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The Two-Column Aerosol Project (TCAP) Science Plan

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Abstract

The Two-Column Aerosol Project (TCAP) field campaign will provide a detailed set of observations with which to (1) perform radiative and cloud condensation nuclei (CCN) closure studies, (2) evaluate a new retrieval algorithm for aerosol optical depth (AOD) in the presence of clouds using passive remote sensing, (3) extend a previously developed technique to investigate aerosol indirect effects, and (4) evaluate the performance of a detailed regional-scale model and a more parameterized global-scale model in simulating particle activation and AOD associated with the aging of anthropogenic aerosols. To meet these science objectives, the Atmospheric Radiation Measurement (ARM) Climate Research Facility will deploy the ARM Mobile Facility (AMF) and the Mobile Aerosol Observing System (MAOS) on Cape Cod, Massachusetts, for a 12-month period starting in the summer of 2012 in order to quantify aerosol properties, radiation, and cloud characteristics at a location subject to both clear and cloudy conditions, and clean and polluted conditions. These observations will be supplemented by two aircraft intensive observation periods (IOPs), one in the summer and a second in the winter. Each IOP will deploy one, and possibly two, aircraft depending on available resources. The first aircraft will be equipped with a suite of in situ instrumentation to provide measurements of aerosol optical properties, particle composition and direct-beam irradiance. The second aircraft will fly directly over the first and use a multi-wavelength high spectral resolution lidar (HSRL) and scanning polarimeter to provide continuous optical and cloud properties in the column below.

Each mission will consist of the aircraft making measurements within two columns of air. One column will be located over the AMF/MAOS Cape Cod site, while a second column will be located a few hundred kilometers east of Cape Cod. The in situ aircraft will make a series of stair-step profiles within the first column, followed by sampling within and above the marine boundary layer as it transits to the second, more remote column for a second set of stair-step profiles. The second aircraft will provide concurrent downward profiling measurements from above.

Acronyms and Abbreviations

4STAR	Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric Research
AAF	ARM Aerial Facility
AATS-14	14- channel Ames airborne tracking sunphotometer
ACSM	aerosol chemistry speciation monitor
AERI	atmospheric emitted radiance interferometer
AeroCom	Aerosol Comparisons between Observations and Models
AERONET	Aerosol Robotic Network
AOD	aerosol optical depth
AMF	ARM Mobile Facility
AMS	aerosol mass spectrometer
AP	asymmetry parameter
ARM	Atmospheric Radiation Measurement
AVHRR	Advanced Very High Resolution Radiometer
CAM	Community Atmospheric Model
CAPS	Cloud, Aerosol, and Precipitation Spectrometer
CARES	Carbonaceous Aerosol and Radiative Effects Study
CCN	cloud condensation nuclei
CCNC	cloud condensation nucleus chamber
CDNC	cloud droplet number concentration
CHAPS	Cumulus Humilis Aerosol Processing Study
CO	carbon monoxide
COSAM	Comparison of Large-Scale Atmospheric Sulphate Aerosol Models
CVI	counter-flow virtual impactor
DOE	Department of Energy
EMSL	Environmental Molecular Science Laboratory
FAA	Federal Aviation Administration
GCM	general circulation model
HRSL	high spectral resolution lidar
HTDMA	humidified tandem differential mobility analyzer
IOP	intensive observation period
IPCC	Intergovernmental Panel on Climate Change
IR	infrared
ISDAC	Indirect and Semi-Direct Aerosol Campaign
MAOS	Mobile Aerosol Observing System
MATRIX	Multiconfiguration Aerosol Tracker of Mixing State
MFRSR	multifilter rotating shadowband radiometer

MILAGRO	Megacity Initiative: Local and Global Research Observations
MOSAIC	Model for Simulating Aerosol Interactions and Chemistry
MVCO	Martha's Vineyard Coastal Observatory
MWR	microwave radiometer
NIMFR	normal incidence multifilter radiometer
PAS	photo-acoustic spectroscopy
PASS	photo-acoustic soot photometer
PI	principal investigator
PILS-IC-WSOC	Particle In Liquid Sampler-Ion Chromatography-Water Soluble Organic Carbon
PNNL	Pacific Northwest National Laboratory
PSAP	particle soot absorption photometer
PTI	photothermal interferometry
RSP	research scanning polarimeter
SP2	Single Particle Soot Photometer
SSA	single scattering albedo
TCAP	Two-Column Aerosol Project
TOA	top of atmosphere
TSI	total sky imager
USCCSP	U.S. Climate Change Science Program

Contents

Figures

Tables

1.0 Introduction

There is a high degree of uncertainty in the understanding of how atmospheric aerosols affect climate (e.g., IPCC 2007, USCCSP 2009, and references therein), including the direct absorption or scattering of radiation ("aerosol direct radiative effects"), and cloud precipitation efficiency, cloud brightness, and cloud lifetime ("aerosol indirect radiative effects"). Adding to the complexity of the problem is the large variability of aerosol composition, hygroscopicity, and particles ranging in size from nanometers to micrometers (e.g., Seinfeld and Pandis 1998). As a result of the computational resources needed to describe this complex variability, a number of simplifications are built into models used to assess the effects of aerosols on radiative forcing and the hydrological cycle. The immediate result of TCAP will be a self-consistent set of measurements with which to evaluate and improve many of these assumptions and thereby reduce the uncertainty in understanding of aerosol-climate interactions.

1.1 Aerosol Radiative Properties (direct effects)

Mixing state refers to the distribution of compounds among a population of particles. In an external mixture, each particle is composed of a single compound. In an internal mixture, each particle of a given size has the same mixture of compounds. Mixing state affects the optical properties of particles, as can be seen in Figure 1, which shows the variation in mass scattering efficiencies for spheres of pure $(NH_4)_2SO_4$, NH_4NO_3, carbon, water, and silica. Hand and Malm (2007), in a comprehensive review of aerosol mass scattering efficiencies evaluated since 1990, note that mass scattering is dependent on particle composition and size distribution, with the refractive index being strongly dependent on composition. Kinne et al. (2006) found in their intercomparison of aerosol optical properties from twenty models that there is a "surprisingly" good agreement between models when predicting AOD, which is a function of mass scattering. However, they argue that the level of agreement of these models was fortuitous because the chemical constituents that compose the aerosol loading differ considerably between models. Furthermore, it is not only composition but vertical distribution that is in need of refinement. In the Comparison of Large-Scale Atmospheric Sulphate Aerosol Models (COSAM) study, which compared 11 general circulation and chemical transport models, Barrie et al. (2001) found an order of magnitude variation in the simulated distribution of combined "SO_x" (defined as the sum of sulfate aerosols + SO_2 gas). These gaps and uncertainty are relevant to climate modeling work because aerosol composition and distribution is closely linked to AOD.

Other observations and modeling studies show that mixing state and morphology of the aerosol constituents can have a strong influence on the optical (for example, Bond et al. 2006, Fuller et al. 1999, Hand et al. 2005, Hopkins et al. 2007) and activation properties of aerosols through their effect on mass scattering and on their effectiveness as CCN (Prenni et al. 2007, Petters et al. 2006). Yet despite the development of detailed chemical process models (e.g., Pacific Northwest National Laboratory's Model for Simulating Aerosol Interactions and Chemistry [MOSAIC] (Zaveri et al. 2008) or Goddard's Multiconfiguration Aerosol Tracker of Mixing State [MATRIX] (Bauer et al., 2008)), there remains a large element of uncertainty in the descriptions of the mixing state of particles and their effect on climate due to a paucity of real-world observations against which these models can be constrained and evaluated.

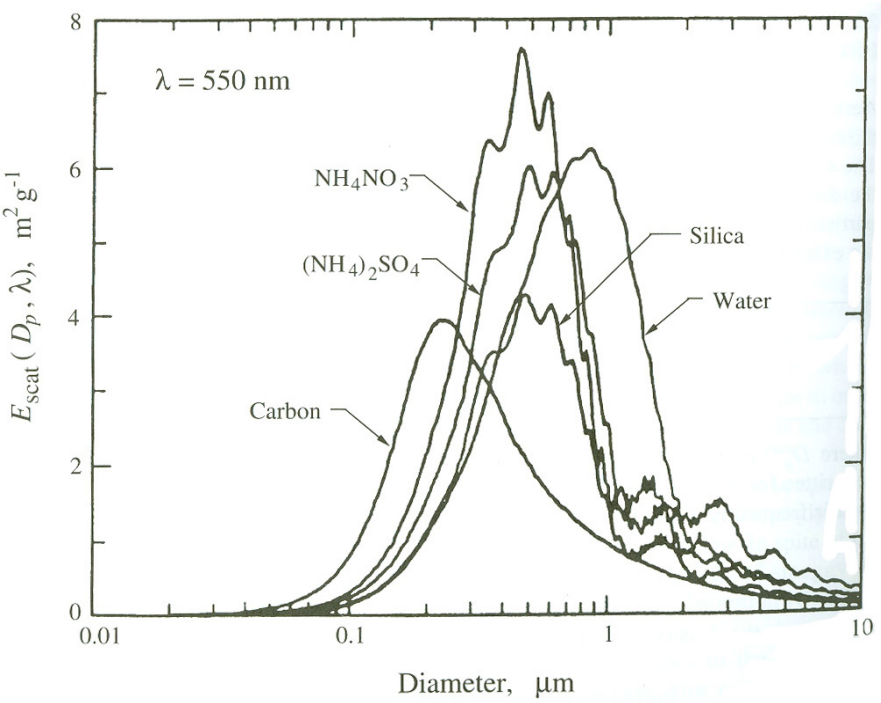

Figure 1. Mass scattering efficiencies of homogeneous spheres (see text). From Seinfeld and Pandis (1998).

