

# AEROSOL PROPERTIES AND PROCESSES

## A Path from Field and Laboratory Measurements to Global Climate Models

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The U.S. Department of Energy strategy for improving the treatment of aerosol properties and processes in global climate models involves building up from the microscale with observational validation at every step.

**T**HE CHALLENGE. Aerosol particles influence climate by modifying both the global energy balance through absorption and scattering of radiation (direct effects), and the reflectance and persistence of clouds and the development and occurrence of precipitation (indirect effects); the term “aerosol” denotes a stable, sparse suspension of microscopical or submicroscopical solid and/or liquid particles in air. Aerosol particles contribute to numerous other climatically important processes, including fertilization of land and oceans through the deposition of nitrates, iron, and other nutrients, ►

Detail of important aerosol processes. See figure 1 on page 1061 for more information.

acidification of lakes and forests through the deposition of sulfates and nitrates, and the reduction of snow and ice albedo through the deposition of black carbon. The uncertainty in the radiative forcing of Earth's radiation budget by anthropogenic aerosols, which are present mainly in the lower troposphere, greatly exceeds that of all other forcing mechanisms combined (Houghton et al. 2001); the term "forcing" denotes a secular change in the flux of an atmospheric radiation component affecting the energy balance of Earth. Quantification of the radiative forcing and of other climate influences of aerosols is therefore of high priority for relating historical climate change to increasing greenhouse gases and determining Earth's climate sensitivity. Although in situ measurements and satellite- and ground-based remote sensing provide essential information regarding aerosol loading, geographical and vertical distribution, and processes and influences, such measurements are necessarily limited in space and time and are limited in their ability to distinguish between natural and anthropogenic aerosol components. Hence, assessment of aerosol influences on climate in simulations by numerical models is essential for interpreting past climate and for projecting future changes for various emission scenarios. This assessment requires accurate representation within global climate models of the physical and chemical properties of the particles that comprise atmospheric aerosols and the processes that influence those properties.

**THE REQUIREMENT.** The climate influences of anthropogenic aerosols are very difficult to determine separately from the effects of natural aerosols using observations alone. Hence, these influences are most effectively determined from climate simulations, specifically by simulations with and without anthropogenic aerosols. Increasingly, it has become recognized that aerosols are highly interactive with other components of the climate system, for example,

influencing and being influenced by clouds and precipitation. This recognition leads to a requirement for climate models that intrinsically incorporates representation of the concentrations and properties of aerosol particles as a function of three-dimensional location and time. Such a representation, in turn, requires an understanding of the governing processes and the dependence of these processes on amounts and properties of particulate matter and gaseous precursors. Direct effects of aerosols on the energy balance of the Earth depend on the optical properties of the aerosol, specifically the extinction coefficient  $\sigma_{\text{ep}}$ , the single scattering albedo  $\omega_0$  (a fraction of the extinction coefficient resulting from scattering), and the asymmetry parameter  $g$  (the mean of the cosine of the scattering angle). Aerosol indirect effects depend on the number concentration of CCN (see appendix for expansion of acronyms) as a function of supersaturation  $s$ ,  $N_{\text{CCN}}(s)$ . These aerosol properties in principle are evaluated as integrals over the properties of the individual particles that comprise the aerosol. The single-particle properties are governed by the particle size and shape, by the properties of the different materials of which the particle is composed, and by the distribution of the materials within the particle. However, because representation of these aerosol properties in such detail is beyond the capability of present climate models, as well as those of the foreseeable future, the challenge of climate modeling is to represent this complexity within the constraint of computational resources. This requires compromises and assumptions that accurately account for the most important effects of aerosols within the constraints of practical application.

Aerosol properties that must be accurately represented include mass concentration, particle size and size-dependent composition, optical properties, solubility, and the ability to serve as nuclei of cloud particles. Key processes (Fig. 1) that must be represented include the emission of primary particles (those emitted directly into the atmosphere), such as mineral dust, sea salt, black carbon, and organic carbon; the emission of aerosol precursor gases, such as DMS, sulfur dioxide, and VOCs; the oxidation of precursor gases; new particle formation; reversible growth of particles by condensation of nonvolatile and semivolatile gases; coagulation; reversible uptake of water vapor; activation to form cloud droplets; in-cloud scavenging; aqueous-phase reactions in cloud droplets; subgrid vertical transport by clouds; cloud-drop evaporation; dry deposition to the surface; and scavenging by falling hydrometeors. Accurate representation of these processes in climate

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*The abstract for this article can be found in this issue, following the table of contents.*

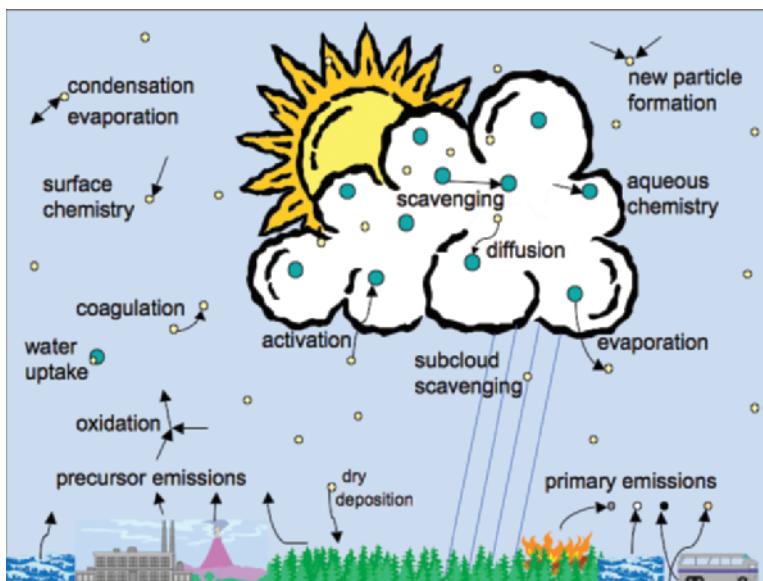
DOI:10.1175/BAMS-88-7-1059

In final form 6 February 2007  
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models rests not only on understanding the pertinent processes, but also on developing efficient ways to represent them in large-scale numerical models.

**A STRATEGY.** Because the treatment of aerosol properties and processes in the present GCMs is highly simplified, opportunities for improving the treatment abound. Four stages in a strategy to represent aerosol processes in climate models and examine aerosol influences on climate are summarized in Table 1. Stage 1 focuses on improving understanding of isolated processes. Stage 2 develops and evaluates modules representing these processes. Stage 3 evaluates the interaction of those modules in integrated aerosol models. Once validated, the modules are incorporated within global climate models in stage 4 and are again evaluated. At the end of this process the models are available to examine the sensitivity of climate to different emissions scenarios.

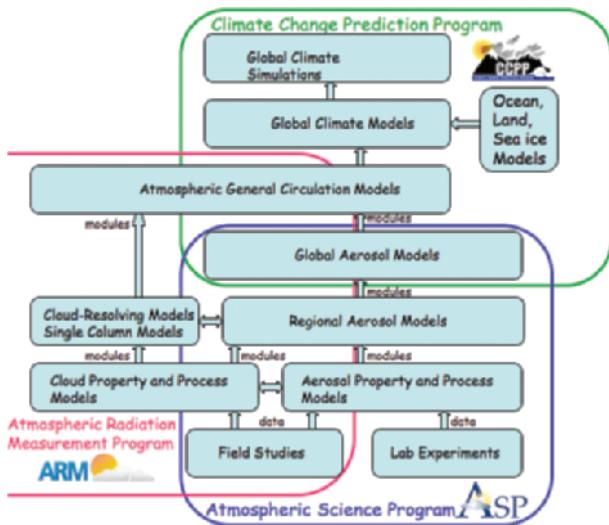
This strategy is illustrated graphically in Fig. 2. Field and laboratory studies provide the measurements on which the strategy is founded. These measurements are used to develop and evaluate models of individual processes and properties of aerosols and clouds (necessary for understanding aerosol indirect effects). The resulting suite of process and property models are then integrated and evaluated in single-column,



**FIG. 1.** Important aerosol processes that influence climate and must be accurately represented in future generations of climate models. Aerosol particles are directly emitted as primary particles and are formed secondarily by oxidation of emitted gaseous precursors. The formation of low-volatility materials in this way results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does not result in new particle formation. Particles age by surface chemistry and coagulation as well as by condensation. With increasing relative humidity particles may accrete water vapor by deliquescence and further hygroscopic growth; with decreasing relative humidity water is lost and ultimately particles may effloresce to the dry state. The uptake of water increases particle size, affecting also the particle optical properties. During cloud formation some fraction of aerosol particles serve as cloud condensation nuclei, by becoming activated, that is, overcoming a free-energy barrier to form cloud droplets. Within clouds interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. If cloud droplets evaporate the particles are resuspended, but if the cloud precipitates the particles are carried below the cloud and reach the surface, unless the precipitating particles completely evaporate. Aerosol particles below precipitating cloud can also be removed from the atmosphere by impaction by precipitating drops and by dry deposition to the surface.

