Improving our fundamental understanding of the role of aerosol–cloud interactions in the climate system

John H. Seinfeld[a,b,1], Christopher Bretherton[c], Kenneth S. Carslaw[d], Hugh Coe[e], Paul J. DeMott[f], Edward J. Dunlea[g], Graham Feingold[h], Steven Ghan[i], Alex B. Guenther[j], Ralph Kahn[k], Ian Kraucunas[i], Sonia M. Kreidenweis[f], Mario J. Molina[l,m], Athanasios Nenes[n,o,p,q], Joyce E. Penner[r], Kimberly A. Prather[l,m], V. Ramanathan[m], Venkatachalam Ramaswamy[s], Philip J. Rasch[i], A. R. Ravishankara[f], Daniel Rosenfeld[t], Graeme Stephens[u], and Robert Wood[c]

The effect of an increase in atmospheric aerosol concentrations on the distribution and radiative properties of Earth's clouds is the most uncertain component of the overall global radiative forcing from preindustrial time. General circulation models (GCMs) are the tool for predicting future climate, but the treatment of aerosols, clouds, and aerosol–cloud radiative effects carries large uncertainties that directly affect GCM predictions, such as climate sensitivity. Predictions are hampered by the large range of scales of interaction between various components that need to be captured. Observation systems (remote sensing, in situ) are increasingly being used to constrain predictions, but significant challenges exist, to some extent because of the large range of scales and the fact that the various measuring systems tend to address different scales. Fine-scale models represent clouds, aerosols, and aerosol–cloud interactions with high fidelity but do not include interactions with the larger scale and are therefore limited from a climatic point of view. We suggest strategies for improving estimates of aerosol–cloud relationships in climate models, for new remote sensing and in situ measurements, and for quantifying and reducing model uncertainty.

climate | aerosol–cloud effects | general circulation models | radiative forcing | satellite observations

Clouds play a key role in Earth's radiation budget, and aerosols serve as the seeds upon which cloud droplets form. Anthropogenic activity has led to an increase in aerosol particle concentrations globally and an increase in those particles that act as cloud condensation nuclei (CCN) and ice nucleating particles (INP). The effect of an increase in aerosols on cloud optical properties, and associated radiative forcing, is the most uncertain component of historical radiative forcing of Earth's climate caused by greenhouse gases (GHGs) and aerosols. The Intergovernmental Panel on Climate Change (IPCC) AR5 assessment of climate forcing factors (Fig. S1) ascribes "high" confidence to the estimate of direct aerosol radiative forcing (mean

[a]Division of Chemistry and Chemical Engineering, California Institute of Technology, Pasadena, CA 91125; [b]Division of Engineering and Applied Science, California Institute of Technology, Pasadena, CA 91125; [c]Department of Atmospheric Science, University of Washington, Seattle, WA 98195; [d]School of Earth and Environment, University of Leeds, Leeds L32 9JT, United Kingdom; [e]School of Earth, Atmospheric and Environmental Sciences, University of Manchester, Manchester M13 9PL, United Kingdom; [f]Department of Atmospheric Science, Colorado State University, Fort Collins, CO 80523; [g]Board on Atmospheric Science and Climate, National Academies of Sciences, Engineering, and Medicine, Washington, DC 20001; [h]Earth System Research Laboratory, National Oceanic and Atmospheric Administration, Boulder, CO 80523; [i]Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, WA 99352; [j]Department of Earth System Science, University of California, Irvine, CA 92697; [k]Sciences and Exploration Directorate, Goddard Space Flight Center, National Aeronautics and Space Administration, Greenbelt, MD 20771; [l]Department of Chemistry and Biochemistry, University of California, San Diego, La Jolla, CA 92093; [m]Scripps Institution of Oceanography, University of California, San Diego, La Jolla, CA 92093; [n]Department of Earth and Atmospheric Science, Georgia Institute of Technology, Atlanta, GA 30332; [o]Department of Chemical and Biological Engineering, Georgia Institute of Technology, Atlanta, GA 30332; [p]Institute of Chemical Engineering Sciences, Foundation for Research and Technology–Hellas, Patras GR-26504, Greece; [q]Institute for Environmental Research and Sustainable Development, National Observatory of Athens, Palea-Pendeli GR-15236, Greece; [r]Climate and Space Sciences and Engineering, University of Michigan, Ann Arbor, MI 48109; [s]Geophysical Fluid Dynamics Laboratory, Princeton University and National Oceanic and Atmospheric Administration, Princeton, NJ 08540; [t]Institute of Earth Sciences, The Hebrew University of Jerusalem, Jerusalem 91904, Israel; and [u]Center for Climate Sciences, Jet Propulsion Laboratory, National Aeronautics and Space Administration, Pasadena, CA 91109

This paper results from the Arthur M. Sackler Colloquium of the National Academy of Sciences, "Improving Our Fundamental Understanding of the Role of Aerosol–Cloud Interactions in the Climate System," held June 23–24, 2015, at the Arnold and Mabel Beckman Center of the National Academies of Sciences and Engineering in Irvine, CA. The complete program and video recordings of most presentations are available on the NAS website at www.nasonline.org/Aerosol_Cloud_Interactions.

Author contributions: J.H.S., C.B., K.S.C., H.C., P.J.D., E.J.D., G.F., S.G., A.B.G., R.K., I.K., S.M.K., M.J.M., A.N., J.E.P., K.A.P., V. Ramanathan, V. Ramaswamy, P.J.R., A.R.R., D.R., G.S., and R.W. wrote the paper.

The authors declare no conflict of interest.

[1]To whom correspondence should be addressed. Email: seinfeld@caltech.edu.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1514043113/-/DCSupplemental.

estimate –0.27 W·m^{-2}), despite the fact that the mean estimate is the result of the difference between two much larger forcings of opposite sign. "Low" confidence is expressed in the forcing associated with cloud adjustments due to aerosols, the so-called "indirect effect" (mean estimate –0.55 W·m^{-2}). Even the high confidence in direct aerosol radiative forcing in the IPCC assessment is based as much on convergence among climate models used as on agreement with actual observational datasets.

Despite an increasing number of laboratory aerosol studies, ambient aerosol/cloud measurements, satellite aerosol datasets, model evaluation and intercomparison studies, and published papers on aerosol–climate interactions over the past 20 y, the uncertainty associated with the estimated radiative forcing attributed to aerosol–cloud interactions has not decreased over the past four IPCC cycles. A critical challenge for projecting future climate is to improve the estimate of aerosol impact on clouds and reduce the uncertainty associated with that estimate.

The importance of this issue and the seeming lack of progress motivated the Arthur M. Sackler Colloquium on "Improving Our Fundamental Understanding of the Role of Aerosol–Cloud Interactions in the Climate System," held June 23–24, 2015, in Irvine, CA: www.nasonline.org/programs/sackler-colloquia/completed_colloquia/Role_of_Aerosol_Cloud_Interactions.html. The Sackler Colloquium posed two big picture questions: Why have the radiative effects of aerosol–cloud interactions been so difficult to constrain and what can be done moving forward to make significant progress?

Participants in the Sackler Colloquium were asked to address the following issues: What are the important aerosol-induced effects involving clouds and their radiative properties that need to be better represented in climate models to produce more accurate prediction? How can the uncertainty associated with computation of these effects be estimated quantitatively? Can existing surface-based, airborne, and remote sensing data already in hand be better used to extract key aspects of the controls on cloud radiative effects? What is an ideal suite of in situ measurements to better constrain the critical parameters and phenomena of interest? If we can measure microphysics accurately in situ, then can one improve satellite retrieval algorithms to translate between satellite-retrieved aerosol optical properties and species-specific aerosol number, mass, and size tracked in aerosol transport and climate models? What are the prospects for new satellite instruments or remote sensing strategies that can constrain the uncertainty associated with key measurements? How can models that estimate the global-to-regional forcing and its impacts be improved to capture the physics and chemistry of the aerosol–cloud interactions more accurately? The present paper, in part, summarizes key aspects of the presentations made at the Sackler Colloquium.