Several recent studies have explored in detail the role of the mixing state for CCN closure studies (Ervens et al. 2009, Wang et al. 2010). These studies concluded that it is only when spatially close to pollution sources that hydrophobic freshly emitted particles are externally mixed and that CCN number concentrations are overestimated if internal mixtures are assumed. In contrast to this result, current large-scale models use fixed time-scales of ~ 1–2 days to convert hydrophobic particles into hygroscopic particles (and thus potential CCN). Although the recent work by Ervens et al. (2009) and Wang et al. (2010) suggest that these time scales are significantly shorter (~ hours), the extent to which these time scales depend on specific conditions, such as photochemical activity, location, and season, has not yet been fully explored.

Closely related to the interest in particle composition and microphysical properties is the geographical extent to which continental aerosols influence the optical depth over marine areas and the ability of regional- and global-scale models to reproduce this feature off the coast of continents, including North America. As noted above, Kinne et al. (2006) in their study of 20 global-scale models, found generally good agreement in AOD, but only for annual global AOD; no such agreement existed over smaller geographical areas. Along the same lines, IPCC (2007) presents an evaluation of simulated AOD from North America. Such studies have produced a wide range of radiative forcing estimates at both the top of the atmosphere (TOA) and at the surface, with surface cooling being about 37% larger than top-of-the-atmosphere cooling. These differences are much smaller than the measurement-based estimate of surface and TOA difference of 60%. USCCSP (2009, page 41) summarizes these problems by noting that "…on a global average, the measurement based estimates of aerosol direct radiative forcing are 55-80% greater than the model-based estimates. The differences are even larger on regional scales." Bellouin et al. (2005) showed a significant discrepancy between the clear-sky direct radiative forcing measured with satellite and that estimated from models (-10.9 Wm^{-2} vs. -0.5 to -0.9Wm^{-2}), while a number of other studies

(e.g., Kinne et al. 2006, Textor et al. 2006) argue that uncertainty in calculations of AOD results in uncertainties in estimates of radiative forcing. More recently, Myhre (2009) has suggested that earlier discrepancies between simulated and observed forcing may be explained by a relatively greater increase in anthropogenic black carbon (BC) aerosols within the total mix of anthropogenic aerosols. Moreover, for direct forcing, the accuracy requirements for aerosol optical properties are stringent, as pointed out in McComiskey et al. (2008), which shows the need to predict AOD to within 0.02 and single scattering albedo (SSA) to within 0.02 in order to reduce local radiative forcing uncertainties to less than 1 Wm^{-2}. For this reason, we are applying state-of-the-art instrumentation and long averaging times to the requisite measurements.

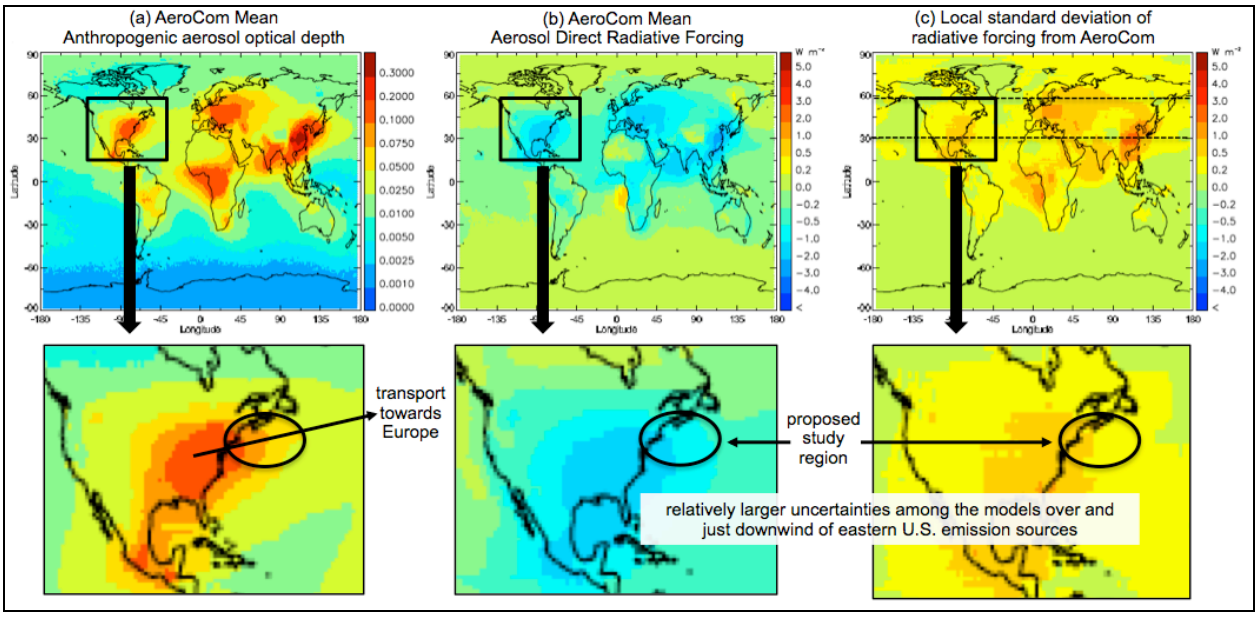

Figure 2. Annual (a) mean anthropogenic AOD, (b) mean direct radiative forcing, and (c) standard deviation of the direct radiative forcing from nine AeroCom models (Aerosol Comparisons between Observations and Models). Adapted from Schultz et al. 2006.

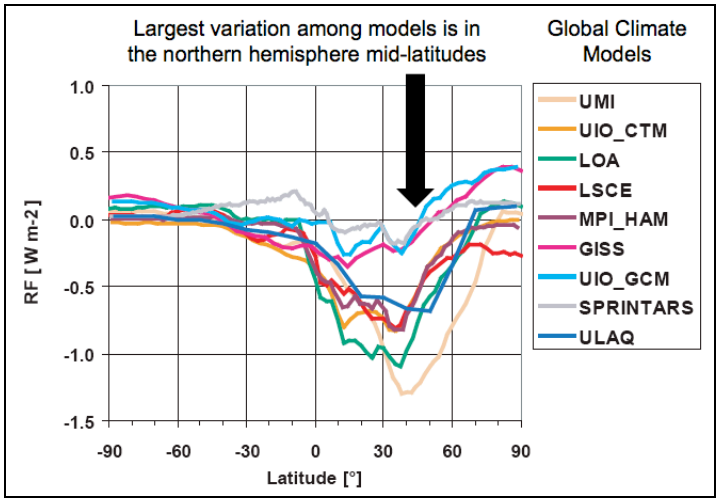

Figure 3. Zonal distribution of total direct radiative forcing for all sky conditions (adapted from Schultz et al. 2006).

Schultz et al. (2006) presented a comparison of several general circulation model predictions of aerosol radiative forcing, illustrating the need to focus on measuring aerosol properties off the eastern coast of North America (Figure 2, above). As expected, regions with the highest anthropogenic emissions also have the highest aerosol loading (Figure 2a) and consequently the greatest cooling associated with aerosol radiative forcing (Figure 2b). The models produce a gradual decrease in AOD and aerosol radiative forcing over the northeastern Atlantic Ocean as particles are transported eastwards towards Europe. However, there are relatively large variations in the magnitude of the predicted aerosol radiative forcing in this region (Figure 2c). The zonal-averaged direct radiative forcing, shown in Figure 3, shows that the midlatitudes (including the proposed study region) have the largest variations of forcing among the climate models.

Recognizing that satellite observations form the basis for many model evaluations, our research team is also interested in the apparent enhancement of AOD in the vicinity of clouds detected by both airborne remote sensing (Su et al. 2008, Redemann et al. 2009) and satellite measurements (Redemann et al. 2009). It has been established that satellite observations of AOD are hampered by issues of effective cloud clearing (Coakley et al. 2005) (i.e., cloud contamination of pixels assumed to be cloud-free) and enhanced scattering in cloud-free pixels due to scattering of sunlight reflected from nearby clouds (Marshak et al. 2008, Kassianov et al. 2009). A recent study by Twohy et al. (2009) used data acquired during the INDOEX campaign to estimate the potential increase in radiative forcing over oceans caused by enhanced scattering of aerosols near clouds. In that study, Twohy et al. estimated the enhancement to scattering and radiative forcing due to swelling of the ambient aerosol in reaction to increasing RH with decreasing distance from cloud. They concluded that aerosol scattering and radiative forcing are larger by 35–65% in partly cloudy environments over that inferred for large (>20 km) cloud-free ocean areas. However, this important study employed many simplifying assumptions with a goal of bounding the effect rather than quantifying it more precisely or exploring other potential contributions such as in-cloud particle production and cloud processing of aerosol. Koren et al. (2007), using AERONET data, determined that visible and near-infrared (IR) AOD was higher by 13% and 22% respectively near cloud relative to AOD away from cloud, and that 30–60% of the free atmosphere is affected by this phenomenon. Using airborne lidar measurements, Su et al. (2008) found that AOD increased by 8% to 17% near cloud (~100 m) relative to measurements made further from cloud (~4.5 km). These studies, in conjunction with the observation that 25% of all MODIS cloud-free pixels are within 500 m of a cloud, and 38% are within 1 km (Lorraine Remer, private correspondence, 2009) imply that this effect can occur over a significant fraction of the Earth with a potentially large increase in global radiative forcing. High spatial resolution observations of the variation of AOD and aerosol microphysical and optical properties as a function of distance from cloud are needed to understand and model the radiative forcing in cloudy environments that cover much of the globe. Making measurements with which to evaluate a recently developed technique to improve AOD calculations in the vicinity of clouds is one of the main operational goals for our field campaign and is described later in this proposal.