**TABLE 1.** Stages of research and model development necessary to examine aerosol influences on climate.

Stage	Activity	Outcome
1)	Conduct process research: field and laboratory studies	Improved understanding of processes
2)	Develop O-D models (modules) representing processes; comparison with process research studies	Modules: model-based representation of understanding
3)	Incorporate modules describing aerosol processes in regional to global aerosol models; production runs; assessment of accuracy of aerosol models	Evaluated aerosol model incorporating processes
4)	Incorporate representation of aerosol processes in climate model; production runs; comparison with observations	Climate relevant runs; assessment of skill of climate model against present and/or prior climate



**FIG. 2.** Department of Energy's strategy for improving the treatment of aerosol properties and processes in global climate models. Field studies and laboratory studies provide the foundation. Aerosol property and process models focus on small numbers of aerosol properties or processes. Regional aerosol models represent important aerosol properties and processes by integrating a suite of property and process models for a limited domain on time scales of days. Global aerosol models are like regional aerosol models, but with a coarser resolution, a global domain, and multiple years of simulation; aerosol processes may or may not feed back on the meteorology. Global atmospheric general circulation models are like global aerosol models, but simulate the meteorology as well as the aerosol. GCMs are composed of atmosphere models coupled to ocean, land, and sea ice models and are typically run from multiple decades to centuries.

cloud-resolving, and regional-scale aerosol models. Once validated for a variety of conditions, the aerosol modules are applied to global models; here, issues of scale need to be addressed. After a global evaluation of the aerosol properties and processes, the models are coupled to models of the land surface, ocean, and sea ice, and the full climate model is applied to climate change simulations.

Many elements of this strategy have been advocated previously. Penner et al. (1994) and Seinfeld et al. (1996) focused on refining estimates of direct and indirect effects of all important aerosol types, calling for closure studies to evaluate the treatment of specific aerosol processes and the evaluation of integrated aerosol models using a combination of surface and aircraft observations and satellite remote sensing. The U.S. Department of Energy (DOE 2001) proposed a Tropospheric Aerosol Program that combined laboratory studies, surface and aircraft measurements, and modeling to improve the understanding and representation of aerosol formation, transformation, properties, and removal. Ramanathan et al. (2002) proposed a National Aerosol–Climate Interactions Program that would use new aircraft and satellite measurements to improve estimates of primary emissions, aerosol effects on clouds and precipitation, and direct and indirect effects of aerosols, with an emphasis on black carbon. In the strategic plan for the U.S. Climate Change Science Program, Mahoney et al. (2003) called for expanded laboratory, surface, airborne, and satellite studies to better characterize aerosol properties and processes and to evaluate and improve their representation in integrated aerosol models and climate models. Diner et al. (2004) and Ackerman et al. (2004) proposed PARAGON, which would integrate data from a variety of aero-

sol measurement platforms into a global modeling framework.

Elements of this strategy have been supported by several U.S. agencies. NASA has deployed a suite of aerosol remote sensing instruments (SAGE, TOMS, MODIS, MISR, OMI, CALIOP) on multiple satellite platforms (*Aqua*, *Terra*, CALIPSO), and NOAA has provided aerosol retrievals from its AVHRR satellite instruments. NASA, NOAA, NSF, and DOE have conducted several large-scale field projects, and notable among them are ACE-1, in the vicinity of Tasmania in 1995 (Bates et al. 1998), ACE-2, in the eastern North Atlantic in 1997 (Raes et al. 2000), and ACE-Asia, in eastern Asia and the western North Pacific in 2001 (Huebert et al. 2003); SCAR-B (Kaufman et al. 1998); TARFOX, over the western North Atlantic in 1996 (Russell et al. 1999); INDOEX in 1999 (Ramanathan et al. 2001); TRACE-P in 2001 (Jacob et al. 2003); ICARTT, in northeastern North America and over the North Atlantic in 2004; and the recent MILAGRO project in and downwind of Mexico City, Mexico, in 2006. Projects such as these have provided in situ measurements and remote sensing retrievals to characterize aerosol properties, improve understanding of aerosol processes, and evaluate aerosol process models and integrated models. Several of these projects have been greatly augmented by international collaboration coordinated through programs such as the WCRP, IGBP, and IGAC. Model evaluations have been coordinated by the IPCC (Penner et al. 2001, 2002) and the Aero-Com project (Kinne et al. 2006).

In parallel with the above work, focusing mainly on climate-related influences of aerosols, much work has focused on the influences of aerosols on air quality. Much of this work is directly pertinent to the

climate influences of aerosols, in the characterization of both aerosol properties and evolution, and more specifically in characterization of aerosol influences on visibility, which is closely related to aerosol direct effects on climate. In the United States much of this work is supported by the EPA and IMPROVE (Malm et al. 2004). Important contributions from the EPA include laboratory studies of gas-to-particle conversion, the treatment of changes in particle size distribution, estimation of emissions of particulate matter and gaseous precursors, and development of models of transport and transformation of aerosols pertinent to their influence on air quality. Much of this model development is directly applicable to climate models.

The U.S. Department of Energy supports three closely linked research programs that contribute to the improvement of the representation of aerosol properties and processes in GCMs: the ASP, ARM, and CCPP. As indicated in Fig. 2, each program plays an essential role in the DOE strategy to improve the accuracy and predictive capability of large-scale climate models. ASP and ARM provide field and laboratory measurements that guide the development of models of specific aerosol properties and processes. The ASP has an explicit charge to improve the understanding of chemical, microphysical, optical, and cloud-nucleating properties of aerosol particles and of the processes that control those properties, and to develop numerical models of these processes that are suitable for inclusion in large-scale models. ARM has a broad mission of understanding and quantifying the atmospheric processes and properties that influence radiation, including both direct and indirect aerosol effects, and of developing and evaluating process models. The CCPP supports the application of process models as modules in GCMs so that the influence of anthropogenic aerosol on climate can be quantified in the context of climate change over the industrial period.

### **GENERATIONS OF CLIMATE MODELS.**

Climate model studies over the past 10 yr have shown that aerosol forcing must be represented in climate models in order for these models to accurately represent temperature change over the industrial period. Early approaches to such representation simply adjusted surface albedo to crudely account for the enhancement of planetary albedo resulting from tropospheric aerosol (Mitchell et al. 1995; Boer et al. 2000; Delworth and Knutson 2000). Subsequently, many models have treated direct effects of aerosol particles using three-dimensional distributions

from “offline” simulations with chemical transport models (e.g., Meehl et al. 2003; Delworth et al. 2006; Collins et al. 2006; Meehl et al. 2006). Some GCMs have actively represented certain aerosol components (usually sulfate) and processes (e.g., Stott et al. 2000; Tett et al. 2002; Hansen et al. 2005; Schmidt et al. 2006). It is now recognized that accurate representation of aerosol influences must take into account phenomena such as correlations of aerosol loading with meteorological variables and the influence of aerosol on clouds and precipitation, and hence that aerosol loading and those properties must be represented actively and interactively in climate models. It is this recognition that is driving much of the current effort to actively represent aerosol processes, properties, and effects in climate models.

Much of the practice of climate modeling has become tied to the production schedules for periodic international assessments of the science of climate change by the IPCC. AR4, to be published in 2007, will be based on simulations completed in 2004 using models that were frozen in 2003. Because these assessments are prepared roughly every 6 yr, it may be anticipated that AR5 will be based on climate simulations completed in about 2010 using models frozen about 2009. Likewise, AR6 will be based on models frozen roughly in 2015. Each IPCC assessment report is therefore based on a successive generation of climate models, with the timing of the model development and application to some extent being governed by the timing of the assessment process. Although climate modeling predates IPCC assessments by many years, recent generations of climate models can be identified by the IPCC report to which they contribute. Thus, we might say that the fourth generation of climate models contributed to the preparation of AR4, and so on.

Inevitably, there is a lag from gaining understanding of processes to representing that understanding in climate models, and this holds true for aerosol processes. Thus, there is a lag from understanding aerosol processes, as represented in zero-dimensional models (box models), to representing this understanding in integrated aerosol models; and there is a further lag in representing this understanding in GCMs that are used in IPCC assessments, which can be as long as a full IPCC cycle or more. There is a further, similar lag of a full IPCC cycle between the representation of aerosol (or other) processes in GCMs and the use of the results of that generation of models in scenario assessments.

The treatment of aerosols in future generations of climate models will rest on an improved understanding of the processes that control aerosol

properties and their evolution as gained in laboratory and field studies carried out in research supported by the DOE ASP and ARM programs, as well as by studies conducted with the support of other U.S. and international agencies. These studies in turn rest on developments in instruments and measurement capabilities to characterize aerosol properties.

Future enhancements will rest also on advances in representing the pertinent aerosol processes in models. Such model development progresses through

a hierarchy of approaches. Initially, a subset of aerosol processes is represented in zero dimensions or, for cloud processes, in one (vertical) dimension. These representations are then commonly incorporated into models that are driven by analyzed meteorological data to allow the representation of these processes to be evaluated by comparison with observations at specific locations and times. These process models subsequently become incorporated as modules in integrated aerosol models that are further tested and evaluated. Ultimately these representations become incorporated in climate models. Because climate models must be run for much greater times (centuries) than is typical for aerosol models, the ability to include aerosol processes will require accurate and efficient representation of aerosol processes and will rest as well on advances in computational hardware and architecture. It is thus clear that representing aerosol influences on climate, at present and for the foreseeable future, will require a suite of approaches. Examples of these approaches are given in the sidebar.

The incorporation of these approaches in present and future generations of climate models is outlined below, together with examples of research needed to support the development and testing of these new approaches. Aerosol properties that need to be represented in climate models to simulate aerosol influences on climate are presented in Table 2, together with approaches to model these properties in each model generation. The approaches representing aerosol processes are given in Table 3.

