improvements, however, will require improvements in the quality of global data sets, in particular:
• Improved emission inventories – better accuracy and better spatial and temporal resolution. Improvements are particularly needed for inventories outside the United States, Canada, and the European Union.
• Improved global data sets for all aerosol properties (e.g., amount, size, absorption and shape) and for all related environmental properties (e.g., optical depth and relative altitude to/of clouds, solar surface albedo, sun position).
• New data sets that exploit synergies among spatial information from satellite remote sensing, long-duration observations from ground-based networks, and closure experiments from field studies.
• Datasets that can be used to examine model simulations of particular aerosol types.
• Improved characterization of data quality and uncertainty.

Lessons from ICARTT
The International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) was a collaboration among several independent research groups, which conducted an intensive field study of tropospheric atmospheric, aerosols, and radiative processes over northeastern North America, the North Atlantic, and Western Europe during the summer of 2004. In the forecast evaluation element of the program, nine air quality models from U.S. and Canadian government agencies and academia were collected at a central facility and used to provide daily air quality forecasts that included the ICARTT study domain. These forecasts were subsequently compared with surface and aloft observations from the field study.

Although ozone was the principal focus of the study, seven of the nine models provided forecasts of PM$_{2.5}$ mass and composition over the northeastern United States and southeastern Canada. Comparing these forecast to the observations, several conclusions were drawn regarding forecast performance and model process deficiencies:
• Six of the seven models showed skill in 24-hour forecasts of 8-hour afternoon average surface PM$_{2.5}$ concentrations. In contrast, the models showed very little skill in forecasting 8-hour ozone concentrations under the same conditions. This performance difference was largely attributable to smaller PM$_{2.5}$ biases in these models.*
• As with ozone forecasts, PM$_{2.5}$ ensemble forecasts showed higher skill than single-model forecasts.
• All models failed to replicate the timing and magnitude of the diurnal variation in PM$_{2.5}$ concentration. Additional modeling experiments demonstrated that the

* This finding does not necessarily contradict the conclusion of the PM modeling presentation that PM models have shown distinct, seasonally varying, biases with under-prediction during the warm season and over-prediction during the cool season. The ICARTT comparisons were for a single summer and a particular region. Figure 6 in the main body of this report, which reports typical diurnal variations in PM surface concentrations as predicted by the models applied in ICARTT, shows considerable variability among the models in terms of bias.
The principal reason for this failure was due to deficiencies in the treatment of planetary boundary layer (PBL) dynamics in the models.

- Both the absolute and relative fraction of organic carbon within PM$_{2.5}$ were significantly underestimated by all models.
- Aqueous-phase oxidation of SO$_2$ into PM$_{2.5}$-sulfate must be included in the models; however, additional research is needed to explain why aqueous-phase oxidation of SO$_2$ too rapid in those models that included it.

**Breakout Sessions**

Following the keynote presentations, workshop participants gathered into breakout sessions to generate recommendations for improving the representation of aerosols in climate and air quality models. These recommendations are summarized as follows:

**New Particle Formation**

The formation of new particles via homogeneous nucleation is a major source of particles under many atmospheric conditions. These particles can affect radiative energy transfer in the atmosphere both directly, through scattering and absorption, and indirectly, through their effects on cloud properties as cloud condensation nuclei (CCN). Two particular issues must be resolved if new particle formation is to be included in aerosol and climate models: First, new microphysical models are needed that can predict new particle formation with sufficient accuracy; and second, new models are needed that can reliably predict the growth rates of freshly nucleated particles. In this context, the session returned the following five recommendations:

1. Develop new empirical models for New Particle Formation (NPF) Rates.
2. Conduct modeling studies to assess importance of NPF to climate.
3. Develop models for growth rates of freshly nucleated particles.
4. Develop methods to measure concentrations of trace species that participate in new particle formation and growth.
5. Carry out collaborative modeling studies and atmospheric observations to assess the skill of models at predicting number distributions.

**Chemistry**

On an annual average, the organic carbon fraction of particulate matter near the Earth’s surface ranges from 10 to 40 percent in North America. This fraction is composed of primary particulate emissions and secondary organic aerosol (SOA), which is created via gas-phase reactions in the atmosphere. In the summer, or on high oxidant days, SOA can be as high as 50 to 90 percent of the fine particle fraction. Given that SOA formation is not particularly well represented in models, a high priority should be given to the improvement of SOA formation modules and to improvement of emission estimates for SOA precursors; therefore, the chemistry breakout session recommended:

1. Develop a database of SOA formation experiments and SOA precursor emissions for use by the modeling community.
2. Design new analytical techniques for speciating organic aerosol matter that do not require filter collections, extractions, or harsh preprocessing conditions.
3. Design experiments to quantify the role of cloud processing in SOA formation.

**Aerosol/Cloud Interactions**

The principal aerosol-related processes that must be considered in terms of cloud/aerosol interactions are 1) aerosol particle activation, 2) aqueous phase reactions in clouds, 3) convective transport and removal, 4) in-cloud and below-cloud scavenging, and how these processes affect cloud droplet size distributions and precipitation processes. Several of these processes, and especially their effect on cloud optical properties, cloud lifecycles, and precipitation, are not well understood or well characterized experimentally.

The ultimate source of cloud-process parameterizations is field observations enlightened by detailed cloud-scale (or smaller) model simulations and occasionally by laboratory experiments. However, obtaining the field data needed to generate parameterizations is a formidable challenge. No single measurement platform, measurement strategy, suite of instruments, or experimental venue is capable of providing the information needed to generate a robust set of parameterizations that represent the full range of aerosol/cloud interactions. Given this situation, the breakout session recommended

1. Use model simulations and observational data to develop cloud-process parameterizations for regional to global-scale aerosol and climate models.
2. Obtain better information on the range in physical and chemical characteristics of atmospheric aerosols.
3. In order to gain a better understanding of the effects of aerosols on precipitation processes, research should give priority to characterizing the effects of aerosols on shallow convection and stratus clouds.
4. In order to better characterize the effects of clouds on aerosols, additional research is needed on 1) vertical transport of aerosols by convection, 2) scavenging and wet deposition of black carbon, and 3) in-cloud chemistry of primary and secondary organic aerosols.
5. Increased attention should be placed on analysis of past field experiments.
6. Additional research is needed on ice nuclei production, ice nucleation, and the effects of aerosols on these processes.

**Emissions**

Emphasis should be placed on improving basic emission information, on improving characterization of the properties of primary particles at the point of emission, and on characterizing “aerosol initial state” (i.e., the state of the aerosol at the model’s minimum scale resolution). The breakout session recommended

1. Develop size-resolved data on chemical composition and mixing state at the point of emission, particularly for particle diameters greater than 50 nm. Information on optical properties would also be useful.
2. Develop and evaluate methodologies for processing aerosol and precursor emissions in order to provide scale-appropriate emission information for models.
3. Develop more accurate emission inventories of primary organic aerosols and organic aerosol precursors. Knowledge of primary emissions of nitrogen compounds (including, but not limited to, soil emissions) should also be improved.

4. Improve data sets for emissions from residential wood combustion, and other biofuels, and on vegetative burning.

5. Integrate the detailed, but necessarily sparse, in situ measurements that provide information on chemical composition with global satellite data.

6. Use North America as a test case for developing data-sharing methodologies.

7. Make significant improvements in the timeliness of emission data availability.

8. Develop methods for estimating future emissions in order to enable studies of their effects on future concentrations of particulate matter.

**Transport**

A significant obstacle to further progress in modeling aerosol fate and transport is the limited ability to model vertical transport and mixing processes in the PBL and above. This observation is particularly true for stable conditions, which frequently occur at night. Material remaining aloft as boundary layer turbulence decays during the late afternoon and evening can enter the free troposphere and be transported considerable distances overnight.

It is often assumed that aerosols are well mixed in the PBL, but this not always true. Fast-reacting trace gas species will often not be well mixed, and this vertical inhomogeneity can affect subsequent chemistry. The result of not reproducing these effects is that the vertical distribution of aerosols and their precursors may not be properly represented in models, which, in turn, can adversely affect both local and regional model results. Moreover, model evaluation exercises can be misleading if vertical transport processes are not accurately represented. The breakout session recommended the research community

1. **Design and carry out studies to examine and characterize vertical transport and mixing processes, in and above the mixed layer, throughout several diurnal cycles, and with particular emphasis on stably stratified conditions in the lower PBL.**

**Aerosol Dry Deposition**

Deposition rates exhibit large uncertainties for submicron particles. Predicted deposition velocities vary by as much as a factor or two or more, and recent models predict a shift in the occurrence of minimum deposition velocity from 0.2 μm to 1 μm particles. Measurements of size-resolved aerosol fluxes over a variety of surfaces could help clarify these uncertainties. The importance of such effects on climate-relevant aerosol properties, such as column mass loading, aerosol optical depth, and radiative forcing is not known at this point, however. Given this uncertainty, the breakout session recommended:
1. Conduct sensitivity studies to assess the importance, for climate modeling, of significant improvements in our ability to characterize deposition fluxes of sub-micron aerosol.

PM Forecasting
The goal of this breakout session was to identify the current challenges in PM process representation for short-term (next-day) forecasting of particulate matter distributions and concentrations. The discussions in this session focused on two broad questions: 1) What are the relevant spatial and temporal scales for short-term (next-day) forecasting of distributions and magnitude of PM concentrations? and 2) What are the current model process representation deficiencies for major constituents of total PM2.5?

Guided by these questions, the breakout session returned eight recommendations. Where possible, synergies with aerosol process research relevant to climate and air pollution assessment studies were also identified.

1. Develop a concurrent evolution in the skill of high-resolution meteorological forecast models in order to meet the short-term goals for accurately forecasting PM2.5, its chemical constituents, and other related species at the desired scales of 1 to 4 km. In particular,
   a. Improve the representation of vertical mixing processes (especially at night) and the modeling of convective clouds at fine scales.
   b. Improve emission estimates at these scales. This effort includes the development of processors for providing scale-appropriate initial-state information.
2. Create an archive of chemical forecast information to provide a continuous record of the forecasted chemical state of the atmosphere.
3. Acquire additional speciated measurements of PM2.5 concentration with hourly resolution in order to adequately assess the skill of the models in representing diurnal variations in PM2.5 concentrations, guide further PM model process representation research, and improve the skill of models.
4. Conduct an assessment to compare estimates of aerosol loading from regional models with those from current global models when aggregated to a common spatial and temporal scale.
5. Improve representation of:
   a. Heterogeneous NO3− formation
   b. Aqueous SO42− formation and characterizing discrepancies between modeled and measured total sulfur
   c. Carbonaceous mass, relative contributions of primary and secondary components to carbonaceous mass, and their seasonal variability.
   d. Fine-coarse mode interactions (e.g., mass transfer between these modes).
6. Perform a rigorous assessment of the feedbacks between chemistry and meteorology and the ability of models to accurately represent these processes.
7. Develop tools for providing emissions from episodic events and for estimating day-to-day emission variability, and place continued emphasis on developing consistent emission data sets for the three countries of North America.
8. Develop an operational global-scale chemistry forecast model and better methods for coupling global and regional forecast models to achieve proper treatment of lateral boundary conditions in regional-scale forecast models.

This final set of recommendations, in particular, illustrates the synergies that could be achieved through coordinated, collaborative research on aerosol processes involving the climate, air quality, and air quality forecast communities.
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Table 3. Treatment of aerosol processes in current (fourth-generation) climate models, likely improvements in the next-generation models, and the role of measurements in evaluating model performance..................................................7
1.0 INTRODUCTION – HISTORY AND PURPOSE OF THE WORKSHOP

In June 2006, NARSTO hosted two back-to-back workshops concerning the state-of-the-science for modeling the lifecycle of atmospheric aerosols as they pertain to air quality management, health effects research, and climate change. The first workshop emphasized improving the representation of the physical and chemical processes involved in the formation, transformation, and removal of atmospheric particles and in estimating the effects of these particles on clouds and on radiative energy transfer. The second workshop focused on evaluating the performance of emission-based (chemical transport) and observation-based (receptor) models in identifying the sources of particulate matter (PM). It also examined the application of these models in health effects research, standards setting, and air quality management. Participation in the workshops was by invitation.

This report summarizes the results and recommendations of the first workshop. Originally, the workshop had been planned to focus on the problems of modeling the processes that govern the lifecycle of atmospheric aerosols and the effects that these aerosols have on radiative energy transfer. However, the problem of modeling lifecycle process is also important to those developing process-based (as opposed to statistical) air quality forecast models. Therefore, a session was added to the workshop to discuss the problems of developing and evaluating aerosol process modules for air quality forecasting.

Within this context, the objectives of the workshop were to

1. Provide a critical review of past model evaluation activities.
2. Assess the potential of high-resolution models (i.e., models with spatial grid resolutions less than 10 km) for simulating concentrations as well as the chemical and physical properties of aerosols, and for estimating the effects of aerosols on radiative energy transfer, with particular emphasis on evaluating model elements that represent aerosol formation, transformation, fate, and effects.
4. Provide guidance to determine which model processes are in greatest need of evaluation and the associated data needs, including observational program designs.
5. Discuss the challenges of providing operational forecasts of aerosol (or PM) concentrations and chemical properties and techniques for improving these forecasts.
6. Discuss the distinct needs of regional climate and air quality models and ways the two modeling communities can complement one another by taking advantage of the research being conducted by both.