State of Knowledge of Aerosol–Cloud Interactions

There is little doubt that the concentration, composition, and spatial distribution of Earth's aerosols have changed since 1750, the date typically used to represent the start of the Industrial Revolution. Owing to the near absence of industrial aerosols at that time, the major direct aerosol sources would have been sea salt, mineral dust, and smoke from wildfires and agricultural burning, with secondary aerosol sources of gas-to-particle conversion of marine and volcanic sulfur-containing gases and biogenic hydrocarbon emissions. Without the benefit of measured preindustrial global aerosol concentrations and composition, it is necessary to estimate the properties of preindustrial aerosols and clouds using global models, to establish the baseline against which the current state can be compared. (As some have suggested, should a different baseline be defined, for which the atmospheric state is better known?)

It has been known for many decades that aerosols influence cloud properties. Twomey (2) argued that an increase in CCN entering shallow, warm clouds will, for a constant cloud liquid water content, lead to more numerous, smaller cloud droplets. This increases the total droplet surface area that interacts with solar radiation, resulting in more sunlight scattered back to space. Changes in CCN also have the potential to influence cloud macrostructure. Albrecht (3) argued that a reduction in the size of cloud droplets would retard and reduce rain formation in shallow marine clouds. This hypothesis has been referred to as the "lifetime effect." That is, the macrostructure of the cloud (such as its spatial extent or liquid water content) is determined by the efficiency with which precipitation develops, which, in turn, is regulated by the aerosol (4). Observations of ship tracks, brighter regions of marine stratocumulus clouds from ship exhaust (5), show that the sign (increase or decrease) and magnitude of the cloud albedo response depend on the mesoscale cloud structure, the free tropospheric humidity, and cloud top height. From the viewpoint of radiative forcing, cloud macrophysics (i.e., liquid water path and cloud fractional coverage) is the dominant contribution to planetary albedo that needs to be simulated in climate models.

The wide diversity in climate model estimates of aerosol–cloud radiative forcing stems largely from two sources. First, although there exists fundamental understanding of aerosol–cloud processes at the scale of a single cloud, the scale disparity between general circulation models (GCMs) and individual cloud processes introduces substantial uncertainties. Second, the change in the aerosol state itself from preindustrial to present day is uncertain (6, 7). Climate models incorporate aerosol–cloud formulations, the parameters of which are usually constrained to achieve a desired balance with the top-of-the-atmosphere radiation measurements.

Fig. 1 shows an example of the disproportionate sensitivity of climate model predictions to the specific submodel describing aerosol–cloud interactions in one prominent GCM, the Geophysical Fluid Dynamics Laboratory (GFDL) climate model. CM3, CM3w, and CM3c represent three different configurations of the submodels impacting aerosol–cloud interactions in the GFDL climate model with alternate, but plausible, parameter choices. The parameter choices considered are the lower bound on updraft velocity variance for CCN activation, the autoconversion threshold radius that governs the conversion of cloud water to rain, and the cloud lifetime as influenced by mixing with the environment. The three different model realizations arise due to the sensitivity of the choice of parameters involved in describing how the clouds respond to precipitation suppression by aerosols. Based on the uncertainties in the microphysics of aerosol–cloud –precipitation, the three realizations produce different warming trajectories. Although the present-day climate is similar in these different configurations, the magnitude of the predicted aerosol indirect forcing differs by as much as –1.2 W·m^{-2}, resulting in significantly different predicted surface temperature evolution over the twentieth century. Interestingly, the simulated temperature evolution that best agrees with the observed trend (CM3w) corresponds to the threshold radius for autoconversion that most poorly reproduces the satellite-observed microphysics. This inconsistency between the "bottom-up" process-based constraint and the "top-down" temperature trend constraint implies that compensating

errors exist in the model, a factor that is a result of the poor constraint on microphysical processes (9). Such inconsistencies are a general feature of global models.

Process-Scale Modeling of Aerosol–Cloud Relationships

Cloud droplet concentration N_d is the key microphysical mediator of aerosol–cloud interactions in warm clouds. The droplet concentration in a nascent warm cloud is governed largely by two factors: the size of those aerosols that activate (the CCN) and the updraft velocity of the air that delivers the aerosols to the altitude of activation. The most advanced GCMs represent warm cloud formation using mechanistic parameterizations that use updraft velocity, particle size distribution, and particle composition to predict N_d and the droplet size distribution.

The atmosphere contains a complex mixture of individual particles of differing size and composition determined by their source as well as subsequent atmospheric processing. The manner in which chemical species are distributed among individual particles is termed the "aerosol mixing state"; if all particles have the same composition, the aerosol is referred to as "internally mixed," and, when individual particles are chemically different, the aerosol is "externally mixed." Ultimately, those particles that can act as CCN are determined by their size and chemical composition (10, 11). Single-particle measurements reveal mixtures of soot, dust, and organic carbon, with secondary species such as sulfate, ammonium, nitrate, and oxidized organics (12–14). Although, in some heavily aged environments, the diversity in particle hygroscopicity is relatively small (15), other studies demonstrate clear impacts of particle mixing state on CCN behavior (16–18).

Although initial cloud formation occurs on the order of minutes, cloud thickness, liquid water path, and altitude can change markedly over the cloud lifetime, which can extend to many hours. Cloud macrophysical properties, such as cloud fractional coverage and thickness, are the most important determinants of albedo variability (19). These properties are governed, for example, by the extent of drizzle and precipitation, entrainment/detrainment of air from above the cloud (20), and boundary layer dynamics below the cloud. Accounting for aerosol absorption effects on clouds raises particularly challenging issues. These include effects on cloud heating by absorbing inclusions in droplets and of absorbing aerosol particles interstitially between droplets (21–25). (Note that the positive aerosol direct forcing contribution in Fig. S1 is also attributed to absorbing aerosols.) Most climate models included in IPCC assessment predict a linear increase in cloud optical depth with increasing aerosol optical depth, because the effect of absorbing aerosols within clouds is not treated. Consequently, they may overestimate aerosol cooling due to clouds.

Explicit calculation of N_d goes beyond CCN alone and depends on aerosol and dynamical parameters. Fig. 2 shows, for a particular GCM simulation, the extent to which N_d variability is dominated by variations in vertical velocity over the continents and certain ocean areas where the aerosol concentration is considerable; other regions are dominated by variations in fine-mode aerosol number concentration. The importance of these parameters for a given region tends to exhibit a strong seasonal cycle.

Turbulent eddy motions are central to the maintenance of almost all cloud regimes, especially boundary layer and cumulus clouds, and impact cloud–precipitation–aerosol interaction within these regimes. Large eddy simulation (LES) and cloud-resolving models (CRMs) are powerful tools for high-resolution numerical modeling of cloud regimes. These approaches explicitly simulate the larger and more energetic turbulent eddies that produce vertical turbulent fluxes of moisture, heat, and aerosol particles, while parameterizing the effects of smaller subgrid eddies. CRMs are typically applied to deep convection, and LES is applied to shallow convection (stratocumulus and cumulus). In some cases, LES and CRMs include a specification of aerosols for activation of cloud droplets and/or ice crystals (27, 28). LES is most frequently applied to 3D simulations of boundary layer clouds, with dominant updraft and downdraft scales of a few hundred meters. A typical LES might have a horizontal grid spacing of 25–100 m, a vertical grid spacing of 5–50 m, and a doubly periodic horizontal domain of 5–100 km on a side, simulating a period of hours to days with a time step of around 1 s (29, 30). CRMs are often applied to deep cumulonimbus convection with updraft and downdraft scales of several kilometers, using horizontal grids of several hundred meters to a few kilometers and vertical grid spacing of a few hundred meters, typically over domains of 100–1,000 km on a side using time steps on the order of 10 s for simulated periods of hours to tens of days (31). Global CRMs have been developed with horizontal grids of 1–10 km and executed for periods of up to a few months, but their application to aerosol–cloud interaction has been limited by the substantial computational demands. "Superparameterization" (32, 33) is a methodology in which a small, computationally efficient CRM is run within each vertical grid column of a GCM in place of the cloud, convection, and turbulence parameterizations within the GCM. An important attribute of LES and CRMs is that they resolve clouds sufficiently well that microphysical parameterizations do not need to include complex representation of subgrid cloud inhomogeneity. Nonetheless, for precipitating boundary layer clouds, and especially