## MULTIPLE APPROACHES

**M**ultiple approaches for multiple applications. Aerosol models

- may examine individual processes or interactions of multiple processes;
- may serve as test beds for new representations of aerosol processes in climate models;
- may be driven by analyzed meteorological data (offline), neglecting aerosol influences on meteorology and climate;
- may produce aerosol distributions that are prescribed in climate simulations;
- may include representation of aerosol influences on meteorology and climate (aerosol feedback);
- may include explicitly resolved or parameterized clouds;
- may examine the consequences of alternative emissions scenarios.

**TABLE 2. Treatment of aerosol properties in fourth-, fifth-, and sixth-generation climate models.**

Treatment			
Property	Fourth generation	Fifth generation	Sixth generation
<b>Mass concentration and composition</b>	Sulfate interactive (online) with climate model dust, sea salt, hydrophilic and hydrophobic OC and BC prescribed from offline aerosol model simulations	Interactive sulfate, dust, sea salt, hydrophilic and hydrophobic OC, BC, nitrate, ammonia	As for fifth generation
<b>Size distribution</b>	Prescribed for each aerosol type except dust; multiple sizes for dust and perhaps sea salt	Variable for each aerosol type (modal)	Variable (sectional, QMOM, or piecewise log-normal)
<b>Mixing state</b>	External	Internal and external mixtures	As for fifth generation
<b>Refractive index</b>	Prescribed	Volume average	Volume average treatment of inclusions
<b>Optical properties</b>	Prescribed, for each aerosol type; function of RH	Parameterized in terms of bulk refractive index and wet effective radius	As for fifth generation
<b>Hygroscopicity</b>	Prescribed	Volume average	Thermodynamic equilibrium
<b>CCN spectrum</b>	Empirical	Köhler theory for external mixtures of internally mixed inorganic and soluble organic salts	As for fifth generation, plus weakly soluble organics and surfactants

**TABLE 3. Treatment of aerosol processes in fourth-, fifth-, and sixth-generation climate models.**

<b>Treatment</b>			
<b>Process</b>	<b>Fourth generation</b>	<b>Fifth generation</b>	<b>Sixth generation</b>
<b>Primary emissions</b>	Prescribed for all species	Sea salt, dust emissions depend on wind speed in host model; also on soil moisture for dust	Sea salt, dust emissions depend on wind speed in host model; also on soil moisture for dust; emission from fires depends on area burned, fuel load, burning efficiency, and emissions factors for each species.
<b>Precursor emissions</b>	Prescribed for all precursor gases	DMS emissions depend on wind speed in host model	DMS emissions depend on wind speed and ocean chemistry in host model
<b>Oxidation of precursors</b>	Reaction of SO <sub>2</sub> , DMS with prescribed oxidant concentrations Instantaneous oxidation of VOC with prescribed yield	Reaction of all precursors with oxidants whose concentrations are calculated in the model Multiple hydrocarbon groups Dependence of yield on total organic aerosol	New hydrocarbon treatment
<b>New particle formation</b>	Neglected	Binary homogeneous nucleation	Ternary nucleation ammonia, organics
<b>Condensation of oxidized precursor gases</b>	Instantaneous condensation	Size-dependent mass transfer treatment	As for fifth generation
<b>Coagulation</b>	Neglected	Brownian coagulation within and between modes	As for fifth generation
<b>Evolution of hygroscopicity of BC, OC and dust (“aging”)</b>	Prescribed hydrophobic-to-hydrophilic conversion time for BC and OC, neglected for dust	Separate treatment of coagulation and condensation effects for BC and OC, condensation effects for dust	Separate treatment of coagulation, condensation, surface chemistry effects
<b>Water uptake</b>	For external mixtures only; no hysteresis in most models; equilibrium	Internal and external; hysteresis treated	Kinetic effects
<b>Aerosol activation</b>	Prescribed number activated	Maximum supersaturation and number activated parameterized in terms of updraft velocity and external mixtures of internally mixed inorganic and soluble organic salts.	Kinetic effects; activation to ice crystal
<b>Aqueous phase reactions in clouds</b>	Bulk treatment (same for all cloud droplets) pH dependence for prescribed ratio of ammonia/sulfate; poorly constrained cumulus cloud fraction	Bulk treatment (same for all cloud droplets); pH dependence for variable ratio of ammonia/sulfate; physically based stratiform and cumulus cloud fraction	Size-dependent cloud drop composition; reactions in hydrated aerosol
<b>Convective transport and removal</b>	Cumulus parameterization Poorly constrained precipitating area	Cumulus parameterization with physically based precipitating area	Statistics from embedded cloud models
<b>In-cloud scavenging</b>	Autoconversion and precipitation rate independent of aerosol Cloud-borne aerosol equals activated aerosol	Autoconversion and precipitation rates depend on aerosol Influence of collision/coalescence on cloud drop number concentration and cloud-borne aerosol; subgrid variability in autoconversion	Statistics from embedded cloud models with microphysics dependent on aerosol scavenging by ice crystals
<b>Subcloud scavenging</b>	Prescribed scavenging efficiency	Size-dependent collection efficiency	Aerosol from evaporated raindrops; precipitation statistics from embedded cloud models

## AEROSOLS IN FOURTH-GENERATION CLIMATE MODELS.

The summaries of treatments of aerosol properties and processes in fourth-generation climate models in Tables 2 and 3 are based largely on the three U.S. climate models that participated in AR4: the National Center for Atmospheric Research CCSM3 (Collins et al. 2006; Barth et al. 2000), the Geophysical Fluid Dynamics Laboratory CM2.1 (Delworth et al. 2006; Ginoux et al. 2006; Tie et al. 2005; Horowitz 2006), and the Goddard Institute for Space Studies Model-E (Hansen et al. 2005; Schmidt et al. 2006). Although the treatment of aerosols differs somewhat from model to model, for most aerosol properties and processes the differences among models within a given generation of climate models are considerably less than the differences between successive generations.

Most of the important substances that comprise the condensed phase of the aerosol [sulfates, hydrophilic (soluble) and hydrophobic (insoluble) OC, hydrophilic and hydrophobic BC, mineral dust, and sea salt] are treated in fourth-generation climate models. Sulfate chemistry is embedded in the climate models, but concentrations of most other aerosol species are prescribed from offline simulations with global aerosol models. Inevitably, this approach for the other species is a compromise, because it does not account for correlations between climatic variables affecting aerosol influences (such as cloudiness) and the concentrations of these species, and further because it does not account for aerosol influences on, for example, the hydrological cycle, which can further influence the concentrations and properties of aerosol particles. The size distribution is prescribed for each aerosol species, so that only the mass concentration of each species is simulated. The aerosol is assumed to be externally mixed, that is, consisting of particles composed wholly of one or another substance, as opposed to individual particles consisting of a mixture of multiple substances. This latter assumption greatly simplifies the representation of aerosol optical properties (absorption coefficient, scattering coefficient, and asymmetry parameter) and their dependence on relative humidity. Optical properties of aerosol particles depend to first order on the refractive index and particle size, and to a lesser extent on particle shape. With the assumption of a prescribed size distribution, and particle shape and composition for each aerosol type, the refractive index and the optical properties and their dependence on relative humidity can be prescribed for each aerosol type; however, this approach cannot account for variability in these properties for a given aerosol type that arises

from differences in particle size distribution, particle shape, or composition. The CCN spectrum  $N_{\text{CCN}}(s)$ , 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 treated using empirical relationships that are suitable at best only for externally mixed aerosols.

Representation of aerosol processes in fourth-generation climate models is highly simplified. Emissions of primary particulate matter and of gaseous precursors of secondary particulate matter are prescribed, rather than treated as being dependent on climate model variables such as wind speed and soil moisture. Concentrations of oxidant species OH, O<sub>3</sub>, and H<sub>2</sub>O<sub>2</sub>, responsible for the conversion of SO<sub>2</sub> and DMS to sulfates, are likewise prescribed, typically at monthly mean values, rather than being generated in the model. In most models VOCs are instantaneously oxidized with a uniform prescribed yield. New particle formation and coagulation are not treated because these processes do not affect aerosol properties when the particle size distribution is prescribed. Condensation of the oxidized precursor gases on particles is assumed to occur instantaneously. If OC and BC are treated as distinct hydrophobic and hydrophilic components, the aging from hydrophobic to hydrophilic is treated using a uniform prescribed conversion time. Aging of dust is neglected. The water content of particles is assumed to be governed by thermodynamic equilibrium with relative humidity, with hysteresis (dependence of the hydration state on the history of its environmental relative humidity) neglected in most models by assuming that particles are always hydrated at humidities above a specified efflorescence point. Aerosol activation is treated by assuming that a prescribed mass fraction of each aerosol type (typically 100% for sulfates, sea salt, and hydrophilic OC and BC, and 0% for dust and hydrophobic OC and BC) forms droplets in clouds, with empirical expressions relating cloud-drop number concentration to mass concentration. A bulk treatment of aqueous-phase chemistry is used, in which oxidation of dissolved SO<sub>2</sub> in cloud droplets is independent of droplet size but depends on droplet pH, with pH diagnosed assuming a prescribed ratio of ammonia to sulfate; cloud properties for chemistry calculations are provided by the host GCM. Vertical transport and removal of aerosol by cumulus clouds is treated using cumulus parameterizations with poorly constrained estimates of precipitation area. In- and below-cloud scavenging are treated using precipitation rates that are not influenced by the aerosol; treatment of in-

cloud scavenging assumes complete of the activated aerosol in clouds.