The information presented and recommendations developed in this workshop are intended to be used by modelers, analysts, and regulators to guide the further development and utility of atmospheric models for pollution management and prevention.
2.0 WORKSHOP ORGANIZATION AND QUESTIONS CONSIDERED

The workshop opened with four invited presentations that described the present status of aerosol process treatment in models. These were designed to set the stage for the in-depth discussions that would take place in subsequent breakout sessions. Several weeks before the workshop, questions were circulated to the workshop attendees to stimulate productive discussions. These questions fell under three main topic areas:

• **Lessons from past model evaluation activities**
  - How well are various aerosol properties (e.g., size distribution, chemical composition, mixing state, phase, hygroscopicity, and refractive index) simulated in current models?
  - What processes seem to result in the largest discrepancies among models and between models and observations?
  - What has been learned from these evaluations – what new insights have been generated regarding the representation of aerosol properties and processes, and what have past activities taught us regarding model evaluation design and implementation?
  - What data or modeling limitations have made it difficult or impossible to improve the representation of critical processes or properties?

• **Performance of high-resolution aerosol models**
  - How well do current high-resolution models simulate aerosol concentrations and properties – and especially vertical profiles of these concentrations and properties – on the regional to mesoscale? What are the principal model sensitivities? What are the additional observational data needs? What spatial resolution is required to adequately treat aerosol processes, especially aerosol/cloud interactions?
  - How important are the shortcomings of regional-scale emission inventories, compared to shortcomings in process modules, in limiting model performance?
  - How well do the models predict the chemical and physical properties of aerosols, including optical properties?
  - How well do models handle the effects of aerosols on cloud optical properties on cloud processes important to understanding the effect of aerosols on the hydrologic cycle?
  - How much improvement do on-line modeling approaches yield in forecasts of air quality and meteorology?
  - What methods are most promising for incorporating aerosols and precursor emissions from long-range transport into regional simulations of PM?
  - What additional real-time chemical observations are needed for PM forecasting?

• **Model process evaluation**
  - Grouping “processes” into 1) emissions, 2) transport 3) new particle formation, condensation and coagulation, 4) mixing state, 5) gas, heterogeneous, aqueous chemistry, 6) aerosol-cloud interactions, 7) dry

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1 The workshop agenda is provided in Appendix A.
and wet deposition. Which processes require fully integrated model evaluation? Which can be isolated from other processes?

- Which processes representations need the most work in order to improve the ability of models to estimate climate forcing? What are the data needs for achieving progress in process evaluation and improvement? What diagnostics are suggested for evaluating these processes?
- Which processes are most important for air quality modeling versus climate change applications? What process evaluations might the air quality and climate communities pursue jointly?

Each breakout session had a chairperson, who refined these questions in order to focus the breakout session discussions. The breakout sessions and the recommendations they generated are discussed in Section 4 of this report.

### 3.0 KEYNOTE PRESENTATIONS

Keynote presentations were delivered by Steven Ghan (Pacific Northwest National Laboratory), and Rohit Mathur (U.S. Environmental Protection Agency Office of Research and Development), Stefan Kinne (Max-Planck-Institute für Meteorologie), Stuart McKeen (NOAA Oceans and Atmospheric Research Branch). Steven Ghan provided an overview of current and proposed approaches for representing aerosol processes in models, and Rohit Mathur discussed the current status and the progress being made in PM forecasting through a joint NOAA/EPA forecast model development program. Stefan Kinne and Stuart McKeen discussed lessons learned from aerosol and PM forecast model evaluation activities conducted by AeroCom (a European-based international global aerosol model evaluation project) and ICARTT.2 Summaries of the major points of the presentations are given below. Copies of the PowerPoint presentations, which contain additional information not covered in the summaries, can be found on the NARSTO website: [http://www.narsto.org/event.src?ID=4](http://www.narsto.org/event.src?ID=4).

### 3.1 Representing Aerosol Processes in Present and Future Climate Models

Since at least the mid 1990s, it has been clear that the effects of aerosols on radiative energy transfer through the atmosphere have to be represented in order to simulate the observed climate changes that have occurred over the past 150 years. Developing the process modules that can properly account for these effects requires a coordinated program of research that includes 1) field measurements and laboratory experiments to develop and test theoretical understanding of aerosol processes and effects, 2) detailed modeling work coupled with experiments to translate theoretical understanding into practical process modules for use in regional and global aerosol models, 3) inclusion in and testing of these modules in aerosol models, and 4) further simplification (in most

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2 ICARTT (the International Consortium for Atmospheric Research on Transport and Transformation) is focused on air quality issues concerning the northeastern United States and southeastern Canada and air pollution transport over the North Atlantic Ocean.

3
cases) of these modules into forms that can be implemented in global climate models. This process is depicted in Table 1.

### Table 1. Stages of model development.

<table>
<thead>
<tr>
<th>Stage</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
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<tbody>
<tr>
<td>Activity</td>
<td>Process research: field and laboratory studies</td>
<td>Development of zero-dimension box models (i.e., Modules) for representing processes; comparison with process research studies</td>
<td>Incorporation of modules describing aerosol processes in regional to global aerosol models. Production runs. Assessment of accuracy of aerosol models</td>
<td>Incorporation of aerosol process modules in climate models; production runs; comparison with observations</td>
</tr>
<tr>
<td>Outcome</td>
<td>Improved understanding of processes</td>
<td>Modules: Model-based representation of understanding</td>
<td>Evaluated aerosol model incorporating processes</td>
<td>Climate-relevant runs; assessment of the skill of climate model against present and/or prior climate</td>
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As this process moves through its successive stages, the details of the chemical and physical processes that affect the lifecycle of aerosols and their effects on radiative processes become increasingly parameterized. Global climate models, for example, must be capable of simulating centuries through model runs lasting no more than weeks to a few months; consequently, aerosol processes must be very parameterized in order to achieve this restraint. On the other hand, regional and global aerosol models, usually simulate much shorter timescales – days to a few years, and regional models, in particular, can be run at much higher spatial resolution with more detailed model physics. Thus, aerosol or regional models can be used to

- Examine individual processes or multiple processes, including radiative feedback to large-scale dynamics
- Provide higher resolution regional or global aerosol distributions for short periods (e.g., five years or less)
- Serve as a test bed for aerosol processes in climate models
- Produce offline aerosol for climate models
- Treat feedback of aerosol on meteorology
- Explicitly resolve or parameterize clouds.

In order to simulate their effects for climate applications, the aerosol properties that need to be represented in space and time are mass concentration and size-dependent composition, the mixing state of the different components that make up the aerosol or particle, solubility, hygroscopicity, aerosol optical properties, and the ability to serve as cloud condensation nuclei. The principal aerosol-related processes (see Figure 1) that must be represented in order to simulate these properties include

- Primary particle emissions
- Emissions of aerosol precursors
- New particle formation
- Coagulation
- Chemical reactions that modify aerosol properties
- Evolution of hygroscopicity
- Water uptake
- Activation of aerosol particles to form cloud droplets and cloud ice-particles
- Aqueous phase reactions in clouds
- Transport by convective clouds
- In-cloud and below-cloud scavenging
- Dry deposition of precursor gases and aerosol particles.

**Figure 1.** Aerosol sources and processes that must be represented in global aerosol and climate models.

Measurements and experiments are critical to every stage of the model development process. Two examples are 1) controlled laboratory experiments for testing basic theory or for developing empirical parameterizations of aerosol processes, and 2) closure experiments that focus on a given process in order to evaluate individual parameterizations of that process. Typical closure experiments might entail measuring parameterization inputs and outputs or comparing the output of parameterizations with
observed output. In addition to evaluating individual parameterizations, it is also important to test the complete, integrated model by comparing model simulations of aerosol formation, transformation, transport, and removal with field observations. These observations often include both in situ and remote sensing data, and they may cover a wide range of spatial and temporal scales. If the measurements are sufficiently reliable, evaluations such as these can reveal limitations in our understanding of the underlying processes, and comparisons based on measurements of many variables can suggest how to improve this understanding.

Over the past decade, the realism with which aerosol effects have been represented in climate models has steadily improved. At first, these effects were approximated by adjusting the surface albedo in order to account for the enhancement of planetary albedo due to tropospheric aerosols. However, these crude representations of the direct aerosol effect were replaced by “offline” simulations of three-dimensional aerosol distributions using chemical transport models. This approach was a significant improvement, but it fails to account for the dynamic interactions between aerosols and meteorology. This recognition is driving much of the current work in improving the treatment of aerosols in the models.

As so much of the activity in climate modeling is dictated by the demands of the periodic assessments of climate science by the Intergovernmental Panel on Climate Change (IPCC), it is convenient to think of the modeling efforts conducted in support of these assessments, which are prepared about every six years, as being undertaken by successive generations of climate models. The fourth IPCC assessment, to be published in 2007, is based on climate simulations that were completed in 2004 using models that were frozen in 2003. Thus, it can be anticipated that the fifth assessment will be based on models that will be frozen around 2009, and the sixth assessment will use models that will be frozen in about 2015.

A summary of aerosol properties included in fourth-generation models, which for the United States are represented by the National Center for Atmospheric Research Community Climate System Model (CCSM3), the Geophysical Fluid Dynamics Laboratory Coupled Model (CM2.1), and the Goddard Institute for Space Studies Model-E, is provided in Table 2. Table 2 also identifies improvements that are expected in the next generation of models, and it notes the role of measurements in evaluating the performance of these improvements.

In the fourth-generation models, most of the important aerosol species are now treated. Sulfate chemistry is internal to the models, but concentrations of most of the other species are simulated offline using global aerosol models. This is an obvious compromise, because it does not include important feedback mechanisms such as the effects of aerosols on cloudiness, cloud albedo, and the hydrological cycle. Size distribution is separately described for each aerosol species, so that only changes in the mass concentration are simulated. The aerosol mixing state, which describes whether individual particles are composed of single species (external mixing) or multiple species (internal mixing) is assumed to be external. Thus, each aerosol type is assumed to
consist of only one chemical species. This assumption greatly simplifies the representation of aerosol optical properties. With the assumptions of a set particle size distribution, particle size and shape, as well as a specified chemical composition, refractive index and other optical properties are in effect prescribed for each aerosol type. However, this approach cannot account for variations in these properties among given aerosol types. Similar to the treatment of aerosol optical properties, water uptake is prescribed as a function of relative humidity for each aerosol type. The cloud condensation nuclei (CCN) spectrum, which is the concentration of aerosol particles that can nucleate cloud droplets as a function of supersaturation with respect to water, is either ignored or estimated by empirical relationships. These relationships are suitable only for externally mixed aerosols.

Table 2. Treatment of aerosol properties in current (fourth-generation) climate models, likely improvements in the next-generation models, and how measurements can be used to evaluate model performance.

<table>
<thead>
<tr>
<th>Property</th>
<th>Current Status (fourth generation)</th>
<th>Next Step</th>
<th>How Measurements Can Help</th>
</tr>
</thead>
<tbody>
<tr>
<td>Components</td>
<td>Sulfate, dust, sea salt, organic carbon, black carbon</td>
<td>Add nitrate, ammonia</td>
<td>Evaluation of integrated models</td>
</tr>
<tr>
<td>Size distribution</td>
<td>Prescribed</td>
<td>Actively modeled; possible approaches are modal sectional moments</td>
<td>New particle formation; Evaluation of integrated models</td>
</tr>
<tr>
<td>Mixing state</td>
<td>External</td>
<td>Internal and external</td>
<td>Evaluation of models</td>
</tr>
<tr>
<td>Cloud-phase (i.e., liquid, solid)</td>
<td>Prescribed</td>
<td>Actively modeled</td>
<td>Evaluation of integrated models</td>
</tr>
<tr>
<td>Refractive index</td>
<td>For external mixtures only</td>
<td>Internal and external</td>
<td>Local radiative closure</td>
</tr>
<tr>
<td>Hygroscopicity</td>
<td>For external mixtures only</td>
<td>Internal and external</td>
<td>Evaluation of diameter growth factor</td>
</tr>
<tr>
<td>CCN activity</td>
<td>Externally mixed inorganic salts</td>
<td>Externally mixed inorganic and soluble organic salts</td>
<td>CCN closure</td>
</tr>
</tbody>
</table>

Representation of aerosol processes in fourth-generation models is also highly simplified. Table 3 provides a summary of how several important processes are currently treated in global climate models, improvements that are contemplated for future-generation models, and the assistance measurements can make in effecting and evaluating these improvements. Considerably more information on current practices and anticipated improvements in representing aerosol processes (as well as the role of measurements in this process) is provided in Ghan Schwartz (2007).

Table 3. Treatment of aerosol processes in current (fourth-generation) climate models, likely improvements in the next-generation models, and the role of measurements in
evaluating model performance.