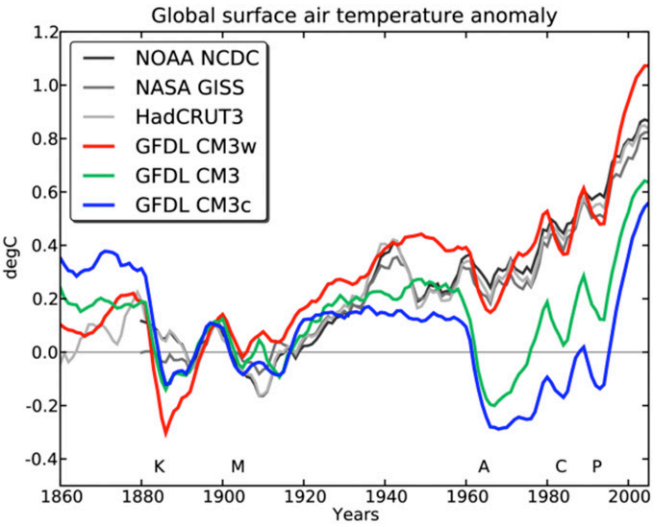

Fig. 1. Sensitivity of predicted global temperature evolution to three versions of the aerosol–cloud submodel in the GFDL climate model: CM3w, CM3, and CM3c (8). In the simulations, emissions of sulfate precursors, organic and black carbon, and GHG concentrations were held constant at their 1990 values. For CM3w, increased cloudiness (the lifetime effect) owing to reduced autoconversion of cloud droplets to rain is partially offset by increased cloud erosion. For CM3c, the increased cloudiness that results from less efficient autoconversion of cloud droplets to precipitation is offset by reducing the lower bound on the vertical velocity variance for CCN activation. The net warming predicted by the three different submodels is: CM3w, 0.57 °C; CM3, 0.22 °C; and CM3c, –0.01 °C. By comparison, predictions of three other GCMs are: NOAA NCDC, 0.59 °C; NASA GISS, 0.53 °C; and HadCRUT3, 0.56 °C.

for mixed- and ice-phase clouds, CRM simulations can be quite sensitive to reasonable choices of microphysical parameterization (34).

A long-standing issue, and one that can potentially address the uncertainty in GCM parameterizations, is how the detailed understanding of aerosol–cloud processes imbedded in LES and CRMs can be scaled up to the GCM. In fact, improved GCM cloud parameterizations and cloud lifetime representations have been derived from suites of simulations with such high-resolution process-scale models, although this process is not always quick or straightforward (35–37).

Ice and Mixed-Phase Clouds

The processes determining aerosol indirect forcing from ice and mixed phase clouds are less well understood than those in warm clouds (38) (Fig. 3). Nominally, only one in a million particles can act as an INP at –20 °C (39–45). If the temperature is reduced sufficiently, then all CCN can potentially act as sites for ice to form on by homogeneous freezing. Heterogeneous freezing becomes far more likely at less than extreme temperatures. Water consumption by growing ice can impede ice nucleation, so updraft and moisture supply can sometimes exert a stronger control on ice formation than the availability of INP. The scarcity of INP compared with CCN profoundly impacts clouds in the mixed-phase regime, as the INP (for cloud droplets) that freeze rapidly grow at the expense of the existing water droplets, leading to increase in the cloud particle size and reduction in their concentration. The ice crystals that result can increase precipitation rates, reduce cloud lifetime, and decrease cloud shortwave reflectivity and longwave emissivity. In pure ice (cirrus) clouds, sufficient, but relatively low, concentrations of INP can reduce or completely inhibit homogeneous freezing, impacting crystal number and size, and hence cloud lifetime and longwave emissivity.

Mineral dust and some biological particles exhibit a high degree of ice nucleation activity. The importance of mineral dust particles as INP is reflected in the high frequency of detection of particles of mineral composition in INP samples and in residues of ice cloud particles. INP contributions from biological particles have been detected in mixed-phase clouds (44), which may arise, in part, from arable soils (46). Biomass burning can act as an INP source (47); less clear at present is the extent to which anthropogenic emissions serve as INP. Global aerosol model simulations of marine organic particle emissions, in combination with measurements isolating INP from within the sea surface microlayer, suggest that marine organic material may be an important source of INP in remote marine ocean environments (48–50). Overall, increases in aerosol number concentration are predicted to generate deeper convective, mixed-phase clouds (51, 52). (Those GCMs in IPCC AR5 that explicitly represent aerosol effects on deep convective and mixed-phase clouds were given stronger weighting in the assessment of forcing.) GCM-generated clouds tend not to reflect sufficient sunlight over remote, high-latitude oceans (53); a possible explanation is inadequate representation of ice formation (Fig. 3). Aerosol composition can play a significant role in mixed-phase and ice clouds. For example, Ault et al. (54) and Fan et al. (55) showed that two mixed-phase storms classified as "atmospheric rivers" with essentially identical meteorology and dynamics—one with clouds seeded with pollution aerosols and the second seeded with dust—produced 40% more precipitation with dust as seeds. Finally, ice production from riming and ice–ice collisions in warm mixed-phase clouds can profoundly impact cloud lifetime and precipitation. The extent to which these processes are important on a climatic scale is generally uncertain (and not included in current GCMs).

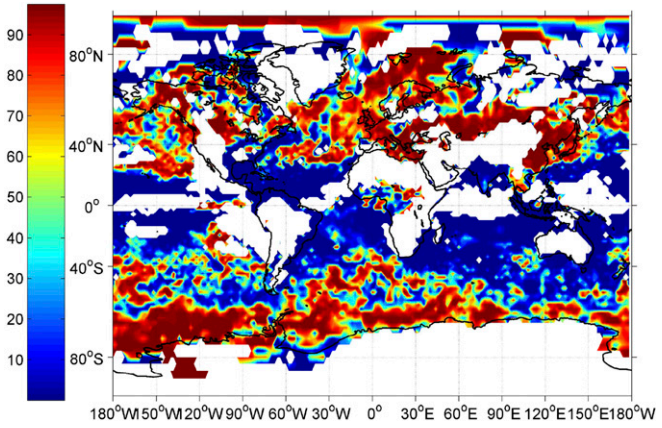

Fig. 2. Percent contribution of vertical velocity variation to the temporal variability in activated cloud droplet number concentration (per cubic centimeter) for all clouds at the 825-hPa level for January 2010 in the NASA Goddard Earth Observing Model Version 5 (26). Values are calculated from the sensitivity of droplet number concentration to vertical velocity, temperature, aerosol modal number concentration, diameter, and chemical composition using the adjoint of the droplet parameterization and the input variances. Vertical velocity variation controls the droplet number concentration variability over continents and large fractions of the ocean with considerable influences from biogenic and anthropogenic emissions. Areas in white correspond to regions where stratus cloud fraction is low.

