ACE-Asia

Asia-Pacific Regional
Aerosol Characterization Experiment

Science and Implementation Plan for the ACE-Asia Network Studies

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24 March 1999

Project Summary

This science and implementation plan (SIP) presents the need for an ACE-Asia ground station network, summarizes the scientific goals and objectives of the experiment, discusses the research issues to be addressed, outlines the plans for implementing the program, and describes some the modeling studies needed integrate our understanding of aerosols and climate. This SIP also briefly describes the connections between the network studies and the three other components of ACE-Asia.

The ACE-Asia network will be composed of two types of stations: basic and enhanced, with routine or intensive studies possible at any of the stations. The basic stations will be outfitted a program-standard aerosol sampler while the enhanced sites will be more highly-instrumented with more sophisticated equipment Four subnetworks of enhanced sites will study (1) detailed aerosol chemical properties, including size-separated aerosol analyses for mineral dust and major ions; organic speciation; organic and elemental carbon; and chemical tracers, (2) aerosol optical properties (such as aerosol light scattering and absorption), aerosol optical depth, and radiative fluxes, (3) vertical structure of the atmosphere, with lidar and other remote sensing devices, and (4) wet deposition rates for aerosols. A systematic approach to quality control, including intercomparisons of instruments and analytical methods will be employed to ensure the results are comparable among stations and to the extent possible with other networks.

The routine network operations will provide information on the spatial and temporal distributions of major chemical constituents and physical properties of surface air within the study region, and by extension provide information on the chemical and physical evolution of the aerosol. The network sites also will be used to assess regional budgets for major chemical species, to constrain overall fluxes, and to provide a context for assessing the "representativeness" of the intensive measurements. This network will provide the geographical coverage needed to test how accurately models (with help from satellite-borne sensors) can calculate aerosol spatial distributions and temporal variability. In particular, models will be used to relate in-depth measurements of aerosol distributions to radiative forcing.

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I. Introduction

This Science and Implementation Plan (SIP) presents the overall strategy for the ACE-Asia Network studies, one of four components of the ACE-Asia Program. The network will provide detailed data relating to aerosol-climate connections for a region of the world where emissions are already high and are expected to increase substantially in the coming years. This document provides a conceptual framework for the scientific operations based on inputs from the various ACE-Asia working groups.

Background

Atmospheric aerosols from both natural and anthropogenic sources directly affect the Earth's radiative balance by scattering and absorbing light, and they also indirectly impact radiative transfer by altering cloud properties. The perturbation of the global radiative balance attributable to anthropogenically produced aerosols is referred to in the ACE-Asia context as “radiative forcing”. The magnitude of forcing by tropospheric aerosols is poorly constrained, and this represents the single greatest uncertainty in assessing climate change (IPCC, 1995). The uncertainties result to some extent from a limited data base on aerosol distributions, but more fundamentally, they are a consequence of our incomplete understanding of the processes responsible for aerosol formation, transport, evolution, and removal relative to their radiative effects.

The Aerosol Characterization Experiments (ACE) have integrated *in situ* measurements, satellite observations, and models to investigate the climate forcing caused by aerosol particles and the roles played by aerosols in biogeochemical cycles. The overall goals of these experiments are:

- to reduce the overall uncertainty in the calculation of climate forcing by aerosols
- to understand the multiphase atmospheric chemical system sufficiently to be able to provide a prognostic analysis of future radiative forcing and climate response

Increasingly, the interest of atmospheric chemists and aerosol scientists has turned to the Asia/Pacific region, first because the aerosol loadings there already have been seriously perturbed by anthropogenic activities, and second because these perturbations are increasing rapidly with time. Studies of aerosol-climate interactions in eastern Asia and the northwestern Pacific will complement and extend the earlier ACE experiments, in large measure due to the unique characteristics of the emissions from Asia.

Substantially more coal and biomass are burned in Asia compared with Europe and North America, and often the emission controls in Asia are minimal or completely lacking. In addition, the oxidizing capacity of the atmosphere over the Asia/Pacific region is changing rapidly as the growing transportation sector in Asia raises the concentrations of nitrogen oxides to levels approaching those in Europe and North America. Dust from the Asian deserts reacts with various trace gases, and in this way the cycles of various chemical constituents and mineral dust become linked. Mixing of aerosol populations and in-cloud processing complicates the situation still

further. The fact that much of the Asian aerosol is advected out over the Pacific Ocean, which has been one of the least polluted regions of the planet, implies significant changes in radiative forcing may occur over a large area of the Earth.

Plans for ACE-Asia consist of four focused components (1) network-based studies of aerosol chemical, optical, and radiative properties described here, (2) an intensive survey of aerosol processes and properties, (3) studies of the direct radiative effects of aerosols, and (4) a set of intensive cloud-aerosol experiments. The strategy of dividing the program into separate components was adopted mainly for practical reasons because it became evident that the various components were in different stages of scientific readiness and that each component had specific instrumental, sampling, meteorological, and logistical needs. Moreover, dividing the program into separate components makes the execution of the program more manageable while still enabling the science team to target specific scientific issues.

II. The Need for an ACE-Asia Aerosol Network

Combined data on aerosol chemical and radiative properties–the kind needed to understand aerosol-climate connections–are particularly scarce in the Asia-Pacific region. Chemical and physical data on aerosols have been collected from only a few organized networks in the Asia, including the JACK (the Japan, China, and Korea) network (Hashimoto et al. 1994), PEACAMPOT (Hatakeyama et al., 1995, UNESCO/IOC/WESTPAC, and PEM-West (special issues in the Journal of Geophysical Research, 1996 and 1997), AIMS (Atmospheric Inputs to the northeastern Asian Marginal Seas, Hong et al., 1998), simultaneous measurements of a single dust event in China and Japan (Fan et al., 1996) and the operation of six island sites in the western North Pacific in cooperation with the SEAREX (Sea/Air Exchange) Asian Dust network (Tsunogai et al., 1985). These programs have been quite narrow in scope, however, and as a result, information on the patterns of variability in aerosol properties over Asia is extremely limited.

The ACE-Asia ground station studies will quantify spatial, seasonal, and interannual variability (e.g., El Niño; Indonesion fires, etc.) of key aerosol properties in near surface air over the study domain. The ACE-Asia datasets will also be used to develop and test regional and hemispheric models that simulate radiative effects of aerosols, and they will be used to better understand the roles aerosols play in biogeochemical cycles. In addition, the network data will be invaluable for planning the ACE Asia intensive investigations and for putting those results in a broader context.

Measurements at the ACE-Asia stations will complement ongoing and planned studies being undertaken for the China Metro-Agro Plex experiment (China MAP), NASA's Transport and Chemistry Experiment-Pacific (TRACE-P), and various national and regional monitoring programs. Coordination among programs will be facilitated by the Coordinating Committee of the East Asia/Pacific Regional Experiment (APARE), an activity of the International Global Atmospheric Chemistry Experiment (IGAC). The ACE-Asia network operations also will be coordinated with other IGAC activities, including ACAPS (the Aerosol Characterization and Processes Study), ACI (Aerosol-Cloud Interactions), DARF (Direct Aerosol Radiative Forcing),

MAGE (Marine Aerosol and Gas Experiment), and SUTA (Stratospheric and Upper Tropospheric Aerosols)

III. ACE-Asia Scientific Objectives and Goals

III.A. ACE-Asia Program Objectives

The overall goal of ACE-Asia is to reduce the uncertainty in climate forcing caused by aerosols over eastern Asia and the northwest Pacific and to develop a quantitative understanding of the multi-phase gas/aerosol particle/cloud system. To achieve these goals, the ACE-Asia Program as a whole will pursue three specific objectives:

Program Objective 1 Determine the physical, chemical, and radiative properties of the major aerosol types in the Eastern Asia and Northwest Pacific region and investigate the relationships among these properties.