<table>
<thead>
<tr>
<th>Process</th>
<th>Current Status</th>
<th>Target Treatment</th>
<th>How measurements can help</th>
</tr>
</thead>
<tbody>
<tr>
<td>New particle formation</td>
<td>Neglected</td>
<td>Ion-induced nucleation Empirical parameterization</td>
<td>New parameterization. Evaluation of parameterization</td>
</tr>
<tr>
<td>Formation of organic carbon</td>
<td>Instantaneous conversion of precursors with prescribed uniform yield</td>
<td>Reaction of precursors with oxidants. Multiple hydrocarbon groups. Dependence of yield on total organic aerosol</td>
<td>Evaluation of process and integrated models</td>
</tr>
<tr>
<td>Aging of black carbon and organic carbon</td>
<td>Prescribed hydrophobic to hydrophilic conversion time</td>
<td>Parameterized treatment of coagulation, condensation and oxidation effects.</td>
<td>Lab experiments. Evaluation of process models</td>
</tr>
<tr>
<td>Water uptake</td>
<td>For external mixtures only. No hysteresis. Equilibrium</td>
<td>Internal and external mixtures. Hysteresis. Kinetic effects.</td>
<td>Evaluation of growth factor</td>
</tr>
<tr>
<td>Activation</td>
<td>Prescribed number activated</td>
<td>Diagnosed number activated</td>
<td>Evaluation of activation models</td>
</tr>
<tr>
<td>Aqueous chemistry</td>
<td>Bulk treatment (same for all cloud droplets). pH dependence</td>
<td>Size dependent</td>
<td>Detailed modeling</td>
</tr>
<tr>
<td>Convective transport and removal</td>
<td>Cumulus parameterization. Ambiguous cloud fraction</td>
<td>Cumulus parameterization with realistic cloud fraction, aqueous chemistry. Statistics from embedded cloud models.</td>
<td>Evaluation of parameterization in integrated models</td>
</tr>
<tr>
<td>In-cloud scavenging</td>
<td>Precipitation rate independent of aerosol</td>
<td>Precipitation rate depends on aerosol. Add influence of collision/coalescence on number. Subgrid variability in autoconversion</td>
<td>Evaluation of collision/coalescence schemes. Evaluation of integrated models.</td>
</tr>
</tbody>
</table>

In summary,

- The representation of aerosol properties and processes in current global climate models is simplified compared to current understanding of aerosol physics and chemistry.
- The climate modeling community recognizes the need to devote additional computational resources to improving the representation of aerosol properties and processes.
- The treatment aerosol processes in climate models could be improved significantly by applying treatments currently used in global aerosol models.
- Field and laboratory research programs can provide the measurements, process modeling, and integrated modeling needed to develop the next generation of parameterizations of aerosol properties and processes.
• Issues of scale, which result from mismatches between the scales resolvable by the models and the scales at which key processes occur, as well as uncertainty need to be addressed.
• Because available funding is always limited (and because many of the issues discussed above are common to other modeling applications), integration of research activities across programs and agencies will enhance the likelihood that the improved parameterizations needed for the next generation of global climate models will be achieved.

3.2 Status of and Progress in the NOAA/EPA PM Forecasting Program

The National Oceanic and Atmospheric Administration and the U.S. Environmental Protection Agency are currently developing a deterministic modeling system for providing daily forecasts of ozone and particulate matter (PM) for the United States. A similar activity is underway in Canada. Although there are differences in the demands placed on forecast models, a number of the problems faced in simulating aerosol (or PM) processes are similar to those faced in climate, and especially, global or regional aerosol modeling.

The ability to forecast local and regional air pollution events is challenging since the processes governing the production and accumulation of ozone and fine particulate matter are complex and non-linear. Comprehensive atmospheric models, by representing in as much detail as possible the various dynamical, physical, and chemical processes regulating the atmospheric fate of pollutants, provide a scientifically sound tool for providing air quality forecast guidance. An increasing number of clinical and epidemiological studies have associated adverse health effects in humans with exposure to fine particulate matter (or PM$_{2.5}$ – particles with aerodynamic diameter less than 2.5 µm). It is thus desirable for local air quality agencies to accurately forecast PM$_{2.5}$ concentrations to alert the sensitive population of the onset, severity and duration of unhealthy air, and to encourage the public and industry to help reduce emission-producing activities. Although model-based forecasts for ozone have publicly been available for some years, forecasts for PM$_{2.5}$ are currently in a developmental stage. Unlike ozone pollution, which occurs primarily during the warm season, elevated PM$_{2.5}$ concentrations are observed throughout the year. Consequently, the challenges for a PM$_{2.5}$ forecast modeling system are enormous – they require accurately predicting the day-to-day variability in ambient PM$_{2.5}$ mass and composition for the full annual range of meteorological and chemical conditions.

Figure 2 depicts a schematic of the currently operational NOAA/EPA air quality forecasting system in the United States. Implementation of the system began during the summers of 2003 and 2004 with ozone forecasts for the northeastern United States. In September 2004, O$_3$ forecast guidance over the northeast domain became fully operational. This domain extended roughly from the western borders of Wisconsin and Illinois to just east of Maine and from central Georgia and Alabama to the northern border of Maine (including parts of southeastern Canada); as of August 31, 2005, the operational domain for O$_3$ forecast guidance was extended to cover the entire eastern United States. In 2006, the domain coverage for experimental O$_3$ predictions was
expanded to cover the entire continental United States. Testing of a PM forecasting capability began in 2004 over the eastern U.S. and was expanded to cover the entire Continental United States in summer 2006. Daily developmental PM forecast runs are currently performed to assess performance issue, but it is not yet operational.

Figure 2. Current (circa 2005) NOAA/EPA Air Quality Forecasting System Operational at NOAA National Weather Service (NWS) [Courtesy: T. Otte, AMD, NERL, EPA].

As illustrated in Figure 2, the core of this air quality forecast guidance modeling system consists of the National Weather Service’s North American Mesoscale (NAM) meteorological forecast model and the U.S. EPA’s Community Multiscale Air Quality (CMAQ) model. Prior to 2006, the NOAA Eta model, run at 12 km horizontal resolution, was used to predict meteorological fields. These predictions were used to drive the chemistry-transport calculations in CMAQ. In 2006, the Weather Research and Forecasting Non-hydrostatic Mesoscale Model (WRF-NMM) model replaced Eta as the NAM. The WRF-NMM is a next-generation mesoscale numerical weather prediction model. WRF is a highly flexible model that is being adopted for a wide range of research applications from regional climate simulation

3 The term “forecast guidance modeling system” is used because the model is intended to provide guidance to forecasters who are ultimately responsible for alerting the public to forecasted air quality conditions.
to atmospheric chemistry. The CMAQ model includes a state-of-the-art atmospheric chemistry system, and it treats cloud processes (mixing and aqueous-phase chemistry) as well as dry and wet deposition. Figure 3 depicts the current CMAQ treatment of aerosols. The aerosol distribution is modeled as a superposition of three lognormal modes that correspond nominally to the Aitkin (diameter \( D_p < 0.1 \mu m \)), accumulation \( (0.1 < D_p < 2.5 \mu m) \), and coarse \( (D_p > 2.5 \mu m) \) modes (Binkowski and Roselle, 2003). In the current implementation, the coarse mode is treated as chemically inert; development efforts are underway to include treatment for sea-salt and interaction between the fine and coarse modes. The model includes representation of both primary and secondary organic aerosols; semi-volatile organic compounds (SVOCs) partition between the gas and aerosol phase. The thermodynamic equilibrium of the \( \text{SO}_4-\text{NO}_3-\text{NH}_4-\text{H}_2\text{O} \) system is represented using the ISORROPIA thermodynamic module (Nenes et al., 1998).

Figure 3. Current treatment of PM/aerosols in CMAQ [Courtesy: P. Bhave, AMD, NERL, EPA].

Once made, air quality forecast predictions are then verified against observations in order to provide feedback to model developers and users. Over the next few years, through a program of phased development and testing, the operational model domain should be expanded to the continental United States and additional pollutant species are expected to be added to the operational forecast products.
Detailed analysis of early developmental PM forecast simulations with models such as the Eta-CMAQ have highlighted several challenges in PM process representation in models used for short-term (next-day) PM forecasting:

- Initial forecast applications with comprehensive regional models show distinct seasonal biases with warm season under-predictions and cool season over-predictions. (e.g., Mathur et al., 2007; Eder and Yu, 2006).
- The warm season under-predictions in part are related to uncertainties in representation of the organic fraction of the airborne fine particle mass and arise from under-prediction of secondary organic aerosols in current models. The cool season over-predictions arise from overestimation of primary unspeciated PM mass (representing crustal material and trace elements) and overestimation of particulate NO$_3^-$.
- Uncertainties in representation of both oxidized and reduced nitrogen cycling in the troposphere influence modeled PM inorganic composition. As airborne NH$_3$ provides a pathway for formation of particulate NO$_3^-$, accurate representation of tropospheric NH$_3$ cycling in models is critical. Significant uncertainties exist both in the spatial and temporal representation of NH$_3$ emissions as well as dry deposition sink processes. Additionally, uncertainties exist in the representation of the reaction probability for heterogeneous N$_2$O$_5$ hydrolysis (e.g., Bhave et al., 2006), which can impact simulated total nitrate and consequently particulate NO$_3^-$ concentrations, especially during the cool seasons.
- Experience with these experimental forecasts has demonstrated that accurate representation of real-time lateral boundary conditions is important in accounting for long-range transport events associated with large wildfires and dust events. Linkage with larger scale models can help improve forecast skill during such episodic transport events provided such models include an accurate description of the sources and origin of such pollution as well as fidelity in representing the long-range three-dimensional transport, chemical processing, and potential sinks of the transported pollutants.
- Chemical data assimilation using near real-time remote sensing data (e.g., aerosol optical depth) can potentially improve forecast model skill during episodic long-range transport events.
- An emerging challenge arising from routine model-based air quality forecasting is “emission forecasting” that is, the ability to accurately specify, in real-time, emissions from episodic events (e.g., wild and prescribed fire events, dust) not reflected in the base year emission inventory. The forecast system development team has found that the use of remote-sensing data to locate and specify wildfire emissions in real time improves forecast skill for both ozone and fine PM. The ability to assimilate information on day-to-day variability in emissions from various anthropogenic sectors (e.g., emissions associated with increased generator use in urban areas during hot days, variability in emissions arising from day-to-day variability in energy and load demand on power plants, temporary shutting-off of facilities, new facilities not represented in base year inventory, variability in mobile emissions associated with large public events and traffic pattern changes, to name a few) is another area that needs further work.
• Comparisons of PM composition with available routine surface network data and aircraft measurements from field intensives (e.g., ICAR TT) have nevertheless helped identify several shortcomings in the ability of existing forecast models to accurately represent the day to day variability in chemical composition of PM$_{2.5}$. In particular, the need for enhancements in representation of process related to gas-particle partitioning of both organic and inorganic constituents were identified. Based on these comparisons, the following process-specific issues are suggested as the priorities for immediate improvement:

1. Representation of heterogeneous NO$_3^-$ formation
2. Aqueous SO$_4^{2-}$ formation and understanding discrepancies between modeled and measured total sulfur
3. Representation of carbonaceous mass, relative contributions of primary and secondary components to carbonaceous mass, and their seasonal variability.
4. Representation of mass-transfer between fine and coarse modes.
5. Improvements in speciation and temporal allocation of primary PM emissions and gaseous precursors.

Many of the process deficiencies identified are not necessarily unique to forecasting applications. Overall performance metrics for predicted total PM$_{2.5}$ and constituent concentrations in forecast models are similar to those in current retrospective applications of such models used for air pollution and climate assessment studies. However, the requirements for spatial and temporal resolution and model forecast duration needed in PM forecast applications are considerably more stringent than those for current model use in assessment studies (see Section 4.5). Continuous analysis of daily forecast applications has provided valuable diagnostic information on the adequacies and inadequacies of the models under varying dynamical and chemical conditions, and this analysis is guiding further development and evolution of these models. These improvements will directly benefit the use of these models in air pollution and climate assessment studies.

Progress in the above-mentioned deficiencies would greatly enhance the ability of current generation air quality forecast models to accurately predict both total PM$_{2.5}$ mass as well as its chemical constituents. It is expected that such models will also need to forecast, with reasonable fidelity, the distributions of coarse mass within the next two to five years. It is also recognized that while several current models predict size and number concentrations, these have not been adequately verified due to lack of sufficient measurements.

### 3.3 Evaluating Aerosol Models: Lessons from AeroCom

The AeroCom initiative ([http://nansen.ipsl.jussieu.fr/AEROCOM](http://nansen.ipsl.jussieu.fr/AEROCOM)) is an open international collaboration among scientists interested in improving simulations of aerosol effects on climate. Specified output of prescribed simulations (or ‘experiments’) is compared among global models and, more importantly, to quality-assured observations. Differences between the models and between the models and observations
are diagnosed in order to stimulate improvement in the representation of aerosol properties and processes.

By the mid 1990s, global modeling of aerosols had entered a new era of complexity. The representation of aerosols in the models began to distinguish aerosol by type (typically sulfate, particulate organic matter, black carbon, dust, and sea salt) and by size (at least accumulation and coarse sizes mode). This level of detail allowed more detailed treatment of aerosol chemistry and aerosol/cloud interactions. However, initial evaluations of these more complex aerosol modules were often subjective and based on limited observational data. In addition, model intercomparisons were rare – especially at the level of aerosol module sub-steps. In order to improve evaluation practice, a number of modeling groups formed the AeroCom initiative. The initiative was organized around four principal goals:

1. Assemble quality-assured observational data sets for global aerosol modeling, including
   a. Data-sets for model evaluation, such as recommended global fields of aerosol optical depth (AOD), from com-bing the individual strengths of available remote sensing data-sets
   b. Data-sets for model input, such as the definition of emission data of aerosol and precursor gases for current (year 2000) and pre-industrial (year 1750) conditions (Dentener et al. 2006).