1.2 Aerosol Activation (indirect effects)

Many aerosol indirect radiative effects stem from the ability of particles to serve as CCN. Twomey (1977) was one of the first investigators to relate an increase in the number of aerosols to a decrease in cloud drop effective radius, r_{eff}, resulting in an increase in cloud optical depth and albedo. A number of factors have since been implicated in relating how cloud albedo varies with increases in aerosol loading, such as the combination of increased cloud absorption of shortwave radiation and enhanced evaporation of cloud

droplets in polluted clouds (Ackerman et al. 2000, Coakley and Walsh 2002). Liu and Daum (2002) showed a systematic increase in the width (e.g. dispersion) of the cloud droplet spectrum associated with increasing number of anthropogenic aerosols. The associated increase in effective drop radius was postulated to negate the effect of increased aerosol concentrations on cloud albedo. Inclusion of this dispersion in a general circulation model (GCM) was shown to reduce the magnitude of the first indirect effect by between 12–35% (Rotstayn and Liu 2003). The simultaneous increase in the relative dispersion and droplet concentration with higher aerosol loading may be due to a number of reasons, including complex chemical heterogeneity of anthropogenic aerosols, broader size distribution, and larger number of smaller drops in polluted clouds (Liu and Daum 2002). Detailed, size-resolved measurements of the chemical composition of aerosols and mixing state are needed to understand these effects and form a central part of the field campaign described in this proposal.

Ghan and Schwartz (2007), in their overview article on aerosol properties and processes, note that "4th Generation Models", defined as models used in the 4th IPCC Report (2007), assume aerosols to be externally mixed. Particles that are emitted directly to the atmosphere ("primary particles") usually are externally mixed but are then subject to coagulation and changes associated with internal chemical processes and the condensation of semi-volatile gases to their surface. These "aging" processes result in a spectrum of mixing-states with a range of climate-affecting hygroscopic properties. These changes are thought to play a major role in the ability of particles to act as CCN. Recent studies have shown that particle aging occurs primarily by physical processes, i.e., coagulation and condensation of semi-volatile species that result in both an increase of particle size and hygroscopic mass. In contrast, chemical aging, i.e., the oxidation of particulate matter resulting in more hygroscopic products, has been shown to be too slow to efficiently convert aerosol mass (Petters et al. 2006, DeGouw and Jimenez 2009).

The results of CCN closure experiments, in which predictions of CCN concentrations are compared with measurements, are highly variable, even in detailed process modeling studies (e.g., Chuang et al. 2000, Cantrell et al. 2001, Stroud et al. 2007). Depending on air mass properties, understanding the particle mixing state is thought to play a key role in the success of these closure studies (see, for example, Ervens et al. 2009 and references therein). Medina et al. (2007), after examining many such CCN closure studies, concluded that there is a strong need for closure studies that cover a wide range of seasons and aerosol types and questioned many simplifying assumptions made in models, including aerosol mixing state and variations of composition with size. They also draw attention to the need for closure studies that include direct measurements of aerosol mixing state and size to allow an improved understanding of the significance of model assumptions used to assess CCN spectra and the resulting properties of clouds forming from these activated particles.

Several studies have shown that uncertainties in CCN studies result in many cases with much smaller errors in predicted cloud droplet numbers (e.g., Sotiropoulou et al. 2006, Ervens et al. 2009). However, simulations done in highly polluted regions (Riverside, California) have shown that for fresh, externally mixed aerosol, the error in cloud droplet number might be as high as for a CCN closure study. Again, this emphasizes the importance of mixing state and the need for its inclusion when studying aerosol effects on clouds.

1.3 Project Description

The preceding discussions have highlighted the importance of chemical composition and mixing state on the radiative properties of the atmosphere and the properties of particle activation. The overarching goal of TCAP is to provide measurements of the radiative, chemical, and microphysical properties of particles within one of two columns of air over 12 months and within both columns of air and a low-altitude transect connecting the two columns during one summer and one winter campaign. The data will be used to evaluate and improve the representation of these observations in both small-scale process models and large-scale climate models. Our goals for the aircraft IOPs and the 12-month deployment of the AMF and MAOS are tightly linked to specific goals of our research team, all of which are focused on quantifying the local radiative uncertainties in a GCM, and which are described in more detail in Section 2 ("Scientific Objectives") of this proposal. We have five broadly defined operational goals associated with the field study.

Aircraft IOPs Goals

- To measure slab-AOD within clear and cloudy columns of air, in both clean and dirty air, at different altitudes in the atmosphere.
- To measure columnar AOD and other key aerosol properties in the vicinity of clouds using aircraft and surface-based remote sensors.
- To measure particle mixing state and activation potential via sampling through a counterflow virtual impactor (CVI) as an aircraft flies through clouds within and directly above the marine boundary layer off the coast of North America.

AMF/MAOS Goals

- To measure the annual cycle of aerosol mixing state, aerosol optical properties, cloud macroscale and microscale properties, and radiation.
- To measure the annual cycle of columnar AOD in both clear and partly cloudy conditions during both clean and polluted periods.

To meet these operational goals, the ARM Facility is providing support for a campaign to include (a) the deployment of the AMF/MAOS for 12 months starting in the summer of 2012, and (b) two aircraft IOPs, one in the summer and one in the winter. The sampling domain will encompass one ~8-km deep column of air over Cape Cod (Massachusetts) and a second ~8-km deep column of air in a semi-remote marine environment to the east of Cape Cod. This depth will be determined by the nominal sampling altitude of the (potential) second, high-altitude aircraft (9 km) with measurements of backscatter and extinction extending up to ~1 km below the aircraft. A four-season deployment of the AMF and (possibly) MAOS at Cape Cod will provide measurements spanning all four seasons of the campaign. The summer IOP is designed to characterize particle composition during warm, photochemically active, relatively cloud-free periods of the year, while the winter IOP is intended to characterize particle composition during cloudy, less photochemically active periods having a different mix of emissions than in the summer.

The basis for requesting two aircraft IOPs in conjunction with an extended deployment of the AMF is simply that AOD near the east coast of North America has a large annual cycle, and understanding the details of AOD for one season does not provide assurance that the same processes dominate in another

season. Along the east coast of North America, the AOD is generally the smallest during the winter and largest during the summer. For example, at the Martha's Vineyard Coastal Observatory (MVCO) Aerosol Robotic Network (AERONET), the summertime AOD values are approximately 80% larger and show much greater variability than their wintertime values (Figure 4). Similar results have been reported in other studies that were based on both surface and satellite measurements (e.g., Prados et al. 2007, Stegmann 2004).

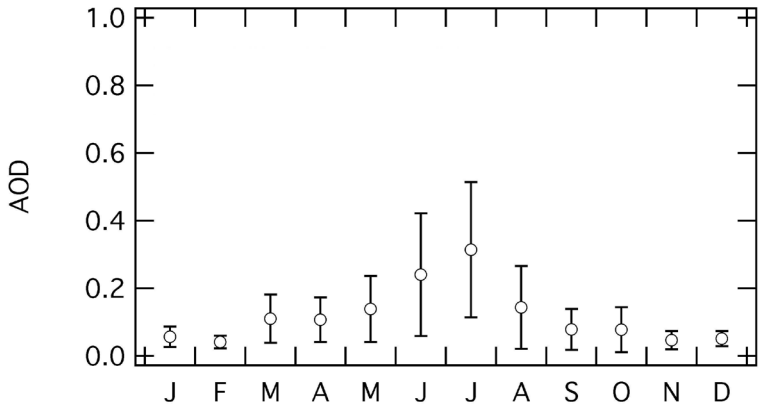

Figure 4. Monthly average AOD at 500 nm observed at the MVCO AERONET site near Martha's Vineyard based on Level 2 data collected during 2004, 2005, and 2008.

Seasonal changes in the AOD could be attributed to changes in the particle loading and do not necessarily indicate a systematic change in the aerosol optical properties. Using retrievals from the Advanced Very High Resolution Radiometer (AVHRR), Higurashi et al. (2000) showed systematic changes in the Ångström Exponent off of the entire east coast of the United States as a function of season, while Stegmann (2004) showed seasonal changes in the Ångström Exponent over a number of specific locations in the north Atlantic. Both studies found the largest values of Ångström Exponent near the east coast of North America during the spring and summer, indicating the presence of an increased number of small particles relative to the number of large particles, and reduced values of Ångström Exponent in the winter and fall. Such behavior can possibly be a result of the increase in accumulation mode particles associated with gas-to-particles conversion. The seasonal changes of the Ångström Exponent indicate changes in the particle size distribution and seasonal variations in the aerosol properties; the change in AOD is not associated simply with an increase in particle loading. These seasonal changes may also reflect changes in the ability of particles to act as CCN. Unfortunately, measurements of only the AOD and Ångström Exponent, as exist from the AERONET or satellite based observations, preclude a determination of the role of each of these factors.

The preceding discussions present arguments for a two-season aircraft campaign that, when combined, will allow for (a) closure studies associated with the different summertime and wintertime aerosol populations, (b) a detailed comparison of the summertime and wintertime aerosol chemical composition and mixing state, (c) evaluating the seasonal dependence of time scales associated with the transition from externally to internally mixed particles, and (d) complement the long-term observations made with the AMF/MAOS.

In the following sections we will describe the sampling strategy and motivation for both the AMF/MAOS deployment and the aircraft IOPs.