**AEROSOLS IN FIFTH-GENERATION CLIMATE MODELS.** The treatment of aerosol properties and processes in fifth-generation models will be much more complete than in fourth-generation models and is increasingly based on understanding of the pertinent processes. More processes will be represented, and in greater detail. With few exceptions, the advances will build on the current generation of global aerosol models (e.g., Easter et al. 2004; Tie et al. 2005; Koch et al. 2006). Because many aspects of the global aerosol models have already been evaluated in comparisons with observations (Ghan et al. 2001a,b; Easter et al. 2004; Tie et al. 2005; Kinne et al. 2006; Koch et al. 2006; Bates et al. 2006), much of the effort involved in upgrading the aerosol treatment in fifth-generation climate models will involve transferring the treatments from the global aerosol models to the global climate models. Within the DOE program this component of the work is carried out largely in the Climate Change Prediction Program. Previous work in the ARM program, which produced treatments of aerosol impacts on clouds (Ghan et al. 1997; Ovtchinnikov and Ghan 2005; Liu et al. 2005) and parameterizations of aerosol radiative properties (Ghan et al. 2001a), will have an impact on the fifth-generation models. The ASP provides field measurements and uses them to evaluate specific processes represented in the models.

Aerosol properties for the fifth generation of climate models will have numerous additional degrees of freedom. New species to be included in the models will include nitrate and ammonia/ammonium, and all aerosol species will be simulated online rather than offline, so that concentrations will vary on hourly rather than monthly time scales and will interact with the meteorology that is simulated in the model. The aerosol particle size distribution will be calculated rather than prescribed. Several different methods (modal, quadrature method of moments, and sectional) are available for representing the particle size distribution and composition and their evolution. The simplest modal method (Whitby and McMurry 1997; Easter et al. 2004; Liu et al. 2005) assumes a lognormal size distribution for each aerosol type and calculates its number and mass concentration from separate conservation equations. The QMOM (McGraw 1997; Wright et al. 2001; Yoon and McGraw 2004a,b) is more general in that it does not assume a lognormal size distribution; aerosol properties and their evolution are calculated from the moments of

the particle size distribution by Gaussian quadratures. The sectional method (Adams and Seinfeld 2002) is most general because the size distribution is explicitly represented, but it requires extensive computational resources in storing, evolving, and transporting large numbers of variables. For this reason the modal method and QMOM are most likely candidates for fifth-generation models. Representation of the aerosol mixing state will accommodate a combination of external and internal mixing so that particles forming by condensation will be capable of being internally mixed with primary particles and/or freshly nucleated particles. Two forms of hydrophobic BC and OC will be treated; one is purely hydrophobic, and the other is internally mixed with sulfates, nitrates, and ammonium. Aging of BC and OC will be expressed in terms of the condensation and coagulation rates, so that fresh BC and OC masses and numbers are transferred to the internally mixed aerosol. Although treating internal mixing can reduce the number of aerosol types, it complicates the representation of optical properties, hygroscopicity, and CCN activity because, as is the case with actual ambient aerosol particles, those properties depend on the now-variable composition. Fifth-generation models will accommodate internal mixing by using mixing rules for refractive index and hygroscopicity pertinent to particle growth with relative humidity and CCN activity. Other mixing rules may be used to represent the optical effects of the inclusion of insoluble material in particles consisting largely of water-soluble material.

Consistent with the online representation of aerosol properties, emissions of DMS, sea salt, and dust will be calculated online using the simulated winds and (for dust) surface moisture and vegetation cover. Likewise, oxidation of aerosol precursor gases will be calculated using oxidant concentrations that are generated online by a gas-phase oxidant chemistry module. VOCs will be separated into multiple classes; the yield of new particulate mass will depend on the total amount of organic aerosol (Chung and Seinfeld 2002; Tie et al. 2005). New particle formation will be introduced as a source of aerosol number, probably using a parameterization of binary nucleation of water and sulfuric acid vapor (Jaeger-Voirol and Mirabel 1989; Harrington and Kreidenweiss 1998; Vehkamäki et al. 2002). Condensation of oxidized precursor gases on existing aerosol particles will be treated using mass transfer theory, so that condensation can be distributed across multiple aerosol types. Coagulation of particles both within each type (which reduces

number and increases mean size) and between types will be represented to accurately represent particle number concentration. Dust will age as condensation of sulfuric acid and secondary organic material onto dust modes changes the bulk hygroscopicity and refractive index of each mode.

Uptake of water by particles will be represented in terms of the bulk hygroscopicity using Köhler theory, with explicit treatment of hysteresis so that dry and hydrated aerosol states are distinguished. Activation of aerosol particles to form cloud droplets will be expressed in terms of updraft velocity and the aerosol particle properties (number, size, and hygroscopicity) for all types (Abdul-Razzak and Ghan 2000; Fountoukis and Nenes 2005), so that aerosol indirect effects and the competition between aerosol types can be treated in a physically based manner. Aqueous chemistry will depend on the pH, calculated from the ratio of sulfate to ammonia, which will be allowed to vary. The cloud fraction will be determined using physically based parameterizations. Convective transport and the removal of aerosol will be improved by using new cumulus parameterizations that diagnose precipitating area and treat cloud microphysics. In-cloud scavenging will be based on the concentrations of the activated particles and will treat the dependence of precipitation development on the number activated. Below-cloud scavenging will use size-dependent collection efficiencies.

In sum, much more detailed representation of aerosol processes and properties is expected in the fifth generation of climate models than in earlier generations. Representation of each of these processes rests on improved understanding of the processes themselves and on improved ability to efficiently and accurately represent this understanding in models, both of which are expected to be greatly advanced by ongoing research.

**AEROSOLS IN SIXTH-GENERATION CLIMATE MODELS.** Although it is difficult to anticipate the treatment of aerosol processes in the sixth generation of climate models so far out into the future, it is clear that this treatment will rely on advances in understanding provided by programs such as the ASP and ARM. For example, recent work has shown that current understanding of the formation of SOA leads to substantial underestimates in the simulated concentrations of OC (Heald et al. 2005; Volkamer et al. 2006). Laboratory and field experiments will provide the foundation for a new generation of models of SOA formation. The challenge will

be to condense that understanding into process models that are simple enough to be used in global climate simulations, but at the same time provide accurate representation of the process.

It is known from field studies that current models based on binary homogeneous nucleation of sulfuric acid and water yield new particle formation rates that are often far smaller than the measured rate (Weber et al. 1999). In such cases new particle formation rates may be better explained by either ternary homogeneous nucleation of sulfuric acid, ammonia, and water (Napari et al. 2002a,b); of sulfuric acid, organic acid, and water (Jimenez et al. 2003; Zhang et al. 2004); or ion-induced nucleation of sulfuric acid and water (Lee et al. 2003; Lovejoy et al. 2004). Laboratory data for multiple precursor gases can be accurately parameterized by the nucleation theorem (McGraw and Wu 2003; McGraw 2005), but further work is needed to account for the influence of background aerosol on the nucleation rate and subsequent particle growth by condensation and coagulation (McMurry et al. 2005). These parameterizations also need to be evaluated and refined using field measurements and, depending on the outcome, incorporated into the sixth-generation models. It seems clear that treatment of subgrid variability will also be required because the nucleation rate is a highly nonlinear function of the precursor gas concentrations. For these reasons much attention needs to be paid to issues of scale.

Representing aerosol particle size distributions and size-dependent composition is essential. Because explicit representation would seem not to be computationally feasible in climate models, alternative approaches must be investigated. Although representation of the particle size distribution by the modal approach is capable of efficiently representing multiple aerosol types under many conditions (Whitby 1978), the inherent assumption of a lognormal size distribution for each mode can break down. For example, aerosol activation in cloud updrafts typically separates the size distribution into activated particles and particles too small to be activated. The resultant discontinuities in the size distributions of the activated and unactivated particles are not well approximated by lognormal functions (Hoppel et al. 1990, 1994; Zhang et al. 2002). Possible solutions to this problem are provided by the more general and more accurate sectional, QMOM, and piecewise lognormal (von Salzen 2005) treatments, with differing computational burdens. The ASP and CCPP can contribute to the comparison and testing of these and other potential approaches.

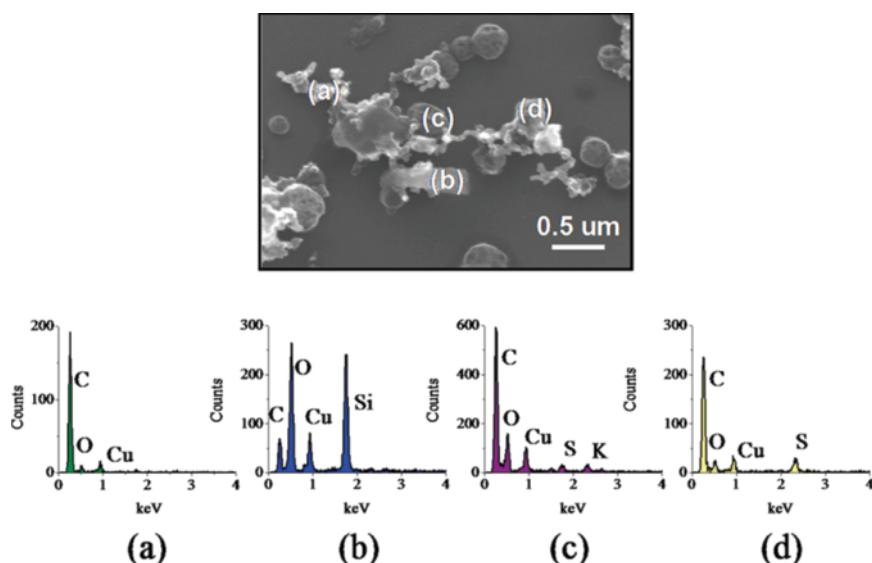
Uptake of water exerts an important and sometimes dominant influence on aerosol particle optical properties. Although the equilibrium Köhler treatment seems appropriate for submicrometer particles, this approach may not work for larger particles, for which the change in particle size may lag changes in relative humidity. Such kinetic effects might be treated by an explicit dynamic form of the Köhler theory. Although this treatment is straightforward for parcel models in which the time dependence of the ambient relative humidity is known, extending it to the Eulerian framework of climate models will require alternate approaches, such as using the turbulence kinetic energy and the vertical gradient of relative humidity.