2. Organize and conduct model evaluation and intercomparison experiments:
   a. Define test simulations (‘experiments’) with requests for specific output.
   b. Provide visualization of the output of individual model results, via a web-tool (http://nansen.ipsl.jussieu.fr/AEROCOM), to allow intercomparisons among models and comparison to data products from remote sensing networks (including satellites).
   c. Conduct regular workshops to discuss results.

3. Produce value-added products for the aerosol modeling and climate communities:
   a. Develop global maps which are characteristic of global modeling results, such as monthly (e.g., model median) fields for aerosol amount, aerosol optical depth, aerosol absorption (via the single scattering albedo, SSA) and aerosol size (via the Angstrom parameter, g) (Kinne et al., 2006).
   b. Support the IPCC by quantifying uncertainties of the effects of aerosols on climate, such as comparisons of simulated anthropogenic aerosol direct ToA (top of the atmosphere) forcing, which ranges from -0.41W/m$^2$ to +0.04W/m$^2$ in current models (Schulz et al., 2006).

4. Serve the aerosol community by
   a. Providing a forum for interactions between aerosol measurement and aerosol modeling groups at regular workshops,
b. Providing standard data-test beds for model evaluations,
c. Archiving data.

Model evaluation requires appropriate, quality-assured data. Thus, the first goal has been one of AeroCom’s more important activities. The assembly of high-quality global data for model evaluation is not a trivial task. Global data are seldom available from a single measurement platform. Moreover, the quantity of interest is usually not measured directly, but rather derived. Aside from corrections that may be needed to offset contaminating effects, derived products often require a-priori assumptions to be made regarding aerosol and/or environmental properties. The appropriateness of these assumptions will vary in both time and space, as will the accuracy of the derived products. Thus, each data product can be expected to have regional and/or seasonal strengths and weaknesses. As a result, the most useful data composites are those that combine regional and seasonal strengths from all available observations. Once obtained, it is essential to determine whether or not data intended for model evaluation are suitable for the task. This means answering such questions as:

- Are the data sufficiently accurate to be useful?
- Are the data representative at the required model resolution?
- Are the data complete in terms of temporal and spatial coverage?

All these factors mean that an essential part of any model evaluation activity is to quantify data uncertainty, document limitations, and communicate them to data users. Unfortunately, the work needed to assure data quality and evaluate the suitability of data products for model evaluation is all too often inadequately supported.

The principal customer for these quality-assured data sets are AeroCom model evaluation experiments (Goal 2). AeroCom intercomparison and model evaluation activities are designed to assess the ability of models to simulate aerosol load, aerosol optical properties, aerosol-cloud interactions, and aerosol radiative effects on different spatial and temporal scales. Over the past several years, AeroCom has conducted four model evaluation experiments:

**Experiment A** in which modelers are asked to run models in their standard configuration and results are reported either for a climatological average (3-10 years), or for the years 1996, 1997, 2000, or 2001.

**Experiment B** in which modelers are asked to use AeroCom-prescribed aerosol and precursor emissions for the year 2000 and year 2000 meteorological fields.

**Experiment Pre** in which modelers are asked to repeat Experiment B, but with AeroCom-prescribed aerosol and pre-cursor emissions for the year 1750.

**Experiment Indi** in which modelers are asked to conduct model sensitivity studies to explore uncertainties regarding aerosol impacts on the hydrological cycle.
Experiment A showed that model simulations of aerosol properties and effects exhibited considerable inter-model variability (or diversity) that is much larger than modeled inter-annual variability (Kinne et al., 2006). For example, Figure 4 displays individual model contributions of five aerosol components to the annual average global aerosol optical depth (AOD). The total combined annual AOD of 17 global models are in general agreement to that of the ground-based AERONET sun-photometer network and to a composite of satellite observations. Good agreement in simulating total AOD, however, does not necessarily translate into good agreement in simulating aerosol radiative forcing.

![Figure 4](image)

**Figure 4.** Individual contributions of five aerosol components (SS-seasalt, DU-dust, POM-particulate organic matter, BC-black carbon, SU-sulfate) to the annual global aerosol optical thickness (at 550 nm). Observed global average AOT is shown for two remote sensing systems: AERONET (a ground-based network) and a satellite-composite, $S^*$, based on MODIS (ocean) and MISR (land). No component apportioning is possible for ‘EH’ due to inter-component mixing. (from Kinne et al., 2006)

Aerosol radiative forcing also depends on aerosol composition, environmental properties, but most importantly on cloud and surface properties. Figure 4 demonstrates that there is considerable model diversity in aerosol composition. This result suggests that there are large differences in aerosol processing among the models. An intercomparison of lifecycles for individual aerosol types (Textor et al., 2006) demonstrated significant model diversity for aerosol sources and sinks, loading, size distribution, and water uptake. In particular, there are significant inter-model variations in removal rate and deposition for sea salt and dust aerosol types. The models strongly disagree on the split between wet and dry deposition and on the split between sedimentation and other dry deposition processes.

In addition, there is high model diversity for the uptake of ambient water vapor, which influences particle size and sink-rate coefficients. Models show little agreement on the partitioning of precipitation scavenging by convective and stratiform rain. Large differences also exist in vertically and horizontally transport among the models, with
greater model diversity in meridional than in vertical dispersion. Significant inter-model differences are also found for natural aerosol emissions. For example, there are large differences in the sizes and surface fluxes of sea-salt and dust aerosol, which are explained by differences in the way the surface fluxes are parameterized and in the aerosol sizes considered. Emission differences for the other aerosol types are smaller, mainly due to the use of similar inventories.

Experiment B was designed to remove the effect of emission difference on model diversity. In Experiment B, emissions for all aerosol types were prescribed. However, the harmonization of aerosol sources surprisingly had only a small impact on the simulated atmospheric burden for all aerosol types (Textor et al. 2007). This result indicates that the simulated fate of atmospheric aerosols depends to a large extent upon the particular aerosol module used and on how it treats various aerosol processes and not on differences in emissions input. This finding does not, however, minimize the importance of good emissions data. The differences in emissions input between Experiments A and B are small compared to the uncertainties in emissions inventories.

Contrasting the results of Experiment B and Experiment Pre provided estimates of direct aerosol forcing by anthropogenic aerosol since the beginning of the industrial revolution (Schulz et al., 2006). Under all-sky conditions, anthropogenic aerosol forcing at the top of the atmosphere (ToA) averages -0.22W/m² (compared to +2.6W/m² by anthropogenically enhanced tropospheric greenhouse gases). However, direct radiative forcing by anthropogenic aerosols exhibits strong regional variations and significant regional variations among models. The variations among models are influenced by considerable model diversity in residence times (controlled by transport and processing), mass extinctions (controlled by aerosol size and water uptake) and forcing efficiency (controlled by composition and environmental properties).

Experiment Indi examined model differences in simulating aerosol-cloud interactions. These comparisons found (Penner et al., 2006) that the largest differences in aerosol indirect radiative ToA forcing (given fixed aerosol sources) were related to the diversity in predicted aerosol concentrations. However, when aerosol concentrations (and the parameterization of droplet concentrations and autoconversion) were prescribed, the models agreed on an estimate of the first indirect effect (droplet radius decrease with increased aerosol loading) of about -0.6W/m² ToA forcing.

AeroCom is continuing its work to improve understanding of global aerosol modeling and processes. However, results to date suggest several key issues that need to be addressed in order to improve current capabilities to estimate the effects of aerosols on climate or to provide reliable lateral boundary conditions for regional forecasting:

• Wet Deposition and its partitioning into stratiform and convective fractions.
• Vertical and horizontal transport.
• Formation of secondary organics formation and their contributions to aerosol mass.
• Representation of aerosol aging (e.g. ability for water uptake, aerosol mixing)
If these issues are to be addressed, a number of data problems must also be dealt with:

- Aerosol emission inventories need to be improved (despite the apparent lack of sensitivity between Exp A and Exp B). Although improvements can be made to inventories in the EU, Canada, and the United States (e.g. NARSTO, 2005), the greatest need for improvement is outside these countries.
- Accurate data sets are needed for all aerosol properties (e.g., amount, size, absorption and shape) and for all related environmental properties (e.g., optical depth and relative altitude to/of clouds, solar surface albedo, sun position).
- New data sets need to be created by exploiting synergies among spatial information from satellite remote sensing, long-duration observations from ground-based networks, and closure experiments from field studies.
- Finally, there is need for datasets that can be used to examine model simulations of particular aerosol types.

In all of this work, data preparation and quality assurance is essential, and these activities are rarely adequately funded. It is critical to remind data users that available data are often (1) inaccurate, (2) the results of other models, (3) inapplicable at scales needed and (4) not adequately characterized in terms of quality and uncertainty. Thus, when designing and executing model evaluations, it is essential to communicate with data providers in order to better understand data quality and limitations, to assure scale comparability (e.g. gridded products at the resolution of modeling, or model sub-sampling to accommodate data), to be specific on data exchange formats and naming conventions, and to have appropriate visualization and evaluation tools readily available.

3.4 Evaluating PM Forecast Models: Lessons from ICARTT/NEAQS-2004

As forecasts of PM$_{2.5}$ aerosol become routinely available to the public, it becomes critical to evaluate the skill of these forecasts and to determine whether or not key processes that govern the behavior of PM$_{2.5}$ are being adequately modeled. During the summer of 2004, a consortium of field programs termed ICARTT (International Consortium for Atmospheric Research on Transport and Transformation) conducted an intensive field study of regional air quality, long-range transport of air pollutants, and the effects of aerosols on radiative energy transfer in the troposphere (see http://www.esrl.noaa.gov/csd/ICARTT/). The ICARTT-2004 field program also included a air pollution forecast model evaluation project in which real-time forecasts of nine air quality models were collected at a central facility and compared with observations (both surface and aloft) from the field study. Although ozone was the primary focus of these forecasts, five research centers also provided forecasts of PM$_{2.5}$ mass and PM$_{2.5}$ composition over the northeastern United States and southeastern Canada. The research groups included two NOAA facilities (the NWS/NCEP CMAQ/Eta model and the ESRL/GSD WRF-Chem model), two groups from the Canadian Meteorological Service (the operational CHRONOS and developmental AURAMS models), and the University of Iowa, which ran the STEM-2K3 model. The centralized collection of seven PM$_{2.5}$ forecasts proved to be useful in quickly identifying

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4 NOAA/ESRL/CSD, National Oceanic and Atmospheric Administration, Environmental Sciences Research Lab, Chemical Sciences Division, Boulder, Colorado.
deficiencies, irregularities and inconsistencies common to all models, as well as for
evaluating the relative performance of the models. Figure 5 summarizes the models and
model domains used in the model performance evaluations.

Figure 5. Models and domains used in the ICARTT/NEAQS-2004 model
evaluation exercise.

Observations from three surface platforms and one upper-air platform were used in the
PM$_{2.5}$ forecast evaluations. The spatial characteristics of regional-scale surface PM$_{2.5}$
mass were provided by hourly PM$_{2.5}$ observations over a 35-day period at 118 AIRNow
surface monitors throughout the eastern United States and Canada. The EPA Speciation
Trends Network provided similar spatial coverage (76 monitors in the eastern
United States and Canada) from 24-hour filter samples that were used to evaluate model surface
PM$_{2.5}$ composition. Detailed PM$_{2.5}$ and gas-phase composition measurements were
available from both the NOAA Research Vessel Ronald H. Brown and the NOAA WP-
3D aircraft as part of the combined New England Air Quality Study (NEAQS) within
ICARTT (http://www.al.noaa.gov/csd/2004). These data were used to assess the
performance of the models aloft.

Several conclusions can be drawn from the results of these evaluations:
• Six out of seven models show skill (when measured against observed persistence) in 24-hour forecasts of 8-hr afternoon average surface PM$_{2.5}$ levels in the eastern United States and Canada. This finding contrasts sharply with ozone forecasts, which exhibit very little skill for similar comparison conditions.
This difference between PM$_{2.5}$ and O$_3$ forecast skill is due to large O$_3$ biases as compared to comparatively smaller relative PM$_{2.5}$ biases in the forecast models.

- Similar to recent findings with ensemble ozone forecasts, the PM$_{2.5}$ ensemble forecast derived from either the geometric or arithmetic mean of all model results exhibits substantially improved correlation coefficients and forecast skill for surface PM$_{2.5}$.

- The typical diurnal pattern of PM$_{2.5}$ for rural and suburban monitors shows maximum values under stable meteorological conditions with one peak at ~9:00 a.m. and another at ~10:00 p.m. local time. As shown in Figure 6, all models fail to replicate both the timing and the relative magnitude of the diurnal variation. Subsequent experiments using alternative planetary boundary layer (PBL) modules within the WRF/Chem model, demonstrated that the principal reason for this failure was due to inadequacies in the treatment of PBL dynamics in each model.

![Figure 6](image-url). Representative diurnal behavior of several PM models compared to average surface observations for three classes of surface measurement sites.
• Both the total mass and relative fraction of organic carbon within PM$_{2.5}$ is significantly underestimated by all of the forecast models. Organic carbon comprises more than half the PM$_{2.5}$ mass for a majority of aircraft observations within the PBL. This characteristic suggests a common deficiency in the formulation of all forecast models. Correlations of organic carbon with elemental carbon from the Speciation Trend Network surface filter measurement data suggest that most of the organic carbon at rural monitors is anthropogenic. However, most of the “rural” monitors used in these evaluations are located relatively close to urban areas; thus, they cannot be considered remote.