Program Objective 2 Quantify the physical and chemical processes controlling the evolution of the major aerosol types and in particular of their physical, chemical, and radiative properties.

Program Objective 3 Develop procedures to extrapolate aerosol properties and processes from local to regional and global scales, and assess the regional direct and indirect radiative forcing by aerosols in the Eastern Asia and Northwest Pacific region.

The science and implementation plan (SIP) presented here is structured around the basic scientific issues that must be addressed to achieve these programmatic objectives.

III.B. ACE-Asia Network Goals

The overall goals of the ACE-Asia network studies are to:

Network Goal 1 Characterize the physical, chemical and radiative properties of the aerosol in the ACE-Asia region and understand the factors controlling the regional and temporal (seasonal to interannual) variability of these properties,

Network Goal 2 Determine the impacts of regional aerosol (organic, ionic, mineral dust) sources and sinks,

Network Goal 3 Intercompare satellite and ground-based measurements of optical depth, and

Network Goal 4 Develop and validate reliable regional- to hemispheric-scale models of aerosol chemical and physical properties.

The ACE-Asia aerosol/radiation network will include two types of stations: basic and enhanced; with either type capable of operating in routine or intensive modes. All basic stations will be outfitted with a standard aerosol sampler. Four subnetworks of enhanced stations will be equipped with comparable sets of instruments to investigate (1) multiphase chemistry, (2) aerosol optics and radiation, (3) aerosol deposition, and (4) vertical structure of the atmosphere. The fourth of these subnetworks will make use of existing lidars and other remote sensing devices.

The subnetworks will be co-located to the maximum extent possible to facilitate the integration and interpretation of physical and chemical information.

The operating plan for the ACE-Asia network is for science teams from the participating countries to purchase their own sampling equipment and, to the extent possible, analyze samples in their own national labs. As in earlier ACE experiments, the individual PIs for the program will solicit funding through the various sponsors available to them. Extra-national support for supplemental instrumentation and analyses would be requested for enhanced sites, for situations in which important scientific gaps exist, and for other areas of research that would make effective use of the network infrastructure. Quality control and quality assurance will be coordinated through APARE and the ACE-Asia National Committees. Some network operations will begin in 2000; full, routine operations should be underway by 2001.

IV. Scientific Issues

ACE-Asia will be a major international collaborative program involving investigators studying a variety of topics related to aerosols, chemistry, optics, radiation, atmospheric physics, climate, and meteorology. Here we briefly review the specific issues to be addressed for the program.

IV.A. Regional and Temporal Variability in Aerosol Properties (Network Goal 1)

Concentrations of both pollution-derived and naturally-occurring aerosols in the ACE-Asia domain are among the highest on earth. For example, the average daily total suspended particle concentrations in some Chinese cities exceeds 400 $\mu g\ m^{-3}$ (UNDP, 1996). Moreover, the aerosol loadings often change rapidly over space and time, making prediction challenging. For example, during a yellow-dust event (called “huang sha" in Chinese, "whangsa” in Korean, “kosa” in Japanese), aerosol mass loadings can change by orders-of-magnitude in a matter of a few hours. Spatial heterogeneity is also evident in remotely sensed images of desert dust and smoke from biomass burning (Husar et al., 1997). The network data also will provide useful benchmarks against which future changes can be quantified.

IV.A.1 Aerosol Mass Loadings and Composition

The chemical composition of the ACE-Asia aerosol will differ from what was measured during previous studies (ACE 1, ACE 2, TARFOX) because the background aerosol, oxidant species, aerosol source material, and combustion practices all differ among regions. The aerosol population in the ACE-Asia region will be a mixture of combustion-derived ionic, organic and soot particles; sea-salt; mineral dust; biogenic sulfur compounds; and poorly characterized organic species of biogenic origin. Speciation and quantification of the aerosol chemical composition provides basic information needed to assess aerosol-radiative forcing and to validate chemical transport models. Quantifying the contributions from the various aerosol sources is needed if we are to develop a reliable predictive capability for aerosol concentrations and climatic impacts under potential future emission scenarios.

Data for aerosol sampled in bulk or for specific size fractions, such as PM-10 or PM-2.5 are useful for characterizing the temporal and spatial variability of the major aerosol species, but data

for chemical composition of aerosols as a function of particle size are required to reliably model aerosol transport, evolution, and radiative properties. Size segregated measurements require more sophisticated equipment (such as cascade impactors), however, and they are considerably more demanding in terms of resources and personnel. While size-selected analyses are not suitable for routine operations, such studies are an example of an especially valuable enhancement for selected stations or for intensive study periods. Similarly, single-particle analysis during intensives will be useful for characterizing aerosol composition and for assessing the degree of mixing of the various chemical components.

IV.A.1.a. Mineral aerosol (Asian dust)

The production and long-range transport of mineral aerosol from Asia impacts the radiative balance over a large region and very likely affects biological productivity in the North Pacific Ocean. While Central Asia is one of the world's largest dust sources, with current estimates of dust production around 800 Tg y^{-1} (Zhang et al., 1997), the magnitude of this source remains highly uncertain. Similarly, the climatic effects of Asian mineral dust are largely unquantified due to the lack of detailed information on space- and time-varying dust properties. Interactions of dust with Earth's radiation field are more complicated than for most other atmospheric aerosols because mineral particles can both scatter and absorb significant quantities of solar and infrared radiation (Sokolik and Toon, 1996), leading to heating under some conditions but cooling under others. Thus we are proposing studies to evaluate the warming vs. cooling effects of Asian dust.

The strongest dust storms occur in spring when vast amounts of dust are lofted into the atmosphere from arid and semi-arid lands in northern and northwestern China. Human activities can increase dust loadings and enlarge the extent of the dust source regions (Tegen and Fung, 1996). Proposed studies of mineral dust during ACE-Asia will provide useful constraints on the proportion of the anthropogenically generated dust that contributes to radiative forcing.

Asian dust typically originates in desert areas far from polluted urban regions, but some dust plumes travel over developed regions, and the chemical and optical properties of the particles are modified by reactions with pollutants and other atmospheric constituents. Thus, dust particles involved in long-range transport can have substantially different radiative properties from those at the sources. In addition, interactions with clouds and other types of aerosols can lead to internal mixtures that also alter the physical and radiative properties of the aerosol population. We propose single particle studies to address this issue.

IV.A.1.b. Carbonaceous aerosol

Much of the aerosol mass over east Asia is likely to be organic owing to the abundance of combustion sources in the region (Ohta and Okita, 1984). In addition to bulk organic and elemental carbon measurements, analytical techniques will be needed to determine organic speciation because that level of detail is needed for closure studies (also called mass accounting studies) and to fully assess the radiative properties of these aerosols. Understanding organic speciation is also at the foundation of methods for assessing the indirect effect of aerosols: the water solubility and surface tension of organic species has a major impact on activation to form

cloud droplets. Detailed characterizations of the organic substances also provide unique information on sources, but it is unlikely that these analyses can be carried out as part of the routine network operations owing to the complexity and expense of these analyses. Instead we will propose investigations of this type at one or more of the enhanced ground stations.

IV.A.1.c. Major ions

Pollution-derived sulfate overwhelms sulfate from natural biogenic sources over eastern Asia and accounts for a substantial fraction of the non-sea salt (nss) sulfate even over the remote North Pacific (Arimoto et al., 1996). Volcanic emissions of sulfate in the Asia/Pacific region, though likely significant, and not well quantified. Particulate nss sulfate and nitrate are known to be correlated at several coastal-continental sites, but significant differences in nss sulfate/nitrate ratios among the sites suggest regional differences in pollutant sources and/or transport patterns as reported by (Akimoto et al., 1994). We are proposing further assessments of the sources for sulfate and nitrate in ACE-Asia because these are major components of the aerosol and because these ions are involved in a variety of chemical reactions.

