

An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts

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[1] The International Global Atmospheric Chemistry Program (IGAC) has conducted a series of Aerosol Characterization Experiments (ACE) that integrate in situ measurements, satellite observations, and models to reduce the uncertainty in calculations of the climate forcing due to aerosol particles. ACE-Asia, the fourth in this series of experiments, consisted of two focused components: (1) An intensive field study that sought to quantify the spatial and vertical distribution of aerosol concentrations and properties, the processes controlling their formation, evolution, and fate, and the column-integrated radiative effect of the aerosol (late March through May 2001). (2) A longer-term network of ground stations that used in situ and column-integrated measurements to quantify the chemical, physical, and optical properties of aerosols in the ACE-Asia study area and to assess their spatial and temporal (seasonal and interannual) variability (2000–2003). The approach of the ACE-Asia science team was to make simultaneous measurements of aerosol chemical, physical, and optical properties and their radiative impacts in a variety of air masses, often coordinated with satellite overpasses. Three aircraft, two research ships, a network of lidars, and many surface sites gathered data on Asian aerosols. Chemical transport models (CTMs) were integrated into the program from the start, being used in a forecast mode during the intensive observation period to identify promising areas for airborne and ship observations and then later as tools for integrating observations. The testing and improvement of a wide range of aerosol models (including microphysical, radiative transfer, CTM, and global climate models) was one important way in which we assessed our understanding of the properties and controlling processes of Asian aerosols. We describe here the scientific goals and objectives of the ACE-Asia experiment, its observational strategies, the types of observations made by the mobile platforms and stationary sites, the models that will integrate our understanding of the climatic effect of aerosol particles, and the types of data that have been generated. Eight scientific questions focus the discussion. The intensive observations took place during a season of unusually heavy dust, so we have a large suite of observations of dust and its interaction with air pollutants. Further information about ACE-Asia can be found on the project Web site at <http://saga.pmel.noaa.gov/aceasia/>.

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1. Introduction

[2] Atmospheric aerosol particles affect the Earth's radiative balance directly by scattering [Charlson *et al.*, 1992] or absorbing [Ramanathan and Vogelmann, 1997] light, and indirectly by acting as cloud condensation nuclei (CCN), thereby influencing the albedo [Twomey, 1991; Charlson *et al.*, 1987], lifetime [Albrecht, 1989], extent and precipitation [Ramanathan *et al.*, 2001a, 2001b] of clouds. In many regions the natural aerosol has been substantially perturbed by anthropogenic activities, particularly by increases of sulfates, nitrates, organic condensates, soot, and soil dust. The present-day global mean radiative forcing due to the direct and first indirect (Twomey effect) effects of tropospheric anthropogenic aerosol particles is estimated to be between -1.1 and -2.7 W m^{-2} (Intergovernmental Panel on Climate Change (IPCC) [2001]: from summing the limits due to five aerosol types on page 351 of the WG1 report), which must be compared with the present-day forcing by greenhouse gases of between $+2.2$ and $+2.6$ W m^{-2} . Furthermore, the global distribution of aerosol particles is extremely inhomogeneous because of their relatively short lifetimes (of the order of days [IPCC, 1994]). As a result the negative forcing is focused in particular regions and subcontinental areas. This uneven forcing can cause continental to hemispheric-scale effects on climate patterns.

[3] Although aerosol particles have this potential climatic importance, they are poorly characterized in global climate models [National Research Council Panel on Aerosol Radiative Forcing and Climate Change, 1996; Anderson *et al.*, 2003b]. Tropospheric aerosols pose the largest uncertainty in model calculations of the climate forcing due to man-made changes in the composition of the atmosphere [IPCC, 2001]. This is a result of a lack of a clear understanding of the processes linking aerosol particles, aerosol precursor emissions, and radiative effects, and a lack of comprehensive global data that could be used to challenge models. How can one verify the utility of nanoscale process parameterizations in hundred-kilometer model grid cells? Aerosol measurements are needed in a globally representative range of natural and anthropogenically perturbed environments for model testing.

[4] Asian aerosol sources are unlike those in Europe and North America: much more coal and biomass are burned (in some cases with minimal emission controls), adding more absorbing soot and organic aerosol to parts of the Asian and Pacific atmospheres. Economic expansion in Asia has unavoidably been accompanied by increases in fossil fuel burning. Although recently sulfur emissions from China have begun to decrease [Carmichael *et al.*, 2002], economic expansion may yet cause the amount of SO_2 , organic matter, and soot emitted into the east Asian atmosphere to increase. The presence of Asian desert dust adds complexity, since it can scatter sunlight back to space (leading to a cooling effect), absorb solar and terrestrial radiation (leading to a warming effect) [Sokolik and Toon, 1996], and serve as an alkaline surface for the uptake of acidic gases [Dentener and Crutzen, 1993; Goodman *et al.*, 2000]. The oxidizing environment of the Asian atmosphere is likely to change as the growing transportation sector raises emissions of nitrogen oxides to levels like those in Europe and North America [van Aardenne *et al.*, 1999]. The fact that much of the Asian

aerosol then blows out over the Pacific implies that significant changes in radiative forcing may be expected over large areas.

[5] The International Global Atmospheric Chemistry Program (IGAC) organized an Asian Aerosol Characterization Experiment to address these questions about Asian aerosols. ACE-Asia included both an intensive observation period in the spring of 2001 and longer-term network observations. Details of the locations and measurements from each aircraft, ship, and surface site can be found in the Technical Appendix¹ to this paper. Further details and data from the program can be found on the project web site at <http://saga.pmel.noaa.gov/aceasia/>.

[6] The goal of ACE-Asia was to determine and understand the properties and controlling factors of the aerosol in the anthropogenically modified atmosphere of east Asia and the Northwest Pacific and to assess their relevance for radiative forcing of climate. To achieve this goal, ACE-Asia pursued three broad objectives: (1) The first objective, "Characterization," was to determine the physical, chemical and optical properties of the major aerosol types in the east Asian and Northwest Pacific region and investigate the relationships between these properties. (2) The second objective, "Radiation," was to quantify the interactions between aerosols and radiation in this region. (3) The third objective, "Processes," was to quantify the physical and chemical processes controlling the evolution of the major aerosol types and of their physical, chemical, and radiative properties and incorporate these into improved models.

[7] Below we discuss the strategies by which observations and modeling were used in the ACE-Asia experiment to reduce these important uncertainties.

2. Scientific Questions

[8] The three ACE-Asia objectives cannot be met without answering a host of more specific scientific questions. Several of these are outlined below, as examples of the kinds of detailed studies that were a part of ACE-Asia. In subsequent sections, we outline the strategies that were used for answering these questions, describe the resources (instruments and platforms) we had at our disposal, and evaluate our success in answering these questions using examples from papers in this special section.

2.1. Objective 1: Characterization

2.1.1. How Do Aerosol Chemical, Physical, and Optical Properties Change With Altitude, Location, and Time in Asia?

[9] The size-dependent chemical composition of aerosols controls everything from direct light scattering and absorption to water uptake ($f(\text{RH})$), the increase in light scattering with humidity) to the radiative properties of clouds. Soot, mineral dust, condensed organics, inorganic anions and cations, sea salt, and a variety of industrial aerosols can either be too dilute to measure, highly

¹ Supporting material is available via Web browser or via Anonymous FTP from <ftp://ftp.agu.org/apend/jd/2003JD003550>. Information on searching and submitting auxiliary material is found at http://www.agu.org/pubs/esupp_about.html.

concentrated, or in between in a given air mass. Because of the relatively short lifetime of particles, their concentrations and composition vary widely on scales of kilometers or less. It is important, therefore, to observe this variability to know not only the reasonable mean values but also the plausible ranges of values for each species in various regions. The mixing state of these various components also has a big impact on light absorption, cloud-nucleating capacity, and removal rates.

[10] The variability of properties with altitude is of particular importance, since many types of aerosol measurements have only been made at the surface. If layering is common and if the nature of the aerosol at altitude is different from that at the surface, this needs to be represented in models. To constrain the major natural and anthropogenic aerosol sources collocated in northeast Asia requires coordinated monitoring (including profile measurements) of complete aerosol characteristics along air mass pathways.

2.1.2. How Absorbing Are Asian Dust and Pollution Aerosols?

[11] Both black carbon (BC) and “yellow dust” particles absorb light in addition to scattering it. Since absorption converts radiation to heat in atmospheric layers, it has the potential to modify atmospheric dynamics and reduce rainfall [Ramanathan *et al.*, 2001a, 2001b; Rosenfeld, 2000; Rotstayn *et al.*, 2000; Chameides and Bergin, 2002; Menon *et al.*, 2002]. Absorption is usually parameterized in models using the single-scattering albedo, $\omega_0(\lambda)$, the fraction of extinction at each wavelength that is scattered rather than absorbed. The $\omega_0(\lambda)$ for dust is a controversial topic, since the small values, ~ 0.87 or less, previously attributed to Saharan dust [e.g., Patterson *et al.*, 1977; Levin *et al.*, 1980; Patterson, 1981; World Meteorological Organization (WMO), 1983] are now thought to be too small. AERONET and satellite reflectance data independently suggest values closer to 0.95 in the mid-visible [Tanre *et al.*, 2001; Kaufman *et al.*, 2001; Dubovik *et al.*, 2002]. However, lower values are expected for Asian dust particles since they are often mixed with absorbing anthropogenic aerosols during transport over the downwind industrial and urban areas [Kim *et al.*, 2001]. Is there an enhancement to the total absorption due to such internal mixing? The modeling of Chuang *et al.* [2003] suggests that the effect is small.

[12] Carbonaceous material from diesel exhaust, residential coal use, and biomass burning can also be strongly absorbing. The absorption per mass (MAE) depends strongly on the morphology of soot and light-absorbing organics and their state of mixing with nonabsorbing sulfates and organics that can enhance their absorption [Fuller *et al.*, 1999]. What values of MAE and $\omega_0(\lambda)$ should modelers use for various Asian aerosol mixtures?

2.1.3. How Do Size and Light Scattering by Asian Aerosols Change With Relative Humidity?

[13] Hygroscopic particles take up water as humidity increases, thereby growing larger ($g(\text{RH})$ is the relative increase in diameter with humidity) and scattering more light ($f(\text{RH})$). Presumably-insoluble mineral dust has a smaller $f(\text{RH})$ than do soluble sulfuric acid and sea-salt aerosols [Tang *et al.*, 1997], although this changes as dust adsorbs soluble vapors like nitric acid or coagulates in clouds with particles containing ionic materials. This is a

critical issue since humidity changes accompany all vertical movements of air and the changes in scattering with RH can be large (factors of several). Models need to be able to describe the evolution of $f(\text{RH})$ and $g(\text{RH})$ for all aerosol types as they age.

2.2. Objective 2: Radiation

2.2.1. With What Accuracy Can in Situ Measurements Be Integrated Over the Depth of the Atmosphere to Predict the Observed Radiative Effects?