• Those models that include aqueous phase oxidation of SO$_2$ into PM$_{2.5}$-sulfate over-predict PM$_{2.5}$ sulfate levels as well as the sulfate fraction of total sulfur (SO$_2$ plus PM$_{2.5}$-sulfate). Models that do not include this oxidation pathway showed either near-zero or negative bias in PM$_{2.5}$ sulfate, but they under-predicted the sulfate fraction of total sulfur. This behavior, which is depicted in Figure 7, illustrates the need for including the aqueous-phase SO$_2$ oxidation mechanism within PM$_{2.5}$ forecast models and also illustrates the need to explain why SO$_2$ oxidation is too fast in those forecast models that include it.

![Figure 7](image-url)

**Figure 7.** Model-predicted and observed SO$_2$/(Total Sulfur) fraction for two air quality forecast models: WRF-Chem and CMAQ/Eta. WRF-Chem does not consider in-cloud SO$_2$ oxidation and often overestimates the SO$_2$/(Total Sulfur) fraction, suggesting that gas-phase conversion alone underestimates total SO$_2$ conversion to sulfate. CMAQ/Eta explicitly calculates in-cloud conversion of SO$_2$, but it often under-predicts the SO$_2$/(Total Sulfur) fraction, implying that the liquid-phase oxidation process within this model is converting SO$_2$ to sulfate too rapidly.
4.0 BREAKOUT SESSIONS

Following the keynote presentations, the workshop reconvened in five breakout sessions:

• New Particle Formation (led by Peter McMurry)
• Chemistry (led by Richard Kamens and Sonia Kreidenweis)
• Aerosol/Cloud Interactions & Wet Deposition (led by Tom Ackerman)
• Emissions, Transport and Dry Deposition (led by Chris Doran and Tami Bond)
• PM Forecasting (led by Rohit Mathur).

Guided by the original questions posed to the workshop attendees (reproduced in Section 2.0), the discussions during the breakout sessions considered the current modeling status and experimental challenges in each of the five areas. Discussion summaries and breakout session recommendations are provided below. A list of breakout session attendees is provided in Appendix B.

4.1 New Particle Formation

The breakout session on new particle formation (NPF) focused on the formation of new particles by homogeneous nucleation in the atmosphere. Homogeneous nucleation in the atmosphere is poorly understood, and it may play an important role in regulating global climate. For readers desiring additional information on the subject, a comprehensive review of the topic was published recently in the Journal of Aerosol Science (Kumala et al., 2004).

Recent observations show that nucleation is a major source of particles in many parts of the atmosphere (Brock et al., 1995; Clarke, 1993; Clarke et al. 1998; Kumala et al., 2004). Particles formed by nucleation can lead to both direct and indirect effects on the earth’s radiation balance (IPCC, 2002). Direct effects arise from scattering or absorption of light by particles, whereas indirect radiative effects are associated with the role particles play as CCN. CCN concentrations affect the extent of cloud cover, the optical properties of clouds, and the cleansing of the atmosphere by rainfall. Cloud optical properties depend on the size distributions and concentrations of cloud droplets, which in turn depend upon CCN concentrations. Observations show that freshly nucleated particles can grow to sizes large enough (~100 nm in diameter) to serve as CCN during the course of a single day. Modeling studies show that new particle formation can significantly affect CCN concentrations (Ghan et al., 2001) and some atmospheric observations have been carried out that also support this conclusion (Kerminen et al., 2005; Laaksonen et al., 2005). Clearly, an understanding of the radiative effects of aerosols will require that the effects of new particle production on aerosol size distributions and CCN concentrations be understood sufficiently to enable these effects to be treated in the models (Pirjolo et al., 2004).

The following results are apparent from atmospheric observations: 1) particles are produced in a variety of locations in the remote marine troposphere and in the clean and polluted continental troposphere, and these particles can account for a significant fraction of all atmospheric particles; 2) H$_2$SO$_4$ vapor often participates in nucleation
(Eisele and McMurry, 1997; Kulmala et al., 2004; Clark et al., 1998); 3) particle production rates, particularly near the earth’s surface, can be orders of magnitude higher than rates predicted by binary nucleation theory as applied to the H$_2$SO$_4$ / H$_2$O system; and 4) once formed, these nanoparticles often grow at a rate two to ten times faster than can be explained by the condensation of H$_2$SO$_4$ vapor and its associated H$_2$O and NH$_3$ (Stolzenburg et al., 2005). The literature provides convincing evidence that both anthropogenic (e.g., SO$_2$) and biogenic (e.g., terpenes, sesquiterpenes, diiodomethane, etc.) sources contribute to new particle formation and growth (O’Dowd et al., 2004; Sakurai et al., 2005; Smith et al., 2005), but the roles of biogenic and anthropogenic precursors in different airsheds are not yet quantitatively understood. Nucleation sometimes follows regular diurnal patterns, with peak particle production rates occurring near midday when solar radiation is most intense (Bradbury and Meuron, 1938; Hogan, 1968; Kontsenogii and Jaenicke, 1994; Went 1964). At other times, nucleation occurs in response to atmospheric perturbations, such as the removal of preexisting particles by cloud processing (Clarke et al., 1998; Hegg, Radke and Hobbs, 1990; Perry and Hobbs, 1991; Radke and Hobbs, 1991) or the addition of gas-phase reactants from a surface source, such as exposed coastal zones (Jimenez et al., 2003; McGovern, Jennings and Oconnor, 1996; McGovern 1999; O’Dowd et al., 1999), smokestacks (Brock et al., 2002), or biogenic sources (Weber et al., 1998). Impactor measurements of aerosol composition suggest that freshly nucleated particles in the Finnish boreal forest are enriched with dimethyl amine (Mäkelä et al., 2001). Several groups have argued that nucleation-mode particles detected in the marine boundary layer are probably produced aloft in free tropospheric cloud outflows and transported to the surface (Covert et al., 1996 a,b). Evidence suggests that nucleation in the upper tropical troposphere is a significant global source of atmospheric particles (Brock et al., 1995; Raes et al., 2000).

Several major research issues must be resolved if new particle formation is to be included in atmospheric models and to quantify its impact on regional and global climate.

- First, microphysical models are needed that predict rates of new particle production with sufficient accuracy. Mechanisms that have been proposed to explain this production include ternary nucleation of sulfuric acid, ammonia and water (Korhonen et al., 1999; Kulmala et al., 1999; Napari et al., 2002; Stanier, Khylstov and Pandis, 2004), nucleation of sulfuric acid and water on negative ions (Lee et al., 2003; Lovejoy, Curtis, and Froyd, 2004), nucleation of products formed by the photooxidation of diiodomethane (Jimenez et al., 2003), and reactive nucleation involving sulfuric acid and organic acids (R.H. Zhang et al., 2004). There is compelling evidence that different mechanisms apply in different regions of the atmosphere, but there is no clear understanding of what these mechanisms may be.

- Second, models are needed that reliably predict growth rates of freshly nucleated particles. Research has shown that the species which form stable molecular clusters can be different from those responsible for particle growth (Park et al., 2004). Indeed, observed growth rates of nucleated nanoparticles are often two to ten times higher than can be explained by the condensation of sulfuric acid (Birmili et al., 2003; Park et al., 2004; Weber et al., 1997, 1998), which is the
most important growth species that has been directly measured to date. There is also evidence that some nucleated particles contain a significant amount of organic material (Q. Zhang et al., 2004; O’Dowd et al., 2002), although the organic compounds that are responsible for their growth are not known with certainty.

**Recommendation No. 4.1.1. Develop new empirical models for New Particle Formation (NPF) Rates.**

Nucleation theories typically predict that rates of stable nuclei formation are very strong functions of the concentration of the nucleating vapors. For example, the binary theory for nucleation of sulfuric acid and water predicts that nucleation rates vary as \([H_2SO_4]^p\) where, typically, 8<p<10. Atmospheric observations, however, suggest that new particle formation rates (i.e., rates at which particles pass through the minimum detectable size, typically ~3 nm) can often be expressed as \(J_{3nm}=A[H_2SO_4]^p\), 1<p<2 (Kulmala, Lehtinen and Laaksonen 2006; Weber et al., 1996). Recent work by Carslaw and coworkers (personal communication to P. McMurry) has shown that the effects of new particle formation on size distributions measured at the Hyytiälä forest in Finland can be reasonably explained by assuming that \(J_{3nm}=A[H_2SO_4]\), where A is an empirical constant based on measurements carried out by Kulmala and coworkers.

More work needs to be done to assess the utility of empirical models, such as these, for NPF rates. First, research needs to be carried out to determine under what conditions such simple expressions successfully reproduce observed rates of new particle formation. This research will require examining existing data sets where particle formation rates and sulfuric acid concentrations were measured, and it may require the acquisition of additional data. Second, factors that affect the value of the coefficient A need to be evaluated. It is likely that A depends on concentrations of species other than H2SO4 that react to produce particles. The concentrations and the identity of those species will almost certainly vary from location-to-location.

**Recommendation No. 4.1.2. Conduct modeling studies to assess importance of NPF to climate.**

A limited amount of work has been done to assess the impact of NPF on CCN concentrations (Ghan et al., 2001; Pirjola et al., 2004); however, a comprehensive understanding of the importance of NPF remains to be developed. Sensitivity calculations should be carried out to assess the sensitivity of radiative forcing to NPF. This work will quantify the accuracy to which new particle formation and growth rates need to be calculated with microphysical models, and it will provide important guidance to those working on the physical chemistry of NPF.

There is now significant empirical information available on the rates at which new particles are formed and grow in a variety of atmospheres (Kulmala et al., 2004a) The previously recommended sensitivity studies should make use of these empirical rates rather than rates calculated using existing theories of dubious validity.
**Recommendation No. 4.1.3.** Develop models for growth rates of freshly nucleated particles.

We know that H$_2$SO$_4$ vapor condenses on particles, contributing to growth. We also know, however, that in most places where measured growth rates are compared with growth rates calculated using measured H$_2$SO$_4$ concentrations (including uptake of H$_2$O and NH$_3$ that occurs when H$_2$SO$_4$ condenses), the measured growth rates are typically five to twenty times higher. It follows that other species in addition to H$_2$SO$_4$ contribute significantly to growth. It is essential that these discrepancies be understood. The higher the value of the growth rate, the greater the likelihood that nucleated particles will grow to sizes that are large enough to produce CCN before they are lost by coagulation. It has been speculated that organics are major contributors to particle growth, and there is some evidence to support this. Our understanding of the effect of organics on growth, however, is far from quantitative or definitive.

It is essential that models for growth rates of freshly nucleated particles be developed and verified by atmospheric measurements. Such models will require an understanding of the gas phase precursors that contribute to growth.

**Recommendation No. 4.1.4.** Develop methods to measure concentrations of trace species that participate in new particle formation and growth.

There is strong evidence that H$_2$SO$_4$ participates in both nucleation and growth processes. In addition, a method has been available for measuring the concentrations of H$_2$SO$_4$ at atmospheric levels (down to $10^4$ molecules cm$^{-3}$) and with high time resolution (30 s) for nearly two decades (Eisele and Tanner 1993; Eisele and Berresheim 1992). Methods for measuring NH$_3$ are also now available (Fehsenfeld et al., 2002; Nowak et al. 2002). However, methods for measuring concentrations of trace organic compounds (e.g., organic acids and amines) that are likely to participate in such processes are not available and are needed.

**Recommendation No. 4.1.5.** Carry out collaborative modeling studies and atmospheric observations to assess the skill of models at predicting number distributions.

Models for atmospheric aerosol processing use moment or sectional approaches to evaluate size distributions. Most efforts to compare model predictions with atmospheric observations have focused on mass. However, in many cases (e.g., radiative transfer and cloud formation) number concentrations are equally important. An effort should be made to carry out collaborative modeling and observational studies to assess and refine the skill of aerosol models for calculating number distributions. Such work would, for example, enable a quantitative assessment of the direct and indirect effects of new particle formation on radiative forcing. However, aerosol number distributions are significantly affected by sources as well as by new particle formation. Thus, this work would also help assess the adequacy of current aerosol emission models.
4.2 Chemistry

The major components of atmospheric PM$_{2.5}$ are sulfates, nitrates, ammonium, organic carbon and elemental carbon (NARSTO, 2004). In general, the organic carbon fraction of particulate matter close to the Earth’s surface throughout North America (on an annual average or long-term averaging basis) varies between 10 and 40 percent (NARSTO, 2004). Secondary organic aerosol (SOA) formation on summer or high oxidant days may account for 50 to 90 percent of the organic fine particle fraction (Lim and Turpin, 2002; Pandis, 2003; Schauer et al., 2002; Turpin and Huntzicker, 1995; Xiao-Feng et al. 2005), as has been observed in Beijing, Los Angeles, Pittsburgh, Atlanta, and other locations. SOA is defined here as organic compounds that reside in the particle phase as a result of gas-phase reactions or heterogeneous reactions on atmospheric particles. SOA coated on existing aerosols may dramatically alter particle geometry and optical properties. Most SOA is to some extent hygroscopic and can thus serve as CCN. Simply stated, however, SOA formation in global, regional and urban models is not adequately represented. Therefore, a high priority should be assigned to the improvement of the representation of SOA in these models, including improvements in emission estimates for SOA precursors as well as in the description of the chemical and physical processes that lead to SOA.