Aerosol nitrate and aluminum-an indicator of mineral dust-were highly correlated in samples from Oahu, Hawaii, but these substances generally not correlated at the PEM-West network of coastal-continental sites in Asia (Arimoto et al., 1996). Thus, either the surface air along the east-Asian coast was chemically distinct from the air transported to the remote Pacific or its chemical composition changed significantly during transport.

Sulfuric and nitric acids are major sources of atmospheric acidity over the ACE-domain, and thus they control important pH-dependent chemical transformations such as S oxidation in sea-salt aerosol and the phase partitioning of ammonia. In addition, submicrometer aerosols impact radiative transfer through both direct and indirect effects. Consequently, reliable predictive tools require knowledge of sources, distributions, and chemical evolution of ionic aerosol constituents. ACE-Asia will collaborate with regional acid deposition programs to determine these characteristics of regional aerosols. One such program the East Asia Network for Environmental Monitoring (EANET) organized by the Japanese EPA; EANET has already begun the collection and analysis of aerosol and precipitation samples from China, Indonesia, Japan, Korea, Malaysia, Mongolia, Philippines, Russia, Thailand, Vietnam. In fact, several national acid deposition programs are also underway in the region, and results published by the WESTPAC office.

IV.A.1.d. Other aerosol components

Trace metal concentrations provide information on aerosol sources, and various radionuclides can be used to trace the histories of air masses. Trace metal concentrations in Asia have been investigated for numerous national and international programs (e.g., Hashimoto et a., 1994), with results indicating that in some areas the levels of some toxic metals (e.g. lead from gasoline) are presently at sufficiently high concentrations to raise public health concerns. Maenhauet et al.(1996) showed that certain trace elements (e.g., K, P, Ca, Mn, Zn, Sr, and I), alone or in combination, can provide information on sources such as biomass burning that are of special interest for ACE-Asia. Stable isotopes of Pb, S, and Nd can be used to characterize sources and source emissions (Mukai et al., 1993) while radionuclides such as ^{7}Be and ^{210}Pb provide

information on the relative strengths of upper tropospheric/lower stratospheric air vs. continental air, respectively (Graustein and Turekian, 1996). Studies of these aerosol components will add to the value of the other network data by virtue of their tracing power and by the unique insights they provide relative to source contributions.

IV.A.2. Aerosol Radiative Properties

Eastern Asia is a major source region of natural and anthropogenic aerosols, readily observed by satellites. However, the effects of Asian aerosols on climate are poorly constrained, and the implications of future increases in aerosol burdens are not known even semi-quantitatively.

One recurrent theme for ACE-Asia is that the climate forcing caused by Asian aerosols will become increasingly important in coming years as emission increase. Accordingly, the ACE-Asia network will address a number of key questions related to radiative impact of Asian aerosols, including:

(1) What chemical species control the optical properties and radiative impact of Asian multicomponent aerosols?

(2) What are the seasonal and interannual variations of Asian aerosol optical and radiative properties? How do aerosol optical depth and single scattering albedo evolve in time?

(3) How do these variations affect the surface radiation budget and surface temperature?

(4) To what extent can chemical transport and radiation transfer models reproduce aerosol impact in the Asian-Pacific region?

An approach to answer these questions will combine long-term aerosol optical and radiation measurements at the network of enhanced sites with short-term intensive field campaigns employing surface sites, ships, aircraft, satellites and mathematical models.

IV.B. Aerosol Sources and Sinks (Network Goal 2)

The sources and sinks of aerosol particles will be investigated because they are central to many other issues being investigated for ACE-Asia, including the oxidation of precursor gases, air-sea exchange, and aerosol evolution. The sources and sinks of substances such as sulfate and nitrate presumably will be investigated for various national acid deposition monitoring programs, and here again, mutual benefits would accrue from coordination between ACE-Asia and the national programs.

We will interpret the chemical data from the basic network coupled with single particle analyses using multivariate statistical methods, including factor analysis, to characterize major sources, i.e., biomass burning, fossil-fuel combustion, etc. Source regions for Asian dust will be characterized based on chemical tracers, trajectory analyses, and satellite imagery. As described in more detail below, the scavenging and removal of aerosols and aerosol precursors *via* precipitation will be quantified in collaboration with existing regional measurement programs.

IV.B.1. Oxidation Mechanisms of Aerosol Precursor Gases

A fundamental uncertainty regarding the role of sulfur compounds in radiative forcing is the fate of anthropogenic and biogenic S gases emitted to the atmosphere, i.e., whether they eventually form new aerosols, become incorporated into existing particles, or are removed before such reactions can occur. While there is general agreement that direct loss to surfaces (dry deposition) and conversion to sulfuric acid are the principal sinks for sulfur gases such as SO_x and DMS, there is no clear consensus as to their relative importance on the global or regional scales. Just as important, but also uncertain, are the relative contributions of homogeneous and multiphase photochemical processes, including reactions occurring in clouds (Chameides et al., 1984; Leaitch et al., 1986, Borys et al., 1988) and sea-salt processing (Chameides and Stelson, 1992; Sievering et al., 1992), for oxidizing SO_2 to H_2SO_4 (Yvon and Saltzman, 1996). These conversions depend critically on oxidants, such as those involved in the HO_x, NO_x, and, in the marine atmosphere, ClO_x and BrO_x cycles. Most of the process-level work on oxidation mechanisms will be conducted during intensives (ship, aircraft, and ground based), but models can tune the relative rates of these processes to match the patterns observed at the network.

IV.B.2. Atmospheric Deposition

Deposition to the surface is the ultimate sink for virtually all atmospheric aerosols, thereby providing an important constraint on chemical cycling. For instance, the average atmospheric lifetime of particulate SO_4^{2-} against deposition is about 4 days corresponding to about 90 turnovers per year of the global particulate S burden (e.g., Chen et al., 1996). The relative importance of wet vs. dry deposition varies spatially and temporally, but under most conditions, scavenging and removal by precipitation is the principal sink for radiatively important aerosols (e.g., Charlson et al., 1992; Penner et al., 1993). On a global scale, wet deposition accounts for 80% to 90% of the particulate SO_4^{2-} sink (e.g., Chen et al., 1996; and references therein). Because of its stochastic nature, deposition *via* precipitation contributes substantially to heterogeneity in the atmospheric burden of aerosols and associated radiative transfer. Consequently, a reliable predictive capability for radiative forcing by aerosols requires explicit consideration of corresponding deposition fluxes. Studies of wet-deposition fields also provide essential constraints for developing and testing regional chemical transport models.

ACE-Asia will focus on quantifying spatial and temporal variability in wet deposition over the study region for four reasons (1) wet-deposition is generally the major sink for radiatively important aerosols, (2) wet-deposition fluxes can be reliably measured at reasonable cost; (3) wet-deposition networks already exist in the ACE-Asia region, and (4) dry deposition is difficult and expensive to measure reliably. We will approach, and to the extent possible, organize regional measurement programs such that comparable data are generated by each network and reported to the project database. We will also initiate an external QA program (see below) to verify data quality. Additional sampling stations will be added as necessary to fill major gaps and shipboard sampling will be implemented during intensives (under the auspices of other ACE-Asia components) to extend deposition fields over the near-coastal ocean.