1.3.1 Sampling Strategy: AMF/MAOS

The combination of the AMF and MAOS will provide measurements of the entire annual cycle of particle composition and optical properties, columnar AOD, cloud microphysical and macrophysical properties, and radiation. We selected Cape Cod for deployment of the AMF and MAOS because it is not subject to large sources of local emissions, as would be a site within or adjacent to Boston Harbor, yet it is within the circulation pattern of the Bermuda High such that it will receive processed emissions from the well-known "Ozone Corridor" of eastern North America. During the course of 12 months we also expect Cape Cod to be subject to a variety of synoptic flow conditions that will provide a corresponding variety of aerosol and cloud conditions, making it the ideal location for an extended deployment which in turn will let us include seasonality and air mass properties in our follow-up research activities (described in Section 2).

The measurements from AMF's multifilter rotating shadowband radiometer (MFRSR) and normal incidence multifilter radiometer (NIMFR) will provide time series of columnar aerosol optical depth in clear and partly cloudy conditions with hourly or better time resolution. One use of these observations will be through a CCN Chemical Closure study (see Section 2) that will be completed using the particle size distributions, chemical composition, and CCN number concentration measured by the instruments included in the MAOS. This particular study will evaluate long-term CCN closure capabilities over the AMF/MAOS site. While this study will be limited to conditions near the surface, the 12-month deployment will enable us to complete the study for a wide range of synoptic weather patterns.

Data from the scanning cloud radar include time series of cloud boundaries, cloud updraft (or downdraft) strength, as well as retrievals of cloud droplet number concentration (CDNC) and cloud droplet effective radius. Many of these retrievals have been used in previous ARM studies but will be augmented here by the detailed measurements associated with the MAOS. These data will be used to investigate cases of the first aerosol indirect effect in the clouds over Cape Cod (see Section 2). Data from the scanning cloud radar can be used to retrieve the liquid water content and effective radius (e.g. Lio and Sassen 1994) in addition to determining the updraft strength and cloud boundaries. With additional assumptions about the size distribution, the cloud droplet number concentration can be retrieved as well (e.g., Frisch et al. 1995). For completely overcast conditions, information about the cloud optical depth measured with a radiometer can be used as a further constraint. The atmospheric emitted radiance interferometer (AERI) can also be used to retrieve droplet effective radius as long as the optical depth of the cloud is less than 60 g m^{-2} (D Turner, personal communication).

1.3.2 Sampling Strategy: Aircraft IOPs

We will sample within two columns of air, each corresponding to simulated columns in a high-resolution regional-scale model (Section 2). One column will be over Cape Cod, Massachusetts, where we will deploy the AMF/MAOS systems, with the other column east of Cape Cod in a semi-remote, open-ocean environment (see Figure 5). The coastal location of the proposed campaign will allow repeated sampling to be done that will sometimes be associated with fresh emissions from Boston, at other times with processed emissions coming from along the coastal "Ozone Corridor" of North America, or with clean maritime air during easterly flow.

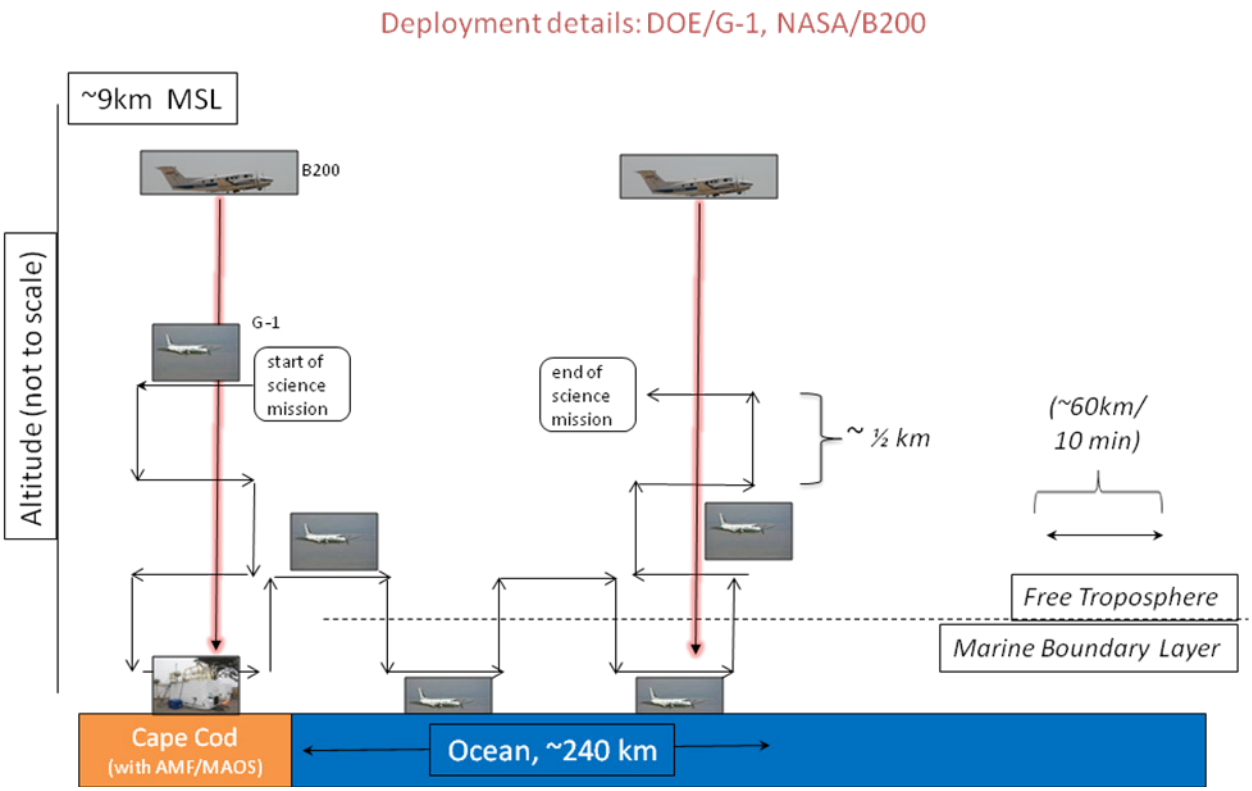

Figure 5. Proposed flight plan of the G-1 and B200 aircraft (or alternative platforms), showing stair-step profiles over the two columns of air and sampling strategy during transit between the columns. Each horizontal arrow represents 10 minutes of flight time. Although the nominal sampling altitude of the B200 is ~9 km, the backscatter profiles extend up from the surface to about 8 km, and the extinction profiles extend up to about 7.5 km.

The G-1 will serve as a platform for a number of in situ instruments needed to characterize the local particle and radiative fields. The second aircraft (the NASA B200) will serve as a platform for two state-of-the-science remote sensing instruments (described below). Our basic flight plan will begin with the G-1 making a series of stair-step profiles over the AMF/MAOS Cape Cod site while the B200 provides continuous profiles of extinction and cloud properties within this column. Each step of the profile will be separated by ~0.5 km. Upon completion of these step profiles, the G-1 will fly to the base of the second (maritime) column of air while collecting information through the cloud condensation nuclei (CCN)/counter-flow virtual impactor (CVI)/mass spectrometer system (described below). This leg will consist of a number of segments alternating between sampling in the marine boundary layer and the free troposphere. Upon arriving at the base of the second (marine) column, the G-1 will begin another stair-step profile with the B200 again providing continuous profiles of extinction and cloud properties within this column. Both aircraft will then return to their base of operation after sampling is completed in the second column. Onboard instrumentation for both aircraft will allow for measurements during both clear-sky and cloudy-sky conditions, both of which are anticipated during the course of the two proposed IOPs.

This one plan, repeated throughout the course of the two IOPS, will provide the information needed to meet the research goals described in Section 2 of this science plan. Our experience suggests that this approach yields a higher return of scientifically useful data and is more likely to receive Federal Aviation Administration (FAA) and military approval in the busy airspace near the coast than a complex set of

strategies that vary from day to day depending on various scientific goal(s) or local weather conditions. Because we are interested primarily in the particle properties within the columns and the relation of these properties to the corresponding optical and cloud properties at the same time and location, we do not plan to make measurements that will explicitly address the evolution of particles as they are transported from any one of the many sources along the east coast of North America. The goal of this study is not to track plumes in a Lagrangian sense. Rather, our interest is to characterize the aerosol plumes at increasing distances from the coast, with an emphasis on simultaneous measurements related to the optical depth and properties of CCN within this domain.

Assuming an aircraft sampling speed of approximately 100 ms^{-1} and a total flight time per mission of about 3 hours (depending on final payload), we plan to have two profiles of four 10-minute legs, giving a time of ~80 minutes to do both stair-step profiles. With three 60-km legs separating the profiles (Figure 5), we will then have a total of 11 legs per mission for a total of about 110 minutes, which will allow ~70 minutes (total of three hours) for changes in altitudes, turns, and a safe return to the base of operations. These are scaling arguments presented only to show that the time and spatial scales proposed for our flights are realistic. Specific details of the flight plan are being worked out with the ARM Facility managers, pilots of the aircraft, military flight controllers, and the FAA.

1.4 Instrumentation Overview: AMF/MAOS

The AMF instrumentation deployed during this study will include the scanning cloud radar, radiation measurements, and lidars. The scanning cloud radar will provide detailed information about the cloud macrophysical and microphysical properties. These measurements will be complemented by the microwave radiometer (MWR), which will provide estimates of the columnar water vapor and liquid water. The total sky imager (TSI) will provide images of the cloud field during daylight hours and can be used to provide an estimate of the cloud fraction. The lidar will provide additional data on cloud-base height as well as aerosol backscatter, and in the case of a HSRL, extinction and AOD. A Doppler lidar will provide information on the sub-cloud updraft velocity, as well as estimates of aerosol backscatter. The MFRSR will be used to measure spectrally resolved irradiances. During clear conditions, the MFRSR can be used to measure AOD. Research currently underway at PNNL is examining ways to extend the MFRSR AOD retrievals to partly cloudy conditions (Section 2). In overcast conditions, the MFRSR can be used to estimate the cloud optical depth and r_{eff}. The AERI will be used to provide profiles of temperature and humidity at high temporal resolution. The AERI will provide estimates of r_{eff} for water clouds with relatively small optical depth. In total, the deployment of the AMF yields two or three independent ways for estimating r_{eff}, using retrievals from the cloud radar, MFRSR (during overcast conditions), and the AERI. Estimates of r_{eff} from all three methods will be compared. During the aircraft IOPs, the cloud microphysical properties retrieved from the various sensors will be compared size distributions measured by the aircraft. The AERI has also been used to derive the columnar amount of carbon monoxide (CO) (Yurganov et al. 2010), but the retrieval is difficult when there are large amounts of water vapor, or in partly cloudy conditions, so while we will look to using the AERI in this fashion, we will not count on its use for our analysis.