In many locations in the United States and western Europe, volatile aromatic compounds comprise up to 45 percent of the urban hydrocarbon mixture in the atmosphere (Fujita et al., 1997; Kurtenbach et al., 1998). Toluene, m- and p-xylene, benzene and trimethyl benzenes, o-xylene and ethylbenzene make up 60 to 75 percent of this load. Model simulations (Makar et al. 2003) of aerosol formation in urban centers in the northeastern United States have shown that the aerosol potential of this class of compounds (Cocker et al., 2001; Edney et al., 2001; Kleindienst et al., 2004; Odum et al. 1997) along with terpenes and other compound classes, are dominated by SOA from aromatics. Like aromatics, terpene emissions from vegetation have long been recognized as potential sources of SOA (Andersson-Sköld and Simpson, 2001; Griffin et al., 1999; Rasmussen, 1972; Went, 1960). Globally, biogenic emissions from vegetation exceed anthropogenic hydrocarbon emissions, and the contribution of biogenics to semi-rural atmospheres can be considerable. Andersson-Sköld and Simpson (2001) estimated that terpene reactions may contribute up to 50 percent of the total organic aerosol in Scandinavia, and this can be greater than the organic aerosol emissions from anthropogenic sources depending on location and season. Also depending on location, α-pinene, β-pinene, d-limonene, and β-caryophyllene emissions may generate more than 65 percent of the SOA from terpene emissions. Sesquiterpene emissions (C15 double-bonded biogenic hydrocarbons) are typically underestimated due to rapid atmospheric reactions and resulting sampling difficulties (Fuentes et al., 2000). Most recently the atmospheric reactions of isoprene, the biogenic building block for terpenes, and the most highly emitted biogenic hydrocarbon, have been shown to lead to SOA formation (Claeys et al., 2004a,b; Edney et al. 2005). Lastly, alkanes have also been shown in recent chamber studies to have considerable SOA potential (Young et al., 2005) and need to be considered in SOA models.
The conceptual framework for current SOA models (Chen and Griffin, 2005; Johnson et al., 2004; Kamens et al., 1999; Kamens and Jaoui, 2001; Odum et al., 1997; Pun et al., 2002; Stroud et al., 2004) is based on trace gas-particle partitioning theory (Pankow, 1994), and most current models ignore the role of particle-phase reactions in producing SOA. Gas-phase reactions of terpene, single-ring aromatics, and C10 and greater paraffins generate low vapor pressure semi-volatile oxygenated products ($10^{-2}$ to $10^{-9}$ torr). These products then thermodynamically distribute between the gas and particle phases. This process appears to be very rapid and occurs on the time scale of seconds. Existing SOA models have been largely evaluated against single component SOA precursor smog chamber experiments and have not been designed or tested against complex organic mixtures.

The amount of organic product that partitions to the particle phase of liquid-like aerosols depends not only on the pure liquid vapor pressure, but also on the amount of absorbing aerosol present and compound activity coefficients (solubility) in the liquid mixture of the particle (Pankow, 1994). The solubilities of partitioning product organic oxygenates and nitrates will differ for particles of different polarities. In the case of sulfate aerosol, ammonium sulfate is often used as “seed” aerosol for SOA and partitioning studies (Cocker et al., 2001a,b; Jang and Kamens, 2001; Odum et al., 2001). Experimental work suggest that wet or dry (NH$_4$)SO$_4$ particles may not significantly impact SOA formation (Cocker et al., 2001a,b) although particle electrolytes can influence α-pinene SOA (Cocker et al., 2001a). Finally, terpenes at part-per-million and sub-part-per-million levels in the presence of ozone or OH, and aromatics in the presence of OH can self-nucleate in environmental chambers. At ambient levels, however, current knowledge implicates sulfuric acid involvement in new particle formation in the atmosphere.

Although a conceptual partitioning framework has been successfully applied to smog chamber studies and used to evaluate parameters in semi-empirical models of SOA formation, it has been recently shown that known aerosol formation reactions are insufficient to explain observed tropospheric organic aerosol concentrations, particularly above the boundary layer. It has also been known for a few years that reactions can occur inside particles that involve volatile molecules converting into less volatile species (Jang and Kamens, 2001; Kalberer et al., 2004; Tolocka et al., 2004). Most recently, lab experiments (Gao et al., 2004; Kalberer et al., 2004; Tolocka et al., 2004) have shown that high-molecular weight-compounds form on multi-hour time scales, and atmospheric observations confirm the presence of high-MW organic aerosol species. However, current models do not consider organic product interactions in particle mixtures, or do not explicitly represent the large fraction of polymer products that may contribute to SOA. The types of particle phase polymers that form are largely unknown. They also do not consider the effects of particle aging and the associated particle “hardening,” or volatility reduction, that appears to occur over time scales of hours and tens of hours. Finally, recent modeling studies, supported by observations, suggest SOA can be formed in cloud water from volatile precursors. Therefore, SOA formation depends not on partitioning alone but also on reactive processes in the particle or aqueous phase, and models of SOA formation need to be upgraded to reflect this new knowledge.
In order to model SOA formation there is an urgent need to characterize gas- and particle-phase reactions that lead to particle-phase polymer formation. It is also important to predict the rates at which these processes occur. A “next” generation of particle mass spectrometers and other approaches coupled with off-line techniques is required so that polymer formation can be quantified. These instruments should be developed and evaluated on smog chamber test atmospheres so that their ability to respond to different kinds of SOA and overall sensitivity can be evaluated before they are moved into the field.

Simple and complex smog chamber mixture experiments are needed for SOA model development. Currently no such data set exists, but such a compilation needs to be made available to the kinetics modeling community. These experiments should be conducted at as low a concentration as possible in the presence of different particle backgrounds. Experiments should include single SOA generating precursors, binary mixtures, and build to complex mixtures that involve terpenes, sesquiterpenes, aromatics and alkanes. These experiments should also be designed so that it is possible to follow and predict polarity changes as aerosols age in these smog chamber systems. Finally, cloud processing of SOA is a very important area and experiments should be developed to quantify the importance of these reactions.

**Recommendation No. 4.2.1. Develop a database of SOA formation experiments and SOA precursor emissions for use by the modeling community.**

Formation of such a database would include the following activities:
- Create a hierarchy of smog chamber oxidation studies for single compounds, binary mixtures, and complex mixtures that involve aromatics, terpenes, sesquiterpenes and alkanes.
- Define the role of preexisting particles in SOA formation.
- Include descriptions of particle ageing and hardening.
- Develop, where possible, parameterizations that may be used in global-scale modeling.
- Improve emission inventories for SOA precursors. Particular interest should be paid to vegetation, urban and regional anthropogenic sources, and to large-scale burning both natural and prescribed.
- Improve understanding of the importance of urban sources of SOAs and SOA precursors on the global scale.
- Explore the use of satellite measurements for providing emission proxy data.

**Recommendation No. 4.2.2. Design new analytical techniques for speciating organic aerosol matter that do not require filter collections, extractions, or harsh preprocessing conditions.**

In past experiments, the composition of SOA has probably been perturbed during sampling, handling and analyses. Eliminating this problem will require the development
of instruments for high-resolution detection of trace species that include oligomers and polymers, and the development of calibration techniques for these compounds.

**Recommendation No. 4.2.3.** Design experiments to quantify the role of cloud processing in SOA formation.

No specific experiments were suggested by the breakout group, but these kinds of experiments would probably need to include both field and laboratory approaches.

### 4.3 Aerosol/Cloud Interactions & Wet Deposition

As discussed in Section 3.1, the principal aerosol-related processes that must be considered in terms of cloud/aerosol interactions are 1) aerosol particle activation, 2) aqueous phase reactions in clouds, 3) convective transport and removal, 4) in-cloud and below-cloud scavenging, and how these processes affect cloud droplet size distributions and precipitation processes. Several of these processes, and especially their effect on cloud optical properties, cloud lifecycles, and precipitation, are not well understood or well characterized experimentally.

The principal problem that complicates both the study and the modeling of cloud processes is the problem of scale and the interactions among these scales. The direct interaction of aerosols and hydrometeors occurs at size ranges considerably less than a meter. The environment in which these interactions occur, in turn, is affected by dynamic and thermodynamic processes on the scale of individual clouds, which is on the order of hundreds of meters. Finally, the environment in which clouds are formed is set by large-scale physics – on the order of tens to hundreds of kilometers. This spectrum of scale interactions, which span at least five orders of magnitude, means that aerosol/cloud interactions in current regional to global aerosol and climate models must be represented by parameterizations that link the aerosol/cloud process or effect of interest with characteristics of the larger scale environment. The mathematical formulation of these parameterizations may change with the size of what is considered the “larger scale environment”. This phenomenon is generally referred to as the “tuning problem” – that is, whenever the size resolution of a model is changed, the process parameterizations have to be adjusted or modified to obtain acceptable performance.

The ultimate source of cloud-process parameterizations is field observations enlightened by detailed cloud-scale (or smaller) model simulations and occasionally by laboratory experiments. However, obtaining the field data needed to generate parameterizations is a formidable challenge. No single measurement platform, measurement strategy, suite of instruments, or experimental venue is capable of providing the information needed to generate a robust set of parameterizations that represent the full range of aerosol/cloud interactions. Aerosol particles are physically and chemically complex and regionally diverse as are the environments in which they are found. The available data do not span this range of complexity, and given the expense of conducting comprehensive, well-designed field experiments, they are never likely to do so. Nevertheless, advancing scientific understanding of aerosol/cloud interactions and developing process modules and parameterizations for representing the effects of these interactions requires field data
generated by deployments of multiple instruments and platforms (ground-based in situ instruments, ground-based remote sensing, satellite-based remote sensing, and aircraft measurements) in a diverse range of climatological settings. Interpretation of these data, development and testing of parameterizations, as well as the design of the experiments themselves, must be informed by theory and by sophisticated high-resolution numerical model simulations. Finally, the resulting aerosol/cloud modules or parameterizations must be tested against observations in the models for which they are designed. In other words, progress in this area can only be made through well thought out integration of theory, field, and modeling work.

With these thoughts in mind, the aerosol/cloud interactions and wet deposition breakout session organized its recommendations around a set of five problem areas:

1. Building parameterizations from cloud-scale observational data.
2. Characterizing aerosol complexity and the effects of aerosols on cloud microphysics.
3. Characterizing the effects of aerosols on precipitation processes.
4. Characterizing the effects of clouds on aerosol composition, transport and removal.
5. Using data from past experiments.

**Recommendation 4.3.1. Use model simulations and observational data to develop cloud-process parameterizations for regional to global-scale aerosol and climate models.**

Climate and aerosol models need parameterizations that include both microphysical processes and cloud-scale dynamics. As it is impossible to develop comprehensive parameterizations from observational data alone, model simulations should be used in conjunction with observations to develop and test characterizations of cloud/aerosol processes and to develop modules (parameterizations) for representing these processes in large-scale models. Cloud resolving models, for example, can incorporate more complete cloud and aerosol microphysics than models with minimum horizontal resolution on the order of kilometers or larger. Cloud resolving models also provide simulations that are easier to test with field observations. However, since field data, and especially in situ or vertical profile data, represent limited temporal and spatial slices through a much larger time and space domain, considerable thought needs to be given as to how these data can be combined with other measurements, such as satellite observations, in order to integrate them over multiple scales. In addition, satellite data can be used to place local measurements in a larger, climatological context. Finally, once process modules have been developed and tested for cloud-resolving models, these modules can be imported to global-scale models that represent cloud processes via embedded cloud-system resolving models (Khairoutdinov et al., 2005). Although these models are too computationally intensive to be used for long-term climate simulations, they could be used to develop the kinds of statistically based parameterizations that are used for coarser-resolution operational climate models. The advantage of using embedded cloud-system resolving models for this task is their ability to generate data
covering a wider range of climatological conditions than is practical from observations alone.

**Recommendation 4.3.2.** Obtain better information on the range in physical and chemical characteristics of atmospheric aerosols.

Characterizing and modeling the effects of aerosols on clouds and cloud processes require knowledge of the size distribution and chemical composition of the aerosols involved. However, the morphology, chemical makeup, and mixing state of atmospheric aerosols are bewilderingly complex, especially when it comes to carbonaceous compounds. One possible approach to dealing with this complexity would be to aggregate particles into general classes of physical characteristics and chemical composition. In fact, a similar approach has been suggested for dealing with the problem of characterizing the health effects of atmospheric aerosols (see, for example, [http://www.narsto.org/files/files/WorkshopSummary.pdf](http://www.narsto.org/files/files/WorkshopSummary.pdf)). Creating a database for such classification will require field measurements of size-resolved hygroscopicity, chemical composition, and mixing state as well as CCN activation and growth kinetics. These measurements would need to be made across a wide range of chemical environments, and they would require sampling from both aircraft and ground-based platforms

**Recommendation 4.3.3.** In order to gain a better understanding of the effects of aerosols on precipitation processes, research should give priority to characterizing the effects of aerosols on shallow convection and stratus clouds.

Three types of studies are suggested: 1) Investigations of gradient transitions in stratus and open cell convection. The focus would be on characterizing the interaction of aerosol number and type with low-cloud formation and drizzle. 2) Field studies in stratus to investigate the autoconversion/precipitation-drizzle problem. Such studies, however, call for better instruments for obtaining cloud droplet size distribution measurements. In particular, there is need for improved accuracy at the smaller end of the size spectrum (around 1 µm), increased sample volume (better statistics), and faster response. 3) Investigations of the comparative role of convective versus stratiform clouds in the Earth’s radiation balance.