IV.B.3. Chemical Reactions and Aerosol Evolution

The particles and gases entering the ACE-Asia study region originate from a variety of sources, in some cases forming distinct layers in the atmosphere. Chemical interactions among the various aerosol constituents have important implications for tropospheric chemistry; for example, the alkalinity of mineral dust may influence the phase partitioning of nitric acid (Song and Carmichael, 1998). Similarly, heterogeneous reactions with calcium carbonate in crustal dust particles may be an important sink for SO_2 in the region (Dentener et al. (1996). Trace metals from natural and anthropogenic sources are ubiquitous components of the aerosol, and some of these metals can catalyze various types of chemical reactions. These chemical transformations significantly alter the composition of the aerosols, and in so doing change their optical properties (Hayasaka et al., 1993). We expect significant gas-to-particle conversion in the near surface troposphere of the ACE-Asia study region owing to the high ambient levels of SO_2 and organic substances.

The chemical and physical evolution of aerosol populations will be characterized as a function of flow regime and season based on corresponding population statistics (means, variabilities, etc.) of constituents (e.g., Moody et al., 1998). Investigations of this type will require the participation of meteorologists and the routine calculation of air-mass trajectories for each site.

Lagrangian experiments, in which air parcels are repeatedly sampled over time, provide a means for studying the chemical and physical processes that control aerosol particle evolution (Huebert et al., 1996). Intensive Lagrangian experiments will be discussed in detail in another of the ACE-Asia SIPs, however, pseudo-Lagrangian conditions may be encountered when air masses pass from one region to another, for example, from Qingdao to Cheju-Island to southern Japan. When, after the fact, trajectories indicate such transport has occurred, the relevant samples will be interpreted accordingly.

IV.C. Intercomparisons of Satellite and Ground-based Measurements (Network Goal 3)

NOTE: We need input from remote sensing working group

IV. D. Model Development and Validation (Network Goal 4)

NOTE: Need input from modeling working group

Mathematical models are an important tool for quantitatively integrating results and evaluating our understanding of physical and chemical processes in the atmosphere. The network data will be analyzed in conjunction with model implementation and evaluation: each particular experimental goal will have associated with it an appropriate physico-chemical model to serve as the test-bed for evaluating the data with respect to our overall understanding of the science. These models will provide a predictive capability for controls on spatial and temporal variability aerosol properties.

In ACE-Asia, for the first time in any large-scale experiment, the chemical evolution of mineral aerosol particles will be rigorously assessed. We hypothesize that high levels of anthropogenic emissions in the region will lead to chemical modification of dust particles and in their radiative and cloud nucleating properties. Prediction of radiative properties of the evolving aerosols

requires knowledge of their size distribution and chemical composition. Models are therefore required that track both gas-phase photochemistry as well as aerosol size and composition. Such models have only fairly recently been developed (Pilinis and Seinfeld, 1988; Meng et al., 1998), and they have been rigorously evaluated with ambient data only for the Los Angeles basin. To the extent possible we will collaborate with related modeling efforts under the auspices of China MAP and TRACE-P.

IV. E. Other Research Topics: Intensive Studies

Beyond the basic network operations, intensive experiments using the network facilities will add another dimension to the program. Here we briefly present several issues amenable to study at the ground stations. The issues presented are not meant to be exclusive, but rather they highlight examples of studies that could be profitably investigated using the network's resources.

IV.E.1. Local Closure Experiments

A key concept behind integrating models and measurements is the closure experiment (Quinn et al., 1996). In such an experiment an overdetermined set of observations is obtained, and the measured value of a dependent variable, such as light scattering by aerosols, is compared with the value calculated from the measured aerosol chemical and physical properties, using an appropriate model, *e.g.* Mie scattering model. A mass-closure analysis addresses the internal consistency of these measurements: Does the chemically analyzed mass account for the total gravimetrically-determined aerosol mass? Is the mass derived from the aerosol chemical size distribution consistent with that from the aerosol number size distribution?

The outcome of closure experiments provides a means for evaluating the uncertainties associated with models and measurements. If the measured and modeled values agree within the range of experimental error and at acceptable level of uncertainty, the model may be considered a suitable representation of the observed system and appropriate for use in higher order models. Poor agreement indicates that there are problems either in the model or measurements that must be corrected before proceeding further.

IV.E.2. Cloud-Aerosol Interactions and Cloud Processing

The changing aerosol burden in the ACE-Asia region has the potential to alter cloud radiative properties, cloud distributions, cloud lifetimes and precipitation patterns (e.g., Hobbs, 1993). There are tentatively four different ACE-Asia experiments planned to investigate cloud-aerosol interactions, and details of those studies will be presented in a separate SIP. The network measurements will provide important constraints on both in-cloud rates of S oxidation and changes in aerosol size spectra that are relevant for the aerosol-cloud component of ACE-Asia.

The aqueous-phase oxidation of S(IV) in clouds is important for sulfur chemistry and sulfate aerosol in particular because in-cloud reactions compete with dry deposition and various oxidation mechanisms as a sink for SO_2. Coagulation of aerosols within clouds may explain significant internal mixing inferred from observations (e.g.., Andreae et al., 1986). Such processes are particularly important for atmospheric dust because cloud processing can add a layer of sulfate

to the particles; changing their cloud nucleating properties, lifetimes, radiative properties, reactivity with other atmospheric constituents, and the solubility of dust-associated trace elements, such as iron.

We propose to conduct a scoping study in advance of a major aerosol/cloud field campaign to obtain data on cloud microphysics, cloud chemistry, and pre-cloud aerosol composition and size distributions. The major objective of this preliminary study is to **<A STATEMENT OF THE OBJECTIVE FOR THE PRELIMINARY STUDY IS NEEDED FROM CLOUD GROUP!>** and to evaluate logistical operations. The scoping study will be conducted at a network station, possibly Cheju Island, and it will run for at least 2 years, starting as early in the program as possible. The plan for preliminary aerosol-cloud interaction studies is another concrete example of the way in which the ACE-Asia network resources can be used to support other parts of the program.

IV.E.3. Cloud Condensation Nuclei

Measurements of cloud condensation nuclei (CCN) are needed to address a central topic of ACE–the indirect aerosol effect. Though the interpretation of CCN measurements is being challenged (Chuang et al., 1997), CCN provide the necessary linkage between aerosol measurements and clouds; CCN spectra provide concentrations of soluble ions within intervals directly related to the likelihood of cloud interactions. Surface measurements provide diurnal and seasonal climatologies that are not possible with aircraft measurements, another unique contribution of the ground stations. These results will be compared with CCN measurements in other parts of the world to assess the indirect effect of Asian aerosols relative to other major source regions. Because of cost and complexity, these measurements may be limited to one or two of the enhanced sites (e.g., Cheju) where CCN numbers can be related to other measurements.

V. Implementation of the ACE-Asia Surface Measurement Network

V.A. Study Domain and Site Locations

A network of surface sites will form the backbone of ACE-Asia. To maximize economies of scale, this network will be designed in collaboration with and build on existing sampling programs in the region including APARE/TRACE-P, China MAP, and various national programs. Some sites from past programs, such as the old SEAREX station at Midway in the North Pacific, will be reactivated while some active sites only need to add a few new instruments to fill-out their existing capabilities.

The influence of Asian dust can be observed every spring at least as far away as the Aleutians and Hawaii, so these will be the northern and eastern boundaries of the ACE-Asia network, respectively. The western boundary will be as close as possible to the main dust source regions in the Chinese deserts. As one main focus of ACE-Asia is on continental outflow, the southern boundary of the study domain will be ~20-30º N to avoid the trade winds that deliver marine aerosols to the continent. To the north, the ACE-Asia domain will extend to ~50º N because most of the pollution sources and outflow are found below this latitude. Several basic sites will be

established outside this domain (e.g., in Singapore or Thailand) to provide useful information for initializing models of the ACE-Asia domain.