### EVALUATION AND IMPROVEMENT.

Although process modules in current global aerosol models can be applied to climate models, these modules have not been evaluated sufficiently to allow them to be used with confidence in climate models, and many such modules need to be improved to provide the basis for the treatment of aerosols in future generations of climate models. Ultimately, improvement of model-based representation of aerosol processes and properties rests on understanding that is grounded in laboratory studies, theory, and field measurements. The approach to developing modules to describe specific aerosol processes is often a cyclical one, involving perhaps initial field observations, such as the widespread occurrence of new particle formation (Komppula et al. 2003) identification of the chemical and physical conditions under which this occurs, examination in the laboratory, identification of unmet measurement needs, and subsequent field studies to compare calculated and measured rates of new particle formation under a variety of conditions. Once such a module is developed, it must be made suitable for incorporation in large-scale aerosol models. This may require considerable effort, both in meeting computational constraints

and in accounting for issues such as subgrid variability, which can greatly affect highly nonlinear processes.

Much of the advance in understanding aerosol processes in recent years has derived from improvements in instruments for determination and characterization of aerosols and aerosol precursors. Table 4 lists some key capabilities in measurements of properties of aerosol particles and precursor gases ( $\text{H}_2\text{SO}_4$ ,  $\text{NH}_3$ , VOC) and oxidants (OH). New techniques for characterizing aerosol composition can provide information that is highly speciated, quantitative, and size resolved for ionic species, organics, and refractory materials such as dust. Methods of electron microscopy and microanalysis, which can characterize the size, morphology, composition, phase, and internal structure of single particles (Fig. 3), can be coupled with time-resolved aerosol collection on substrates to follow temporal variations of specific types of field-collected individual particles. Other techniques, such as PILS, AMS, and SPMS provide real-time measurements with a time resolution of seconds (Fig. 4). Although PILS provides little size resolution, it provides accurate concentration measurements of soluble ionic aerosol species. AMS provides size resolution of composition (except for refractory material such as BC, sea salt, and mineral dust) for particle sizes between 30 nm and 2.5  $\mu\text{m}$ . SPMS provides composition information for individual particles as small as 50 nm.



**FIG. 3.** (a), (b), (c), (d) Analysis of four different locations on a single soot particle collected in Mexico City in 2003 shows complex submicrometer morphology and varying elemental composition as determined by SEM-EDX analysis. During the residence time of the particle in the atmosphere the carbon has become internally mixed with sulfur, silicon, and potassium; copper is an artifact of the substrate grid. [From Johnson et al. (2005).]

**TABLE 4. Advanced techniques for characterization of properties of aerosols, precursor gases, and other key quantities.**

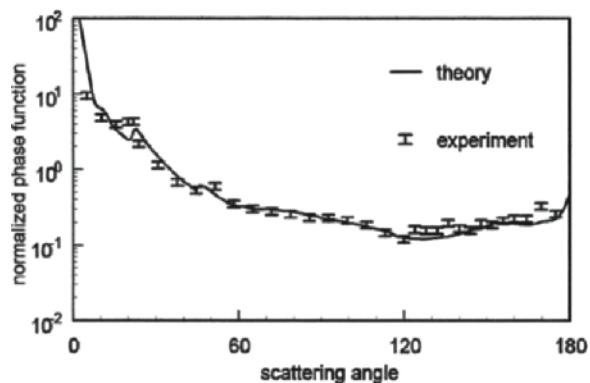
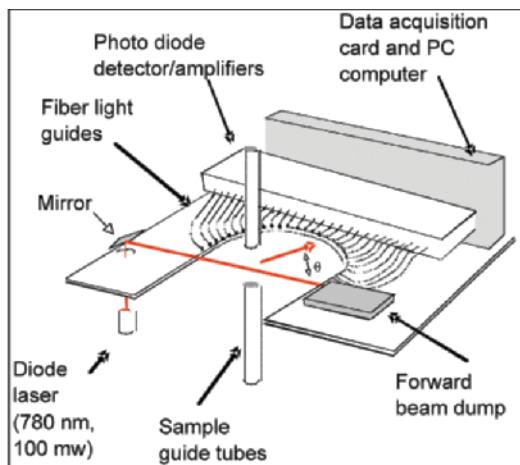
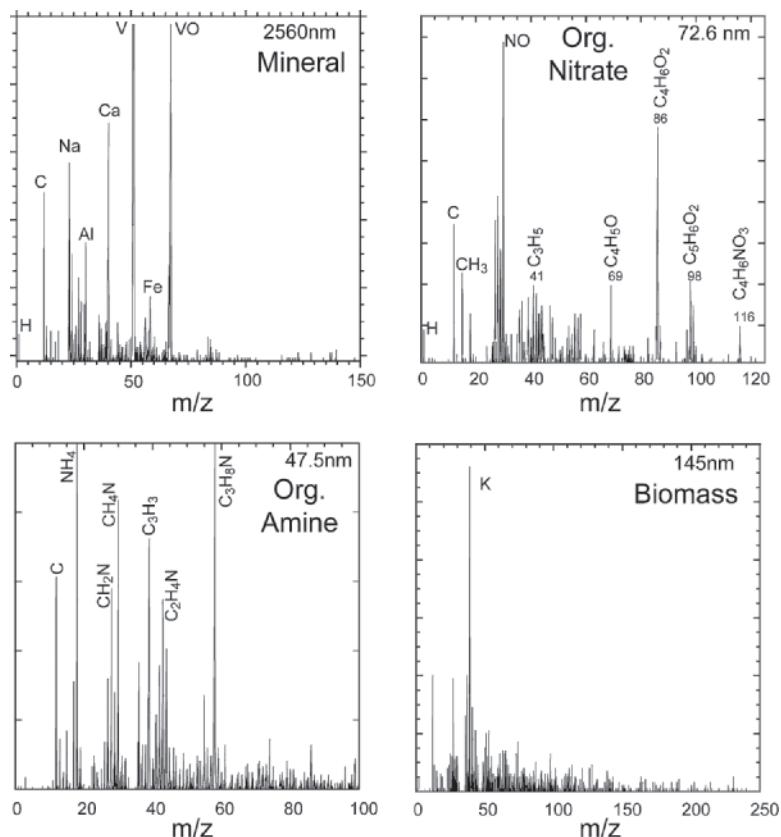
Property	Instrument/technique: reference
H <sub>2</sub> SO <sub>4</sub> gas concentration	CIMS: Eisele and Berresheim (1992)
NH <sub>3</sub> gas concentration	CIMS: Nowak et al. (2002), Fehsenfeld et al. (2002)
OH gas concentration	Laser-induced fluorescence: Martinez et al. (2003)
VOC gas concentration	PTR-ITMS: Prazeller et al. (2003)
Concentration of ionic species in aerosols	PILS: Weber et al. (2001) + ion chromatography
Size-resolved composition	AMS (thermal vaporization mass spectrometry): Jayne et al. (2000)
Single-particle composition	SPMS (laser desorption mass spectrometry): Thomson et al. (2000), Su et al. (2004), Zelenyuk and Imre (2005)
Single-particles morphology, composition, phase, internal structure, hygroscopicity	TEM, SEM, EDX microanalysis: Buseck and Anderson (1998), Fletcher et al. (2001), De Bock and Van Grieken (1999), Laskin et al. (2006), Twohy et al. (2005); environmental SEM and TEM: Ebert et al. (2002), Laskin et al. (2006), Wise et al. (2005)
BC concentration	Thermal-optical: Chow et al. (1993)
Dust concentration	PIXE-Johansson et al. (1995)
Ultrafine number concentration	CNC: Stolzenburg and McMurry (1991)
Particle size distribution	SMPS: Wang and Flagan (1990); nano-SMPS: Woo et al. (2001); DMA: Knutson and Whitby (1975); PCASP: Liu and Daum (2000); APS: Stanier et al. (2004)
Mixing state	H-TDMA: McMurry and Stolzenburg (1989); SPMS, SEM, TEM, scanning transmission X-ray microscopy
Activated particle size distribution and composition	CVI: Noone et al. (1988) + CCN + DMA + PCASP + AMS +TEM: Twohy et al. (2001, 2005)
Angular distribution of light scattering, size distribution, and real and imaginary components of refractive index	Polar nephelometer: Grams et al. (1974), Barkey et al. (2002)
Absorption coefficient	Aethelometer: Hansen et al. (1984); photoacoustic: Arnott et al. (1999, 2006); cavity ring-down extinction–nephelometer scattering: Zalicki and Zare (1995), Strawa et al. (2003, 2006); multiangle absorption photometry: Petzold et al. (2005)
Scattering coefficient; backscatter fraction; multiple wavelengths; multiple relative humidities	Integrating nephelometer
Extinction coefficient (multiple) wavelengths)	Raman lidar, cavity ring down, vertical derivative of optical depth measured by airborne sun photometer: Schmid et al. (2006)
Hygroscopic growth	H-TDMA: Radar and McMurry (1986), Gasparini et al. (2006b)
CCN spectrum	CCN spectrometer: Hudson (1989), Gasparini et al. (2006a)
Droplet size distribution	CAPS: Baumgardner et al. (2001)
Ground-based and satellite-based remote sensing of aerosol optical depth and column properties	AERONET: Holben et al. (1998), Dubovik and King (2000), Kaufman et al. (2002)
Ground-based and satellite-based remote sensing of vertical distribution of aerosol properties	Aerosol lidar: Goldsmith et al. (1998), Turner et al. (2001), Müller et al. (2001), Shipley et al. (1983); Satellite: Kaufman et al. (2003), Winker et al. (2004)
Updraft velocity	Gust probe: Lenschow (1986), Conant et al. (2004)