[14] The accuracy with which in situ measurements can be integrated over the depth of the atmosphere to predict the observed radiative effects is really a test of the understanding of the entire aerosol/radiation system. Direct measurements of aerosol optical depth (AOD) and radiative fluxes across the IR, visible, and UV ranges can be used to test everything from our ability to sample aerosols of various sizes to our analytical methods and the models that link observed aerosol properties to their impact on radiative fluxes. We must be able to accurately represent the column values of AOD and radiant fluxes using models based on our measured values of aerosol concentration, $f(\text{RH})$, and ω_0 . These radiative flux measurements also provide a measure of the radiative forcing by aerosols, the “bottom line” in aerosol/climate forcing studies. If models can predict the forcing in a wide variety of air mass types, we gain confidence that they are realistic where we can’t make measurements.

2.2.2. How Much Quantitative Information About Aerosols and Their Radiative Impacts Can Be Derived From Satellites and Other Remote Sensors?

[15] Spaceborne observations allow us to look over long times at places where we cannot make in situ measurements [Kaufman *et al.*, 2002]. However, since they only measure radiances at the top of the atmosphere (TOA), models must be used to invert these radiances into the aerosol fields that could have produced them [Gordon, 1997]. The resulting aerosol products are thus a strong function of the assumptions in those models: for a given TOA radiance, a model assuming a highly reflective aerosol would infer a smaller concentration than would a model assuming a more absorbing aerosol. One of the most widely used satellite aerosol products, the TOMS absorbing aerosol index, is insensitive to aerosols near the surface so it can completely miss significant amounts of low-lying aerosol. It is critical, therefore, that in situ measurements be used to test and improve the algorithms used to derive aerosol products from satellites and to quantify the limitations of these products. Although satellite calibration and validation were not primary objectives of ACE-Asia, many ACE-Asia measurements can be of tremendous value for this purpose. In turn, validated satellite aerosol products (e.g., regional fields of multiwavelength optical depth, indicators of column size and absorption) can benefit ACE-Asia by providing a multifaceted picture of aerosol events during and surrounding the ACE-Asia time frame. Satellite data can also be used to extrapolate in situ observations over regions that appear to be homogeneous. Can these retrievals be made sufficiently accurate to detect trends?

[16] Lidars and sky radiometers are also widely used for making remote aerosol measurements. Often the measured light is analyzed to obtain estimates of aerosol properties

versus altitude (lidar) or column-integrated size distributions (sky radiometers). It is important to test these approaches against in situ measurements. Comparisons with in situ aerosol data can be used to identify indicators for selecting the most reasonable constraints to use in inversions of remote observations.

2.2.3. What Is the Radiative Forcing by Asian Aerosols and How Does It Change With Emissions, Time, and Distance From the Continent?

[17] This bottom line is what policymakers need, and it depends on understanding all the issues discussed above. They need to know the sensitivities of the radiative forcing to changes in controllable emissions like anthropogenic BC and SO₂ or to land use changes and agricultural practices that could alter the dust flux. The net forcing includes aerosol/cloud effects and direct effects in cloudy skies (as from layers of dust above clouds), both of which received less attention than did clear-sky direct radiative forcing in ACE-Asia.

2.3. Objective 3: Processes and Modeling

2.3.1. Are the Available Source Inventories for Asian Primary Aerosols and Gas-Phase Precursors Sufficiently Realistic to Support Assessments of Control Strategies?

[18] Since all the mass that models eventually put into aerosols has to be emitted from some source, aerosol models cannot be any better than the source terms they use. For some species (i.e., BC, SO₂, VOC, NO_x) these emission estimates are themselves derived from complex models of fuel usage, combustion conditions, and sociological factors [Streets *et al.*, 2001]. In other cases (i.e., sea salt and dust) emissions are estimated using parameterizations that approximate what we know about the influence of factors like wind speed and agricultural practices on primary particle generation [Woodcock, 1953; Gillette and Hanson, 1989]. These models rely on maps of soil types and vegetative cover, which may change seasonally or become outdated because of land-use changes. Some sources are highly variable in time (i.e., volcanoes and biomass burning), making it a challenge for models to accurately describe the emissions that may have affected the air observed on any particular day. Since the ambient concentrations are controlled by both sources and sinks, how well can observations constrain these sources?

2.3.2. How Are Aerosol Properties and Dynamics Modified by the Uptake of Gases?

[19] Emissions of volatile organics, NO_x, and SO₂ from the Asian continent undergo photo-oxidation as air masses are advected eastward over the Pacific, forming condensable organics, sulfates, and HNO₃ that can enlarge existing particles. If alkaline dust absorbs SO₂ and HNO₃ vapors it will also make the dust particles more hygroscopic, shortening their lifetime and increasing their impact on clouds. A key issue is the extent to which particles of continental origin retain their source identity in the face of gas-to-particle conversion and cloud processing. Dust no doubt also evolves in clouds and fogs, where coagulation mixes soot and industrial ash with dust and photochemical products to make a multicomponent aerosol of great complexity and variety.

2.4. Addendum

[20] Obviously this is not an exhaustive list of the scientific questions addressed during ACE-Asia. They do represent,

however, the kinds of issues for which we hoped to reduce uncertainties regarding the climatic impact of Asian aerosols.

3. Research Strategies

[21] We employed a wide variety of strategies for addressing these questions.

3.1. Making as Many Related Observations as Possible on Each Platform

[22] To characterize the regional and temporal distribution of aerosol properties in clean, polluted, and dusty air masses required comprehensive, coupled measurements of size-dependent aerosol chemical, optical, and physical properties from ships, aircraft, and surface sites during both a springtime intensive period and from a few surface sites over a period of years. Sampling and analytical artifacts were studied carefully and minimized whenever possible.

[23] We tried to measure the variables that control other variables so we could study their relationships. Size distributions and chemical composition, for example, affect scattering and absorption, so we measured all four simultaneously. Likewise scattering, absorption, and $f(\text{RH})$ control the extinction in a layer, so we measured them at the same time we measured the AOD and radiative flux changes above and below layers [Schmid *et al.*, 2003]. Closure experiments (see section 3.6) depend on the simultaneous measurements of related factors.

[24] Related measurements were also used to identify potentially contaminated or unreasonable points in data sets, as in an isolated case where the EC (elemental carbon) was very high but aerosol light absorption was not. Dramatic deviations from the relationships noted throughout the rest of the program can be a useful QA/QC flag. Corrections for measurement nonidealities often benefit from having related measurements. A good example is the use of optical particle counter (OPC) and aerodynamic particle sizer (APS) derived size distributions to compute the angular truncation error for the integrating nephelometers. On the C-130 we had 5 independent measures of sulfate aerosol: the rapid PILS sampler (which does not sample large particles), a Moudi cascade impactor (which suffered from hard-to-quantify tubing losses), a total aerosol sampler (TAS, which is slow but free of inlet or plumbing biases), an impactor/FTIR system (which is sensitive to only a few inorganic species), and samplers for electron microscopic analysis of individual particle composition (which is less quantitative). Each has its limitations but each also supplies unique information that helps to explain the total picture.

[25] Making all these related measurements clearly requires the participation of more groups than would be required if only one measure of sulfate and one measure of OC was needed. This strategy naturally fits into a large experiment like ACE-Asia, in which the instruments and personnel of many countries can be brought into play at the same time. Its success also requires that the project have an open data policy, so that all participants can freely compare their values with those of other groups.

3.2. Making Time Series Measurements at a Variety of Surface Sites

[26] Continuous time series measurements are one of the most powerful ways of testing chemical transport models.

Do the models show the correct seasonal cycle? Do they correctly forecast the arrival of particular air masses, pollution plumes, dust storms, etc.? Is there a diurnal variability? Are the statistics of the modeled values similar to those of the observations? Models need to reproduce observations on several timescales to be realistic.

[27] Eulerian time series observations were made at several dozen surface sites, as described in section 4 and in the Technical Appendix. These included a lidar network and instrumented sites in China, Taipei, Korea, Japan, Hong Kong, Hawaii, and the mainland United States. The Gosan site on Jeju Island, Korea, was among the most heavily instrumented surface sites.

3.3. Evaluating Spatial Variability Using Surface Networks, Ships, and Long Aircraft Flights

[28] We measured the same variables in many different places to test the ability of models to describe the spatial heterogeneity of aerosol concentrations and intensive properties. One challenge here is to intercompare or standardize the various measurements well enough to know how much of the apparent difference between sites could be caused by measurement uncertainty or bias, so that the remaining differences can confidently be ascribed to real spatial gradients. For some species (EC and OC on the mobile platforms and at Gosan are a good example) intercomparison experiments have quantified the agreement between sites and instruments [Schauer *et al.*, 2003]. For a few others (such as the Improve samplers, the UC Davis rotating drum impactors, and the EC measurements in the Japanese VMAP network) the sampling systems are identical in each location to ensure comparability. However, between these two examples lies a chasm: the VMAP EC was collected and analyzed using totally different physical principles from the NIOSH/Sunset Labs method used on the aircraft, the R/V *Ronald H. Brown*, and at Gosan. Trying to fit models to both these “EC” data sets could be seriously misleading.

[29] To study spatial variability, we also made three C-130 flights that covered large areas. One circled Japan, one circled Korea, and one flew a large circuit to the south of Japan and up the Chinese coast past Shanghai. In each case, the flight track was designed to characterize large-scale regional gradients and to define interfaces between air masses that models should be able to describe. In this case, there is no concern about calibration differences between instruments, but measurements in the various grid cells were not perfectly simultaneous.

3.4. Measuring the Altitude Profiles of Aerosols Using in Situ Measurements From Aircraft

[30] Measuring the altitude profiles using in situ measurements from aircraft is critical for determining how representative the more frequent surface measurements of aerosol physical and chemical properties are of aerosol layers in the column. We used three aircraft (two with comprehensive aerosol instrument packages) to measure aerosol concentrations and properties above the surface. Our profiles included rapid ascents and descents to get nearly-simultaneous measurements of AOD and a few other rapidly-measured parameters (SO₂, ozone, CN, scattering) at many altitudes, followed by longer sampling legs at a few interesting altitudes so the slower-responding instruments

(OC, EC, ionic concentrations, size distributions, absorption) had enough time to characterize those layers. We did profiles near a variety of surface sites, to add the vertical dimension to their time series measurements. We made numerous profiles near the Gosan site, flew past and above the R/V *Ronald H. Brown* several times, and profiled near the lidars in the Tokyo region.

[31] One of the things that made it possible to measure altitude profiles of dust is the new low-turbulence inlet (LTI) that was used on the C-130. This device, which was developed at Denver University (C. Wilson, personal communication, 2003), uses boundary layer-removal to eliminate turbulence in the inlet. Roughly eighty percent of the air entering the tip is sucked through the porous walls of the inlet cone, so that turbulence cannot propagate into the remaining flow. It enhances large particle concentrations, but does so in a calculable manner so that the relationship between ambient and post-inlet size distributions can be constrained.