Sampling will be implemented at regionally representative locations with local logistical support. We anticipate establishing approximately twenty basic network stations in the study region. Data from sites not strongly impacted by local emissions, especially mountain or island sites, will be particularly valuable for model evaluations, and remote sites of that nature will be most suitable for the enhanced ACE-Asia measurements. Potential sites will be visited to evaluate their suitability well in advance of the commencement of sampling operations. Several possible enhanced sites have been identified, including Shapuotou (37.5°N, 105°E) near the Asian dust source region; Qingdao (36°N, 120°E), on the east Asian coast; Kosan (34°N, 126.5°E), on the island of Cheju, south of the Korean peninsula, and several possible sites in Japan operated by M. Uematsu, including Rishiri (45 °N, 142 °E), Sado (38 °N, 138 °E), Hachijo (33 °N, 140 °E), Haha-jima (27 °N, 142 °E), Minami-torishima (24 °N, 154 °E). In addition, aerosol sampling is being conducted at Okinawa (27 °N, 128 °E) and Oki (36 °N, 133 °E) by H. Akimoto, with analses by S. Tanaka. Other Japanese groups (such as NIES) may provide aerosol data complementary to ACE-Asia, but they may not use the IMPROVE aerosol sampler. Open ocean enhanced sites will include Midway (28°N, 117.5°W) and possibly Oahu (21.5°N, 117.5°W).

The strategy of emphasizing remote sites for also is being adopted for TRACE-P, with the rationale that the remote sites will provide a more regionally representative picture of atmospheric conditions. Sites more strongly affected by local sources would be most useful for the basic network and for targeted, most likely intensive, studies. Principal investigators from any site within the ACE-Asia domain who are able to secure funding for basic measurements and who are willing to abide by the guidelines for submitting data to the archive will be encouraged to participate in the program and to become involved in the interpretation of the network data.

V.B. Station Operations

The network with be composed of two types of stations: basic and enhanced (Table 1). The basic stations will be outfitted a program standard aerosol sampler while the enhanced sites will be more highly-instrumented with more sophisticated equipment for measuring radiative fluxes, aerosol optical depth, and aerosol chemical, microphysical, and radiative properties. This dual-density network would provide the geographical coverage needed (1) to test how accurately models calculate aerosol spatial distributions and temporal variability and (2) to link aerosol distributions to radiative forcing.

Activities among network sites affiliated with participating programs (China MAP, EANET, TRACE-P, ACE-ASIA) will be coordinated to the extent possible, i.e., sampling protocols will be standardized and analytical methods intercompared. A quality assurance program will also be implemented. All ACE-Asia data will be archived in a central location to facilitate the exchange of information among participants and programs. This comprehensive regional data base will have benefits that extend long past the ACE-Asia time frame.

The network studies must be of a sufficient duration to characterize seasonal variability in major sources and processes. An absolute minimum of two annual cycles is needed; four to five years would be far more desirable for providing a context for the intensive studies. The long-term monitoring component of ACE-Asia also will be coordinated with WCRP activities (WMO Scientific Advisory Group on Aerosols and Aerosol Optical Depth); this will allow tracking of changes in climate forcing from Asian aerosols long after the intensive field operations of ACE-Asia have been completed.

Table 1. Network Stations and Modes of Operation

Type of Station	Mode of Operations	Enhancements	Types of Measurements
Basic	Routine	---	Routine measurements with an IMPROVE-type aerosol sampler following common protocols. Meteorology (continuous temperature, relative humidity, barometric pressure, surface wind speed and direction)
	Intensive	Various	Daily sampling, if possible, during March and April 2001 and other intensives. Specialized instruments, specialized protocols, limited sampling campaigns
Enhanced	Routine	Enhanced Chemistry Subnetwork	Basic measurements plus enhancements for the determination of size-separated aerosol chemistry, organic and elemental carbon, organic speciation
	Routine	Aerosol Optics and Radiation Subnetwork	Basic measurements plus enhancements for radiative fluxes and optical properties (see below for details)
	Routine	Deposition Subnetwork	Basic measurements plus enhancements for wet/dry deposition measurements
	Intensive Observations	Various	Specialized instruments, specialized protocols, limited sampling campaigns
LIDAR	Routine	---	Aerosol lidars

V.B.1. Basic Stations

V.B.1.a. Aerosol samplers

For the network data to be most useful, at least one common measurement needs to be made at each site, and to the extent possible on the mobile platforms. Participants at the Second ACE-Asia Planning Meeting in Cheju, Korea recommended that the IMPROVE-type sampler (IMPROVE stands for Interagency Monitoring of Protected Visual Environments) be deployed at all basic sites. The IMPROVE-Equivalent International Aerosol Sampler incorporates the California Air and Industrial Hygiene Laboratory's 23 l min^{-1} cyclone. This device collects aerosols smaller than 2.5 μm in diameter (PM-2.5), and it was adopted for the use in the IMPROVE network after side-by-side tests with other samplers. The sampler was recommended by the UN's WMO Global Atmospheric Watch Panel on Quality Assurance of GAW data (1993) and for GAW's Middle Eastern Network (1994-1998), and the sampler has been adopted for use by many other groups. In November, 1996, the US Environmental Protection Agency deemed IMPROVE the standard for all non-urban US sites. At this point, roughly 300 such samples are in active use with another 225 on order for emplacement in the US in Spring 1999.

<THE NETWORK WORK GROUP NEEDS TO DECIDE ON THE SELECTION OF AN AEROSOL SAMPLER. THERE ARE A NUMBER OF THINGS TO BE CONSIDERED, INCLUDING WHAT TYPES OF SAMPLERS WILL BE USED FOR OTHER NETWORKS. INPUT FROM MODELERS ALSO WOULD BE EXTREMELY HELPFUL.>

In the IMPROVE configuration, the sampler has three 2.5 μm channels (see Table 2), each leading to the appropriate filter substrate designed for a particular analysis. All channels are supported by a single pump, a 1/3 hp (roughly 250 watt) GAST double-piston pump, available in either 110V or 220V. The flow for each is reduced to 7.7 l min^{-1} by critical orifices, checked by a vacuum gauge on the pump, and validated by total flow measured by the pressure drop across the cyclone. The sampler also has a channel for 10 μm particles available, and it is flexible enough to allow alternative measurements *via* the fourth port. One design uses a low flow rate onto a Nuclepore® filter that allows for microscopic examination of single particles.

The aerosol sampling units are made in the machine shops of the Crocker Nuclear Laboratory, UC Davis; at actual cost. The total cost per unit (before shipping) is roughly $1250, and this includes a $350 pump. Filter cassettes are needed, and the cost of these is 6 for $120, but surplus cassettes may be available at nominal cost as IMPROVE is moving to a different system. The samplers normally take about two to three months to construct.

Twenty-hour hours will be the standard interval over which all 'basic' aerosol samples will be collected. However, sampling frequencies across the network may vary spatially and temporally based on resources available from participating national programs. We propose a standard in which a minimum of two 24-hr samples are collected on the same days each week at each site. More frequent sampling will be instituted during springtime intensive experiments, and different sampling intervals will be used for specific experiments, such as diel studies.