Other measurement highlights include aerosol size distribution to particle diameters as low as 3 nm, water uptake on particles of a selected size as a function of relative humidity, angular distribution of light scattering intensity (Fig. 5), and properties of particles that have been scavenged by cloud droplets (by aerodynamically separating them from surrounding

gases and unactivated particles; Noone et al. 1988; Twohy et al. 2001, 2005). Thorough reviews of the technology of in situ aerosol measurement are provided by McMurry et al. (2000, 2004) and Sullivan and Prather (2005). Further information on AMS and SPMS systems is available (online at <http://cires.colorado.edu/~jjose/ams.html>).

Each of these measurements can be used to evaluate different aspects of the treatment of aerosol properties that must be represented in large-scale aerosol models. The ASP conducts field studies designed to obtain such measurements. A key approach to the analysis of field measurements is the so-called closure experiment, which consists of a comparison of measured and modeled quantities, where the modeled quantity is determined using a process model and the measurements are other quantities as input variables. Several such closure experiments are given in Table 5. An example of such a closure study is shown in Fig. 6, which shows several comparisons of CCN measured as a function of supersaturation and calculated based on measurements of particle size distribution and composition (Gasparini et al. 2006a). Other examples of property closures include mass concentration and concentrations of individual substances, refractive index, and scattering, absorption, and extinction coefficients.

The same closure approach can also be used to examine processes, particularly those processes that dominate the relationship between properties, that can be measured. For example, under some conditions the aerosol activation process dominates the relationship between subcloud aerosol and activated aerosol. By measuring the updraft velocity and the properties of the subcloud aerosol and the aerosol activated within cloud droplets, understanding the activation process and representation of that understanding in

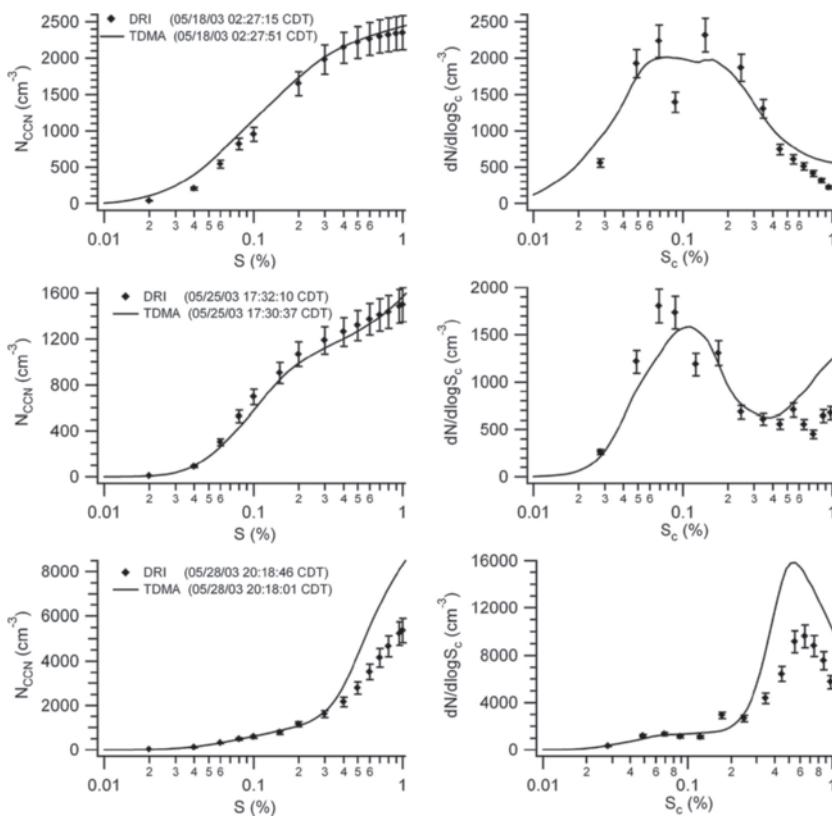


**FIG. 4 (TOP RIGHT).** Examples of mass spectra of individual atmospheric aerosol particles sampled by SPLAT (Zelenyuk and Imre 2005) during a field study in Houston, TX, August, 2000. Indicated size is vacuum aerodynamic diameter. [From Zelenyuk and Imre (2005).]

**FIG. 5 (LOWER RIGHT).** Polar nephelometer to measure angular distribution of light scattering and index of refraction of ambient aerosols. Lower panel shows measured angular distribution of light scattering intensity from laboratory-generated smoke, yielding a size distribution with a median diameter of 86 nm and variance of 0.113; unpublished results from B. Barkey (UCLA, 2006, personal communication; upper panel modified from Barkey et al. 2002).

**TABLE 5. Closure experiments on aerosol properties and processes.**

Closure experiment	Measurements	Reference
Aerosol mass concentration	Ions, BC, organic dust, total mass	Zhang et al. (2005)
Refractive index	Ions, BC, organic, mixing state, refractive index	—
Radiative absorption	BC, dust, aerosol size distribution, mixing state, absorption	—
Radiative scattering	Ions, BC, organic, dust, aerosol size distribution, relative humidity, mixing state, scattering	Clarke et al. (2002)
Radiative extinction	Ions, BC, organic, dust, aerosol size distribution, relative humidity, mixing state, extinction	Wang et al. (2002)
New particle formation	Condensation nuclei concentration, ultrafine size distribution, H <sub>2</sub> SO <sub>4</sub> , NH <sub>3</sub> , organic vapor	McMurry et al. (2005)
CCN concentration	Ions, BC, organic, dust, aerosol size distribution, CCN concentration	Cantrell et al. (2001), Gasparini et al. (2006a), Rissman et al. (2006)
Water uptake	Ions, BC, organic, dust, aerosol size distribution, humidification size factor, extinction factor	Swietlicki et al. (1999), Dick et al. (2000)
Aerosol activation/droplet nucleation	CCN spectrum, ions, BC, organic, dust, aerosol size distribution, updraft velocity, droplet number, activated aerosol	Conant et al. (2004), Meskhidze et al. (2005)
Droplet collision/coalescence	Droplet size distribution, drizzle size distribution	Wood (2005)



**FIG. 6. Comparison of (left) cumulative and (right) differential CCN spectra measured as functions of supersaturation during the ARM-ASP aerosol IOP with the DRI CCN spectrometer with spectra calculated from TDMA measurements of the size distribution of aerosol number and hygroscopicity. [From Gasparini et al. (2006).]**

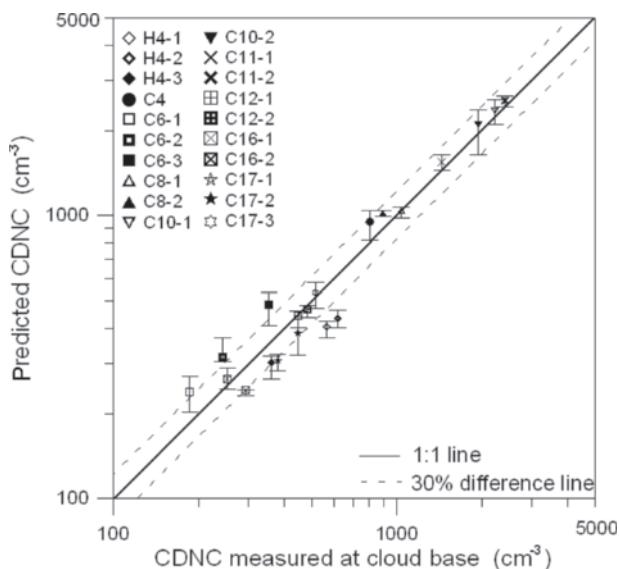
process models can be tested using the updraft velocity and subcloud properties as input to an activation model and using the activated aerosol measurements to evaluate the activation simulation. Figure 7 illustrates such an evaluation. Similar closure experiments can be applied to new particle formation, water uptake, droplet collision/coalescence, and in-cloud scavenging.