Connecting Process-Scale Models to Climate Models

A persistent challenge in characterizing aerosol–cloud radiative effects is the untangling of aerosol and meteorological controls on cloud properties. Aerosol–warm cloud interactions have been expressed formally as a chain of processes that relate changes in emissions (E) of CCN to changes in cloud droplet number concentration N_d to all sky shortwave cloud forcing R (56–58),

$$\frac{d\ln R}{d\ln E}=\frac{d\ln R}{d\ln N_d}\frac{d\ln N_d}{d\ln CCN}\frac{d\ln CCN}{d\ln E} \quad [1]$$

$$\frac{d\ln R}{d\ln N_d}=\frac{d\ln C}{d\ln N_d}+\frac{d\ln R_c}{d\ln N_d} \quad [2]$$

$$\frac{d\ln R_c}{d\ln N_d}=\frac{d\ln R_c}{d\ln \tau}\frac{d\ln \tau}{d\ln N_d} \quad [3]$$

$$\frac{d\ln \tau}{d\ln N_d}=\frac{d\ln L}{d\ln N_d}-\frac{d\ln r_e}{d\ln N_d} \quad [4]$$

where C is cloud fraction, R_c is in-cloud cloud radiative forcing, L is cloud liquid water path, and cloud optical depth $\tau \approx L/r_e$, with r_e as cloud droplet effective radius. Eqs. **1**–**4** can be merged into the composite relation

$$\frac{d\ln R}{d\ln E}=\left[\frac{d\ln C}{d\ln N_d}+\frac{d\ln R_c}{d\ln \tau}\left(\frac{d\ln L}{d\ln N_d}-\frac{d\ln r_e}{d\ln N_d}\right)\right]\frac{d\ln N_d}{d\ln CCN}\frac{d\ln CCN}{d\ln E}. \quad [5]$$

Ghan et al. (59) estimate the structural uncertainty in representations like Eq. **5** using a suite of atmospheric models. Ideally,

we would want to constrain these microphysical and macrophysical responses with present-day observations. Feingold et al. (60) discuss just how difficult this path is—both practically (measurement errors, scale issues, etc.) and conceptually (e.g., quantifying partial derivatives in an adjusting system). Still, even if this chain of responses can be quantified in the present-day atmosphere, quantification of overall aerosol–cloud forcing requires knowledge of the change in CCN since preindustrial periods, which remains a major source of uncertainty (6).

Estimating Uncertainty in Climate Model Predictions

The uncertainty associated with the predictions of a full GCM is a convolution of the uncertainties arising from many individual submodels describing the underlying processes. Owing to model complexity and observational limitations, GCM uncertainty is often assessed solely on the basis of model diversity—the spread of predictions of several models. International efforts to compare and evaluate aerosol and cloud processes among global models, such as AeroCom (aerocom.met.no/Welcome.html), have been successful in understanding some of the causes of the diversity. Recently, advanced statistical approaches have been used to sample the combined effects of many sources of global model uncertainty for CCN (61). Combinations of multimodel and statistical approaches have the promise to provide deeper insights into the causes of model spread.

Estimates of climate model robustness have focused mainly on the extent to which overall GCM output agrees with available in situ and satellite measurements. However, more complete approaches that evaluate the convolution of many model uncertainties show that model–observation agreement may give a misleading impression of the reliability of a model for calculating forcing. Lee et al. (62) show how, in a model as complex as a GCM, essentially identical predictions of the aerosol state (in their study, CCN and particle concentrations) can result from a spectrum of different parameters, or even submodels, a situation termed "equifinality." As model complexity increases in the effort to attain more realistic treatment of cloud processes, that complexity can only be expected to compound further the problem of estimating the overall uncertainty associated with model predictions.

Techniques to comprehensively explore uncertainty in computationally expensive global models are just emerging. These enable us to generate a wide range of model predictions whose plausibility can be tested against observations. A major challenge will be to understand how our current observing systems allow the range of predictions of these models to be constrained, and what this means for forcing uncertainty. To date, observations have been used mainly to test the extent to which particular models simulate the present atmosphere, but, according to Lee et al. (62), such model–observation agreement may not be directly informative about model uncertainty. With more advanced approaches, it will be possible to relate measurements to the effect they have on constraining model uncertainty, as defined by the range of plausible models. With such information, it will also be possible to better understand where and how future measurements should be made. So far, uncertainty quantification approaches have been used mostly for global models, but they may also provide a way to explore uncertainty in aerosol–cloud interactions in models confined to represent smaller scales (63). Adjoint sensitivity approaches provide another pathway to obtain uncertainty information (64, 65).

Although aerosols have been identified in the IPCC assessments as one of the most important sources of uncertainty in predicting climate change, it has been noted that (*i*) aerosol effects on climate should be anticipated to occur mainly in relatively pristine environments seldom seen in the present day (66) and (*ii*) an overall aerosol radiative forcing more negative than –1.0 $W{\cdot}m^{-2}$ is implausible, as it implies that none of the ~0.3 °C temperature rise between 1850 and 1959 can be attributed to Northern Hemisphere forcing (67). On the other hand, a number of features of observed climate change can be explained only if aerosol forcing offsets a portion of GHG forcing. The most notable of these are precipitation effects, especially in the tropics and the Northern Hemisphere as a whole (68, 69), for which GHG forcing alone cannot explain the observed signatures in precipitation (the sign of the changes itself would be wrong). Moreover, the aerosol offset to GHG forcing consistent with these effects cannot be a result of direct forcing only, as the estimated magnitude of direct aerosol radiative forcing is too small to explain the counteraction of GHG effects. Even if ambient aerosol concentrations are deemed to be sufficiently high to reduce the susceptibility of present-day clouds to further increases, this does not diminish the importance of understanding and characterizing their historical role in countering warming by GHGs, and in confounding the scientific understanding of the planet's response to changes in forcing agents in the future.

Observations of Aerosol–Cloud Relationships

Observational data form the foundation on which GCM performance is evaluated. Such data include essential cloud properties like liquid water path and cloud fraction, as well as radiative

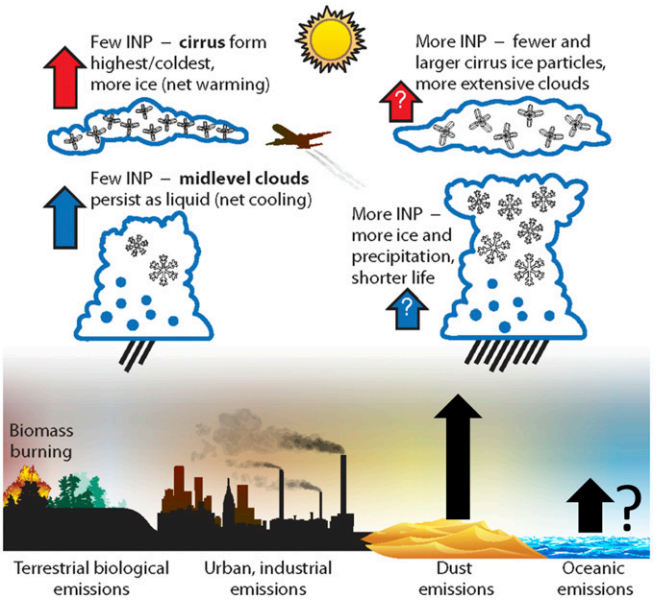

Fig. 3. Climate relevance of ice nucleation by atmospheric aerosols via altered cloud radiative properties and precipitation (41). Heterogeneous INP are responsible for ice crystal initiation in clouds entirely warmer than –38 °C (midlevel clouds here), and INP compete with spontaneous freezing of wet aerosols in cirrus clouds in a nonintuitive manner. Various possible aerosol sources of INP are indicated, although the contributions of many aside from mineral dust are poorly constrained. Since 2010, the potential contribution of arable soil dust is also now recognized. The likely but uncertain change in the magnitude of the general cooling impact (blue arrows) of midlevel clouds and warming impact (red arrows) of high cirrus clouds in response to increases in the relative number concentrations of INP is suggested.

properties. Although currently not possible, precise, and simultaneous, global satellite observations of CCN and updraft velocity, together with cloud microphysical and radiative properties, would provide strong constraints on modeling these processes in detailed (LES, CRM) models and in global model parameterizations where virtually all these features are subgridscale. Current satellite and suborbital remote sensing data alone cannot constrain the microphysical, optical, and chemical properties of airborne particles sufficiently to estimate CCN concentrations. Mass Extinction Efficiencies (MEEs), used to translate between remote sensing-derived particle optical properties and aerosol mass, must be obtained from in situ measurements, estimated from modeled particle composition and size distributions, or simply assumed. Similarly, aerosol hygroscopicity, required to account for humidity-dependent particle optical property changes as well as conditions that initiate cloud formation, cannot be derived from remote sensing observations except under special conditions (70). Finally, data can be used to determine correlations only; to establish cause and effect, a physical model is needed.