**Recommendation 4.3.4.** In order to better characterize the effects of clouds on aerosols, additional research is needed on 1) vertical transport of aerosols by convection, 2) scavenging and wet deposition of black carbon, and 3) in-cloud chemistry of primary and secondary organic aerosols.

With respect to vertical transport, organization of a ground-based lidar network combined with measurements from the CALIPSO satellite would provide needed data for documenting the vertical distributions of aerosols and for evaluating vertical transport computations used in global aerosol models. This information would also provide valuable information on intercontinental transport of particulate matter air pollution. In order to gain better understanding of black carbon wet deposition, enhanced precipitation collection arrays (and analysis for carbon species) are needed.
Scavenging studies should include in-cloud and below-cloud droplet sampling and measurements of the hygroscopicity of black carbon as a function of aerosol sulfur coating. In terms of in-cloud chemistry, we need to move beyond sulfur chemistry to a more complete understanding of the aqueous-phase chemistry of organic aerosols.

**Recommendation 4.3.5. Increased attention should be placed on analysis of past field experiments.**

Comprehensive field experiments are expensive, and data from past field experiments can be valuable sources of information for addressing many of the research priorities listed above. But three problems, in particular, stand out when considering historical data. First, there is need for a good catalogue of past experimental data in order to provide researchers information on which datasets might meet their needs. Second, this data catalogue needs to provide information on data availability and comprehensive information on data quality. Third, methods for producing derived data products must be well documented, as there can be issues of consistency and accuracy in data analysis among seemingly similar derived datasets.

**Recommendation 4.3.6. Additional research is needed on ice nuclei production, ice nucleation, and the effects of aerosols on these processes.**

The Atmospheric Radiation Measurement (ARM) program’s North Slope of Alaska (NSA) site is an excellent venue for such studies, and joint research with ARM on these matters is highly encouraged. In fact, ARM will be conducting a field experiment at the NSA site in April 2008 that specifically addresses interactions between aerosols and mixed-phase clouds. This experiment follows and earlier study, the Mixed-Phase Arctic Cloud Experiment (M-PACE), which was conducted in October 2004.

### 4.4. Emissions, Transport, and Dry Deposition

Characterizing the effects of aerosols on climate, as well as improving our ability to provide operational forecasts of particulate matter air quality, require improvements in current emission information. From the perspective of modeling the effects of aerosols on climate, the breakout group recommended placing a priority on improving information on the properties of primary particles at the point of emission and on characterizing the dependence of “aerosol initial state” upon the characteristics of the model that will be using this information.

#### 4.4.1 Emissions

The physical properties of aerosols – including hygroscopicity, surface tension, refractive index and size distribution – govern their direct and indirect effects on climate. The working group assumed that current research will lead to the development of process modules (and their installation in aerosol and climate models) that adequately describe the evolution of the chemical and physical properties of aerosols as well as their effects on radiative energy transfer. Once this capability is available, these models will require a description of the initial state of the aerosol as model input. As the following two recommendations point out, this information will be needed not only at the point of
emission to the atmosphere, but also at the minimum resolution scale of the model in question.

**Recommendation No. 4.4.1 (particle properties at emission).** Develop size-resolved data on chemical composition and mixing state at the point of emission, particularly for particle diameters greater than 50 nm. As these measurements are made, it will also be useful to determine other properties, such as total scattering, absorption and phase function. These properties can serve as calibration points for connecting composition and physical properties. Emission inventories for secondary aerosol precursors are also needed, and all these inventories need to have global coverage.

**Recommendation No. 4.4.2 (aerosol initial state).** Develop and evaluate methodologies for processing aerosol and precursor emissions in order to provide scale-appropriate emission information for models. Particular attention needs to be devoted to transport and transformation processes with time scales of a few tens of minutes to a few hours. Development of these tools is also important for PM forecasting applications.

Aerosols emitted from combustion sources are extremely dynamic and evolve substantially in composition and size on a time scale of minutes to hours. Thus, the specification of “as-emitted” depends on the nature of the model involved. The larger the spatial scale, the more preprocessing is required in order to get the appropriate initial state, because the apparent emission rates of primary and secondary aerosols are sensitive to the minimum model resolution. For local and regional-scale models, emission information (which is provided at the point of release to the atmosphere) must be processed so that it is representative of fresh emissions mixed on a scale that can range from a hundred meters or less to a few kilometers. In contrast, global models may require emission information appropriate to scales of a hundred kilometers or more. Thus, different emission inventories may be needed for models with different scales: tailpipe, roadway, neighborhood, urban, regional, continental and global. This requirement introduces, in turn, a range of temporal scales over which emission behavior must be understood.

Providing this kind of information requires improvement in our understanding of the evolution of primary aerosols and the formation of secondary aerosols on the timescales representative of transformation between initial emissions and grid-scale resolution. As these processes are not well understood, both experiments (such as those that have occurred near roadways and in laboratories) and models (such as large-eddy simulations) should be brought to bear on this matter. Since “aerosol initial state” is so scale dependent, providing this level of emissions detail should be an inherent part of the modeling procedure.

**Recommendation No. 4.4.3 (focus species)** Develop more accurate emission inventories of primary organic aerosols and organic aerosol precursors. Knowledge of primary emissions of nitrogen compounds (including, but not limited to, soil emissions) should also be improved.
There are several reasons for the choice of priorities in this recommendation. The organic fraction of particulate matter typically accounts for about half of the total particle mass, but its composition and properties are poorly understood. At urban scales, the source of about one-third to one-half of the organic matter remains unknown. Direct radiative forcing by organic aerosols may be either positive or negative, depending upon composition, and OC probably influences formation of CCN.

As one means of improving organic carbon emission estimates, the working group recommends mass closure studies at sites that have no significant upwind sources (e.g., Edmonton), using upwind and downwind measurements and source characterization of both primary organic carbon and precursors. Measurements that can partition the relative contributions of anthropogenic and biogenic emissions, or primary and secondary emissions, should be included in such studies and further developed. Such measurements include use of $^{14}$C and of fragment markers used in mass spectrometry.

**Recommendation No. 4.4.4 (source categories).** Improve data sets for emissions from residential wood combustion, and other biofuels, and on vegetative burning.

Measurements of size-resolved chemical composition of primary emissions are needed for all classes of biomass burning. This type of combustion exhibits substantial variability, which must be characterized and attributed to governing factors when possible. For example, a variety of vegetation classes or fire intensities may be related to open burning emissions. In addition to biomass burning, a recent survey of organic carbon emission rates (Bond et al., 2004) identified gasoline vehicles and the use of solid fuel (wood or coal) in industry as source categories requiring improved emissions information. Although the importance of these emitters varied by region, residential wood combustion was significant in all world regions.

The breakout group was unable to recommend characterization of specific precursor gases because the reactions and precursors of large amounts of secondary organic aerosol have not been identified. Within the group, this inability came to be known as a “chicken and egg” problem: the source community is ready to quantify sources once the needed precursors are identified, and the process community is ready to study the chemical mechanisms once the precursors are recognized. It is essential to determine some classification of both precursors and mechanisms, even if it is not fully precise, so that both communities can move forward.

**Recommendation No. 4.4.5 (data synthesis).** Integrate the detailed, but necessarily sparse, in situ measurements that provide information on chemical composition with global satellite data. These efforts may include data assimilation into numerical models or statistical analysis. Extensive data synthesis should also be applied to evaluate regional models and their representation of vertical transport processes (see Recommendation 4.4.9). Relevant instruments may include satellites, ground-based lidar, radar wind profilers, and tower-mounted instruments.
Whenever possible, inverse modeling using both in situ and satellite data should be applied to improve global emission inventories and to evaluate models. Satellite measurements of reactive gases, which are concentrated near source regions, may help to evaluate both activity and emissions. Retrieved parameters such as spectral extinction, optical depth, estimates of aerosol size, and air mass classification can also provide constraints; some satellite measurements (e.g., multi-angle imaging spectroradiometer, or MISR) can provide information on plume dynamics.

**Recommendation No. 4.4.6** *(miscellaneous emission data development needs)*. *Use North America as a test case for developing data-sharing methodologies.*

Because of the global nature of the climate problem, emission inventories must be global. However, the working group recognized the difficulty of obtaining emission information from all relevant countries. Because NARSTO has already fostered international cooperation within North America in estimating emissions, it was suggested that this model should be cultivated, and the lessons learned from this experience used to work towards improving global emission inventories. Close NARSTO collaboration with international working groups, for example the Global Emissions Inventory Activity (GEIA),\(^5\) will facilitate this knowledge transfer.

**Recommendation No. 4.4.7** *(miscellaneous emission data development needs)*. *Make significant improvements in the timeliness of emission data availability.*

Both the forecasting and analysis phases of field measurement campaigns need accurate, up-to-date data. These data are difficult to obtain even for North America.

**Recommendation 4.4.8** *(miscellaneous emission data development needs)*. *Develop methods for estimating future emissions in order to enable studies of their effects on future concentrations of particulate matter.*

These estimates are needed for both anthropogenic and biogenic sources.

Because of time constraints, the breakout group focused on a few topics of significance rather than attempting to cover all relevant issues. For that reason, several topics are not detailed in the recommendations given above. For many of these topics, there are large and active research communities, and their absence in the detailed discussion should not be construed as implying a lack of importance. Some of these neglected topics are

- Sea salt and mineral dust are often considered “natural” aerosols. Whether they are natural or not, these categories must be included in models when these models are being evaluated by comparison with measured aerosol optical depth.
- Black carbon is often co-emitted with organic carbon, and the presence of this absorbing aerosol may alter the sign of direct radiative forcing. Studies on chemical composition of organic carbon emissions will also likely yield information on black carbon.

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\(^5\) [http://geiacenter.org/](http://geiacenter.org/)
• Biogenic emissions of secondary precursors could be largely responsible for the missing organic carbon.
• Biogenic primary particles may be important, and they are not well understood. Sensitivity studies of their potential importance are recommended.
• Airplane contrails may have either a net warming or net cooling effect, depending on the season. Contrail effects are potentially large.

4.4.2 Transport
A significant obstacle to further progress in modeling aerosol fate and transport is the limited ability to model vertical transport and mixing processes in the atmosphere, both in the PBL and above. In particular, an adequate description of mixing in stable conditions, such as frequently occur at night, is still lacking. Material remaining aloft as boundary layer turbulence decays during the late afternoon and evening can enter the free troposphere and be advected considerable distances overnight.

It is often assumed that aerosols are well mixed in the PBL, but this need not be the case. Fast-reacting trace gas species will often not be well mixed, and this vertical inhomogeneity can affect subsequent chemistry. The result of not reproducing these effects is that the vertical distribution of aerosols and their precursors may not be properly represented in models, which, in turn, can adversely affect both local and regional model results. Moreover, model evaluation exercises can be misleading if vertical transport processes are not accurately represented.

Recommendation No. 4.4.9. Design and carry out to examine and characterize vertical transport and mixing processes, in and above the mixed layer, throughout several diurnal cycles, and with particular emphasis on stably stratified conditions in the lower PBL.

4.4.3 Aerosol Dry deposition
Deposition rates exhibit large uncertainties for submicron particles. Predicted deposition velocities vary by as much as a factor or two or more, and recent models predict a shift in the occurrence of minimum deposition velocity from 0.2 µm to 1 µm particles. Measurements of size-resolved aerosol fluxes over a variety of surfaces could help clarify these uncertainties. The importance of such effects on climate-relevant aerosol properties, such as column mass loading, aerosol optical depth, and radiative forcing is not known at this point, however.

Recommendation No. 4.4.10. Conduct sensitivity studies to assess the importance, for climate modeling, of significant improvements in our ability to characterize deposition fluxes of sub-micron aerosol.

4.5 PM Forecasting
The overall goal of this breakout session was to identify the current challenges in PM process representation for short-term (next-day) forecasting of particulate matter
The discussions in this session focused on addressing two broad questions:

1. What are the relevant spatial and temporal scales for short-term (next-day) forecasting of distributions and magnitude of PM concentrations?
2. What are the current model process representation deficiencies for major constituents of total PM$_{2.5}$?

Each question was further subdivided into several sub-themes, which were then discussed in detail and short-term priorities were identified. Where applicable, synergy with aerosol process research relevant to climate and air pollution assessment studies were also identified.

### 4.5.1 Relevant spatial and temporal scales for PM forecasting

The spatial and temporal resolution of current air quality forecast models is largely dictated by the resolution of the driving meteorological model. The spatial resolution currently modeled is about 12 km. However, a target spatial resolution of 1 to 4 km is needed to adequately capture the urban regions. It was also recognized that the accuracy of the “chemical forecast” model can only be as good as that of the driving “meteorological forecast” model.

Forecasters responsible for issuing local air quality forecasts use forecast model guidance as one of their forecasting tools. A key piece of information in developing these forecasts is information on the temporal variability and buildup of pollution over time. Forecasters often need guidance on diurnal variations in ambient PM$_{2.5}$ levels. Thus adequate skill in the forecast model’s ability is needed not only to represent the day-to-day variability in ambient PM$_{2.5}$ levels, but also variations at the shorter scales. The ability to resolve the intra-day variability in PM$_{2.5}$ is desirable from a forecaster’s perspective and poses significant scientific challenges. Improvements in meteorological models as well as emission inventory data at finer temporal and spatial scales, in conjunction with continuous assessment of process representation improvements, are critical in order to achieve this goal.