3.5. Giving a High Priority to Intercalibrations, Intercomparisons Between Platforms, and Harmonizing of Similar Measurements for Flight Time and Other Resources

[32] It is unfortunately very easy to make misleading measurements of aerosols. For instance, most of the dust mass is in supermicron particles, which can easily be lost in inlets [Huebert *et al.*, 1990] or can settle out in sampling lines. Airborne sampling of particles is particularly difficult [Baumgardner and Huebert, 1993], both because of losses due to the turbulence that accompanies the rapid acceleration of air as it is sampled and because this acceleration causes an unavoidable dynamic heating that can evaporate semivolatile species. Stationary inlets also modify size distributions (often down to sizes of a few microns [Howell *et al.*, 1998]), making it hard to sample ambient distributions of large particles without artifacts.

[33] For many measurements the analytical methods are also sources of uncertainty. Light absorption by aerosols, for instance, is commonly measured by collecting particles on a filter and measuring changes in the transmission or reflection of light by the filter. However, the filter matrix and other collected particles cause multiple scattering that can increase the apparent absorption above what the ambient aerosols actually caused [Bond *et al.*, 1999]. Correcting for matrix effects in the presence of heavy dust is a challenge.

[34] Carbonaceous aerosols are hard even to classify, since “organic carbon” and “elemental carbon” are operationally defined [Lim *et al.*, 2003]. EC is often thought of as graphitic soot, but in most publications it is actually defined as the carbon that is burned off during heating in oxygen above some selected temperature. (BC or black carbon often refers to EC measured via light absorption.) Various instruments and techniques use different temperatures and analytical conditions, so their EC “concentrations” are not comparable. How can measurements in many locations by many groups using several methods be rationalized?

[35] We used several approaches to harmonize observations. Prior to the intensive phase, most of our TSI aerodynamic particle sizers were taken to the TSI facility to be cleaned, tuned up, calibrated, and intercompared. Many surface sampling sites used identical Improve samplers, to minimize inlet and sampling differences between them. As

noted above several groups used identical Sunset Labs OC/EC analyzers and the NIOSH analysis protocol, to ensure a common definition of EC and OC from the aircraft, the R/V *Ronald H. Brown*, and Gosan. They also did a blind analysis of punches from several exposed quartz filters to test the comparability of this analytical method in eight laboratories [Schauer *et al.*, 2003]. Likewise, inorganic ion unknowns were sent to nine laboratories to ensure that analytical calibration differences could be constrained.

[36] However, sampling issues could still lead identical instruments to different results, especially if one was on an aircraft with their (often acute) inlet issues and the other was on a ship or a tower with a different potential for contamination or loss of particles. We therefore did side-by-side intercomparisons between the aircraft and surface sites (flybys). These present a challenge in part because of the different speeds of the platforms: the C-130 aircraft would sample a 120 km long swath in 20 minutes, while it would take almost 7 hours for that same 120 km of air to reach the ship at 5 m/s wind speed. During those 7 hours any change in wind speed or direction could cause the ship to sample air that was different from what the integrating samplers on the aircraft had sampled. This problem is less acute, fortunately, for faster instruments that can make simultaneous measurements at the moment they pass the ship. The C-130 did 4 intercomparisons with the R/V *Ronald Brown*, 2 with the Twin Otter, 7 with Gosan, 1 with the Kingair, and 2 with the NASA P-3. The Twin Otter also made 3 comparisons with the *Brown*. As is the case in any such experiment, there were some instruments that were not functioning properly during each of these intercomparisons. The effort to interpret these experiments and to “harmonize” the measurements on the various platforms is still underway as of this writing.

[37] We coordinated two C-130 flights with the NASA P-3 aircraft, which was a part of the TRACE-P program [Jacob *et al.*, 2003]. The P-3’s payload included several measurements that were identical to those on the C-130 (hydrocarbons, SO₂, aerosol light scattering and absorption, and PILS anions and cations), so that by intercomparing we were able to tie together data sets that extended over a greater temporal and spatial domain. The P-3 also carried some photochemical species measurements (OH, H₂SO₄ vapor, and NO_x, to name a few) that provided us with valuable complimentary data. In like manner our more detailed aerosol measurements supplemented their data set. Our first two flights and their last two flights from Japan involved coordinated wingtip-to-wingtip flight legs. As a result of this careful intercalibration, A. Clarke *et al.* (unpublished manuscript, 2003) were able to see clear regional differences in the altitude profiles of optical properties: similar surface values of ω_0 in the northern (ACE-Asia) region and southern (TRACE-P) region were tied to larger ω_0 values aloft in the dusty north, and smaller ω_0 values aloft in the biomass-burning-dominated south. Biomass burning aerosols were only occasionally identified in the ACE-Asia flights.

3.6. Using Local Closure and Column Closure Experiments to Test the Consistency of Models and Measurements

[38] In a closure experiment, one attempts to fully characterize a system through measurements of both its integral properties (such as AOD) and its components (such as the

vertical distribution of aerosol properties), from which one should be able to predict the integral properties. If the predicted and observed values of the integral property agree to within measurement uncertainties, closure is achieved. Obviously this provides a stronger test the smaller the measurement uncertainties. Achieving closure in a variety of circumstances suggests that both the measurements and models are meaningful, whereas a failure to achieve closure could be the result of any one measurement or model not being correct. Closure experiments are often separated into local closure (between variables measured simultaneously in one location) and column closure (between a profile of in situ measurements and column-integrated properties such as AOD).

[39] One simple local closure experiment is the summation of measured masses of ions, OC, EC, and insoluble material from integrated filter or impactor samples, to derive a total aerosol mass to compare with one that has been determined gravimetrically. Missing mass or excess mass suggests that one or more constituents are not being measured properly. This type of closure experiment was conducted on the R/V *Ronald Brown* (P. K. Quinn *et al.*, Aerosol optical properties measured on board the R/V *Ronald H. Brown* during ACE-Asia as a function of aerosol chemical composition and source region, submitted to *Journal of Geophysical Research*, 2003). Unfortunately, because of the short sampling times on level flight legs, neither aircraft attempted to measure the gravimetric mass of particles. We did local closure experiments on optical properties (measured versus computed from composition and size using Mie theory) and on $f(\text{RH})$ (measured using humidity-controlled nephelometers versus computed from composition and thermodynamics), among others.

[40] Column closure experiments were done on extinction. Values derived from Sun photometer-measured AOD profiles were compared to those from in situ measurements of absorption and scattering [Schmid *et al.*, 2003; Redemann *et al.*, 2003], or from in situ measurements of size distribution and composition [Wang *et al.*, 2002]. Since Sun photometry does not rely on aerosol inlets, this type of experiment has proven to be of value for identifying times when inlet losses are more likely than others (compare Redemann *et al.* [2003] with Schmid *et al.* [2003]).

3.7. Measuring Direct and Diffuse Upwelling and Downwelling Radiative Fluxes at the Surface and Versus Altitude to Enable Closure Experiments and Direct Computations of Aerosol Radiative Forcing

[41] Ultimately, the impact of aerosols is a perturbation of the energy reaching the surface, leaving the top of the atmosphere, and being absorbed in layers of BC or dust. Measurements of these radiant fluxes [Bush and Valero, 2003] alongside the measurements of the aerosols doing the forcing allows for the two to be related and modeled.

3.8. Using Chemical Transport Models in a Forecast Mode to Predict the Locations of Interesting Features for Flight Planning and Ship Movements

[42] We used three models for mission planning (NCAR’s MATCH [Rasch *et al.*, 2001], the University of Iowa/Kyushu University CFORS model [Uno *et al.*, 2003], and NASA Goddard’s GOCART [Chin *et al.*, 2000]). Each

provided daily forecasts of dust and pollution outbreaks, enabling us to focus our sampling on these features that contained high concentrations or interesting gradients of aerosols. (Of course, this limits our ability to claim that our aerosol measurements are regionally representative.) In addition to the obvious benefit of getting more useful measurements than if we sampled randomly, this integrated these models into the experiment from the start. As a result, the process of improving the models on the basis of observations literally began while we were still in the field.

3.9. Coordinating in Situ Observations With Satellite Overpasses to Allow the Spatial Extrapolation of the in Situ Observations and to Improve Satellite Retrieval Algorithms

[43] The vast majority of C-130 and T/O flights included profiles timed to coincide with some satellite overpass (Table 1). These profiles usually included having the aircraft near the surface at the time of the overpass, followed by a rapid profile climb to the aircraft ceiling (or at least to get above most of the observed AOD) and then legs at several altitudes to allow for better characterization of the layers that caused the AOD. The satellites have been used to define the region over which the in situ observations might logically be extrapolated [Nakajima *et al.*, 2003], and the in situ observations are being used to perform calibration and validation of the satellite sensors and retrieval algorithms [J. Wang *et al.*, 2003; R. Kahn and C. Hsu, personal communications, 2003].

3.10. Evaluating Process Rates by Comparing Observed and Modeled Concentrations

[44] While process rates can be quantified using evolution experiments, we did not have that option. The few exceptions were times when we encountered by chance an air mass that had been sampled at a different location the day before (C. McNaughton *et al.*, The spatial distribution and size evolution of particles in Asian outflow: The significance of primary and secondary aerosol during ACE-Asia and TRACE-P, submitted to *Journal of Geophysical Research*, 2003). We also have a few measurements of composition in source regions from which to approximate the initial amount of various anions in dust that was sampled off the coast some days later. Another approach for assessing the uptake of SO₂ by dust is comparing measured concentrations of SO₂ and coarse-mode sulfate with model predictions using various uptake rates, to see which agrees best. The same is true for nitric acid uptake by alkaline dust, for which we lack time-separated data in an identified air mass with which to assess the evolution of this relationship.

[45] Thus observed spatial and temporal patterns of concentrations are our primary vehicle for testing and improving models. Of course, chemical transport models of this complexity include assumptions about sources, transport, evolution, and removal, so this “tuning” process unfortunately has a large number of parameters that can be varied. This limits to some extent our ability to constrain the rate of any one process by this method.

3.11. Opportunities

[46] April 2001 was a very dusty month in east Asia, so we had ample opportunities to employ these strategies on dust and dust/pollution mixtures. Many papers refer to a

Table 1. Number of Coordinated Flights in Satellite Scenes

| Coordinated Flights | C-130 | Twin Otter |
|---------------------|-------|------------|
| MODIS | 9 | 6 |
| MISR | 6 | 4 |
| MISR/Local mode | 3 | unknown |
| SeaWiFS | 7 | 8 |
| NOAA 14 | 2 | 2 |
| NOAA 16 | 4 | 0 |

pair of dust storms sometimes referred to as “the perfect dust storm,” in the period 5–15 April. Two intense pulses of dust from the Gobi Desert passed out of China, first into the northern Sea of Japan and later into the Yellow Sea. These were sampled by the ship and aircraft on multiple occasions. The concentrations on 11 April over the Yellow Sea were on the order of 1000 µg/m³ and were in places mixed with pollutants from the Beijing and Qingdao areas, so this case has been the focus of several papers.