Table 2. Sampling Channels for the IMPROVE-Equivalent International Aerosol Sampler

Channel	Denuder	Filter	Analyses
A	None	25 mm Teflon®	Mass (gravimetric), Elemental analyses (including Al or Si etc. for dust), Optical absorption
B	Carbonate	25 mm nylon	Ion chromatography, SO_4^{2-}, NO_3^-, Cl^-, $CH_3SO_3^-$, NH_4^+, Na^+, Mg^{2+}, K^+, Ca^{2+}
C	None	25 mm quartz	Carbon by combustion, Organic C (4 temps), Elemental C (3 temps)

Table 3. Sampling Duration and Frequency and Chemical Analyses of the ACE-Asia Network Aerosol Samples

Species or Parameter	Station Type*	Possible Analytical Technique(s)	Sampling Duration and Frequency	Accuracy	Precision	Nominal LOD
Mass	Basic	Gravimetric Analysis	24 hrs 2 per week	TBD	TBD	TBD
Major ions	Basic	Ion Chromatography	24 hrs 2 per week	TBD	TBD	TBD
Mineral Aerosol (and Trace Elements)	Basic	Mass, XRF, PIXE, ICP-MS, INAA	24 hrs 2 per week	TBD	TBD	TBD
Organic/Elemental Carbon	Basic	Thermo-optical Techniques	24 hrs 2 per week	TBD	TBD	TBD
Size-Separated Dust	Enhanced	Mass, XRF, PIXE, ICP-MS, INAA	TBD	TBD	TBD	TBD
Size-Separated Anions	Enhanced	Ion Chromatography	TBD	TBD	TBD	TBD
Organic Species	Enhanced	Gas Chromatography-Mass Spectrometry	24 hrs 2 per week	TBD	TBD	TBD
Radionuclides	Enhanced	d-Spectrometry	24 hrs 2 per week	TBD	TBD	TBD
Single Particle Analysis	Enhanced	SEM, EMP, TEM	TBD	TBD	TBD	TBD
OTHERS?						

* All measurements made at the basic stations also will be made at the enhanced stations.

V.B.1.b. Aerosol analyses

One of the fundamental properties of the aerosol that can be determined with a reasonable amount of effort is the aerosol mass loading (Table 3). This property also is the basis for the PM-10 and PM-2.5 air pollution standards promulgated in the U.S., and the inclusion of this measurement for total suspended particles and/or the PM size fractions in the ACE-Asia studies will ensure comparability to large data bases in the United States and elsewhere. The gravimetric data also will provide a basis for normalizing other types of measurements, e.g., micrograms sulfate to micrograms total aerosol mass. Another important use of the gravimetric data will be for mass closure studies, in which the sum of the masses of all analytes will be compared with the total measured quantity. As the aerosol loadings in Asia will be quite high, the sensitivity of the gravimetric methods should not be an issue, but of course the proper handling and treatment of the filters is necessary, requiring some training for station operators.

A chemical measurement essential for the ACE-Asia studies is the determination of mineral dust concentrations (Table 3); this measurement will be made at all of the network stations. There are several approaches for doing this, generally based on the analysis of an indicator element such as Al or Si, although there are some interferences such as coal fly ash. Techniques used for the analyses include instrumental neutron activation, proton-induced X-ray emission, X-ray fluorescence, or inductively-coupled mass spectrometry, etc. Determining the ash free dry weights of the aerosol samples is an inexpensive and easy way to estimate the mineral aerosol loadings, but there are disadvantages to this approach because the chemical techniques will provide data for other substances, including sea salt and certain types of pollution aerosol.

As illustrated in Table 3, ion chromatography (IC) will be used for the routine analysis of the aerosol samples from all network sites. This is a well established technique used for the determination of a suite of anions, including ammonium, nitrate, nitrite, sodium, chloride, sulfate, and methanesulfonate in aqueous extracts of aerosol samples. Sodium and other cation concentrations will be determined either by ion chromatography or by an elemental method. Elemental carbon/organic carbon loadings for a groups of sites will be determined by M. Uematsu and his group from the University of Tokyo.

V.B.2. Enhanced Stations

V.B.2.a. Chemical properties–enhanced measurements

The measurements of aerosol composition over the basic network will be supplemented by the chemical analysis of size-separated aerosols at the enhanced stations (Table 3). Various types of cascade impactors can be used to sample size-separated aerosols for chemical analyses; for ACE-Asia the types impactor used for specific applications will be dictated by the amount of material needed for analysis, required integration times, size-cuts of interest, etc. One advantage of impactor samples is that they provide information on aerosol composition as a function of aerodynamic size, which has obvious relevance for evaluating transport processes and for relating the chemical data to physical properties.

Single particles will also be sampled and analyzed to provide a measure of the size distribution of the various mineral components of Asian dust, information absolutely required for a thorough evaluation of the optical properties of the dust particles. Single-particle analyses also have shown that aerosol populations are markedly heterogeneous (Anderson et al., 1996), a characteristic that is impossible to assess based on analysis of bulk samples. Analytical techniques for single particles include automated scanning electron microscopes, electron microprobes, and transmission electron microscopes.

^{7}Be and ^{210}Pb activities will be determined to trace air mass history and evaluate sources, specifically the relative influences of upper tropospheric/lower stratospheric vs. continental sources. These two naturally occurring radionuclides can be readily determined in bulk high-volume aerosol samples by direct gamma counting (e.g., Graustein and Turekian 1996).

V.B.2.b. Aerosol optical and radiation measurements

A subnetwork of enhanced sites will provide the ground-based measurements of aerosol optical and radiative properties needed to develop an aerosol climatology in the ACE-Asia study region and to quantify aerosol impact on atmospheric chemistry and climate.

The key aerosol optical characteristics required both for the assessment of radiative forcing as well as for satellite retrieval validations are spectral aerosol optical depth, aerosol light scattering coefficient, and aerosol light absorption coefficient. The latter two are needed for the calculation single scattering albedo, which is a crucial parameter, indicating the heating or cooling effects of aerosols. The measurements of these optical aerosol characteristics are currently performed at diverse monitoring stations around the world, and therefore, commercial instruments and standard operating procedures are readily available.

Complementary to aerosol optical measurements, a subnetwork of enhanced stations will perform the radiation measurements of integral solar direct, diffuse, and global radiation; integral infrared radiation; and sun brightness. The integral radiation measurements will be used to quantify aerosol radiative forcing at the surface and to constrain model simulations. The sun brightness measurements will be used to retrieve the particle size distribution covering larger sizes, which are not readily available from others measurements.

Table 4 lists the recommended instruments to measure optical and radiative characteristics of atmospheric aerosols at the subnetwork stations. Some instrument description is given in Appendix A.

Aerosol optical depth is measured by a sunphotometer. At a minimum, three-wavelength sunphotometers must be installed at the subnetwork stations. The recommended wavelengths are 340, 550, and 880 nm. It is required that measurements of scattering and absorption coefficients be performed at similar wavelengths.

The recommended integral solar and thermal radiation instruments (see Table 4) are relatively inexpensive and are easy to operate and maintain. These instruments are currently used at the Baseline Surface Radiation Network (BSRN) stations, which is sponsored by the World Climate Research Programme. Close collaboration with WMO/BSRN will be beneficial for both programs.

It is recommended that aerosol optical and radiation measurements be coordinated in time and be reported as hourly means. At selected stations, these measurements must be supplemented by measurements of size-resolved composition of the aerosol particles. A better understanding the relationships between various aerosol properties established from measurements is urgently needed.

It is desirable that some of the enhanced stations be co-located with existing lidar installations. Lidar measurements will provide valuable information about the aerosol vertical structure, which can be used for interpretation of other aerosol optical and radiation measurements as well as for radiation transfer models

During intensive field campaigns, ships and aircraft can be used to extend the measurements out over the western Pacific Ocean and through the vertical column. At these times the network stations will be further enhanced with instruments too complex or too expensive to operate on a continuous basis, but needed to provide a complete characterization of aerosol radiative forcing. The more comprehensive aerosol optical and radiation measurements will include aerosol light scattering coefficient at different relative humidities, aerosol backscattering coefficient, scattering phase function, sky and sun brightness, spectral global and diffuse solar radiation, and spectral UV radiation.