In Situ Observations. In situ observations are vital for establishing the necessary process understanding required to include the important physics and chemistry in models. Although satellites provide long-term, global coverage of a wide range of aerosol and cloud properties and so offer direct comparison with, and challenge to, global models, important information such as the vertical distribution of stacked aerosol and cloud layers, the optical properties of absorbing particles, and updraft speeds may not be obtained. Aircraft platforms provide a means for describing detailed microphysical properties and their horizontal and vertical distributions, and, although field experiments typically focus on specific regions for limited time periods, they complement satellite measurements by providing detailed in situ information. Ground-based remote sensing, such as the Aerosol Robotic Network (aeronet.gsfc.nasa.gov) of Sun- and sky-scanning photometers, contribute frequent, long-term observations at key locations. Surface-based radiation measurements offer a reliable means to infer the effects of aerosols on surface fluxes, which, in turn, influence vertical and horizontal advection, surface evaporation, and the hydrologic cycle. Aerosol–cloud interactions can also be addressed from surface measurements, because aerosol and cloud properties can be measured simultaneously in the same column (as opposed to satellites that measure aerosol in cloud-free areas adjacent to clouds). Finally, it is difficult to measure important cloud properties like liquid water path and cloud optical depth, particularly in broken cloud scenes and for clouds with low liquid water path.

Challenges remain to measure certain aerosol properties, notably absorption and density; large particles, particularly dust and bioparticles with sizes above 1 μm, are difficult to capture through inlets on rapidly moving platforms. Although turbulence measurements are regularly performed from aircraft, quantifying updraft speeds and statistically meaningful entrainment rates remains difficult; finally, INPs remain a challenging measurement, especially from aircraft, where most inlets place limits on the size of particles sampled and can alter their properties (via heating) compared with the ambient atmosphere. Remotely piloted or unmanned airborne vehicles will play a role in this new generation of in situ measurements.

Focused experiments in geographical areas that are critical in climate response provide a means of constraining model representations. Model sensitivity analyses can help to define processes that can be evaluated and constrained by measurements in different regions (62). Many key portions of the globe remain undersampled, for example, the Southern Ocean, the maritime and continental tropics, much of Asia, and both polar regions. In an effort to replicate preindustrial aerosol–cloud relationships, it is helpful to study those regions of the present-day atmosphere that approximate preindustrial conditions (71). In such regions, clouds are particularly susceptible to small increases in CCN levels (72); the remote Southern Ocean is a prime example. Although much of the North Atlantic is in a state that can be classified as polluted, conditions in which present-day aerosol has been scrubbed by upstream processes could, in theory, replicate the preindustrial state.

Despite the cost and complexity of large-scale, multiplatform experiments, the benefits of the datasets delivered are substantial, and their legacy is significant. An example of such a large-scale experiment is the Variability of the American Monsoon Systems (VAMOS) Ocean-Cloud-Atmosphere-Land Study Regional Experiment, which focused on the stratocumulus cloud deck of the southeast Pacific, its predictability, and its interaction with anthropogenic aerosol from the South American coastal region (73, 74). To achieve successful execution of such experiments, interagency and cross-country coordination is important. See Box 1.

Satellite Measurements. Satellite measurements are an essential component of an observational strategy to constrain aerosol-cloud relationships. Current capabilities and limitations of satellite observations are summarized in Box 2.

Aerosol optical properties that can be measured from satellites are aerosol optical depth (AOD) and its spectral dependence at solar wavelengths, which provide general information on particle size and, in some circumstances, absorption. Observations that contain multiangle and/or polarization measurements can be used to retrieve additional information about particle size, shape, single scattering albedo, and indices of refraction, although sensitivity to these quantities varies depending on observing conditions.

The number of activated cloud drops at cloud base depends on the aerosol CCN supersaturation (S) activation spectrum and on cloud base updraft velocity (W_b). AOD is correlated to CCN (75). Owing to lack of better options, AOD has been used as a common surrogate for CCN, despite a number of drawbacks: (*i*) Aerosols smaller than ∼0.1 μm in diameter are indistinguishable from molecular scattering from air molecules, whereas aerosols larger than ∼0.05 μm often serve as CCN, especially in pristine environments, where small absolute changes in aerosol loading lead to large relative changes in cloud properties; (*ii*) aerosol swelling at high relative humidity causes uncertainty in determining the size distribution and total dry mass of particulate matter (76); (*iii*) cloud contamination of aerosol retrievals can exist (77); (*iv*) cloud-scattered light can alias nearby aerosol retrievals; (*v*) the observed AOD are column totals and may come from aerosol layers other than those that interact with cloud base; and (*vi*) clouds obscure the aerosol signal beneath them (78).

The desired information about CCN is mainly the number concentration of particles, whereas AOD depends heavily on their optical cross-section. All else being equal, a particle size distribution dominated by smaller particles has a steeper AOD spectral slope. Because more numerous smaller particles are needed to produce the same AOD, the aerosol index (AI), the product of AOD and a quantity related to the variation of AOD with wavelength, exhibits the desired qualitative effect of yielding a larger value when many small particles are present. Moderate Resolution

Box 1 – Some Large-Scale Field Experiments Planned to Address Aerosol–Cloud–Climate Interactions

Large-scale field studies will take place in 2016–2018 in the southeast Atlantic region. This region hosts one of the most extensive, semipermanent stratocumulus cloud decks on the planet, arising as a result of the cold, upwelling Benguela current along the southern African coast and the strong inversion at boundary layer top resulting from dry air aloft driven by the descending arm of the Hadley circulation. The cloud field modulates the heating of the subtropical Atlantic Ocean, impacts the hemispheric heat budget, and has a substantial influence on large-scale weather systems. To the north, the stratocumulus transitions to trade cumulus in the Gulf of Guinea.

From July to October, large plumes of radiatively absorbing aerosol emitted from biomass burning across Southern Africa advect over the southern Atlantic above the inversion to the west of Namibia and Angola. Farther offshore, as the biomass burning layers descend around the high pressure, regions of entrainment into clouds may occur. This situation offers an ideal natural laboratory to study direct, semidirect (that attributable to heating from sunlight absorption by carbonaceous particles), and indirect aerosol–cloud effects. Close to shore, the absorbing aerosol aloft increases heating above the cloud deck and reduces shortwave heating at cloud top, reinforcing the stratocumulus cloud below. Farther offshore, entrainment of absorbing aerosol may darken clouds and heat the boundary layer, leading to a reduction in cloud thickness, if not complete evaporation.

The NASA Earth Venture Suborbital-2 program Observations of Aerosols Above Clouds and Their Interactions will base a P-3 aircraft in Namibia in a different month within the biomass-burning period for each year between 2016 and 2018. In 2016, this program will be supplemented by the NASA ER-2 aircraft to investigate the radiative budget through the column using stacked aircraft with a range of in situ and remote sensing instruments. The United Kingdom will deploy its research aircraft, a BAe-146, during August–September 2016 as part of the Clouds and Aerosol Radiative Impacts and Forcing: Year 2016 (CLARIFY) project. In addition, lidars, radars, and radiometers will be deployed on St. Helena. The focus of CLARIFY is on processes taking place closer to the coast than in the larger-scale NASA experiment. The US Department of Energy will deploy an Atmospheric Radiation Measurement (ARM) mobile facility on Ascension Island from June 2016 to May 2017 during its Layered Atlantic Smoke Interactions with Clouds program. The array of radiometers and lidars will be used to explore the cumulus regime in this offshore region, where entrainment of biomass smoke layers is likely. These studies will be complemented by the Aerosol Radiation and Clouds in Southern Africa project, a French experiment to provide long-term surface-based measurements both in situ and of the aerosol column. In 2016, the French F20 Falcon aircraft, equipped with high-resolution lidar and polarimetric capability, will be deployed in the region.