The MAOS provides measurements of the aerosol chemical and optical properties. These measurements will allow us to document changes in number of aerosol parameters as a function of season. The single particle soot photometer (SP2) provides critical measurements of not only the amount of black carbon in

individual particles, but also information about the mixing state of the particles with respect to the amount of black carbon mixed with other components. Thus we will be able to document changes in the aerosol composition as a function of season. The aerosol chemistry speciation monitor (ACSM) will be used to measure the non-refractory aerosol mass (including ammonium, nitrate, sulfate, chloride, and organic mass). The ACSM does not see refractory materials, such as sea-salt particles and black carbon. To address this shortcoming, the Particle In Liquid Sampler-Ion Chromatography-Water Soluble Organic Carbon (PILS-IC-WSOC) will be used to estimate the mass loading of sea-salt particles, and the SP2 will be used to estimate the amount of black carbon. Aerosol optical properties will be measured with a 3-wavelength nephelometer. Aerosol absorption will be measured with a particle soot absorption photometer (PSAP), photo-acoustic soot photometer (PASS), and SP2. The light scattering and absorption measurements will be combined to provide the SSA. The hydroscopic growth factor (representing changes in aerosol scattering associated with changes in relative humidity) will also be made using the MAOS humidigraph. Aerosol size distributions will be measured for sizes ranging from 15 nm to 1 μm.

1.5 Aircraft IOPs

The G-1 will provide in situ measurements of aerosol physical and radiative properties, size, and direct measurements of the mixing states of aerosols with an airborne particle mass spectrometer capable of providing information on the size and mixing state of individual particles with sizes down to ~100 nm. The G-1 platform will also include (among other instruments) a newly developed sun-tracking/sky-scanning photometer ("4STAR") that measures the optical depth from the altitude of the aircraft to the top of the atmosphere. Measurements of effective cloud droplet radius (r_{eff}) from both the AMF and the airborne Cloud, Aerosol, and Precipitation Spectrometer (CAPS) probe during cloudy conditions will be an integral part of the model evaluation studies proposed as part of our post-campaign analysis to quantify aerosol indirect effects (see Section 2).

During the 2007 Cumulus Humilis Aerosol Processing Study (CHAPS, described by Berg et al., 2009a) an aerosol mass spectrometer (AMS) was plumbed to allow sampling through either an isokinetic inlet (which precluded large particles) or a counter-flow virtual impactor (CVI) which only allowed cloud droplets into the AMS. For the campaign described here, we plan to add a cloud condensation nucleus chamber (CCNC), the Single Particle "SPLAT II" mass spectrometer (described below), and an Aerodyne AMS so that under clear-sky conditions we can characterize the chemical composition of particles that can act as CCN at specified supersaturations. The sampling airstream for this system will parallel the design used during CHAPS that allowed easy switching between the isokinetic and CVI lines.

The Aerodyne AMS instrument flown during CHAPS measured only non-refractory particle components (e.g., sulfates and organics). SPLAT II measures these and refractory materials (e.g., sea salt, black carbon, and mineral dust). Because the focus of our CCN study is on the activation of aerosols, which requires a universal detection of aerosol components, we have chosen the SPLAT II instrument for this research project. However, the Aerodyne AMS provides quantitative measurements of sulfate, organics, ammonium, nitrate, and chlorides, which are needed for our high-resolution modeling study; hence, we are requesting both mass spectrometers. We note that both a single-particle mass spectrometer (similar to SPLAT II) and an Aerodyne MS were flown during the 2010 Carbonaceous Aerosol and Radiative Effects Study (CARES) study. We mention this because data collected during CARES will let us compare the performance of these instruments, and with such a comparison we will be able to compare SPLAT II data from our proposed campaign with observations from previous studies (e.g., CHAPS).

Whereas the Aerodyne mass spectrometers are relatively well known throughout the scientific community (e.g., see http://www.aerodyne.com/products/aerosol_mass_spectrometer.htm), less information is available on SPLAT II, so we present a short overview here. Characterization of particle size, composition, and morphology will be accomplished with this instrument, a tool to come from DOE's Environmental Molecular Science Laboratory (Zelenyuk et al. 2009). SPLAT II provides in real time the size and internal composition of individual particles in the 50-nm to 3-µm size range, characterizing both refractory and non-refractory aerosol fractions in each particle. It sizes up to 2000 particles per second and chemically characterizes 20–50 particles per second. In addition, this instrument provides information on aerosol number concentration and asphericity with 1-second temporal resolution and size distribution with resolution of 60 seconds or less, depending on aerosol loading. These measurements also yield average aerosol density and densities for particle classes with different mixing states (McFarquhar et al. 2010; Vaden et al. 2010a, 2010b). We have had experience in making airborne measurements with SPLAT II during the 2008 Indirect and Semi-Direct Aerosol Campaign (ISDAC) (Zelenyuk et al. 2009).

A key component of the instrument payload will be a dual-column CCNC, described in Roberts and Nenes (2005). The sampling strategy will be to have one CCNC column scan stepwise over a range of supersaturation values; the number density of drop-forming aerosols will be measured at a distinct saturation for ~4 minutes. Then a new saturation will be set (~1 min), and drop-forming aerosol number density will then be measured at the new saturation for ~5 minutes. A total of three saturations will be used for a total scan time of ~15 minutes; for this reason, level flight legs are advantageous. The second CCNC column will be set at a single supersaturation that will allow for determination of features on a shorter (seconds) time scale.

We plan to conduct a second, somewhat novel experiment using the CCNC during the campaign. SPLAT II will be used to characterize the ambient aerosol and the residue of cloud water droplets using the CVI. Alternately, the droplet output of the CCNC will be separated from the unactivated aerosol using a pumped CVI (Boulter et al. 2006), and this flow will be used as input for the single particle mass spectrometer. In this way, we will be able to produce a chemical closure experiment on droplet formation; measurements of the ambient aerosol, the droplet residue, and the CCN-forming aerosol as a function of supersaturation will be performed. It is noteworthy that this concept has been utilized in ground-based studies (such as one performed by Slowik and Abbatt in 2007 (http://www.chem.utoronto.ca/staff/ABBATT/Egbert2007/Egbert%20Study.htm), but never, to our knowledge, from an aircraft or in a marine or free-tropospheric environment. These two CCN studies are designed to be complementary, with the decision to operate either the standard CVI or pumped CVI to be made before each flight.

The Radiation Closure study will combine in situ measurements made with the G-1 and data collected with the recently developed Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric Research (4STAR). This study will make extensive use of measurements from the single particle mass spectrometer to be made during the stair-step profiles within our two columns of interest. Measurements to be made during the these profiles will quantify aerosol light scattering and absorption, mixing state, the presence of water-soluble organic carbon (through the PILS instrumentation), total aerosol mass, and hygroscopic growth factors.

4STAR will directly yield atmospheric direct-beam transmittance, with the stair-step ascending or descending aircraft flight patterns providing optical depth and extinction between the stair-step segments. Full spectral measurements provide improved retrieval of trace gases (e.g., Cede et al. 2006, 2008;

Chance et al. 1998, 2002; Kroon et al. 2008; Pilewskie et al. 2000; Shetter and Muller 1999; Shetter et al. 2003; Swartz et al. 2005) and improve aerosol optical depth and aerosol extinction measurements (as residuals of the total optical depth after subtraction of contributions from molecular and trace gases). The addition of sky-scanning measurements will enable retrievals of size distribution, complex refractive index, and shape (Dubovik and King 2000; Dubovik et al. 2000)—all provided by the first AERONET-like measurements from aircraft. In addition, modeling studies (Barker et al. 2002) have shown that cloud property retrievals from airborne spectral zenith radiance coupled with upwelling spectral flux are more robustly constrained than studies based on the ground-based counterparts. Marshak et al. (2008) have presented analytical techniques suited for use with airborne measurements under broken or partly skies, which will be considered for use in our follow-up analysis. The primary motivation for carrying 4STAR is to have an exact match in "slabs" sampled by the 4STAR with the in situ instruments aboard the G-1, which will let us greatly tighten closure results and eliminate sampling discrepancies associated with comparisons of airborne measurements with either ground-based or space-based remote sensing products.