**Recommendation 4.5.1** Develop a concurrent evolution in the skill of high-resolution meteorological forecast models in order to meet the short-term goals for accurately forecasting PM$_{2.5}$, its chemical constituents, and other related species at the desired scales of 1 to 4 km. In particular,

- Improve the representation of vertical mixing processes (especially at night) and the modeling of convective clouds at fine scales. (See Recommendations 4.3.4 and 4.4.9)
- Improve emission estimates at these scales. This effort includes the development of processors for providing scale-appropriate initial-state information. (see Recommendation 4.4.2).

Currently, the skill of forecast models is assessed in terms of their ability to provide the next-day forecast. However, the ability to provide a two- to three-day forecast was identified to be a desirable short-term goal (i.e., within five years). It was also
recognized that since most chemical forecasts are initialized from a previous forecast cycle, an archive of daily chemical forecast information can provide a valuable continuous record of the chemical state of the atmosphere. With continuous improvement in the model skill and resolution, such an archive would potentially provide a unique source of information for analysis of long-term trends. The ability to analyze seasonal and potentially inter-annual variability, once the archive is populated with several years of data, would be useful for both air pollution as well as climate assessment studies.

**Recommendation 4.5.2.** Create an archive of chemical forecast information in order to provide a continuous record of the forecasted chemical state of the atmosphere. As forecast models improve and with increased use of data assimilation from improved air chemistry monitoring networks, this record could be an important source of chemical climatology reanalysis products.

The need for additional observations of PM$_{2.5}$ and its constituents was also identified to be a major short-term need. The choice of a forecast metric is largely dictated by available measurements against which model forecasts can be verified. Current routine PM measurements are available only at the surface and are spatially sparse. While hourly total PM$_{2.5}$ data is available, models forecast skill and the adequacy of PM process representations can only be adequately assessed for daily average values since speciated data are only available at that temporal scale.

**Recommendation 4.5.3.** Acquire additional speciated measurements of PM$_{2.5}$ concentration with hourly resolution in order to adequately assess the skill of the models in representing diurnal variations in PM$_{2.5}$ concentrations, guide further PM model process representation research, and improve the skill of models.

The requirements for spatial and temporal resolution and model forecast duration needed to improve PM forecasting are considerably more stringent than those for the current models used in air quality assessment studies. Consequently, achievement of the above goals is expected to directly contribute to overall evolution of air quality models and their use in both forecast and assessment studies. The ability to simulate finer spatial and temporal scales in regional models can also provide useful “climate relevant” process guidance, since it is recognized that the PM process challenges are similar for all applications, though scales and priorities are different. The ability to improve representation of spatial heterogeneity in regional PM loading and its compositional characteristics is expected to improve representation and more accurate estimation of aerosol radiative impacts.

**Recommendation 4.5.4.** Conduct an assessment to compare estimates of aerosol loading from regional models with those from current global models when aggregated to a common spatial and temporal scale. The breakout group was not aware of any rigorous comparisons study of this nature.
4.5.2 **Current model process representation deficiencies for major constituents of total PM$_{2.5}$**

As discussed above, detailed verification of modeled PM compositional characteristics is currently limited by availability of observational data sets. While current speciated measurement networks have reasonable spatial coverage, most provide only daily-average measurements of PM chemical components. Hence temporal variability verification of processes regulating PM composition can at best only be currently done for average impacts over a day. Initial comparisons of diurnal variations in total PM$_{2.5}$ predictions from several PM forecast models indicate that most models do not reproduce the observed diurnal cycle at urban and suburban monitor locations, particularly the nighttime to early morning transition (McKeen et al., 2006), and raises questions on the ability of the models to simulate diurnal variations in individual components, even though the daily average compositional predictions are reasonable. While current speciated measurements have been useful in assessing the ability of current models in simulating seasonal variations in PM compositional characteristics, greater frequency in measurements is needed to support rigorous verification of daily predictions from PM forecast models. Breakout group discussions also focused on identifying available high time resolution speciated measurements. Aircraft measurements from intensive field experiments could potentially be used to investigate spatial variability in PM composition at shorter time-scales; it is suggested that this idea be explored to characterize spatial variability in PM composition from high time resolution measurements. Possibilities also exist for innovative analysis of available surface network data to extract the inherent spatial signature in these daily average measurements since daily average measured values, especially at rural locations are likely to represent a larger spatial signature.

Comparisons of PM composition with available routine surface network data and aircraft measurements from field intensives (e.g., ICARTT) have nevertheless helped identify several shortcomings in the ability of existing forecast models to accurately represent the day to day variability in chemical composition of PM$_{2.5}$. In particular, improvements are needed in the representation of process related to gas-particle partitioning of both organic and inorganic constituents.

**Recommendation 4.5.5. Improve representation of:**

- Heterogeneous NO$_3^-$ formation
- Aqueous SO$_4^{2-}$ formation and characterizing discrepancies between modeled and measured total sulfur
- Carbonaceous mass, relative contributions of primary and secondary components to carbonaceous mass, and their seasonal variability.
- Fine-coarse mode interactions (e.g., dynamic mass transfer approach).

Progress in the above-mentioned deficiencies would greatly enhance the ability of current-generation air quality forecast models to accurately predict both total PM$_{2.5}$ mass as well as its chemical constituents. It is expected that within the next two to five years, such models will also need to forecast the distributions of coarse mass (PM$_{10-2.5}$) with reasonable fidelity. It is also recognized that while several current models predict size and number concentrations, these predictions have not been adequately verified due to
lack of sufficient measured data. Verifying the skill of regional PM modes to predict number concentrations and size distribution is also needed for health and climate assessment applications.

Another improvement and area of development for forecast models relates to on-line integrated meteorology and chemistry modeling. Several initial efforts (e.g., GATOR-GCMOM, MM5-MAQSIP, WRF-Chem) have illustrated the potential advantages associated with consistent use of data (and in some cases more frequent) in meteorological and chemistry calculations through on-line coupling. The approach also facilitates the investigation of potentially important feedback effects related to radiative forcing of aerosols. Initial experiments incorporating direct radiative forcing of aerosols in such models have shown that including feedbacks can impact (a) representation of photolysis rates through either attenuation or enhancements, and (b) simulated meteorological field such as temperature and boundary layer heights. As an example, scattering aerosols could potentially reduce photolysis rates. Such reductions could result in reduced secondary pollutants. On the other hand, cooling effects can suppress mixing heights which can then increase predicted concentrations.

**Recommendation 4.5.6.** Perform a rigorous assessment of the feedbacks between chemistry and meteorology and the ability of models to accurately represent these processes. Simulating radiative forcing will require forecast models to include representation of the optical properties of aerosols. This enhancement would enable these models to be used for predicting visibility degradation arising from pollution as well as for climate assessment studies.

Assessment of drawbacks in PM process modules must however be done in conjunction with an assessment of drawbacks and deficiencies (including sampling artifacts) in existing emission estimates and inventories used in the models. For example, discrepancies in prediction of the relative amount of reduced and oxidized nitrogen compounds and their gas/particle partitioning are associated with not only uncertainties in representation of heterogeneous NO$_3^-$ formation, but also by uncertainties in current NH$_3$ emissions and their spatial and temporal allocation as used in the models. Additionally, uncertainties in representation of bi-directional surface exchange processes for NH$_3$ and also sulfur budgets can regulate the simulated NH$_3$ availability and impact model estimates of oxidized nitrogen budgets. The assessment of simulated total sulfur must also include an examination of the processes related to the accurate representation of clouds, dissolution of gaseous species regulating aqueous oxidation of SO$_2$ (e.g., H$_2$O$_2$), and relative to uncertainties in representation of SO$_2$ oxidation sub-grid plumes. Finally, the relative importance of improvements in kinetic parameters, yields, and partitioning coefficients, and improvements in representation of gas and aqueous secondary organic aerosols, should also be investigated in conjunction with improvements in emission estimates for primary organic species and aerosols.

The challenges of providing short-term air quality forecasts also highlights the need for providing short-term, episodic “emission forecasting” tools. For example, emissions from episodic events (e.g., wild and prescribed fire events, dust, etc.), which are
reflected in base-year emission inventories, must be estimated in real-time and pose challenges in air quality forecasting that are not encountered in retrospective assessment studies. It is also necessary to assimilate information on day-to-day variability in emissions from various anthropogenic sectors, such as

- Emissions associated with increased generator use in urban areas during hot days
- Variability in emissions arising from day-to-day variability in energy and load demand on power plants
- Temporary shutdown of facilities
- Addition of new facilities not represented in base year inventory
- Variability in mobile emissions associated with large public events and traffic pattern changes.

These and other emission inventory development needs must be coordinated across North America. There is a clear need to develop an updated and consistent emission data set for the United States, Canada, and Mexico that can be used for forecast models applied to North America.

**Recommendation 4.5.7.** Develop tools for providing emissions from episodic events and for estimating day-to-day emission variability, and place continued emphasis on developing consistent emission data sets for the three countries of North America.

Finally, there is the increasing need to account for long-range transport beyond the continental scale. The experience in running long-term simulations over regional domains that has arisen from forecast applications has highlighted the importance of specifying accurate lateral boundary conditions and the importance of long-range, hemispheric transport in affecting these conditions. One way to provide this information is by coupling regional models to global ones. Initial experiments involving linkage of regional models with global models have shown mixed results, and they have highlighted the need for development of methods that facilitate consistent coupling between these models at different scales. Even if the methods are developed, they will be of little help without the development of a compatible global-scale chemistry forecast model. It would be highly advantageous if the development of such a model could be coordinated with on-going research to develop and improve global aerosol and climate models.

**Recommendation 4.5.8.** Develop an operational global-scale chemistry forecast model and better methods for coupling global and regional forecast models to achieve proper treatment of lateral boundary conditions in regional-scale forecast models.
5.0 REFERENCES


APPENDIX A

AGENDA: WORKSHOP ON PM SIMULATION AND PROCESSES EVALUATION

June 27, 2006

8:00 – 8:30 Welcome and Review of Workshop Objectives
8:30 – 8:45 Questions

Lessons from Past Performance Evaluation Activities (Plenary Session)

8:45 – 9:30 Lessons from AEROCOM – Stefan Kinne
9:30 – 10:15 Lessons from ICARTT – Stuart McKeen
10:15 – 10:30 Break
10:30 – 11:30 Representing aerosol processes in models – Now and in the Future – Steve Ghan
11:30 – 12:15 Status and progress in PM forecasting – Rohit Mathur
12:15 – 1:30 Lunch

Revisiting Objectives and Charge to Breakout Sessions (Plenary Session)

1:30 – 2:00 Objectives, Charge and Organizational Issues

Breakout

2:00 – 3:00 Breakout Sessions – everyone deals with measurements
   1. New Particle Formation and Chemistry – Peter McMurry and Richard Kamens
   2. Aerosol/Cloud Interactions & Wet Deposition – Tom Ackerman
   3. PM Forecasting – Rohit Mathur
   4. Emissions, Transport and Dry Deposition – Chris Doran and Tami Bond

3:00 – 3:30 Break

3:30 – 5:30 Breakout Sessions Continued

6:00 – 7:00 Reception

June 28, 2006
8:00 – 10:00  Breakout Sessions Continued

10:00 – 10:30  Break

**Breakout Session Reports and Next Steps** (Plenary Session)

10:30 – 12:30  Breakout Session Reports

12:30 – 1:30  Lunch

1:30 – 2:30  Where do we go from here? (Assign session report authors)
APPENDIX B

BREAKOUT SESSION PARTICIPANTS

New Particle Formation and Chemistry
Jeff Brook (Environment Canada)
Tom Grahame (DOE Office of Fossil Energy)
George Hidy (Envair/Aerochem)
John Jansen (Southern Company)
Richard Kamens (University of North Carolina)
Eladio Knipping (EPRI)
Sonia Kreidenweis (Colorado State University)
Qianfeng “Jeff” Li (University of North Carolina)
Bob McGraw (Brookhaven National Laboratory)
Peter McMurry (University of Minnesota)
Peter Mueller (TropChem)
Ted Russell (Georgia Tech University)
Rich Scheffe (EPA Office of Air Quality Planning and Standards)
Ivar Tombach (Consultant)
Ashley Williamson (DOE Office of Science)

Aerosol/Cloud Interactions and Wet Deposition
Tom Ackerman (Pacific Northwest National Laboratory/University of Washington)
Mary Barth (National Center for Atmospheric Research)
Pete Daum (Brookhaven National Laboratory)
Steve Ghan (Pacific Northwest National Laboratory)
Wanmin Gong (Environment Canada)
Ralph Kahn (NASA Jet Propulsion Laboratory)
Stefan Kinne (Max-Planck-Institut für Meteorologie)
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Markus Petters (Colorado State University)
Ashley Williamson (DOE Office of Science)

Emissions, Transport, and Dry Deposition
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Tami Bond (University of Illinois)
Jim Boylan (Georgia Department of Natural Resources)
Chuck Brock (NOAA Earth System Research Laboratory)
Jeff Brook (Environment Canada)
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Ralph Kahn (NASA Jet Propulsion Laboratory)
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Serena Chung (NOAA Earth Systems Laboratory)
Paula Davidson (NOAA National Weather Service)
Colleen Farrell (Environment Canada)
Jerome Fast (Pacific Northwest National Laboratory)
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Rohit Mathur (NOAA/EPA Office of Research and Development)
Stu McKeen (NOAA Office of Oceanic and Atmospheric Research)
Richard Menard (Environment Canada)
Mireya Moya (Universidad Nacional Autónoma de México)
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