Special attention must be paid to the interpretation of the data collected in the ACE-Asia region. For instance, a nephelometer, which is used to measure the aerosol scattering coefficient, is typically calibrated with non-absorbing spherical latex particles. When dust or black carbon are dominant aerosol constituents, the measured scattering coefficients must be corrected to account for non-sphericity and strong absorption which are typical for these aerosols. Developing of adequate algorithms for the analysis and interpretation of measurements conducted at the network stations will be required.

Table 4. Aerosol Optical and Radiation Measurements Recommended for the Enhanced Stations

Parameter	Instrument
Aerosol spectral optical depth Aerosol light scattering coefficient	Sunphotometer Nephelometer
Aerosol light absorption coefficient	Aethalometer (or Photometer)
Direct solar radiation	Pyrheliometer
Global solar radiation	Pyranometer
Diffuse solar radiation	Shaded pyranometer
Long-wave radiation	Pyrgeometer
Sun brightness	Solar aureole photometer

V.B.2.c. LIDAR observations

WE NEED INPUT/TEXT FROM REMOTE SENSING WORK GROUP

(BH suggests the following:

(1) Objective: Obtain a climatology of backscatter in 3D with simultaneous lidar observation at many sites throughout Asia"

(2) Objective: Provide support for intensive observations from aircraft and ships by identifying transport pathways of dust clouds and layers in real time.

Much can be learned about the potential for long-range transport and the extent of impacts from continental emissions by understanding how aerosol loadings vary with altitude. This is a crucial area of research, but a comprehensive program to study vertical distributions of all important chemical species throughout the year would be prohibitively expensive. Detailed snapshots of the 3-dimensional structure of the aerosol burdens will be obtained from aircraft missions in intensive experiments for ACE-Asia, TRACE-P, and possibly other programs. Long-term observations made with aerosol lidars will be a potent complement to the more sporadic *in situ* observations of vertical structure.

These instruments can generate long-term backscatter data, but they cannot identify the chemical species involved. Sky radiance measurements can be used to infer size distributions, but require assumptions about the nature of the aerosol. This approach could be built around existing lidar installations in Chiba, Tokyo, Tsukuba, Anhui, Beijing, Shapato, Hong Kong, and Seoul. An effort is now underway to organize their observations into a network. A lidar group in Japan is testing their shipboard lidar onboard the RV Mirai, research vessel that will be used during the intensive studies. By 2003 lidar observations from satellites may be possible.

V.B.2.d. Wet deposition

Several programs are currently (or will soon begin) measuring and reporting wet-deposition fluxes in the ACE-Asia region (Ayers et al, 1996; B. Hicks, NOAA Air Resources Laboratory, personal communication, 1999) (Table 5). (GROUP, PLEASE ADD TO THE TABLE IF YOU CAN. ALSO, PLEASE SEND R. ARIMOTO CONTACT INFORMATION FOR ASSOCIATED PROGRAM MANAGERS.) Although data precision varies somewhat among programs, available information indicates that all quantify water deposition and concentrations of major inorganic chemical constituents of samples (H^+, Ca^{2+}, Mg^{2+}, K^+, Na^+, NH_4^+, NO_3^-, Cl^-, and SO_4^{2-}) without significant bias. However, because sampling protocols (wet-only versus bulk), preservation techniques, and integration times (daily to monthly) vary among these programs, all data from all programs may not be directly comparable. For instance, microbial growth in inadequately preserved precipitation samples can result in significant losses of carboxylic species, H^+, NH_4^+, and NO_3^- between collection and analysis (e.g., Mueller et al., 1982; Keene et al., 1983; Keene and Galloway, 1984; Herlihy et al., 1987). Cl-, SO_4^{2-} and base cations are less subject to such artifacts. In addition, the long integration times (weekly to monthly) employed by most programs preclude detailed analysis of source-receptor relationships.

Despite these potential limitations, all programs generate deposition data that would be useful for investigations planned as part of ACE-Asia. Consequently, we invite all programs in the region to participate in this research effort. Those that agree to collaborate will be asked (1) to provide data in a timely fashion (i.e., within about 6 months after sample collection) for incorporation into a common ACE-Asia data base and (2) to participate in a central, quality-assurance program involving periodic (every 2 months) analysis of external audit solutions and field blanks. After finalizing the regional coverage provided by collaborating programs, the ACE-Asia modeling community may recommend instrumenting additional sites to fill major gaps or to extend coverage to more remote island locations (in parallel with aerosol sampling described above). (NOTE: WE REQUEST INPUT FROM REGIONAL-SCALE MODELERS ON THIS POINT.) To maximize the utility of resulting data, wet-only precipitation will be sampled on a daily basis at any new stations added specifically for ACE-Asia. Samples will be preserved with a biocide immediately after collection and subsequently analyzed for $CH_3SO_3^-$ and carboxylic species ($HCOO^-$ and CH_3COO^-) in addition to the major inorganic constituents mentioned above. During intensives associated with other components of ACE-Asia, precipitation will also be sampled from ships to extend coverage of deposition fluxes over the coastal ocean; $CH_3SO_3^-$ will be measured in these samples to constrain fluxes of nss SO_4^{2-} originating from oceanic $(CH_3)_2S$ emissions..

V.B.3. Intensive Studies

Cloud condensation nuclei (CCN) studies

The feasibility of undertaking long-term measurements of CCN needs to be assessed for the basic stations, but these measurements would be valuable at the enhanced stations or in intensive experiments. Whenever they are made, the CCN measurements should continuously cover the supersaturation range 0.02 to 1%, with resolution of at least ten supersaturations. Sample processing for the CCN studies also should be done, but mostly during intensive study periods. The analyses for the CCN studies will include volatility and size vs. supersaturation, and particle-size-resolved chemistry. Sizing can be achieved with a differential mobility analyzer (DMA) located upstream of the CCN spectrometer. The results obtained with the DMA can be related to other aerosol size measurements as a means of determining the relative solubility of the particles. Volatility can be evaluated by heating the sample to various temperatures as a means of indirectly determining particle composition on a real time basis. Size-resolved chemistry can be achieved with a MOUDI or other type of impactor, which collects size-separated particles on special substrates. The CCN spectra from each MOUDI stage can be continuously monitored (cycling through the various size stages) so that it can be compared to the time-integrated sized-resolved chemistry, which should include both elemental and organic carbon analysis. Conservation of soluble ions or mass can be used to relate the two measurements. As carbon is the principal insoluble component of CCN, data for elemental and organic carbon could provide a degree of closure with the size vs. supersaturation measurements.

V.C. Quality Assurance/Quality Control

Need input from working groups

V.C.1. Data Quality Assurance

To ensure comparability of results among stations, we propose that one facility serve as a central reference laboratory for each subnetwork. The model we propose for this important exercise would have each country participating in ACE-Asia designate a national reference laboratory for the program, with the following comparisons between labs

- Periodic analysis by all labs of audit solutions provided by the central lab
- Periodic analysis by the central lab of field-sample and field-blank splits from the national labs
- Periodic analysis by the national labs of field-sample and field-blank splits from each participant lab
- Routine analysis of field blanks by all labs.
- Routine analysis of lab splits by all labs.
- Periodic summary reports by the central lab detailing analytical performance by each national lab.
- Periodic summary reports by each national lab detailing analytical performance by each of their respective participant labs.

This approach would have the advantage of minimizing the duplication of analytical cross-checks and from a more practical standpoint, strategically using the resources available for station operations.