Imaging Spectroradiometer (MODIS) retrieves size distributions over water from which CCN estimates based on the AI can be derived. In addition to the qualitative nature of the CCN–AI relationship, another critical issue is distributing the aerosol column amount in the vertical, relative to the clouds. Finally, AODs are very small in remote regions of the ocean; however, these are the regions where cloud susceptibility to CCN is largest.

An alternative approach to satellite measurement of CCN(S) has been proposed by Rosenfeld et al. (79) using clouds as natural CCN chambers. A CCN chamber measures CCN(S) by applying a known supersaturation to an air sample and counting the number of activated cloud droplets. At present, direct satellite measurements of updraft speeds do not exist. An indirect inference of W_b was developed recently (80, 81) by retrieving the properties that propel air vertically, such as ground skin versus air temperature and cloud base height (82). Satellite retrieval of CCN(S) is made possible by combining the inferred W_b with retrieved convective cloud base drop concentrations (83), for example, multispectral data as taken by the Visible Infrared Imaging Radiometer Suite onboard the Suomi National Polar-orbiting Partnership polar orbiting satellite (84). This strategy approaches disentangling the effects of CCN and meteorology on satellite-derived aerosol–cloud interaction.

Strategies for Improving Estimates of Aerosol–Cloud Forcing in Climate Models

Climate Models.

i) Incorporate better representation of clouds themselves into climate models, followed by testing these models on cloud lifecycle time scales using high spatial resolution models, such as LES and CRMs. The twin targets of increased complexity and resolution in producing global estimates have to be attended to judiciously because an excess march in one direction may not necessarily produce better estimates. Interactions between convection, microphysics, and radiation in models must be tested against field measurements wherever possible. An important metric is the extent to which the aerosol–cloud–precipitation cycle is simulated compared with observations, e.g., CloudSat measurements. The simultaneous comparison of modeled surface temperature evolution, top-of-the-atmosphere and surface radiation balance, and precipitation-related microphysical statistics with satellite and other observations forms a stringent evaluation test of global models. The execution of well-designed, comprehensive, multiplatform numerical simulations to map out aerosol–cloud responses for all climatically important cloud and aerosol types is needed to evaluate the performance of GCM parameterizations.

ii) Continue to develop and test models for mixed-phase and ice cloud formation and their behavior when imbedded in climate models. In particular, the different pathways for ice cloud formation and dissipation, and resulting precipitation, have to be critically compared with available observational estimates.

iii) Develop a modern suite of consensus metrics for evaluating aerosol–cloud interactions in GCMs against a spectrum of observations and process model results.

iv) Develop new methodologies that enable model uncertainty to be efficiently quantified, and apply these techniques across multiple models to establish causes of intermodel diversity. Establish traceable links between observations and their effects on model uncertainty, and determine where new observations are most needed to allow improved model constraint. Use observation metrics to evaluate and constrain model behavior (e.g., aerosol–cloud relations) as well as gross changes in aerosol and cloud properties of relevance to forcing over decadal and centennial periods.

Remote Sensing Measurements.

i) Carry out suborbital measurements of aerosol microphysical and chemical properties for the major aerosol types globally, at the level of detail required to represent CCN and INP behavior in aerosol–cloud interactions. This includes liquid water path and cloud fraction.

ii) Assure at least continuity of, and preferably enhance, global-scale aerosol and cloud measuring capabilities, including both polar-orbiting and geostationary platforms.

Box 2 – Satellite Contributions to Aerosol–Cloud Interaction Studies

Satellite Instrument Measurement Capabilities

- Polar orbiting imagers provide frequent, global coverage.
- Geostationary platforms offer high temporal resolution.
- Multiangle imagers offer aerosol plume height and cloud-top mapping.
- Multiangle and multiplatform imagers offer cloud-top rising rates.
- Passive instruments can retrieve total-column aerosol optical properties (AOD).
- Active instruments determine aerosol and some cloud vertical structure.
- UV imagers and active sensors can retrieve aerosol above cloud.
- Multiangle, multispectral, and polarized imagers obtain some aerosol type information.
- Active sensors can obtain some aerosol type information, day and night.
- Satellite trace-gas retrievals offer clues about aerosol sources and evolution.
- Visible (VIS)-IR imagers can retrieve cloud properties, including cloud fraction, cloud top temperature, liquid water path (LWP), r_e, and N_d.
- High-resolution (<375 m) VIS-IR imagers can retrieve W_b, N_d, and CCN(S) for certain cloud types.
- Passive microwave imagers retrieve cloud LWP, water vapor, and surface wind speed.
- Spaceborne radar and passive microwave sensors retrieve precipitation characteristics.

Satellite Instrument Limitations

- Polar orbiters provide snapshots only.
- It is difficult to retrieve aerosols that are colocated with cloud.
- Instruments can rarely detect aerosol in droplet-formation region below clouds.
- The practical particle size limit for remote sensing of aerosols is about 0.1 μm diameter. The practical limit on AOD detection (or accuracy) over dark water is ~0.02; over land, the limit might approach 0.02 but is not as good, in general. Currently, the lower limit on AOD required for aerosol-type identification (at the level of small, medium, or large; absorbing vs. nonabsorbing; or spherical vs. nonspherical) is 0.15 to 0.2 under good observing conditions. Polarization might improve this under some circumstances.
- Horizontal resolution is typically ~100s of meters or poorer.
- Passive instruments (imagers) offer limited vertical information.
- Active instruments (e.g., lidar) offer little spatial coverage.
- There is limited information about aerosol particle microphysical properties; aerosol hygroscopicity must be deduced from qualitative aerosol "type."
- Cloud microphysical and macrophysical retrievals are subject to biases related to cloud inhomogeneity.
- Satellites provide very limited information about cloud dynamical properties.
- Aerosol retrievals are aliased by the presence of clouds and vice versa.
- It is difficult to separate the effects of meteorological factors that covary.

iii) Develop techniques for remote sensing of updraft velocity and for measurement of the rate of dry air entrainment into clouds. Coordinate with in situ measurements.

iv) Develop new satellite instruments that take advantage of the aerosol type discrimination possible from multiangle, multispectral imagers with polarization channels, so satellite-derived aerosol type can be associated as specifically as possible with the microphysical detail obtained from in situ measurements. Improved methods for relating observations from space to the needed microphysical quantities will allow extrapolation of satellite data to areas where in situ observations are lacking.

v) Develop next-generation remote sensing aerosol retrieval algorithms that take advantage of multiple instruments and transport model simulations.

In Situ Measurements.

i) Carry out process-level LES or CRM simulations in conjunction with routine observations at supersites like the US Department of Energy Southern Great Plains facility, and at other locations where models indicate cloud sensitivity to aerosols, over a variety of meteorological and aerosol regimes to observationally constrain aerosol–cloud effects and to evaluate the performance of GCM parameterizations.

ii) Design large-scale field programs, in conjunction with satellite remote sensing, to provide comprehensive data for aerosol–cloud model testing and evaluation (Box 1).

iii) Carry out INP measurements to cover the full INP size distribution over continents and remote ocean regions, for different cloud base heights and relative humidity.

iv) Develop better understanding of how aerosols from different sources with different compositions influence cloud microphysics and macrophysics through in situ measurements of size-resolved mixing state. Compare aerosols available to act as CCN and INP.

v) Perform studies that compare and ultimately integrate in situ measurements of cloud microphysics, composition/source, and processes with models and satellite measurements.

IPCC Report Procedure. The Coupled Model Intercomparison Project, which feeds into IPCC assessments, has become a very substantial undertaking for the climate modeling community, in terms of both model preparation and computational resources. Climate models are very complex. To first order, they must satisfy overall constraints (replicating the current climate in all its observational dimensions, as well as historical climate change and variability) as well as the process-level constraints against which individual parameterizations are tested. Model tuning and compensating errors are inevitable, as are well-conceived parameterization refinements that have to be tabled because they do not improve overall model performance. With respect to aerosol–cloud interactions, there is, in addition, a diversity of scientific opinion about the extent to which current parameterizations are sufficiently robust to apply over a diversity of conditions. To meet imposed deadlines, considerable effort is put into tuning models to match the available data. As our understanding of model uncertainty increases, it is apparent that model tuning may give a misleading impression of the total uncertainty in aerosol–cloud–climate effects, which may contribute to persistent model diversity. For the aerosol–cloud system, better progress would be facilitated if the model development cycle allowed more-robust advances to be incorporated and tested in models. This would involve improved representation of basic physics and chemistry and a deeper investigation of model uncertainties and observational constraints.