4. Tools and Resources

[47] We had access to a variety of mobile platforms and fixed sites for conducting the ACE-Asia observations. Details of investigators, instruments on each platform, flight plans, ship tracks, and the location of each data archive can be found in the Technical Appendix to this paper.

4.1. Aircraft

[48] The C-130 and Twin Otter aircraft operated from the Iwakuni MCAS in Iwakuni, Japan. This was also the site of our Operations Center, where flight and ship operations were planned each day with the aid of the three chemical transport modeling groups.

[49] The largest aircraft was a Lockheed C-130A owned by the U.S. National Science Foundation and operated by the Research Aviation Facility of the National Center for Atmospheric Research. This plane carried a comprehensive package of measurements of precursor gases (SO₂, hydrocarbons, and ozone), tracer gases (halocarbons, Hg vapor, CO, and CO₂), aerosol chemistry (OC, EC, inorganics), aerosol morphology (SEM and TEM), number size distributions (SMPS, OPCs, APSs), optical properties (scattering, absorption, and lidar extinction/backscatter ratio), f(RH), lidar, AOD(λ), and radiant fluxes. The range of the C-130 allowed it to operate near Gosan, over the Yellow Sea, and throughout the Sea of Japan, with a ceiling of around 6 km. Most of the aerosol instruments sampled air from a pair of Low Turbulence Inlets (LTIs), which reduced the sampling artifacts that have plagued previous airborne studies of supermicron particles. The C-130 conducted 19 research flights from Iwakuni, between 31 March and 4 May 2001 (Figure 1).

[50] The U.S. Navy’s CIRPAS Twin Otter also operated out of Iwakuni. Its measurements were similar to those on the C-130, but it lacked a lidar, hydrocarbon/halocarbon measurements, CO, CO₂, and SO₂. However, it carried an Aerodyne aerosol mass spectrometer, which permitted it to make rapid measurements of sulfates and other species. The Twin Otter relied on a single long inlet above the cockpit to deliver sample to most of its instruments. The ceiling of this unpressurized plane was about 3.7 km. The Twin Otter also flew 19 flights (Figure 2) between 31 March and 30 April 2001.

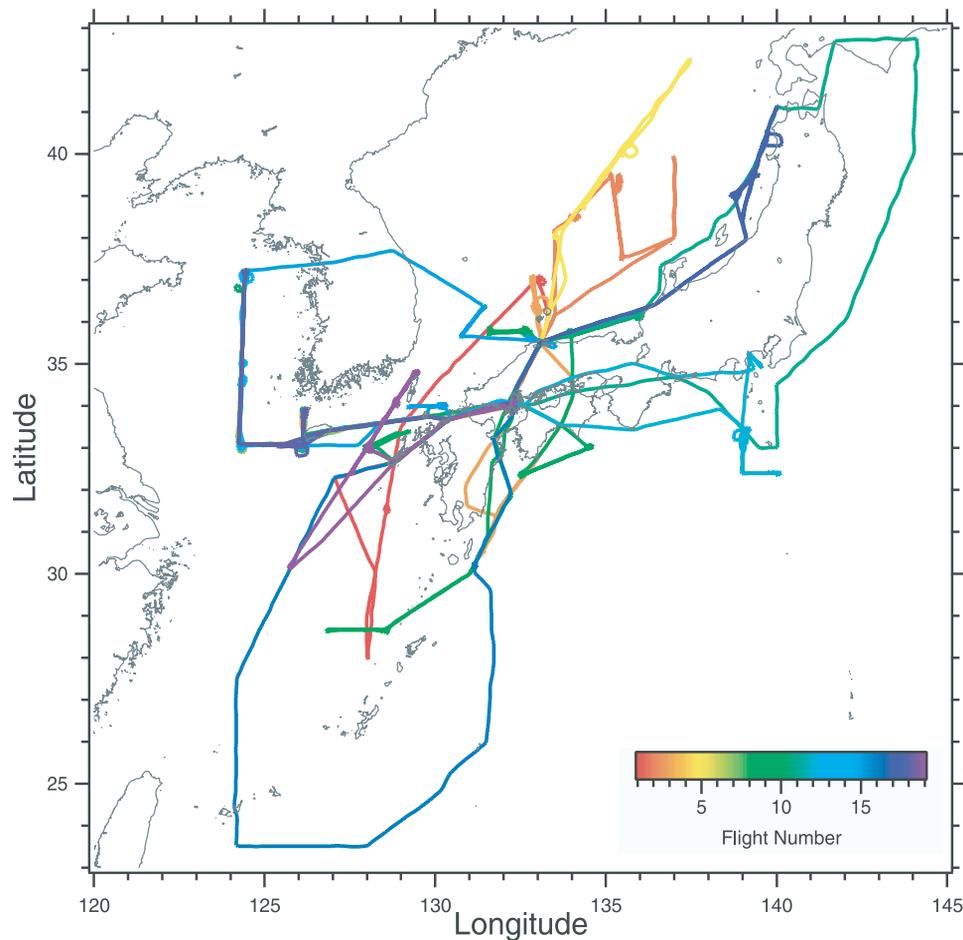


Figure 1. All 19 NSF/NCAR C-130 flight tracks. Colors represent time, starting with red through the rainbow to violet. Individual tracks are shown in the Technical Appendix.

[51] The third aircraft was the Australian ARA Kingair, which was supported in part by the Japanese APEX experiment. The Kingair operated from the Kagoshima Airport and focused more on the impact of aerosols on clouds. Its package was optimized for cloud physics, with a few aerosol measurements. Most of the Kingair operations were over the East China Sea between Kyushu and Amami Ohshima, where the APEX experiment had intensive surface observations. They flew 12 flights between 18 April and 1 May 2001.

4.2. Ships

[52] The NOAA R/V *Ronald H. Brown* departed Hawaii on 14 March 2001 carrying a very comprehensive package of aerosol precursor gas measurements, aerosol chemical, physical, and optical measurements, a lidar, some seawater chemistry measurements, and an extensive suite of radiant flux measurements. The air masses sampled during the first 10 days of the cruise had not been in contact with any continent for the past 3–6 days and were classified as background marine. On 25 March the air masses reaching the ship showed evidence of Asian continental air. Figure 3 shows the cruise track in the vicinity of Asia. The *Brown* sampled the plume of the Miyakejima volcano on 29–31 March east of Japan and again on 9–10 April in the Sea of Japan. The air sampled in the Sea of Japan (7–10 April)

was highly polluted with back trajectories crossing Japan. On April 11 a cold front passed the ship bringing high concentrations of dust. These conditions continued through 14 April. The *Brown* sampled moderately polluted air southwest of Japan from 14 to 18 April and ended the cruise in Yokosuka, Japan, on 20 April.

[53] The JAMSTEC R/V *Mirai* made continuous in situ measurements during a 14–27 May cruise (Figure 4) [Yoshikawa *et al.*, 2002; K. Miura *et al.*, manuscripts in preparation, 2003]. K. Miura *et al.* (manuscripts in preparation, 2003) inferred the lifetime of aerosols using size distributions and anthropogenic tracers. After passing a low-pressure system on 18 May radon and supermicron aerosol concentration increased, as predicted by the CFORS model [Uno *et al.*, 2003]. Mixtures of soil dust and anthropogenic particles caused relatively high scattering and absorption coefficients and high concentrations of nss-SO_4^{2-} , Al, Fe, and EC [Fujitani *et al.*, 2002].

[54] Lidar observations were performed alongside a 95-GHz cloud profiling radar throughout the cruise. Plumes of dust were observed on 20–21 and 25–26 May. On 26 May a layered structure of dust and anthropogenic aerosols were observed [Sugimoto *et al.*, 2002]. These plumes were also measured with an OPC sonde on a kytoon. The CFORS model suggests that this air mass was a sulfate aerosol plume from the Asian continent. H. Okamoto *et al.*

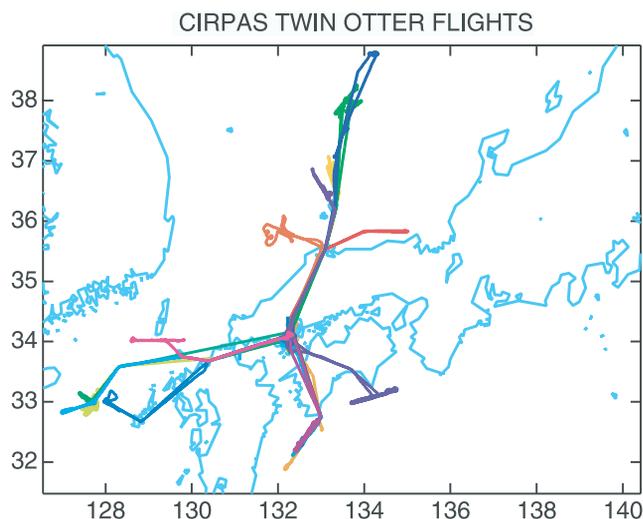


Figure 2. All Twin Otter flight tracks. Individual tracks are shown in the Technical Appendix.

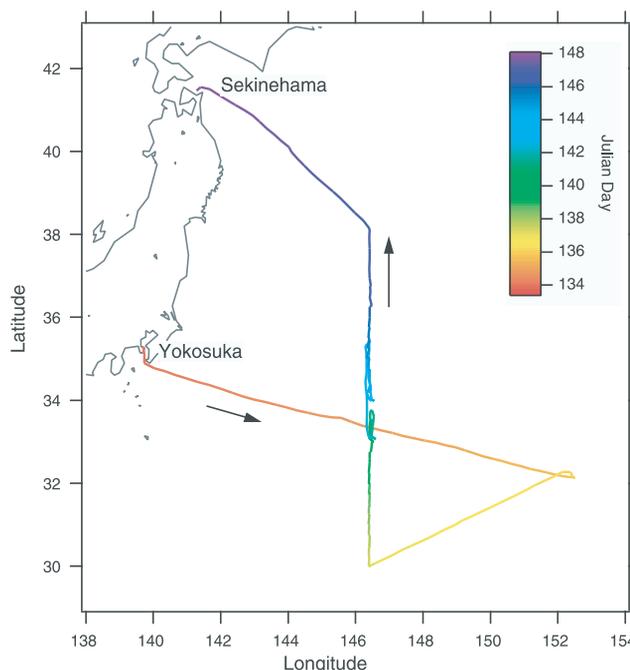


Figure 4. R/V Mirai cruise tracks.