Our Columnar Radiative Closure Study (Section 2) will make extensive use of the two instruments deployed on the King Air B200. The first is NASA's second-generation HSRL (HSRL-2), which uses the HSRL technique to independently retrieve aerosol (and tenuous cloud) extinction and backscatter (Grund and Eloranta 1991, She et al. 1992, Shipley et al. 1983) without a priori assumptions on aerosol type or extinction-to-backscatter ratio. The HSRL-2 is advanced version of the HSRL-1 system (Hair et al. 2008). HSRL-1, which has flown over 800 hours on the NASA King Air B200 in 10 field campaigns since March 2006, measures profiles of aerosol backscatter and depolarization (532 nm and 1064 nm) and aerosol extinction at 532 nm. HSRL-2 provides the same measurements as HSRL-1 and adds measurements of aerosol backscattering, extinction, and depolarization in the UV (355 nm). Additional UV measurements to be made onboard the B200 will enable the lidar to readily differentiate biomass burning smoke from urban aerosols and will permit the use of advanced techniques to retrieve profiles of aerosol effective radius, aerosol surface, and volume concentrations; aerosol refractive index; and single scatter albedo. The second key instrument requested for the B200 (or equivalent aircraft) is NASA's Research Scanning Polarimeter (RSP), which measures the intensity and degree of linear polarization over a broad spectral (400–2250 nm) and angular (±60° from nadir) range. RSP measurements also permit columnar retrievals of key aerosol properties (optical depth, location and width of both modes of a bimodal size distribution, refractive index) and can also provide profiles of extinction coefficient, cloud optical depth, effective radius and effective variance of cloud droplet size distribution, liquid water path, and droplet number concentration parameters.

2.0 Scientific Objectives

2.1 CCN Chemical Closure Study (a local closure study), Cziczo, Ervens, Zaveri, Zelenyuk

One of our first post-campaign analyses will examine the chemical composition of particles that either have been activated or have the potential to be activated, leading to the formation of cloud droplets. As illustrated in Figure 5, the measurement strategy calls for performing level sampling legs within and above the marine boundary layer, with more extensive sampling within the lower part of the free troposphere. An emphasis in our post-campaign analysis will be placed on opportune interception of any layers of material, including biomass burning, mineral dust, or industrial plumes.

The CCN Activation Study will make use of in situ particle observations at different altitudes of CCN activation at a variety of water supersaturations. This analysis will use the physical and chemical properties of particles directly measured onboard the G-1 as input to the particle-resolved version of the comprehensive aerosol module MOSAIC (Zaveri et al. 2008, 2010a) to simulate CCN distributions directly measured onboard the G-1 and compare these simulated CCN properties with the directly measured values from the G-1, thus providing a measure of the uncertainty of this aspect of model performance.

2.2 Radiation Closure Study (a local closure study), Kassianov, Flynn, Zaveri

The AOD for any slab in the atmosphere can be defined using data from the 4STAR, HSRL, in situ measurements of aerosol optical properties or aerosol size distribution, and chemical composition. The airborne instrument 4STAR is an updated version of the 14-channel (NASA) Ames airborne tracking sunphotometer (AATS-14). Here, we will repeat the procedure previously made for the airborne AATS-14 (e.g., Schmid et al. 2006). The aerosol optical depth measured by the 4STAR at any altitude represents an attenuation of the solar beam in a layer located between the top-of-atmosphere (TOA) and the altitude at which the measurement takes place. The difference between AOD measured at two altitudes yields the AOD of the atmospheric slab. The AOD for a slab of atmosphere can also be obtained by integrating the extinction profile measured by the HSRL between the altitudes of interest. The slab AOD can also be determined by integrating the in situ aircraft measurements of the aerosol scattering coefficient measured by an integrating nephelometer (with appropriate humidity corrections), and the aerosol absorption coefficient. The slab AOD can also be computed using the measured particle size distributions, mixing state, and shape to be provided by the aerosol mass spectrometer, SPLAT II. We anticipate using the method described by Kleinman et al. (2007) to define the particle refractive index using AMS measurements. As with the CCN closure studies, aerosol composition and mixing state data will be used to constrain the particle-resolved version of the comprehensive aerosol module MOSAIC (Zaveri et al. 2008, 2010a) to investigate the effect of aerosol mixing state and aging on the optical properties.

In addition to the radiative closure study defined above, the in situ measurements of scattering and absorption will be used to compute the SSA and asymmetry parameter (AP). The measured SSA and AP will also be compared to values of SSA and AP derived from the size distributions and the AMS-based refractive index. In addition, the observed AP will be compared to values obtained using parameterizations that have appeared in the literature (e.g. Wiscombe and Grams 1976, Andrews et al. 2006).

2.3 Aerosol Type Comparison Study (column study), Ferrare, Hostetler, Cairns

This study will focus on assessing inferences of aerosol type made from multiwavelength lidar data. Model intercomparisons conducted as part of the Aerosol Comparisons between Observations and Models (AeroCom) project have shown large differences in how models represent the vertical distribution of aerosols (Textor et al. 2006) and aerosol composition (Kinne et al. 2006) even when similar emissions are used (Textor et al. 2007). Consequently, there is a strong need for vertical profiles of aerosol type and

optical properties to evaluate and improve aerosol transport models (Textor et al. 2006). The HSRL data sets will provide valuable information regarding aerosol type as a function of time and altitude.

The HSRL measurements of aerosol intensive optical properties (i.e., depolarization [532nm, 1064nm], extinction/backscatter ratio ["lidar ratio"], backscatter color ratio [532nm/1064nm]) provide qualitative information about the aerosol physical properties. The HSRL measurements of aerosol intensive parameters and aerosol optical depth have been used to identify aerosol types and apportioned aerosol optical depth to the various aerosol types. The HSRL aerosol type analysis has proven valuable for identifying smoke and urban plumes during recent missions. The in situ measurements of particle size and composition (SPLAT II) on the G-1 will be used to evaluate these remote sensing inferences of aerosol type.

These HSRL aerosol type analyses have also shown that there is significant variability in aerosol types with altitude. Preliminary analyses of aerosol types derived from HSRL measurements acquired over several years indicate that that only in approximately 15% of cases is 90% or more of the AOD in the column attributed to a single aerosol type and that a single aerosol type accounts for 75% or more of AOD in only about 37% of cases. Therefore, this vertically resolved aerosol type analysis will be important for assessing aerosol types inferred from the column retrievals acquired by the 4STAR and RSP instruments described in the next section.

2.4 Radiation Closure Study (column study), Kassianov, Flynn

We plan to carry out two studies related to the radiation budget within the two columns of air to be studied. The first experiment will integrate profiles of AOD measured by the 4STAR, HSRL, and in situ measurements of particle scattering and absorption over the vertical span of the G-1 flight pattern. These measured values will be compared to the integrated values of AOD measured by the ground-based MFRSR and/or those derived from airborne-based NASA/GISS RSP observations. The second experiment will contain an appropriate integration of profiles of SSA and AP. Similar to Andrews et al. (2006), we will determine the column-integrated values of AP by weighting the individual values of AP with measured profiles of the scattering coefficient. We will determine the column-integrated values of SSA by weighting the individual values of SSA with measured profiles of the extinction coefficient. We will contrast these column-integrated values with the corresponding values provided by MFRSR (e.g., Kassianov et al. 2007) and/or RSP.

These experiments will be carried out for both clear-sky and partly-cloudy conditions. As noted earlier, cloud contamination impacts on the aerosol retrieval could be substantial for cloudy cases. For example, cloud-induced enhancement of clear-sky reflectance can lead to significant overestimation of retrieved AOD (e.g., Wen et al. 2006). We have developed a new method (Kassianov and Ovtchinnikov 2008; Kassianov et al. 2009) that can substantially reduce such enhancement. Our method is based on unpolarized reflectances in the visible and near-infrared spectral ranges. Since the RSP measures polarized reflectances in the visible and near-infrared spectral ranges (Waquet et al. 2009) and polarized reflectance is sensitive to cloud-induced effects in a same way that unpolarized reflectance (Cornet et al. 2010), appropriate adjustments of our methods will be made. These adjustments will involve simulations and analysis of the RSP observations.

An original version of our MFRSR method has been developed for clear-sky conditions and has been successfully applied to sample the SSA and AP for different aerosol types at diverse locations, such as Niamey, Niger (e.g., Slingo et al. 2006) and Mexico City (Kassianov et al. 2005). Here, we will extend the original version to partly cloudy conditions (Kassianov et al. 2010). In particular, we will examine the reduction of the impact of broken clouds on the aerosol retrievals (SSA and AP) using a well-known framework (Long and Ackerman 2000) developed for selection of clear skies from surface-measured broadband irradiances.

2.5 RP5: Cloud-Aerosol Interactions (column study) Berg and Berkowitz

A number of studies have documented evidence of the first aerosol indirect effects in warm stratocumulus clouds over oceans (e.g. Radke et al. 1989, Durkee et al. 2000), over land (Kim et al. 2008), or shallow cumuli in the vicinity of both large (Liu et al. 2008) and small (Berg et al. 2009b) cities. All of these studies were short-duration studies designed to investigate a specific cloud type. Feingold (2003) conducted a study at the ARM Southern Great Plains site to investigate aerosol indirect effects in warm, non-precipitating clouds. Their study utilized data from the Raman lidar, MWR, radar, and optical particle counter.

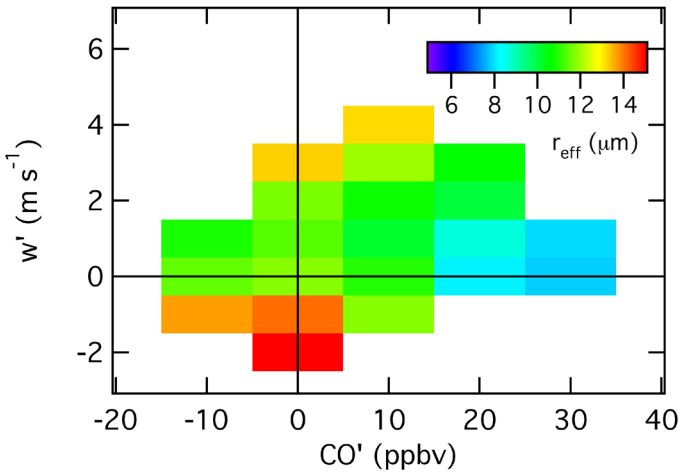

Figure 6. Change in average r_{eff} (color) with increasing perturbation CO and increasing perturbation vertical velocity (w).