Such closure experiments provide valuable information about the cause of model shortcomings because, unlike evaluations of integrated models, differences between simulations and observations can be attributed to specific processes. In contrast, when large differences are found in comparisons of simulations by integrated models with field measurements, the differences could arise from errors in the representation of any of many different processes or boundary conditions. Closure experiments focus attention

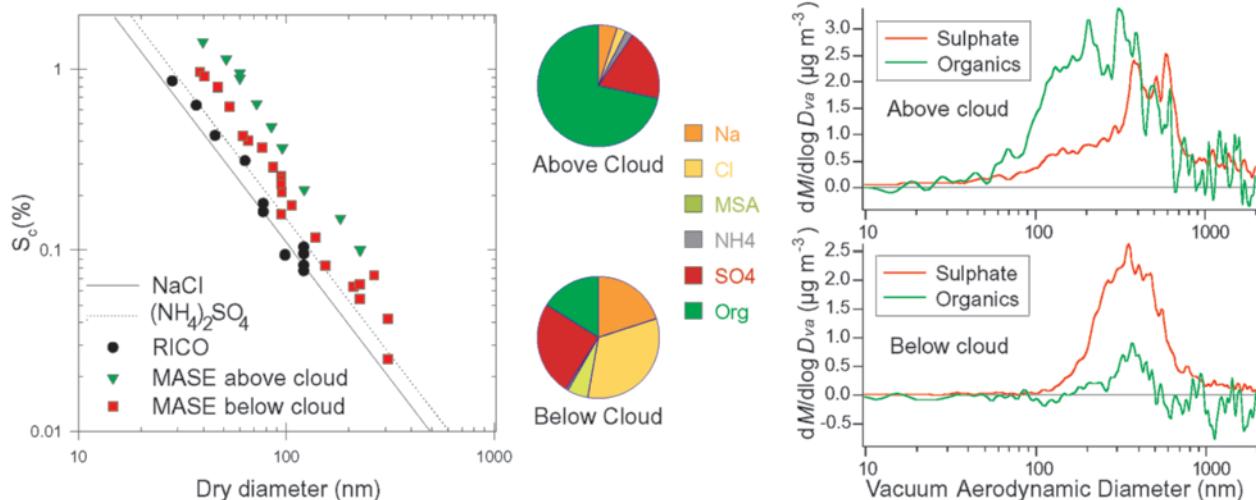
on isolated processes, so that the cause of the problem is much more evident.

Evaluations such as these serve two purposes. First, they provide valuable information about the current level of uncertainty in the treatment of individual processes and in the aerosol simulated by integrated models. Second, they identify problems that need to be addressed and suggest directions for future improvement. For example, Wood's (2005) recent evaluation of droplet collision/coalescence schemes identifies weakness in all schemes and suggests how they might be improved.

Increasingly it is becoming appreciated that advances in understanding of aerosol processes from field measurements are much more likely when multiple measurements of aerosol properties are made simultaneously. An example is shown in Fig. 8. Physical measurements of the dependence of critical supersaturation of particles as a function of their diameter show marked differences above and below a shallow stratus deck in the vicinity of Pt. Reyes, California. Activation of the above-cloud particles of the same size requires a greater supersaturation, and activation of particles at both altitudes requires



**FIG. 7.** Comparison of cloud droplet number concentration measured on 20 different aircraft flights (denoted by symbols) with that parameterized in terms of measured updraft velocity and measured aerosol size distribution during the CRYSTAL-FACE field experiment (modified from Meskhidze et al. 2005; reproduced with permission of A. Nenes).



**FIG. 8.** Example of difference in CCN activity of aerosols and relation to composition below (110–170 m) and above (400–470 m) clouds during an Atmospheric Science Program field study (MASE) conducted off the coast of California, north of San Francisco, on 25 Jul 2005. (left) Critical supersaturation as a function of particle size from measurements; also shown for comparison are measurements made in clean maritime air in the eastern Caribbean boundary layer (RICO project) and the theoretical dependence for two soluble salts, sodium chloride and ammonium sulfate (J. Hudson, Desert Research Institute, 2006, personal communication; Hudson 1989; Hudson and Da 1996). Pie charts (middle) show ionic composition measured by PILS and organic fraction inferred by difference from total volume, inferred from light scattering at low relative humidity and assumed mass scattering efficiency of  $3.3 \text{ m}^2 \text{ g}^{-1}$ ; below cloud mass concentration  $8.1 \pm 0.3 \mu\text{g m}^{-3}$ ; above cloud,  $3.8 \pm 0.2 \mu\text{g m}^{-3}$  (Y.-N. Lee, Brookhaven, National Laboratory, 2006, personal communication). (right) The distribution of sulfate and organic mass with particle size (top) above cloud and (bottom) below cloud measured by AMS (M. Alexander, Pacific Northwest National Laboratory, 2006, personal communication).

a greater supersaturation than would be expected for particles consisting entirely of inorganic salts; also shown for reference are measurements made in the eastern Caribbean, which are consistent with an inorganic salt composition. Simultaneous measurements of bulk composition, by PILS, show a greater organic fraction above clouds than below. The AMS measurements confirm that this organic fraction is much greater in the diameter range corresponding to the CCN measurements of 40–200 nm. In the absence of the chemical measurements the reasons for the differences in critical supersaturation would not be known; in the absence of the physical measurements the consequences of the differences in composition would not be known.

In some instances results of field studies might suggest a return to laboratory experiments. For example, a recent study examining production in Mexico City, Mexico, showed that amounts of secondary organic aerosol produced from anthropogenic volatile organic carbon were as much as eight-fold greater than that predicted by current models, with much of the excess secondary organic aerosol formed from first-generation oxidation products (Volkamer et al. 2006). Heald et al. (2005) had likewise found that concentrations of secondary organic aerosol measured in the free troposphere during the ACE-Asia campaign exceeded modeled concentrations by factors of 10–100. As the understanding of the gas-to-particle conversion processes represented in the models derives largely from laboratory experiments, it is clear that new laboratory experiments would need to be conducted to examine reaction systems that have not been represented in previous models. It is likely that new chemical mechanisms will have to be developed, based on such new experiments to account for the underestimated yield and to provide the basis for simplified treatment of these processes in global climate models. This new understanding would in turn need to be evaluated by further field measurements.

**REGIONAL- AND GLOBAL-SCALE AEROSOL MODELS.** Once modules treating a particular aerosol property or process have been developed and evaluated using laboratory and field measurements, the next challenge is to apply them in regional and global models. Regional models provide an important test bed for evaluating an integrated package of process modules by comparing simulations with field measurements. Much of the research in the DOE ASP is focused on this activity. A second important staging

ground for such modules is global aerosol models, which are run at grid resolutions comparable to that of global climate models (typically 100 km) but for only a few years rather than for hundreds of years. Such short simulations provide a test bed for evaluating aerosol packages at the grid resolution of global climate models without the high cost of running climate simulations to equilibrium. Applying aerosol packages to simulations at 100-km grid resolution introduces issues of scale, because aerosol packages that have been developed in a regional modeling framework with grid resolutions of 1–10 km may not perform well at grid resolutions of 100 km. Subgrid variability in processes at scales between 10 and 100 km may need to be represented if the package is to perform well at 100-km grid resolution. This challenge can be addressed in a regional modeling framework, but the limited spatial degrees of freedom in a regional simulation at 100-km grid resolution may not provide a rigorous test of the treatment of the influence of scale. Global aerosol models can be evaluated against the rich set of global satellite retrievals of a variety of gas, aerosol, and cloud properties (Schoeberl et al. 2006; Kaufman et al. 2003; Bellouin et al. 2005; Sekiguchi et al. 2003; Spinhirne et al. 2005). Although intensive field experiments, regular surface-based in situ measurements at NOAA Climate Monitoring and Diagnostics Laboratory stations, twice-weekly in situ measurements above the ARM Southern Great Plains site (Andrews et al. 2004), and surface-based aerosol retrievals by the ARM Raman lidar (Ferrare et al. 2006), AERONET (Holben et al. 1998), MPLNET (Welton et al. 2002), and EARLINET (Bösenberg et al. 2003; Mattis et al. 2004) provide limited horizontal sampling, they also provide accuracy and vertical resolution lacking in satellite retrievals. These datasets have been used successfully in evaluations of global aerosol model simulations of the spatial and temporal distributions of aerosol particle mass concentration, composition, size distribution, and optical properties (Jacobson 2001; Kinne et al. 2003; Easter et al. 2004; Liu et al. 2005; Heald et al. 2005; Tie et al. 2005; Kinne et al. 2006; Koch et al. 2006; Bates et al. 2006). Retrievals from the recently launched CloudSat (Stephens et al. 2002) and CALIOP (Winker et al. 2004) instruments are systematically providing vertical profiles from space of clouds (by radar; information online at [www.cloudsat.cira.colostate.edu/](http://www.cloudsat.cira.colostate.edu/)) and of aerosols (by lidar; information online at [www-calipso.larc.nasa.gov/products/lidar/](http://www-calipso.larc.nasa.gov/products/lidar/)), literally adding another dimension to the evaluation of aerosol models.

## **APPLICATION IN GLOBAL CLIMATE**

**MODELS.** Once aerosol packages have been evaluated in global aerosol models they are ready for incorporation into global climate models. Such incorporation can be straightforward if the packages have been designed from the outset for ultimate application to global climate models, and even more so if the global aerosol model that served as the staging ground is the atmospheric component of a global climate model. However, further work and testing is required if, for example, the climate model resolution differs from that of the aerosol model, or if the treatment of clouds in the global aerosol model differs substantially from the treatment in the climate model. Such differences can have profound impacts on the spatial distributions of aerosol simulated in the global climate model and on the resulting climate influences. Attention must be devoted also to ensure that the global energy balance is maintained. For these reasons, several climate modeling groups are using the atmospheric component of their climate model as the staging ground for the development and evaluation of new aerosol packages.