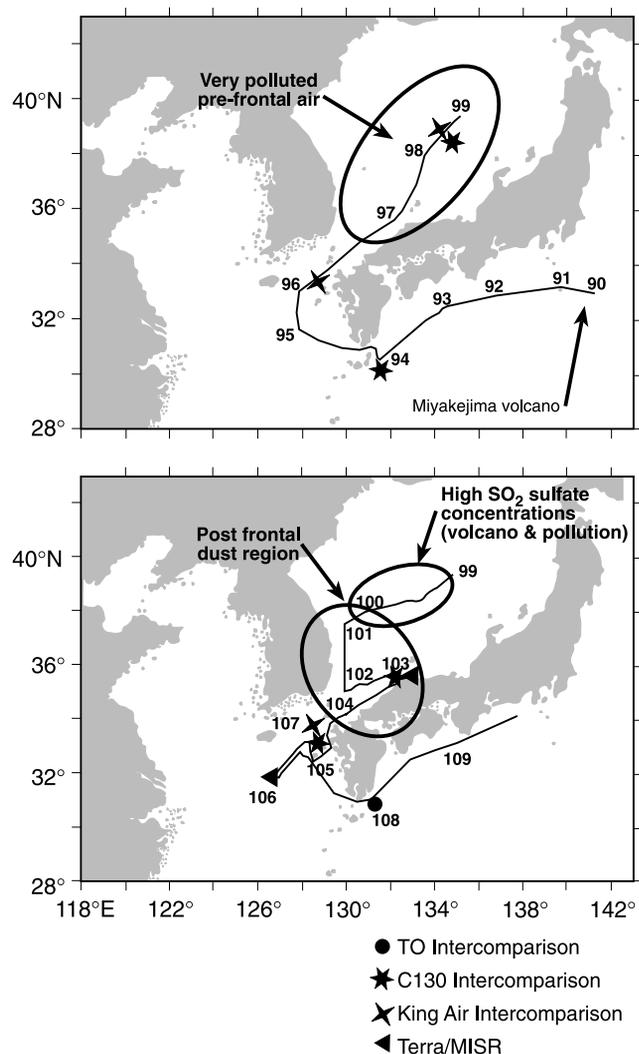


Figure 3. R/V Ronald Brown cruise tracks near Asia.

(manuscript in preparation, 2003) used the lidar and cloud radar to study microphysical parameters of ice and water clouds. Bistatic lidar experiments were also performed to measure water cloud particle size [Sugimoto *et al.*, 2001].

4.3. Surface Network Sites

[55] A variety of surface observations were made (Figure 5), including some in networks that extended over large distances.

4.3.1. Gosan

[56] The Gosan super site (126°10'E, 33°17'N) on Jeju Island, Korea, is located between major aerosol source regions. It supported a wide variety of aerosol and radiation measurements, in addition to two lidars. Monitoring of physico-chemical and optical properties of aerosols and air mass back-trajectory analysis revealed that different aerosol characteristics were observed depending on the meteorological conditions. Aerosol characteristics observed at Gosan showed that it was frequently impacted by Asian desert dust storms and anthropogenic sources located in China, Korea, and Japan depending on its air mass history. Two major Asian Dust outbreaks were observed at Gosan super site, approximately 11–13 April and approximately 25–26 April during the ACE-Asia IOP. Results of air mass back trajectory analysis confirmed by model simulations showed that two dust events originated from different source regions. The first one originated from the northwestern Chinese desert region and traveled through Chinese coastal industrial areas and over the Yellow sea. The latter one originated from the sandy area in northeastern China and passed over the Korean peninsula before it reached Gosan.

4.3.2. People's Republic of China

[57] Observations of aerosol chemical and physical properties were made during the AEC-Asia IOP at the China Dust Storm Research network of ground stations in hyper-arid, semi-arid, urban, rural, and coastal areas of China [Zhang *et al.*, 2003; Gong *et al.*, 2003]. The sampling sites



Figure 5. ACE-Asia Surface and Network sites.

included Aksu, Dunhuang, Zhenbeitai, Xi'an, Changwu, and Beijing. Dust from western sources contained 12% Ca by mass, a number which has subsequently been used to infer dust mass from Ca measurements on the aircraft. In the source regions 1.7% of dust mass was in particles $<2.5 \mu\text{m}$ and 30% was $>16 \mu\text{m}$. Five major transport pathways of Asian dust storm were identified during this period through back trajectory analyses. The Dunhuang and Zhenbeitai sites also hosted extensive radiation and remote sensing measurements [Zhao *et al.*, 2003; Iwasaka *et al.*, 2003; Y. S. Kim *et al.*, 2003].

4.3.3. Variation of Marine Aerosol Properties (VMAP)

[58] The Japanese network VMAP allowed for latitudinal gradient measurements of aerosols along $141^\circ\text{--}142^\circ\text{E}$: Rishiri (45°N), Sado (38°N), Hachijo (33°N), and Chichi-jima (27°N). This provided an opportunity to look at fine and coarse major ions, trace elements, and OC/EC [Matsumoto *et al.*, 2003] versus latitude. Several other species were measured, but not at all sites. The variety of settings, from northern rural Japan to urban to remote islands is excellent for testing models.

4.3.4. APEX

[59] The Amami Ohshima site supported a wide array of aerosol and radiation measurements. The chemical composition time series has been used to understand the air mass characteristics over Amami-Ohshima. Between 20 and 30% of the aerosols were carbonaceous aerosols (S. Ohta *et al.*, unpublished manuscript, 2003), which is consistent with CFORS and SPRINTARS aerosol models [Uno *et al.*, 2003; Takemura *et al.*, 2003]. Comparison of aerosol chemical composition with an aerosol type classification from SeaWiFS [Higurashi and Nakajima, 2002] looks very promising. Aerosol radiative forcing at Amami-Ohshima has been compared with that of the Gosan site by D.-H. Kim *et al.* [2003]. The Gosan site aerosols have a smaller forcing compared with that of Amami-Ohshima from surface radiation measurements [Nakajima *et al.*, 2003].

4.3.5. Asian Dust Network (AD-Net)

[60] Asian dust emission and transport were studied with lidar network observations coupled to the CFORS model. This analysis suggests that most of the Asian dust trans-

ported to Japan originated in Inner Mongolia and/or Mongolia [N. Sugimoto *et al.*, unpublished manuscript, 2003; Shimizu *et al.*, 2003; Uno *et al.*, 2003]. In the free troposphere, however, the dust from Tarim basin was sometimes significant up to 10 km. By contrast, polarization lidar observations suggest that most of anthropogenic aerosols were transported below 3 km.

[61] Around 10 April the optical characteristics of a dust layer above 4 km (during the Perfect Dust Storm case) were studied with the AD-Net lidar network, a MPL on the R/V *Ron Brown*, radiometers, and satellite data. T. Murayama *et al.* (private communication, 2002) estimated radiative forcing due to elevated dust above cumulus clouds, a potential warming situation. On 23 April the ground-based lidars in the Tokyo area were compared to the in situ airborne instruments on NCAR C-130. The extinction profiles obtained with the lidars and the airborne instruments (a tracking Sun photometer, nephelometers, and PSAP) were in good agreement [Murayama *et al.*, 2003] except near the surface, where horizontal heterogeneity in the Tokyo area was a factor. The mixing state of dust and sulfate inferred from polarization lidar observation was also well explained by the airborne optical and chemical measurements. The extinction-backscatter ratio of dust obtained by Raman lidar agreed with the airborne nephelometric lidar ratio measurement. Intercomparison studies between lidars, Sun photometers and airborne instruments were also performed near Jeju Island [S.-C. Yoon *et al.*, unpublished manuscript, 2003; Hong *et al.*, 2003].

4.3.6. Hong Kong Site

[62] As a cooperative effort with the TRACE-P and ACE-Asia intensive campaigns in spring 2001, trace gases [T. Wang *et al.*, 2003] and aerosols were measured at the Hong Kong Polytechnic University's Atmospheric Research Station at Hok Tsui in southeastern Hong Kong ($22^\circ13'\text{N}$, $114^\circ15'\text{E}$, elevation: 60 m above the sea level [Wang *et al.*, 1997]). The main objective of the measurement program was to provide continuous ground-based data in the subtropical region of east Asia as a contrast to the more dust-dominated sites in northern China. The field measurement was a collaborative effort of scientists from the Hong Kong Poly-

technic University, University of California at Irvine, and the Australian Nuclear Science and Technology Organization.

5. Discussion

[63] How successful were these strategies and platforms in answering the eight scientific questions posed above? Furthermore, are there lessons or changes in strategy that could make future aerosol/climate studies more productive?

5.1. How Do Chemical, Physical, and Optical Aerosol Properties Change With Altitude, Location, and Time in Asia?

[64] The measurements made during ACE-Asia revealed intensive layering of aerosols advecting out of Asia with multiple layers of dust and or pollution separated by clean layers [Redemann *et al.*, 2003; Bahreini *et al.*, 2003]. Using a 1 μm dry (55% RH or less) aerodynamic diameter cut point for many of the measurements effectively separated the two dominant and independent modes of the size distribution [Anderson *et al.*, 2003a]. Dust, which dominated the coarse mode, was present throughout the column while pollution aerosols (organic and ionic species), which dominated the accumulation mode, were generally confined to the boundary layer ($< \sim 1.5\text{--}2.0$ km) [Anderson *et al.*, 2003a; Chin *et al.*, 2003]. Although the two dominant modes of the size distribution were distinctly different in the relative amounts of different chemical species, dust also was present in the submicron (accumulation) mode and sulfate, nitrate, organic carbon and elemental carbon also were present on the dust particles in the coarse mode, presumably from heterogeneous reactions on the dust and coagulation of pollution aerosol with dust during long range transport [Alfaro *et al.*, 2003; Chuang *et al.*, 2003]. The relative amount of absorbing aerosols associated with the coarse-mode dust decreased with altitude (A. Clarke *et al.*, unpublished manuscript, 2003). The single-scattering albedo increased with altitude during ACE-Asia because of the highly scattering/low-absorbing dust aloft. In contrast, the single-scattering albedo decreased with altitude during TRACE-P (A. Clarke *et al.*, unpublished manuscript, 2003), perhaps because they encountered much less dust. Surface values of the single-scattering albedo at 550 nm for both experiments, north and south of 25°N were similar (0.88 ± 0.03 in pollution and 0.96 ± 0.01 for dust).

[65] The Aerodyne Aerosol Mass Spectrometer (AMS) aboard CIRPAS's Twin Otter measured distinct layers of submicron, nonrefractory aerosols from the surface to an altitude of ~ 3700 m [Bahreini *et al.*, 2003]. The composition of the nonrefractory aerosols was dominated by sulfate and organics. Mass-weighted size distributions of sulfate aerosols showed little variation from day to day and layer to layer. Higher organic concentrations were measured when the AMS sampled air masses from Korea rather than from northern China.

[66] The physical, chemical and optical properties of Asian dust aerosols at Kwangju, Korea [D.-H. Kim *et al.*, 2003], varied depending on the source region and transport path of the air mass. Major air mass pathways of Asian dust storms were from either northwestern Chinese desert regions or northeastern Chinese sandy areas during the ACE-Asia IOP. This variation of composition with source region was found at most time series sites.