Acknowledgments

The authors acknowledge Dr. Kristina Pistone for taking meticulous notes during the Colloquium. The Pacific Northwest National Laboratory (PNNL) is operated for the US Department of Energy (DOE) by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830. Work at PNNL was supported by the US DOE Decadal and Regional Climate Prediction using Earth System Models program and by the US DOE Earth System Modeling program.

1 IPCC (2013) Summary for Policymakers. *Climate Change 2013: The Physical Science Basis*, eds Stocker T, et al. (Cambridge Univ Press, New York), pp 1–29.
2 Twomey S (1977) Influence of pollution on shortwave albedo of clouds. *J Atmos Sci* 34(7):1149–1152.
3 Albrecht BA (1989) Aerosols, cloud microphysics, and fractional cloudiness. *Science* 245(4923):1227–1230.
4 Stevens B, Feingold G (2009) Untangling aerosol effects on clouds and precipitation in a buffered system. *Nature* 461(7264):607–613.
5 Christensen MW, Stephens GL (2011) Microphysical and macrophysical responses of marine stratocumulus polluted by underlying ships: Evidence of cloud deepening. *J Geophys Res* 116(D3):D03201.
6 Carslaw KS, et al. (2013) Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature* 503(7474):67–71.
7 Wilcox LJ, Highwood EJ, Booth BBB, Carslaw KS (2015) Quantifying sources of inter-model diversity in the cloud albedo effect. *Geophys Res Lett* 42(5): 1568–1575.
8 Golaz JC, et al. (2011) Sensitivity of the aerosol indirect effect to subgrid variability in the cloud parameterization of the GFDL atmosphere general circulation model AM3. *J Clim* 24(13):3145–3160.
9 Suzuki K, Golaz JC, Stephens GL (2013) Evaluating cloud tuning in a climate model with satellite observations. *Geophys Res Lett* 40(16):4464–4468.
10 McFiggans G, et al. (2006) The effect of physical and chemical aerosol properties on warm cloud droplet activation. *Atmos Chem Phys* 6(9):2593–2649.
11 Farmer DK, Cappa CD, Kreidenweis SM (2015) Atmospheric processes and their controlling influence on cloud condensation nuclei activity. *Chem Rev* 115(10): 4199–4217.
12 Furutani H, Dall'osto M, Roberts GC, Prather KA (2008) Assessment of the relative importance of atmospheric aging on CCN activity derived from field observations. *Atmos Environ* 42(13):3130–3142.
13 Sareen N, Schwier AN, Lathem TL, Nenes A, McNeill VF (2013) Surfactants from the gas phase may promote cloud droplet formation. *Proc Natl Acad Sci USA* 110(8):2723–2728.
14 Fierce L, Riemer N, Bond TC (2013) When is cloud condensation nuclei activity sensitive to particle characteristics at emission? *J Geophys Res* 118(24): 13476–13488.
15 Dusek U, et al. (2006) Size matters more than chemistry for cloud-nucleating ability of aerosol particles. *Science* 312(5778):1375–1378.
16 Cubison MJ, et al. (2008) The influence of chemical composition and mixing state of Los Angeles urban aerosol on CCN number and cloud properties. *Atmos Chem Phys* 8(18):5649–5667.
17 Padro LT, et al. (2012) Mixing state and compositional effects on CCN activity and droplet growth kinetics of size-resolved CCN in an urban environment. *Atmos Chem Phys* 12(21):10239–10255.
18 Collins DB, et al. (2013) Impact of marine biogeochemistry on the chemical mixing state and cloud forming ability of nascent sea spray aerosol. *J Geophys Res* 118(15):8553–8565.
19 George RC, Wood R (2010) Subseasonal variability of low cloud radiative properties over the southeast Pacific Ocean. *Atmos Chem Phys* 10(8):4047–4063.
20 Ackerman AS, Kirkpatrick MP, Stevens DE, Toon OB (2004) The impact of humidity above stratiform clouds on indirect aerosol climate forcing. *Nature* 432(7020): 1014–1017.
21 Koren I, Kaufman YJ, Remer LA, Martins JV (2004) Measurement of the effect of Amazon smoke on inhibition of cloud formation. *Science* 303(5662):1342–1345.
22 Kaufman YJ, Koren I (2006) Smoke and pollution aerosol effect on cloud cover. *Science* 313(5787):655–658.
23 Koren I, Martins JV, Remer LA, Afargan H (2008) Smoke invigoration versus inhibition of clouds over the Amazon. *Science* 321(5891):946–949.
24 Jacobson MZ (2012) Investigating cloud absorption effects: Global absorption properties of black carbon, tar balls, and soil dust in clouds and aerosols. *J Geophys Res* 117(D6):D06025.
25 Ten Hoeve JE, Jacobson MZ, Remer L (2012) Comparing results from a physical model with satellite and in situ observations to determine whether biomass burning aerosols over the Amazon brighten or burn off clouds. *J Geophys Res* 117(D8):D08203.
26 Sullivan SC, Lee D, Oreopoulos L, Nenes A (2016) Role of updraft velocity in temporal variability of global cloud hydrometeor number. *Proc Natl Acad Sci USA* 113:5791–5796.
27 Fridlind AM, et al. (2012) A comparison of TWP-ICE observational data with cloud-resolving model results. *J Geophys Res* 117(D5):D05204.
28 Kazil J, et al. (2011) Modeling chemical and aerosol processes in the transition from closed to open cells during VOCALS-REx. *Atmos Chem Phys* 11(15): 7491–7514.
29 Berner A, Bretherton CS, Wood R, Muhlbauer A (2013) Marine boundary layer cloud regimes and POC formation in an LES coupled to a bulk aerosol scheme. *Atmos Chem Phys* 13(24):12549–12572.
30 Blossey PN, et al. (2013) Marine low cloud sensitivity to an idealized climate change: The CGILS LES intercomparison. *J Adv Model Earth Syst* 5(2):234–258.
31 Miyamoto Y, et al. (2013) Deep moist atmospheric convection in a subkilometer global simulation. *Geophys Res Lett* 40(18):4922–4926.
32 Grabowski WW (2001) Coupling cloud processes with the large-scale dynamics using the cloud-resolving convective parameterization (CRCP). *J Atmos Sci* 58(9): 978–997.
33 Khairoutdinov MF, Randall DA (2001) A cloud-resolving model as a cloud parameterization in the NCAR Community Climate System Model: Preliminary results. *Geophys Res Lett* 28(18):3617–3620.
34 VanZanten M, et al. (2011) Controls on precipitation and cloudiness in simulations of trade-wind cumulus as observed during RICO. *J Adv Model Earth Syst* 3(2): M06001.
35 Bretherton CS, Park S (2009) A new moist turbulence parameterization in the Community Atmosphere Model. *J Clim* 22(12):3422–3448.
36 Bogenschutz PA, et al. (2012) Unified parameterization of the planetary boundary layer and shallow convection with a higher-order turbulence closure in the Community Atmosphere Model: Single-column experiments. *Geosci Model Dev* 5(6):1407–1423.
37 Wang H, Feingold G (2009) Modeling mesoscale cellular structures and drizzle in marine stratocumulus. Part II: The microphysics and dynamics of the boundary regions between open and closed cells. *J Atmos Sci* 66(11):3257–3275.
38 Lohmann U, Feichter J (2005) Global indirect aerosol effects: A review. *Atmos Chem Phys* 5(3):715–737.
39 Hoose C, Mohler O (2012) Heterogeneous ice nucleation on atmospheric aerosols: A review of results from laboratory experiments. *Atmos Chem Phys* 12(20): 9817–9854.
40 Murray BJ, O'Sullivan D, Atkinson JD, Webb ME (2012) Ice nucleation by particles immersed in supercooled cloud droplets. *Chem Soc Rev* 41(19):6519–6554.
41 DeMott PJ, et al. (2010) Predicting global atmospheric ice nuclei distributions and their impacts on climate. *Proc Natl Acad Sci USA* 107(25):11217–11222.
42 Rosenfeld D, et al. (2011) Glaciation temperatures of convective clouds ingesting desert dust, air pollution and smoke from forest fires. *Geophys Res Lett* 38(21): L21804.
43 Burrows SM, Hoose C, Poschl U, Lawrence MG (2013) Ice nuclei in marine air: Bioparticles or dust? *Atmos Chem Phys* 13(1):245–267.
44 Creamean JM, et al. (2013) Dust and biological aerosols from the Sahara and Asia influence precipitation in the western U.S. *Science* 339(6127):1572–1578.
45 Cziczo DJ, et al. (2013) Clarifying the dominant sources and mechanisms of cirrus cloud formation. *Science* 340(6138):1320–1324.
46 Tobo Y, et al. (2014) Organic matter matters for ice nuclei of agricultural soil origin. *Atmos Chem Phys* 14(16):8521–8531.
47 McCluskey CS, et al. (2014) Characteristics of atmospheric ice nucleating particles associated with biomass burning in the US: Prescribed burns and wildfires. *J Geophys Res* 119(17):10458–10470.
48 Prather KA, et al. (2013) Bringing the ocean into the laboratory to probe the chemical complexity of sea spray aerosol. *Proc Natl Acad Sci USA* 110(19):7550–7555.
49 Wilson TW, et al. (2015) A marine biogenic source of atmospheric ice-nucleating particles. *Nature* 525(7568):234–238.
50 DeMott PJ, et al. (2016) Sea spray aerosol as a unique source of ice nucleating particles. *Proc Natl Acad Sci USA* 113:5797–5803.