**INTEGRATION.** Development of new aerosol packages for global climate models involves many steps, from laboratory and field measurements to process modeling, evaluation, and refinement; integrated modeling evaluation and refinement; and scaling to global models, evaluation, and again refinement. Laboratory and field measurements provide the foundation upon which understanding of aerosol properties and processes is based. Process models provide test beds for evaluating both the full expression of that understanding in detailed models and for simpler representations suitable for application to global models. Integrated aerosol models provide test beds for treatment of the full set of processes that control aerosol properties. Global aerosol models provide the setting for evaluating treatments of the subgrid variability in aerosol processes for global conditions that can be sampled from instruments on satellite platforms. Global climate models are the ultimate destination of aerosol process and property modules. Although the development process from laboratory and field measurements to regional and global aerosol models, and ultimately to global climate models, requires much effort, this process is essential to the accurate determination of the climate influences of atmospheric aerosols. Integration of all these components is essential to ensure that the understanding of processes that is developed in labo-

ratory studies and field measurements is accurately and effectively represented in successive generations of climate models.

**CLOSING REMARKS.** Global climate models are essential tools for understanding climate change and for the development of policy regarding future emissions of greenhouse gases, primary aerosol particles, and aerosol precursor gases. As such, these models must include accurate representation of radiative forcing and other influences of anthropogenic tropospheric aerosols that are significant contributors to climate change over the industrial period. Unlike most greenhouse gases, tropospheric aerosol particles are quite dynamic, undergoing transformations that modify their properties that affect atmospheric radiation, clouds, and the hydrological cycle. The concentrations and properties of these aerosol particles are quite variable spatially and temporally in large part because of their interactions with other elements of the climate system. These interactions go in both directions— aerosols influencing other climatic variables and other climatic variables influencing the concentrations and properties of aerosol particles. For these reasons it is increasingly becoming recognized that accurate determination of aerosol influences on climate requires actively modeling the loading and properties of aerosols in climate models, rather than calculating these quantities in aerosol models, offline from the climate model. Here a sequential approach has been outlined whereby increasingly detailed representation of aerosol properties and processes will be incorporated into future generations of climate models. Successful implementation of this approach will rely on advances in understanding these processes in laboratory and field studies, and advances in representing these processes in aerosol models from regional to global scales. This strategy is being implemented by the several programs within the Department of Energy's Climate Change Research Division, and substantial advances in understanding and quantification of these phenomena can be expected over the next several generations of climate models.

**ACKNOWLEDGMENTS.** This work was supported primarily by the U.S. Department of Energy's Atmospheric Science Program, with partial support from the DOE Atmospheric Radiation Measurement program and the DOE Climate Change Prediction Program, all of which are components of the DOE Climate Change Research Division. Helpful comments were provided by Rickey Petty,

Anjali Bamzai, Wanda Ferrell, David Bader, and Warren Wiscombe. We also thank William Collins, Richard Easter, Richard Ferrare, Paul Ginoux, Chris Hostetler, Alexander Laskin, Yin-Nan Lee, Athanasios Nenes, Peter McMurry, and Gavin Schmidt for comments and suggestions on the manuscript. Jeffrey Rosenfeld, Anne Thompson, and two anonymous reviewers also provided helpful

comments during the review process. Mikaela Alexander, Brian Barkey, Roberto Gasparini, James Hudson, Kirsten Johnson, Yin-Nan Lee, and Athanasios Nenes kindly provided figures. PNNL is operated for the DOE by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830; BNL is operated for DOE by Brookhaven Science Associates, LLC under Contract DE-AC02-98CH10886.

**APPENDIX.** Acronyms and (for some programs and experiments) Web sites.

ACE	Aerosol Characterization Experiment
AERONET	Aerosol Robotic Network (online at <a href="http://www.nas.nasa.gov/Groups/WAN/aeronet/aeronet.html">www.nas.nasa.gov/Groups/WAN/aeronet/aeronet.html</a> )
AMS	Aerosol mass spectrometry
ARM	Atmospheric Radiation Measurement (online at <a href="http://www.arm.gov/">www.arm.gov/</a> )
APS	Aerodynamic particle sizer
AR $n$	$n$ th assessment report of IPCC Working Group 1
ASP	Atmospheric Science Program (online at <a href="http://www.asp.bnl.gov/">www.asp.bnl.gov/</a> )
AVHRR	Advanced Very-High Resolution Radiometer
BC	Black carbon
CALIOP	Cloud–Aerosol Lidar with Orthogonal Polarization (online at <a href="http://www-calipso.larc.nasa.gov/products/lidar">www-calipso.larc.nasa.gov/products/lidar</a> )
CALIPSO	Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observation
CAPS	Cloud Aerosol and Precipitation Spectrometer
CCN	Cloud condensation nuclei
CCPP	Climate Change Prediction Program (online at <a href="http://ccpp.llnl.gov/">http://ccpp.llnl.gov/</a> )
CCSM	Community Climate System Model (online at <a href="http://www.cesm.ucar.edu/index.html">www.cesm.ucar.edu/index.html</a> )
CIMS	Chemical ionization mass spectrometry
CloudSat	A polar-orbiting 94-GHz nadir-looking cloud radar (online at <a href="http://cloudsat.atmos.colostate.edu/">cloudsat.atmos.colostate.edu/</a> )
CM2.1	Coupled Model version 2.1
CNC	Condensation nuclei counter
CRYSTAL-FACE	Cirrus Regional Study of Tropical Anvils and Cirrus Layers-Florida Area Cirrus Experiment
CVI	Counterflow virtual impactor
DMA	Differential mobility analyzer
DMS	Dimethylsulfide
DOE	Department of Energy
EARLINET	European Aerosol Research Lidar Network
EDX	Energy Dispersive X-ray
EPA	Environmental Protection Agency
GCM	Global climate model
H-TDMA	Humidified Tandem Differential Mobility Analyzer
ICARTT	International Consortium for Atmospheric Research on Transport and Transformation (online at <a href="http://www.al.noaa.gov/ICARTT/index.shtml">www.al.noaa.gov/ICARTT/index.shtml</a> )
IGAC	International Global Atmospheric Chemistry (online at <a href="http://www.igac.noaa.gov/">www.igac.noaa.gov/</a> )
IGBP	International Geosphere–Biosphere Program (online at <a href="http://www.igbp.kva.se/">www.igbp.kva.se/</a> )
IMPROVE	Interagency Monitoring of Protected Visual Environments (online at <a href="http://vista.cira.colostate.edu/improve/">http://vista.cira.colostate.edu/improve/</a> )
INDOEX	Indian Ocean Experiment (online at <a href="http://www-indoex.ucsd.edu/">www-indoex.ucsd.edu/</a> )
IOP	Intensive operational period
IPCC	Intergovernmental Panel on Climate Change (online at <a href="http://www.ipcc.ch/">www.ipcc.ch/</a> )
MASE	Marine Stratus Experiment
MILAGRO	Megacity Initiative: Local and Global Research Observations (online at <a href="http://www.windows.ucar.edu/tour/link=/milagro/milagro_intro.html">www.windows.ucar.edu/tour/link=/milagro/milagro_intro.html</a> )

MISR	Multiangle imaging spectroradiometer (online at <a href="http://www-misr.jpl.nasa.gov/">www-misr.jpl.nasa.gov/</a> )
MODIS	Moderate Resolution Imaging Spectroradiometer (online at <a href="http://modis.gsfc.nasa.gov/">modis.gsfc.nasa.gov/</a> )
MPLNET	Micro-Pulse Lidar Network (online at <a href="http://mplnet.gsfc.nasa.gov/">http://mplnet.gsfc.nasa.gov/</a> )
NASA	National Aeronautics and Space Administration
NSF	National Science Foundation
NOAA	National Oceanic and Atmospheric Administration
OC	Organic carbon
OMI	Ozone Monitoring Instrument (online at <a href="http://aura.gsfc.nasa.gov/instruments/omi/index.html">http://aura.gsfc.nasa.gov/instruments/omi/index.html</a> )
PARAGON	Progressive Aerosol Retrieval and Assimilation Global Observing Network
PCASP	Passive Cavity Aerosol Spectrometer Probe
PILS	Particle Into Liquid Sampler
PIXE	Particle Induced X-ray Emission
PTR-ITMS	Proton Transfer Reaction Ion Trap Mass Spectrometer
QMOM	Quadrature method of moments
RICO	Rain in Cumulus over the Ocean
SAGE	Stratospheric Aerosol and Gas Experiment (online at <a href="http://eosweb.larc.nasa.gov/GUIDE/campaign_documents/sage1_project.html">http://eosweb.larc.nasa.gov/GUIDE/campaign_documents/sage1_project.html</a> )
SCAR-B	Smoke, Clouds, Aerosols, Radiation-Brazil (online at <a href="http://eosweb.larc.nasa.gov/GUIDE/campaign_documents/scar_project.html">http://eosweb.larc.nasa.gov/GUIDE/campaign_documents/scar_project.html</a> )
SEM	Scanning electron microscopy
SMPS	Scanning Mobility Particle Sizer
SPLAT	Single-particle laser ablation time-of-flight mass spectrometry
SPMS	Single-particle mass spectrometry
SOA	Secondary organic aerosol
TARFOX	Tropospheric Aerosol Radiative Forcing Observational Experiment (online at <a href="http://geo.arc.nasa.gov/sgg/tarfox/">http://geo.arc.nasa.gov/sgg/tarfox/</a> )
TDMA	Tandem differential mobility analyzer
TOMS	Total Ozone Mapping Spectrometer (online at <a href="http://jwocky.gsfc.nasa.gov/">http://jwocky.gsfc.nasa.gov/</a> )
TEM	Transmission electron microscope
TRACE-P	Transport and Chemical Evolution over the Pacific
VOC	Volatile organic carbon
WCRP	World Climate Research Programme

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