[67] Dust had a marked influence on the size distributions of several species. For example, low-molecular-weight dicarboxylic acids were predominantly on fine-mode particles under nondusty conditions. However, during a strong dust event oxalic and other diacids were also found in the coarse mode, suggesting a significant modification of organic aerosols by alkaline dust particles [Kawamura *et al.*, 2003; Mochida *et al.*, 2003a]. Water-soluble organic species were dominated by oxalic acid followed by malonic or succinic acid [Matsunaga *et al.*, 2002]. Diacids in the fine mode may be evaporated and then deposited on the coarse dust particles that contain alkaline minerals (e.g., CaCO_3), or they may deposit on this large surface area immediately after photochemical formation. These same compounds served as indicators of the spatial influence of Asian emissions: dicarboxylic acids were found to be more abundant off the coasts of east Asia, especially in the Sea of Japan and East China Sea, decreasing toward the central Pacific [Mochida *et al.*, 2003b]. The atmospheric aerosol composition in the western North Pacific is heavily influenced by the photochemical oxidation of organic precursors from Asian source regions.

[68] Although we gained significant insights, there is room for improvement. While the new Low Turbulence Inlet on the NSF C-130 provided for the first time a quantitative supermicron sample for the instruments inside the plane, many in situ measurements on the various platforms were limited in their ability to measure dust. There is evidence from both the C-130 and the R/V *Ronald H. Brown* that there was bounce in the impactors used for the chemical measurements. The Aerodynamic Particle Sizer (APS), our main instrument on all platforms for measuring the physical size distribution, appears to undercount or undersize dust particles. This is likely a result of the nonsphericity of the particles. Future experiments should include a laboratory phase where instruments are characterized and tested for the conditions expected in the field. In hindsight, there should have been an effort to generate dust and test the response of the sizing instruments. Similarly, we should have made dust aerosol in the laboratory, tested impactors for bounce, and tested strategies to minimize this artifact (e.g., noncontaminating coatings, etc.). In addition, since a large focus of this experiment was dust, there should have been a standard chemical measure of dust on each platform, something that was particularly lacking on the aircraft.

[69] An additional surface super site (as heavily instrumented as the Gosan site) in the dust source regions would have provided a valuable measure of chemical composition and optical characteristics of Asian dust aerosols before they were transported to polluted areas. Since no aircraft measurements were made over the dust source regions, we lack in situ vertical profile measurements of physical, chemical and radiative properties that could initialize the chemical transport models and thus support more defensible conclusions about process rates (section 5.8, below).

5.2. How Absorbing Are Asian Dust and Pollution Aerosols?

[70] The absorbing aerosols were somewhat surprising. Chuang *et al.* [2003] found that roughly 40% of the EC they measured was in supermicron particles when dust was

present, apparently the result of coagulation between soot and dust during transport. Despite this, the dust aerosol was observed to be relatively nonabsorbing. For example, airborne measurements indicated that the mean SSA of the supermicron aerosol during dust events was 0.96 [Anderson *et al.*, 2003a].

[71] It is difficult to measure absorption well, in part because most devices collect absorbing aerosols on a filter and measure transmittance or reflectance. Filter-based methods are calibrated with simple, laboratory aerosols and it is possible that the response to complex ambient aerosols is different [Bond *et al.*, 1999]. We did not have either photoacoustic or extinction cell approaches for measuring absorption of still-suspended particles, which would have provided valuable comparisons with the filter-based methods. Most of the in situ absorption results analyzed to date are at a single-wavelength, which leaves us ignorant about its wavelength dependence. It is true most of the solar energy is in the visible (where all the PSAPs made their measurements), but we are unable to observationally constrain flux changes in either the IR or the UV (which have important actinic implications for the photochemistry that creates both condensable species and oxidants). The TRACE-P data suggests that these actinic changes can be significant [Lefer *et al.*, 2003; Tang *et al.*, 2003].

[72] However, we did have airborne measurements of spectrally resolved radiant fluxes [e.g., Pilewskie *et al.*, 2003], which can be used to derive spectra of aerosol layer absorption and ω_0 (the latter when layer AOD spectra are combined with the absorption spectra in a radiative transfer model). The same types of measurements have recently been used to derive aerosol absorption and ω_0 spectra for African dust and smoke aerosols [P. Pilewskie *et al.*, unpublished manuscript, 2003; Bergstrom *et al.*, 2003]. Corresponding analyses of the ACE-Asia absorption spectra are underway and will be reported in a later issue. These measurements describe absorption by aerosols in their ambient state, without the sampling issues mentioned above.

[73] Single-scattering albedos based on these PSAP and nephelometric measurements suggest that Asian dust itself is not as strongly absorbing in the mid-visible as some early results for African dust [e.g., Patterson *et al.*, 1977; Levin *et al.*, 1980; Patterson, 1981; WMO-IAMAP, 1983]. The ω_0 of coarse-mode aerosol sampled by aircraft during dust-dominated events fell within the range of 0.95 to 0.97 [Anderson *et al.*, 2003b]; the most significant absorption by Asian dust seems to be in the UV and IR [Sokolik, 2002]. Recent analyses of ground-based Sun/sky photometry and of satellite reflectance [Tanre *et al.*, 2001; Kaufman *et al.*, 2001; Dubovik *et al.*, 2002] independently yield absorption by Saharan dust that is much weaker than the earlier results cited above. On the other hand, the single-scattering albedo (ω_0) of atmospheric aerosol at Kwangju increased from 0.85 for nondust periods to 0.87–0.95 during dust storms. Indeed, the ω_0 in the coarse mode was as low as 0.81 during one Asian dust storm period when fine soot particles had agglomerated to coarse dust particles during transport over industrial/urban areas [D.-H. Kim *et al.*, 2003].

[74] To model absorption one needs to know the mass of the absorber. We used two principal instruments for measuring EC in ACE-Asia: an R&P semi-continuous thermal

EC/OC analyzer in the VMAP network [Matsumoto *et al.*, 2003] and the Sunset Labs thermal/optical NIOSH 5040 method [Birch, 1999] on the April 2001 intensive mobile platforms and at Gosan. While we have intercompared Sunset instruments used by eight groups [Schauer *et al.*, 2003], we have not resolved the differences between the R&P and NIOSH methods. The former collects only particles above 0.15 μm and apports evolved carbon to EC or OC on the basis of a fixed temperature, while the latter collects all sizes on a quartz filter and uses a thermal/optical approach for setting the EC/OC split. Furthermore, we lacked sufficient sensitivity during short free tropospheric sampling legs on the aircraft to measure the altitude profile of EC, a critically important absorber. This sensitivity needs to be improved through development work and flight trials.

[75] The relationship between absorption and the mass of EC is important both for radiative transfer models and for instruments that use absorption to infer EC mass. Using nearly identical absorption and EC measurement methods, the mass absorption efficiency found on the C-130 (4–12 m^2/g , Timothy Bertram, personal communication 2003) was substantially smaller than that found at Gosan (13 m^2/g [Chuang *et al.*, 2003]) and on the R/V *Ronald Brown* (9–12 m^2/g (P. Quinn, personal communication, 2003)). It is not clear how much of this difference can be attributed to sample loading issues, as opposed to the potential that the C-130 actually encountered elevated aerosols unlike those sampled at the surface. Chuang *et al.* found a significantly higher MAE in submicron than supermicron particles, which they attribute to internal mixtures of submicron EC with sulfates and other condensed material.

5.3. How Do Size and Light Scattering by Asian Aerosols Change With Relative Humidity?

[76] Aerosol hygroscopicity varied from strongly deliquescent with hysteresis (marine) to a smooth monotonic dependence on RH with little hysteresis (volcanic) [Carrico *et al.*, 2003]. Defined as the ratio of particle scattering extinction coefficients σ_{sp} (RH = ambient) to σ_{sp} (RH = 19%), the average $f(\text{RH} = \text{ambient})$ in the marine boundary layer (as measured aboard the R/V *Ronald Brown*) ranged from 1.25 (dust) to 2.88 (volcanic) [Carrico *et al.*, 2003]. In situ optical measurements from the C-130 showed the fine-mode aerosol to be only moderately hygroscopic ($f(\text{RH}) = 1.7 \pm 0.2$) and the coarse-mode aerosol to be nearly nonhygroscopic ($f(\text{RH}) = 1.1 \pm 0.1$ [Anderson *et al.*, 2003a]), using a somewhat different definition of $f(\text{RH})$ (85% versus lower than 40% RH). Standardization of methods is needed.

5.4. With What Accuracy Can in Situ Measurements Be Integrated Over the Depth of the Atmosphere to Predict the Observed Radiative Effects?

[77] Several ACE-Asia papers [Wang *et al.*, 2002; Murayama *et al.*, 2003; Redemann *et al.*, 2003; Schmid *et al.*, 2003] assess column closure by comparing extinction profiles derived by four techniques: (1) airborne Sun photometry, (2) airborne nephelometry and filter-based absorption photometry, (3) airborne particle size and composition measurements, and (4) surface-based lidar. Comparing aerosol extinction σ_{ep} (550 nm) from the four techniques yields good agreement for the vertical distribution of aerosol

layers. However, the level of agreement in extinction values varies by aerosol type and measurement technique.

[78] The σ_{ep} (550 nm) computed from Twin Otter in situ size distribution and composition measurements shows good agreement with airborne Sun photometry in the marine boundary layer but is considerably less in layers dominated by dust if the particles are assumed to be spherical [Wang *et al.*, 2002; Schmid *et al.*, 2003]. This difference appears not to be caused by the large size of dust (since no correlation was found between aerosol size and extinction difference), but instead by dust nonspherical shape (which causes underestimation both in aerodynamically derived particle size and in extinction-to-volume ratios). σ_{ep} (550 nm) values from in situ scattering and absorption measurements on 14 Twin Otter profiles were about $\sim 13\%$ less than values from airborne Sun photometry, but again no correlation between aerosol size and extinction difference was found [Schmid *et al.*, 2003]. The corresponding aerosol closure study using in situ scattering and absorption measurements on 28 C-130 profiles led to slightly better agreement with airborne Sun photometer-derived values. This agreement is very noteworthy, given that the higher speeds of the C-130 pose a more difficult challenge for measurements of large particles. This suggests that the C-130 LTI was successful in sampling the large particles responsible for 550-nm extinction, and that over-sampling by aerodynamic focusing (B. J. Huebert *et al.*, PELTI: Measuring the passing efficiency of an airborne low turbulence aerosol inlet, submitted to *Aerosol Science and Technology*, 2003) was largely compensated by line losses downstream. It also points to the need, in future aerosol experiments, for careful attention to design and construction of inlet and plumbing systems that take into account aircraft speed and expected aerosol size.