51 Fan J, et al. (2013) Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds. *Proc Natl Acad Sci USA* 110(48): E4581–E4590.
52 Wang Y, et al. (2014) Assessing the effects of anthropogenic aerosols on Pacific storm track using a multiscale global climate model. *Proc Natl Acad Sci USA* 111(19):6894–6899.
53 Trenberth KE, Fasullo JT (2010) Simulation of present-day and twenty-first-century energy budgets of the Southern Oceans. *J Clim* 23(2):440–454.
54 Ault AP, et al. (2011) Detection of Asian dust in California orographic precipitation. *J Geophys Res* 116(D16):D16205.
55 Fan J, et al. (2014) Aerosol impacts on California winter clouds and precipitation during CalWater 2011: Local pollution versus long-range transported dust. *Atmos Chem Phys* 14(1):81–101.
56 Ackerman AS, et al. (2000) Effects of aerosols on cloud albedo: Evaluation of Twomey's parameterization of cloud susceptibility using measurements of ship tracks. *J Atmos Sci* 57(16):2684–2695.
57 Feingold G, Siebert H (2009) Cloud-aerosol interactions from the micro to the cloud scale. *Clouds in the Perturbed Climate System*, eds Heintzenberg J, Charlson RJ (MIT Press, Cambridge, MA), pp 319–338.
58 Wood R (2007) Cancellation of aerosol indirect effects in marine stratocumulus by cloud thinning. *J Atmos Sci* 64(7):2657–2669.
59 Ghan S, et al. (2016) Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability. *Proc Natl Acad Sci USA* 113:5804–5811.
60 Feingold G, et al. (2016) New approaches to quantifying aerosol influence on the cloud radiative effect. *Proc Natl Acad Sci USA* 113:5812–5819.
61 Lee LA, et al. (2013) The magnitude and causes of uncertainty in global model simulations of cloud condensation nuclei. *Atmos Chem Phys* 13(17):8879–8914.
62 Lee LA, Reddington CL, Carslaw KS (2016) On the relationship between aerosol model uncertainty and radiative forcing uncertainty. *Proc Natl Acad Sci USA* 113:5820–5827.
63 Johnson JS, et al. (2015) Evaluating uncertainty in convective cloud microphysics using statistical emulation. *J Adv Model Earth Syst* 7(1):162–187.
64 Morales-Betancourt R, Nenes A (2014) Understanding the contributions of aerosol properties and parameterization discrepancies to droplet number variability in a global climate model. *Atmos Chem Phys* 14(9):4809–4826.
65 Sullivan SC, Morales-Betancourt R, Barahona D, Nenes A (2016) Understanding cirrus ice crystal number variability for different heterogeneous ice nucleation spectra. *Atmos Chem Phys* 16(4):2611–2629.
66 Stevens B (2013) Aerosols: Uncertain then, irrelevant now. *Nature* 503(7474):47–48.
67 Stevens B (2015) Rethinking the lower bound on aerosol radiative forcing. *J Clim* 28(12):4794–4819.
68 Bollasina MA, Ming Y, Ramaswamy V (2011) Anthropogenic aerosols and the weakening of the South Asian summer monsoon. *Science* 334(6055):502–505.
69 Polson D, Bollasina M, Hegerl GC, Wilcox LJ (2014) Decreased monsoon precipitation in the Northern Hemisphere due to anthropogenic aerosols. *Geophys Res Lett* 41(16):6023–6029.
70 Pahlow M, et al. (2006) Comparison between lidar and nephelometer measurements of aerosol hygroscopicity at the Southern Great Plains Atmospheric Radiation Measurement site. *J Geophys Res* 111(D5):D05S15.
71 Hamilton DS, et al. (2014) Occurrence of pristine aerosol environments on a polluted planet. *Proc Natl Acad Sci USA* 111(52):18466–18471.
72 Koren I, Dagan G, Altaratz O (2014) From aerosol-limited to invigoration of warm convective clouds. *Science* 344(6188):1143–1146.
73 Wood R, et al. (2011) The VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-REx): Goals, platforms, and field operations. *Atmos Chem Phys* 11(2):627–654.
74 Mechoso CR, et al. (2014) Ocean–cloud–atmosphere–land interactions in the southeastern Pacific. *Bull Am Meteorol Soc* 95(3):357–375.
75 Andreae MO (2009) Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions. *Atmos Chem Phys* 9(2):543–556.
76 Kapustin VN, et al. (2006) On the determination of cloud condensation nuclei from satellite: Challenges and possibilities. *J Geophys Res* 111(D4):D04202.
77 Koren I, Oreopoulos L, Feingold G, Remer LA, Altaratz O (2008) How small is a small cloud? *Atmos Chem Phys* 8(14):3855–3864.
78 Zhu Y, Rosenfeld D, Yu X, Li Z (2015) Separating aerosol microphysical effects and satellite measurement artifacts of the relationships between warm rain onset height and aerosol optical depth. *J Geophys Res* 120(15):7726–7736.
79 Rosenfeld D, et al. (2016) Satellite retrieval of cloud condensation nuclei concentrations by using clouds as CCN chambers. *Proc Natl Acad Sci USA* 113:5828–5834.
80 Zheng Y, Rosenfeld D, Li Z (2015) Satellite inference of thermals and cloud-base updraft speeds based on retrieved surface and cloud-base temperatures. *J Atmos Sci* 72(6):2411–2428.
81 Zheng Y, Rosenfeld D (2015) Linear relation between convective cloud base height and updrafts and application to satellite retrievals. *Geophys Res Lett* 42(15): 6485–6491.
82 Zhu Y, et al. (2014) Satellite retrieval of convective cloud base temperature based on the NPP/VIIRS Imager. *Geophys Res Lett* 4(4):1308–1313.
83 Rosenfeld D, Fischman B, Zheng Y, Goren T, Giguzin D (2014) Combined satellite and radar retrievals of drop concentration and CCN at convective cloud base. *Geophys Res Lett* 41(9):3259–3265.
84 Rosenfeld D, et al. (2014) High resolution (375 m) cloud microstructure as seen from the NPP/VIIRS Satellite imager. *Atmos Chem Phys* 14(5):2479–2496.