In an earlier study (the CHAPS campaign, described in Berg et al. 2009b), r_{eff} was grouped according to the vertical velocity and the amount of perturbation CO (observed CO with mean and trend removed, where CO was used as a surrogate for aerosol loading; see Figure 6). A systematic decrease in r_{eff} was found with increasing amount of perturbation CO. These observations were based on a limited number of aircraft flights for shallow convective clouds downwind of Oklahoma City, but demonstrate the utility of coincident measurements of cloud microphysics, cloud dynamics, and particle loading. A similar analysis is anticipated using a more complete set of observations from the proposed campaign.

In addition, a methodology similar to that employed by Feingold (2003) and Kim et al. (2008) will be employed in our analysis of the boundary-layer clouds above the AMF/MAOS. The ARM cloud radar

will provide estimates of the cloud boundaries, cloud microphysics, and cloud dynamics (inferred from the vertical velocity). The effective radius will also be derived from the AERI, to provide an additional measure of the effective radius. The liquid water content will be derived from the microwave radiometer. Observations from these instruments will be supplemented by direct measurements from instruments onboard the G-1 (e.g., direct measurements of w' from the gust probe or cloud droplet number from the CAPS). As highlighted by Kim et al. (2008), accounting for variation in the liquid water path is critical for an examination of the indirect affect. This study is different from previous studies in several ways. First, there will be a significant amount of information gathered about the chemical composition of the particles. These data will allow us to determine if the cloud microphysical structure shows any sensitivity to the aerosol composition and mixing state of black carbon (determined from the SP2) rather than just the aerosol loading. Second, the study will be conducted over a full year, allowing for the examination of many more cases than was possible in previous studies. During the aircraft IOPs, the cloud radar data will be augmented with measurements of cloud drop-size distributions and turbulence made on board the aircraft using methods similar to those employed by Berg et al. (2009a).

2.6 High-Resolution Modeling of the Measurements, Fast, Zaveri

Dr. Jerome Fast will lead the high-resolution modeling study using advanced aerosol and cloud treatments implemented in the chemistry version of the Weather Research and Forecasting (WRF-Chem) community model to simulate the evolution of aerosols and their effect on CCN and aerosol direct and indirect radiative forcing. This work will differ from the direct model/observation comparison that is noted above by focusing on how radiative forcing within the two columns was affected by secondary organic aerosols, aerosol mixing state, and grid-resolution.

We anticipate using a nested grid configuration for the proposed campaign, with an outer grid encompassing much of eastern North America and the western Atlantic Ocean and an inner grid over the study area using a horizontal grid spacing of ~3 km to resolve much of the cloud distributions in the region. Boundary conditions for the model will be obtained from simulations we perform with the global Community Atmospheric Model (CAM) climate model described below.

WRF-Chem will be run with a variety of treatments for aerosol processes, ranging from simple to more complex representations. The aerosol treatments will be systematically evaluated using the field campaign data and the methodology of the Aerosol Modeling Testbed (Fast et al. 2010, http://www.pnl.gov/atmospheric/research/aci/amt/intex.stm). We will quantify whether the improved performance associated with research versions is worth the additional computation expense. The summer and winter measurement intensive periods will provide a means of contrasting model performance as a function of seasons. The processes we will examine in the model include:

- *Impact of regional-scale particulate distributions on radiative forcing:* Since current particulate models often significantly underestimate the amount of organic aerosol mass by an order of magnitude or more (Volkamer et al. 2006), this missing mass may be a large source of uncertainty in estimates of aerosol radiative forcing. A recently proposed "volatility basis set" approach proposed by Shrivastava et al. (2008), Robinson et al. (2007), and Donahue et al. (2006) has been implemented in WRF-Chem and is currently being evaluated using Megacity Initiative: Local and Global Research Observations (MILAGRO) and CARES field campaign data. Preliminary results indicate that organic aerosol mass is represented much better using the volatility basis set than more traditional approaches. For TCAP, we intend to compare simulated total organic aerosols with those obtained from the

Aerodyne AMS and from observations at the MAOS ground site in addition to evaluating simulating simulated aerosol composition and size distribution from the other instrumentation. Aerosols and aerosol precursors will also be evaluated using routine monitoring data collected over the eastern U.S. and Canada, and simulated extinction and backscatter profiles will be evaluated using data from the HRSL. After the model has been evaluated using the available data, and the uncertainties in predicted aerosol mass, composition, and size distribution have been quantified, we will compute simulated regional variations in aerosol radiative forcing. The model will enable us to quantify the relative role of organic aerosols in aerosol direct radiative forcing in the region in relation to other aerosol compositions and examine the impact of organic aerosols on indirect radiative forcing by performing sensitivity simulations in which organic aerosols are not present.

- *Treatment of the aerosol mixing state and its impact on radiative forcing:* Rather than assuming either external or internal mixtures of aerosols as do most models, a new formulation is being developed at PNNL that will treat the evolution of the aerosol mixing state more realistically based on the results from a explicit particle-resolved model (Riemer et al. 2009, Zaveri et al. 2010a). In this model, external mixing is assumed for primary emissions with the resulting aerosol plume becoming more internally mixed with time. The model also uses the mixing state to compute CCN and aerosol optical properties. The measurements from the aerosol mass spectrometers and SP2 to be deployed at the AMF/MAOS and the G-1 aircraft will be used to quantify mixing state of anthropogenic plumes over the western North Atlantic Ocean, thus providing the basis for evaluating this new model. The new model will be compared to conventional treatments to assess whether accounting for evolving mixing state is significant in terms of regional scale modeling.
- *Effects of subgrid scale variability and estimates of radiative forcing:* A recent study by Qian et al. (2010) has shown that the coarse grid spacing employed by global climate models does not adequately represent the evolution of some aerosol species downwind of Mexico City. We will quantify the subgrid scale variability (in terms of a global climate) model for aerosol properties, cloud properties (including cloud-aerosol interactions), and aerosol radiative forcing computed from WRF-Chem, and compare these results with other work done at PNNL using global-climate scale models. The new theory on evaluating AOD in the presence of clouds will be utilized, since a large fraction of the WRF-chem model domain will have regions near cloud boundaries.

2.7 Global-climate Scale modeling of the Measurements, Rasch

Dr. Phil Rasch has lead the development of the CAM model and will lead a team of PNNL modelers that will evaluate the CAM5 model in representing anthropogenic aerosols over the Atlantic Ocean. CAM5, released in July 2010, contains new physics parameterizations intended for the next generation of climate projections. In our study, CAM5 will be run in a case study mode based on the AMF/MAOS data and aircraft IOPs described above that assimilates meteorological fields derived from global analyses into the model so that the CAM5 fields will follow the observed conditions. In this way, uncertainties in the representation of meteorology (except for clouds) will be minimized. As described above, we anticipate sampling within two columns of air and making extensive use of these observations. However, our evaluation of CAM5 will also use other data sources (e.g., AERONET, satellite, surface air-monitoring networks) to evaluate spatial variations in aerosol mass, composition, AOD and other optical properties. Two types of simulations will be performed:

- *Representing horizontal and vertical variability of anthropogenic aerosols and their impacts:* CAM5 will be run several months prior to the field campaign and for the duration of the AMF and MAOS deployment. CAM5 grid cells will be evaluated with the detailed in situ and remote sensing measurements from both the surface-based and aircraft platforms. We will determine how well the coarse grid spacing is able to represent the aerosol properties, cloud properties, and aerosol radiative forcing as anthropogenic aerosol plumes are transported over the Atlantic Ocean. The CAM5 results will be compared with those from higher-resolution WRF-Chem simulations to help identify regional processes that may be contributing to the changing aerosol and cloud properties. The long-term deployment of AMF/MOAS will be used to assess how well a climate model performs in a region of relatively high aerosol loading in an urban environment, in contrast to previous evaluations of CAM using long-term data derived from ARM's North Slope of Alaska, Southern Great Plains, and Tropical Western Pacific sites, where the effect of anthropogenic aerosols is relatively small. The vertical profiles of aerosol extinction and aerosol intensive parameters derived from the HSRL measurements from the B200 will be used to help evaluate the ability of models to reproduce aerosol extinction and optical thickness profiles as well as to help determine how well models can represent horizontal and vertical variations in aerosol types.
- *Assessing simulated radiative forcing in a column:* Our past experience on many modeling projects suggests that we will find differences between simulated aerosol direct radiative forcing and cloud-aerosol interactions and measurements. A series of simulations will be performed to assess the cause of any such differences, e.g., whether these differences result from simulated aerosol and cloud properties or the treatments of aerosol-radiation-chemistry-cloud interactions. Closure studies will be performed using CAM5 run in its single-column mode, which replaces simulated aerosol and cloud properties with measured values. Some of the observed cloud properties will be obtained using the ARM value-added products[1] derived from AMF and MAOS measurements. The resulting aerosol direct radiative forcing and cloud-aerosol interactions will be compared with the previous CAM5 simulations and measurements to assess the relative uncertainties in a manner similar to the methodology used by Barnard et al. (2009) for the WRF-Chem model that evaluates the aerosol optical property treatments.

2.7.1 Data Processing and Expected Final Products

The AMF team will be responsible for processing the AMF data throughout the course of its one-year deployment. Data from the G-1 will be made available through the ARM Aerial Facility team, under the supervision of Dr. Beat Schmid. The final products and their relevance to TCAP science issues have been described in Section 2 section of this document.

2.7.2 Project Management

The field campaign has been designed to provide data for a number of closely related projects with all of the investigators having a strong hand in the design of the campaign. Members of our team also have a long history of working together and with others in the DOE community, and look forward to continuing this tradition in the following projects.