[79] The large suite of profiles from the C-130 and Twin Otter where we can compare in situ and remote measurements of light extinction are the most comprehensive data sets to date on column closure. This allowed us to test for discrepancies between the two with a high degree of confidence in the results. Also, it allowed us to look for dependence of discrepancies on such factors as ambient-RH and fine-mode fraction of the aerosol, yielding insight to the reasons for the discrepancies.

5.5. How Much Quantitative Information About Aerosols and Their Radiative Impacts Can Be Derived From Satellites and Other Remote Sensors?

[80] Two approaches in lidar observations were employed during the ACE-Asia IOP with the Asian Dust network (AD-Net) consisting of 12 lidars in China, Korea, and Japan (N. Sugimoto *et al.*, unpublished manuscript, 2003). One is continuous observation of vertical profiles with automated simple lidars and the other is accurate measurement of optical characteristics of atmospheric aerosol with high-performance lidars. The former approach combined with a chemical transport model is useful for understanding the outline of an aerosol event.

[81] The polarization lidars in ACE-Asia proved extremely valuable in distinguishing dust from pollution aerosols and in showing the tendency for dust layers to extend to greater heights than pollution layers, sometimes reaching 10 km [e.g., N. Sugimoto *et al.*, unpublished manuscript, 2003; Murayama *et al.*, 2003]. This ability to distinguish dust from

pollution, and from mixtures of the two, by using lidar polarization was for the first time demonstrated by comparison to airborne in situ measurements of aerosol composition [e.g., Murayama *et al.*, 2003]. The lidar measurements, which are more frequent than the aircraft flights, then proved very useful in testing model predictions of dust and pollution transport, and helped identify missing dust source regions in the models. Lidar and aircraft measurements commonly captured three types of layers in the atmosphere [Murayama *et al.*, 2003]: a boundary layer (surface to 1.2–1.5 km) where accumulation-mode particles with a low depolarization ratio dominated; an intermediate layer up to ~ 3.5 km where fine and coarse particles were moderately externally mixed, giving moderate depolarization ratio; and an upper layer above where mineral dust dominated, giving a high depolarization ratio.

[82] Quantitative comparisons between extinction profiles derived by lidar, airborne Sun photometry, and in situ measurements showed agreement in layer heights, but extinction values sometimes differed by more than expected uncertainties [e.g., N. Sugimoto *et al.*, unpublished manuscript, 2003; Murayama *et al.*, 2003; Schmid *et al.*, 2003]. Reasons for the disagreement include imperfect collocation combined with strong spatial variability, especially in the boundary layer near sources, plus use of standard or height-independent extinction-to-backscatter ratios in conditions where aerosol properties (including size, shape, and composition) varied strongly with height.

[83] Ground-based Sun/sky radiometry provided large data sets that have been inverted to yield optical depth spectra, column size distributions, complex refractive indexes, and single-scattering albedos [e.g., Sano *et al.*, 2003]. These large data sets are very valuable in documenting the hourly-to-monthly evolution in aerosol column optical depth and other properties that are very useful in deriving aerosol radiative forcing. However, comparisons of derived column size distributions and other properties with airborne in situ measurements in the column remain to be done. The inversion algorithm's assumption of size-independent complex refractive index is not consistent with the size-dependent composition typically observed in ACE-Asia [e.g., Wang *et al.*, 2002]. Attempting inversions that allow complex refractive indices to differ in large and small size modes may be a useful step forward.

[84] ACE-Asia's extensive measurements of aerosol optical depth spectra, single-scattering albedos, fine-mode fractions, and other properties provide a wealth of information for testing satellite retrievals of these and related properties (R. Kahn, personal communication, 2003). The bulk of this work will be reported in future issues.

5.6. What Is the Net Direct and Indirect Radiative Forcing by Asian Aerosols and How Do They Change With Emissions, Time, and Distance From the Continent?

[85] ACE Asia's measurements and analyses found the radiative budget in the east Asian region to be strongly perturbed by aerosol loading. While many studies consider only the cooling effects at short wavelengths, studies from the Ronald H. Brown demonstrate that dust also has important warming effects at thermal IR wavelengths. High-resolution spectra were used to observationally determine

the aerosol IR radiative forcing at the surface [Vogelmann *et al.*, 2003]. They found that the aerosol IR forcing is significant: often a few W m^{-2} and up to 10 W m^{-2} for large aerosol loadings. Calculations using an IR aerosol model [Markowicz *et al.*, 2003b] produced good agreement with these observations, as well as with pyrgeometer downwelling fluxes, and IR satellite (CERES) measurements. The IR aerosol radiative forcing can be 10–25% of the shortwave aerosol forcing at the surface, and up to 19% at TOA [Markowicz *et al.*, 2003b].

[86] In addition to aerosol effects on longwave and on photosynthetically active radiation (PAR - an extension of the visible), several papers report effects on total solar radiation. Bush and Valero [2003] found a 24 hour clear sky shortwave radiative forcing of -30.5 W m^{-2} at surface with a mean optical thickness of 0.41 at wavelength 500 nm, τ_{500} , from ground-based radiometer measurements at the Gosan site (33.28°N, 127.17°E) as averages from 25 March to 4 May 2001. The maximum forcing reached -52.1 W m^{-2} . Markowicz *et al.* [2003a] found a mean surface forcing of -26 W m^{-2} with $\tau_{500} = 0.43$ from the R/V *Ronald Brown* measurements over the Sea of Japan. Conant *et al.* [2003] found an area-averaged forcing of -22 W m^{-2} at the surface and -12 W m^{-2} at TOA for the region of 20°N–50°N, 100°E–150°E by using the CFORS mesoscale model with mean $\tau_{500} = 0.40$. Nakajima *et al.* [2003] summarized the range of April monthly mean estimates of 24 hour clear sky shortwave ARF from various methods as $-10.1 \pm 1.5 \text{ W m}^{-2}$ at TOA and $-24.5 \pm 4.8 \text{ W m}^{-2}$ at surface with $\tau_{500} = 0.38 \pm 0.06$ at Gosan and $-9.1 \pm 1.3 \text{ W m}^{-2}$ at TOA and $-22.8 \pm 6.1 \text{ W m}^{-2}$ at surface with $\tau_{500} = 0.36 \pm 0.03$ at Amami.

[87] Combining these forcings and optical depths yields values for the forcing efficiency factor, β ($\text{W m}^{-2}/\tau_{500}$). Results at the surface are -74 for the Gosan site, -60 for the Sea of Japan, and -55 for the east Asian area. These values are similar to or smaller than the reported β values of -75 in INDOEX [Ramanathan *et al.*, 2001a, 2001b] and -70 in TARFOX [Russell *et al.*, 1999]. On the other hand, Nakajima *et al.* [2003] obtain rather scattered values of the efficiency factor, ranging from -50 to -80 at Gosan and Amami-Oshima (28.15°N, 129.30°E), when using several different methods, including the SeaWiFS satellite, surface radiation measurements, and the SPRINTARS GCM-model [Takemura *et al.*, 2003]. There is still a substantial difference in the evaluation of the forcing depending on the methods. Optical thickness values simulated by SPRINTARS and CFORS are smaller than observed values from sky/Sun photometer and satellite-derived values, producing small model forcing values.

[88] The forcing efficiency factor depends strongly on the ω_0 value assumed in the model and satellite evaluations. Ramanathan *et al.* [2001b] proposed an ω_0 range from 0.85 to 0.90 in the INDOEX region. The CFORS model generated east Asian area-averaged values from 0.91 to 0.93 depending on internal/external mixing methods [Conant *et al.*, 2003], whereas SPRINTARS generated a slightly larger value of 0.95 as the average at Gosan and Amami-Oshima [Nakajima *et al.*, 2003]. On the other hand, the observed ω_0 values have large variability depending on time and location in the ACE-Asian experiment period. Twin Otter and R/V *Ronald H. Brown* measured ω_0 in heavily polluted condi-

tions of 0.80 for the dry aerosol and 0.86 for aerosol at 55% relative humidity [Markowicz *et al.*, 2003a; Mader *et al.*, 2002]. D.-H. Kim *et al.* [2003] evaluated the monthly mean ω_0 in April 2001 as 0.88 at Gosan and 0.84 at Amami-Oshima, by simultaneous analysis of diffuse/direct radiative fluxes and sky/Sun photometer data. Höller *et al.* [2003] measured ω_0 from ~ 0.7 to 0.99 at Yasaka, Japan, near the coast of the Sea of Japan. By using ω_0 values from the small end of the observed range it is possible to generate forcing efficiency factor values more negative than -100 at the surface. Combining these with observed optical depths yields strong surface forcing values of more than -50 W m^{-2} . These small values of ω_0 yield very small TOA forcing values less than -3 W m^{-2} . Nakajima *et al.* [2003] proposed the forcing efficiency factor varies largely from -50 to -80 at Amami-Oshima with 75% contribution from change in the single-scattering albedo and 25% from the asymmetry factor change.

[89] It is important to recognize that after interacting with pollution, dust can attain small values of ω_0 (i.e., $\omega_0 < 0.9$). Such values, observed in the downwind side of the continent, contrast strongly with ω_0 values of fresh dust particles, e.g., 0.95 to 0.98 observed in the Sea of Japan [Conant *et al.*, 2003] and suggested from satellite and sky/Sun photometer measurements [Kaufman *et al.*, 2001]. D.-H. Kim *et al.* [2003] showed that during the heavy dust event of 9–13 April at Gosan, ω_0 actually decreased to 0.84 from initial values that were near the Gosan April mean of 0.88. Bush and Valero's values in the Gosan dust event show a large forcing of -30 W m^{-2} and -50 W m^{-2} at the surface, with β values around -73 to -95 , suggesting that the Gosan column ω_0 must have been relatively small during that time.

[90] Indications that low ω_0 values are caused by the dominance of BC particles, both in dust storm events and in the normal condition at Amami Oshima, are supported by a CFORS simulation showing a tongue of carbonaceous aerosol plume extending from Taiwan to Amami Oshima [Conant *et al.*, 2003] and by aerosol type detection by the SeaWiFS satellite sensor [Higurashi and Nakajima, 2002].

[91] Nakajima *et al.* [2003] obtained an aerosol indirect forcing of -1 to -3 W m^{-2} by using a satellite method and the SPRINTARS model at the surface and TOA. They found a corresponding decrease in the effective radius of low-level clouds of order 2 microns. Their indirect forcing value is smaller than the -6 W m^{-2} derived by Ramanathan *et al.* [2001b] as the sum of indirect and semi-indirect forcings in the INDOEX region. This suggests that the indirect forcing value differs between cloud systems in the tropics and middle latitudes.

5.7. Are the Available Source Inventories for Asian Primary Aerosols and Gas-Phase Precursors Sufficiently Realistic to Support Assessments of Control Strategies?