V.C.2. Instrument Intercomparisons

Defendable, comparable measurements of the aerosols' chemical composition from surface and airborne platforms will be critical for achieving a number of our objectives. The use of a standard aerosol sampler at each of the network sites will obviate the need for extensive intercomparisons of the aerosol samplers used at the ACE-Asia sites, but some comparisons of this type may still be useful, especially if one of the other large networks uses a different sampler from the one used for ACE-Asia. A standard optical sensor for the sites would still require periodic calibrations.

The quality of the data will depend directly on how many groups take part in intercomparison experiments, to get their CN counters, particle sizers, chemical samplers, optical and radiative instruments tuned up to perform similarly. Those data that can be traced to intercompared instruments might be given a "quality- checked" flag in the data base. This will enable modelers to know which apparent concentration differences are the least likely to be the result of instrumental calibration variations. Without these quality control and intercomparison checks, the simultaneous data collection from a variety of sites would be of lesser value. Working groups have been formed to address several intercomparison issues, and these groups will focus on technique development and standardization prior to or in the initial phase of the experiment.

V.C.3. Analytical Intercomparisons

The intercomparisons must evaluate sample preparation methods (e.g., splits of acid or aqueous extracts) as well as the most commonly used instrumental methods. In addition, comparisons of different techniques used to determine the same substance also would be valuable. For example, mineral aerosol data will in all likelihood be obtained through individual particle analyses and by bulk chemical analyses. These techniques produce different yet complementary kinds of data: chemical analyses of bulk and cascade impactor samples produce mass concentrations for the dust while single-particle methods produce number concentrations plus size distributions. Converting to mass concentrations from the single particle data is not trivial owing to the presence of particles with complex shapes, the two-dimensional nature of the EM analysis of small particles, and the common occurrence of multi-phase aggregates. Even so, the comparison of single particle vs. bulk methods will provide a measure of the internal consistency in the two sets of results.

Several groups within ACE-Asia have the capability of doing single particle analyses, and if multiple groups are involved in the program, some differences will likely occur due to the methods used for sampling, the instrument's capabilities, and the approach used for data reduction. It is important to make any such differences known to the scientists, especially the modelers, who will use these data.

V.C. Implementation of Modeling Efforts

Need input from modeling work group.

V.E. Operational Issues Covered in the Project Prospectus

Several important sections that relate to the ACE-Asia Network are covered in the Project Prospectus: these are included in Section V–Project and Data Management and deal with (1) management structure, (2) data archive, (3) tentative schedule, and (4) world wide web.

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APPENDIX A. Instruments recommended for aerosol optical and radiation measurements.

Aethalometer.

Manufacturer: Magee Scientific.
Information is available on website: http://www.mageesci.com.
Model AE-30 operates at seven wavelengths from 450 to 900 nm.
Model AE-16 operates at a single channel of 880 nm will be appropriate.
The price range for these instruments is approximately $10K-$14K.

Multiwavelength Nephelometer.

Manufacturers: TSI or Radiance Research Corporation
Information is available on website: http://www.tsi.com

Broadband radiation instruments.

Manufacturer: THE EPPLEY LABORATORY, INC.
Information is available on website: http://www.eppleylab.com
PRECISION SPECTRAL PYRANOMETER (Model PSP)

The Precision Spectral Pyranometer is a World Meteorological Organization First Class Radiometer designed for the measurement of sun and sky radiation, totally or in defined broad wavelength bands. It comprises a circular multi-junction wire-wound Eppley thermopile which has the ability to withstand severe mechanical vibration and shock. Its receiver is coated with Parson's black lacquer (non-wavelength selective absorption). This instrument is supplied with a pair of removable precision ground and polished hemispheres of Schott optical glass. Both hemispheres are made of clear WG295 glass which is uniformly transparent to energy between 0.285 to 2.8µm. For special applications, other Schott glasses and Infrasil II quartz hemispheres are available. Included is a spirit level, adjustable leveling screws and a desiccator which can be readily inspected. The instrument has a cast bronze body with a white enameled guard disk (shield) and comes with a transit/storage case. A calibration certificate traceable to the World Radiation Reference and a temperature compensation curve is included.

SPECIFICATIONS

Sensitivity: approx. 9 µV/W m^{-2}.
Impedance: approx. 650 Ohms.
Temperature Dependence: ±1% over ambient temperature range -20 to +40°C (temperature compensation of sensitivity can be supplied over other ranges at additional charge).
Linearity: ±0.5% from 0 to 2800 W m^{-2}.
Response time: 1 second (1/e signal).
Cosine:
±1% from normalization 0-70° zenith angle;
±3% 70-80° zenith angle.
Mechanical Vibration: tested up to 20 g's without damage.
Calibration: integrating hemisphere.
Size: 5.75 inch diameter, 3.75 inches high.
Weight: 7 pounds.
Orientation: Performance is not affected by orientation or tilt.

INCIDENCE PYRHELIOMETER (Model NIP):

The Eppley Normal Incidence Pyrheliometer is a World Meteorological Organization First Class Pyrheliometer designed, as its name implies, for the measurement of solar radiation at normal incidence.

The NIP incorporates a wire-wound thermopile at the base of a tube, the aperature of which bears a ratio to its length of 1 to 10, subtending an angle of 5°43'30". The inside of this brass tube is blackened and suitably diaphragmed. The tube is filled with dry air at atmospheric pressure and sealed at the viewing end by an insert carrying a 1 mm thick, Infrasil II window. Two flanges, one at each end of the tube, are provided with a sighting arrangement for aiming the pyrheliometer directly at the sun. A manually rotatable wheel (not shown) which can accommodate three filters, while leaving one aperature free, is provided. The pyrheliometer is mounted on a power-driven equatorial mount for continuous readings. Please see Solar Trackers.

A calibration certificate traceable to the World Radiation Reference and a temperature compensation curve are included.

SPECIFICATIONS:

Sensitivity: approx. 8 µV/W m^{-2}.
Impedance: approx. 200 Ohms.
Temperature Dependence: ±1% over ambient temperature range -20 to +40°C.
(temp. compensation can be supplied over other ranges at additional charge.)
Linearity: ±0.5% from 0 to 1400 W m^{-2}.
Response time: 1 second (1/e signal).
Mechanical Vibration: tested up to 20 g's without damage.
Calibration: reference Eppley primary standard group of pyrheliometers.
Size: 11 inches long.
Weight: 5 pounds.

PRECISION INFRARED RADIOMETER (Model PIR):

The Precision Infrared Radiometer, Pyrgeometer, is intended for unidirectional operation in the measurement, separately, of incoming or outgoing terrestrial radiation as distinct from net long-wave flux. The PIR comprises a circular multi-junction wire-wound Eppley thermopile which has the ability to withstand severe mechanical vibration and shock. Its receiver is coated with Parson's black lacquer (non-wavelength selective absorption). Temperature compensation of detector response is incorporated. Radiation emitted by the detector in its corresponding orientation is automatically compensated, eliminating that portion of the signal. A battery voltage, precisely controlled by a thermistor which senses detector temperature continuously, is introduced into the principle electrical circuit.

Isolation of long-wave radiation from solar short-wave radiation in daytime is accomplished by using a silicone dome. The inner surface of this hemisphere has a vacuum-deposited interference filter with a transmission range of approximately 3.5 to 50 µm.

SPECIFICATIONS

Sensitivity: approx. 4 µV/W m^{-2}.
Impedance: approx. 700 Ohms.
Temperature Dependence: ±1% over ambient temperature range -20 to +40°C.
Linearity: ±1% from 0 to 700 W m^{-2}.
Response time: 2 seconds (1/e signal).
Cosine: better than 5%.
Mechanical Vibration: tested up to 20 g's without damage.

Calibration: blackbody reference.
Size: 5.75 inch diameter, 3.5 inches high.
Weight: 7 pounds.
Orientation: Performance is not affected by orientation or tilt.