[1] described at http://www.arm.gov/data/vaps

- Carl Berkowitz will be the project's administrative lead, working with other ARM personnel as needed, leading the coordination of field campaign activities, and be responsible for overall management of the field campaign. Larry Berg is the co-lead for these activities.
- John Shilling, Barbara Ervens, Rahul Zaveri, and Alla Zelenyuk will assume primary responsibility for the data collection and post-campaign analysis of observations made in support of our Cloud Condensation Nuclei Chemical Closure Study.
- Evgueni Kassianov, Connor Flynn, and Rahul Zaveri will have primary responsibility for observations and post-campaign analysis of observations made to support our local Radiation Closure Study.
- Chris Ferrare, Rich Hostetler, and Brian Cairns will take the lead in working with the HSRL observations that we hope to have from the NASA B200 aircraft. Funding for this aspect of TCAP was pending as of June 9, 2011.
- Evgueni Kassianov and Connor Flynn are the principal investigators (PIs) for our two Column Radiation Closure Studies.
- Larry Berg and Carl Berkowitz will continue their studies on cloud-aerosol interactions, building on past work done during the CHAPS campaign.
- Jerome Fast and Rahul Zaveri are co-leading our high-resolution modeling studies. Jerome is leading our WRF-Chem analysis. Rahul is taking the lead for our modeling studies to examine particle mixing state, CCN, and aerosol optical properties.
- Phil Rasch will lead the modeling team to evaluate the CAM5 model in representing anthropogenic aerosols over the Atlantic Ocean.

3.0 ARM Critical Instruments

3.1 ARM Aerial Facility

This section identifies facilities, instrumentation, logistical support, and data being anticipated to be available from the ARM Facility. The assumption that the following instruments are available for deployment comes primarily from information presented at the ARM Facility webpage (http://www.arm.gov/sites/aaf) or the description of new instruments acquired with funds through the Recovery Act (http://www.arm.gov/about/recovery-act/instruments). Instruments available through the Recovery Act are identified by "(RA)" following the facility associated with the instrument, e.g., "AAF (RA)." An overview of the airborne sun-tracking spectrometer (4STAR) is provided at http://stm.arm.gov/2008/presentations/0310/2Flynn_4STAR.pdf, with additional information provided via citations in Section 4, "References." An overview of the AMS is provided in Zelenyuk et al. 2009, with a general description of its past airborne deployment at http://www.emsl.pnl.gov/news/newsletter/20081006.pdf .

Time frame: We are planning to have one three-week aircraft IOP during the summer of 2012 and a second three-week IOP that winter. Preliminary dates are for July 2012 and February 2013. The most current information may be obtained from the ARM Aerial Facility.

Location: Selection of a ground site is still underway as of July 2011. There are a number of airfields on or near Cape Cod (Massachusetts) that would be suitable; the ARM Aerial Facility (AAF) is in the process of locating a site that will be consistent with the flight plan described in this document (illustrated in Figure 5). The most current information may be obtained from the ARM Aerial Facility.

Mission Length: The actual hours per mission will, of course, strongly depend on the final weight and power needs of the instruments. For planning purposes, we have assumed three-hour missions for both the G-1 (or larger) aircraft and the King Air B200 (or comparable aircraft). The most current information may be obtained from the ARM Aerial Facility.

Number of Missions per IOP: Although this will depend on available resources, we anticipate making missions on two sequential days followed by a planned down day for rest and basic instrument maintenance. Thus, for each three-week campaign, we plan to have on the order of 14 or 15 flights. Our experience is that a smaller number of flights would actually occur as a result of weather or instrument problems. The most current information may be obtained from the ARM Aerial Facility.

Aircraft Platforms: We anticipate deployment of the DOE Gulfstream-1 aircraft and the NASA King Air B200.

Instrumentation for Aircraft:

- Although the list below gives our list of requested instruments and lead scientists, the final set of instruments expected to be deployed is still being finalized by the ARM Aerial Facility. Please contact Dr. Beat Schmid for the latest information.
- We use the 'greater than' sign (">") to indicate instrument priority where applicable.

Table 1. Instruments Requested for DOE G-1 Deployment

Campaign	Measurement	Instrument	Facility/Contact
All analysis: Basic state variables from in situ aircraft (G-1 or equivalent)	5-port air motion sensing: • True air speed • Altitude • Angle-of-attack • Side-slip and temperature • Relative humidity	Aircraft Integrated Meteorological Measurement System (AIMMS-20)	AAF (RA)
Aerosol Indirect Effects (Berg, Berkowitz)	• Tracers of anthropogenic plume • Chemical aging time scales	Research-grade trace gas observations, with the following priority: CO >SO_2> NO_x & NO_y > O3 (Trace Gas Instrument System)	AAF (RA)
• Aerosol Indirect Effects (Berg, Berkowitz) • CCN Chemical Closure Study (Cziczo, Zaveri)	Cloud droplet spectra and total liquid water content	Cloud, Aerosol, and Precipitation Spectrometer (CAPS)	AAF
Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	For the in situ aircraft (nominally the Gulfstream G-1): airborne direct beam and angularly resolved sky radiance	4STAR	PNNL
• CCN Chemical Closure Study (Cziczo, Zaveri) • Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Aerosol mixing state, on a real-time, particle-by-particle basis, with simultaneous measurements of aerosol size, density, and shape	Single Particle Mass Spectrometer (SPLAT II) (high-priority instrument)	PNNL/EMSL
• CCN Chemical Closure Study (Cziczo, Zaveri) • Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	• Aerosol chemical composition • Anions • Cations • Water-soluble organic carbon	Particle in Liquid auto Sampler (PILS) with online IC analyzer (for both positive and negative ions)	AAF (RA)
• CCN Chemical Closure Study (Cziczo, Zaveri) • Local Radiative Closure Study (Kassianov, Flynn, Zaveri) • High-Resolution Modeling (Fast, Zaveri)	Size-resolved aerosol composition	Aerodyne HR-ToF-AMS (lower priority than SPLAT II and PILS, but highly desirable)	PNNL
• CCN Chemical Closure Study (Cziczo, Zaveri) • Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Number concentration (0.05–1 micrometers)	Ultra High Sensitivity Aerosol Spectrometer	AAF (RA)

Campaign	Measurement	Instrument	Facility/Contact
Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Aerosol scattering coefficient as a function of relative humidity	Humidigraph (3 Relative Humidities with single wavelength nephelometers)	AAF (RA)
Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Aerosol light absorption	Following priority: PTI > PASS-3 > PSAP	AAF
• CCN Chemical Closure Study (Cziczo, Zaveri) • Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Number concentration (>10 nm)	TSI-3010 Condensation Particle Counter (CPC)	AAF
• CCN Chemical Closure Study (Cziczo, Zaveri) Local Radiative Closure Study (Kassianov, Flynn, Zaveri)	Number concentration (>3 nm)	TSI-3025 Ultra-fine Condensation Particle Counter (CPC)	AAF
CCN Chemical Closure Study (Cziczo, Zaveri)	• Liquid water content • Particle number concentration	Isokinetic Inlet and Counter-Flow Virtual Impactor (CVI)	AAF (RA)
CCN Chemical Closure Study (Cziczo, Zaveri)	• Total condensed atmospheric water content measurement • Droplet size spectrum 2–50 µm	Cloud Spectrometer and Impactor (CSI)	AAF
CCN Chemical Closure Study (Cziczo, Zaveri)	Aerosol hygroscopic properties	Humidified Tandem Differential Mobility Analyzer (from AOS or MAOS, adapted to aircraft)	Cziczo (ARM instrument mentor for the HTDMA) proposes to modify the instrument presently on the Aerosol Observing System of AMF2 to make it airworthy. (RA)
CCN Chemical Closure Study (Cziczo, Zaveri)	Number of aerosols that activate to become cloud condensation nuclei at two independently selectable supersaturations	Dual Column Cloud Condensation Nuclei Counter (CCN) and Ultra-High Sensitivity Aerosol Spectrometer (for particle size distribution out of cloud)	AAF (RA)

Table 2. Instruments Requested for King Air B200 Deployment

Campaign	Measurement	Instrument	Facility/Contact
• High-resolution modeling analysis (Fast, Zaveri) • Cloudy-sky AOD retrieval evaluation(Kassianov, Ferrare, Hostetler)	Profiles of aerosol backscatter (532 and 1064 nm), extinction (532 nm), and depolarization (532 and 1064 nm)	NASA/Langley High Spectral Resolution LIDAR	NASA/Langley
• High-resolution modeling analysis (Fast, Berg) • Cloudy-sky AOD retrieval evaluation (Kassianov, Ferrare, Hostetler)	• Retrievals of aerosol optical depth, effective radius and variance of size distribution, index of refraction, and single-scatter albedo • Retrievals of cloud droplet effective radius and variance of size distribution, droplet number concentration, liquid water path, optical depth	NASA/GISS Research Scanning Polarimeter (RSP)	NASA/Langley

Instruments for large-scale model evaluation

No additional measurements are needed for this aspect of the post-campaign analysis (Rasch, Russell, Fast, Zaveri).

3.2 ARM Mobile Facility

The AMF and the MAOS ground-based systems will be deployed at or very near Cape Cod, Massachusetts, for a continuous period to span both the summer and winter IOPs.

Time frame: We are anticipating a 12-month deployment of the AMF beginning in the summer of 2012 and extending through the following spring. The timing will coincide with the two aircraft IOPs previously described. Discussions are still underway regarding the timing for deployment of the MAOS.

Location: The final selection of sites for the AMF/MAOS units is still under discussion as of July 2011. However, we anticipate they will be deployed towards the tip of Cape Cod, past Truro and within the national seashore.

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