[92] New estimates of emissions from east Asia were prepared specifically for the spring 2001 experiments [Streets *et al.*, 2003]. Estimated emissions for April 2002 are shown in Table 2, along with estimates of uncertainty. The overall uncertainties in emissions (measured as 95% confidence intervals) range from: $\pm 16\%$ (SO_2), $\pm 31\%$ (CO_2), to $\pm 360\%$ (BC), and $\pm 450\%$ (OC). Since the model calculations are driven by the emissions, a comparison of the

Table 2. Estimated Total Emissions for April 2001 in East Asia

| | Anthropogenic, Gg/yr | Biomass Burning Gg/yr | Total, Gg/yr | Uncertainty, % |
|--------------------------|-------------------------|--------------------------|-----------------|-------------------|
| SO ₂ | 2701 | 55 | 2756 | ±16 |
| NO _x | 1863 | 407 | 2270 | ±37 |
| CO | 17,369 | 10,053 | 27,422 | ±185 |
| NMVOC | 3309 | 1791 | 5100 | ±130 |
| CH ₄ | 8377 | 460 | 8838 | ±65 |
| NH ₃ | 2186 | 136 | 2323 | ±72 |
| CO ₂ | 709 | 167 | 877 | ±31 |
| BC | 166 | 67 | 233 | ±360 |
| OC | 578 | 498 | 1076 | ±450 |
| Wind-blown dust | | | 240,000–650,000 | ±500 |
| Anthro-PM ₁₀ | 2429 | 1264 | 3694 | ±500 |
| Anthro-PM _{2.5} | 1434 | 743 | 2178 | ±500 |

predictions with the observations provides a first-order estimate of the quality of the emissions. Comparison of observed ratios of species (e.g., OC/BC, BC/CO) to those estimated from the emissions inventory and those calculated by models provide additional insights into the quality of the emission estimates and can provide insights into source sectors (such as emissions from domestic heating using biofuels). From analysis of ACE-Asia data the following insights into emissions of aerosols and gaseous have emerged.

[93] From the analysis of surface EC and OC observations (R/V *Ronald Brown* and VMAP) we find that the calculated values are consistent (mean values within ±30%) with the observations, suggesting that the emissions are accurate to within the stated uncertainties. This is somewhat surprising in light of the likelihood that reported values are very dependent on the analysis method, and different techniques were used on the R/V *Ronald Brown* and in VMAP as discussed above. One important issue that has emerged from the analysis of the surface data is related to how well the surface observations constrain BC and OC emission estimates, given that the bulk of the mass flux of BC and OC occur at altitudes well above the surface layer [Uno *et al.*, 2003]. A completely different picture emerges when the same models are compared to aircraft filter data. Here the calculated values are significantly lower than the observations (in terms of mean values, by factors of 3–5; G. Carmichael, personal communication, 2003). The largest differences are found for low-altitude flights in the Yellow Sea, close to the Chinese sources! A similar tendency is found in certain gas phase species; for example, CO is also underpredicted at low altitudes in the Yellow Sea. Recent inverse model studies have shown a need to increase China's CO emissions [Kasibhatla *et al.*, 2002; Palmer *et al.*, 2003]. Yet observed ratios of BC/CO are reasonably well represented by the emissions inventory. This suggests that there may be a problem in the inventories of CO, BC and OC within a given sector. The domestic sector (and specifically domestic coal) is a likely candidate, and analysis suggests that this sector may be underestimated by at least a factor of 2 [Carmichael *et al.*, 2003]. The next step toward improving BC and OC emission estimates, will be to use the ACE-Asia data in formal inverse studies as has been done for CO.

[94] The four-dimensional measurements of dust associated with several large dust outbreaks, obtained during ACE-Asia, have provided valuable insights into our ability

to estimate dust emissions. Dust emissions are inherently difficult to quantify, as the emission rates are dependent on complicated interactions between the surface and the lowest layers of the atmosphere, and there are no well-established constraints on the total quantity emitted (unlike the case of coal where we have information on total coal consumed for a region). Having multiple models involved in the forecast and subsequent analysis of the dust data, each using different emission algorithms, has shed important insights into our capabilities to estimate dust emissions, and led to improvements. For example, we identified drought-stricken regions in Liaoning Province (northeast of Beijing) as a new region of dust emissions. Size resolved measurements of airborne dust over the extent of the size distribution provided for the first time information with which to test and improve the size dependency of the emission algorithms. The various algorithms performed well in terms of predicting the frequency and general location of the dust emissions associated with large events. However, estimates of the dust emitted vary greatly; for example, current estimates of dust emitted during ACE-Asia range from: ~250 Mt of soil dust of particles with diameter <40 μm between 1 March and 31 May 2001 [Zhao *et al.*, 2003], to 643 Mt (from 5 to 15 April) of dust (<36 μm) [Liu *et al.*, 2003]. Yet with these large differences in emissions used by the various models, each model produces dust distributions that are consistent with observed features.

[95] So there remains the critical question of whether current models are able to discriminate/constrain dust emission estimates. Can measurements help? The dry deposition quantity of dust at Zhenbentai was estimated by Zhang *et al.* [2003] and Gong *et al.* [2003]. If this number is assumed to hold for all of east Asia, a dry deposition flux for spring 2001 is ~150 Mt dust (upper limit). Models estimate that in this region about 50% of the dust emitted is deposited by dry deposition pathways. This implies an emission estimate of ~300 Mt, a value within the range predicted by the models.

[96] The ACE-Asia experiment has produced a comprehensive evaluation of existing emissions and led to improved estimates. The data have also been used to develop new inventories. For example a Hg emissions inventory for Asia has been developed on the basis of H. Friedli's measurements. The current inventories perform well for most gas phase species (e.g., nonmethane hydrocarbons, CO, NO_x, and SO₂) when input into atmospheric

models and compared with observations (well within the uncertainty limits). In terms of aerosol quantities, current models using these emissions are able to explain the large latitudinal gradients and time variations in observed dust, sulfate, BC and OC distributions. However, the analysis has also identified problems.

[97] So are the available source inventories for Asian primary aerosols and gas-phase precursors sufficiently realistic to support assessments of control strategies? The answer is a qualified yes. The models that can be used for assessments have been improved using ACE-Asia observations. That said, there remain large uncertainties in Asian primary aerosol emissions. Further improvements will require additional analyses of the ACE-Asia data as well as follow-on studies.

5.8. How Are Aerosol Properties and Dynamics Modified by the Uptake of Gases?

[98] It is very important to know whether alkaline dust shunts acidic gases away from other possible reaction pathways. Unfortunately, our data may not be able to provide a conclusive answer.

[99] There is no doubt that calcium (presumably from dust) and nitrate are on the same size particles and that a fraction of the sulfate that is not sea salt is at times associated with dust (B. Huebert, unpublished data, 2003; P. Quinn, personal communication, 2003). The first implies HNO_3 , NO_2 , NO_3 , or N_2O_5 uptake by alkaline dust, while the second could be due either to SO_2 uptake or to coagulation of dust particles with sulfate particles in clouds. The temporal and particle-size associations between calcium and nitrate are well established.

[100] However, how do we rule out the possibility that some of these are primary sulfate and nitrate particles? Calcium nitrate is a commonly applied fertilizer in China, so it may be present in soils and agriculturally derived dust. Gypsum is also a common soil mineral that could be the source of some fraction of the coarse-mode NSS. The morphology of primary versus secondary salts may be hard to distinguish with an electron microscope, since they may have been through multiple cloud cycles followed by drying in the vacuum of the microscope.

[101] What kind of evidence would it take to conclude that gas uptake really did happen? For starters, it would be desirable to measure both the precursor gases and the aerosol species on the same timescale. Neither NO_y nor HNO_3 was measured in ACE-Asia, so we cannot assess the precursor gas:aerosol ratio for nitrates.

[102] The most definitive way to study uptake is to look for a change in the precursor gas:aerosol ratio with time. For example, if much of the nitrate was primary, we wouldn't expect to see much of a change in the Ca/NO_3 ratio in most size ranges with aging or passage through urban areas. For nitric acid and sulfur dioxide the lifetimes are a day or so, so one would need to measure these ratios in air masses at a half, one, and two days downwind of major urban source areas. In the Yellow Sea and Sea of Japan we often found air masses with ppbs of SO_2 that still had not reacted, so the formation of sulfate would have been easily measurable. The TRACE-P data suggest that there was often enough NO_x and HNO_3 (200–1500 pptv) to make a measurable change in aerosol nitrate concentrations upon oxidation and

condensation. Evolution studies would be a good approach for future experiments.

5.9. Harmony

[103] There are substantial disagreements among authors about several issues, such as the numerical value of the ω_0 of dust and dust/pollution mixtures. The EC specific absorption measured on the *Ron Brown* is different from that measured using nearly identical methods on the C-130. As of this writing it is not clear whether these (and other) difference are real or are due to measurement artifacts. A harmony effort (S. Masonis, personal communication, 2003) is studying intercomparison periods in an attempt to resolve differences.

[104] In reality, however, side-by-side measurements often produce unexplainable differences that may be due to either measurement biases or the failure of the instruments to sample exactly the same air. An important conclusion from these exercises is that actual uncertainties are often larger than investigators expect. This may explain why different groups, applying different degrees of corrections to their data, could write papers containing disparate values for critical parameters.

[105] How accurately do we need to know aerosol radiative forcing of climate? Would one tenth of a watt per square meter uncertainty be adequate? Half a watt per square meter? The latter is likely to be too lax for many assessments. Yet the radiative forcing estimates cited above for the ACE-Asia region in April of 2001 vary by tens of watts per square meter. Is this good enough for the needs of groups like IPCC? Probably not: a few tenths of a watt per square meter can cause critical changes. If we assume that polluted, noncloudy regions cover 5% of the Earth, we would need to constrain the direct forcing in these regions to within $\pm 4 \text{ W m}^{-2}$ to achieve an uncertainty of $\pm 0.2 \text{ W m}^{-2}$ globally. While our results certainly advance the state of the science, even more accurate radiative forcing estimates are needed.

[106] Future experiments can do better. More instrument testing and intercalibration should precede any field program, which requires earlier funding decisions. Investigators need to have resources far enough in advance that they can go into the field knowing how comparable their measurement systems are, rather than finding out after the fact. Every group that wants their measurement to be taken seriously needs to participate in laboratory and field intercalibration exercises. Reviewers must insist that calibration and quality assurance metadata accompany each set of data. Ultimately, review articles that evaluate the degree of testing and correction employed by each group must be written, so that nonspecialists can understand which data sets to rely most heavily upon.

6. Conclusions

[107] It is evident from ACE-Asia observations that Asian aerosols can have a wide range of physical and optical properties, depending on the mixture of mineral dust (which is less absorbing than expected) with pollutants such as black carbon, sulfates, nitrates, and organics. Dust clearly perturbs both the radiative environment (including actinic fluxes) and the chemical one, serving as an alkaline surface

upon which acidic gases may absorb. Yet dust is not just dust: most of the dust we encountered was intimately mixed with pollution, creating a multicomponent aerosol of great complexity and variety that will not be fairly described by average optical properties or an assumption of sphericity.

[108] We were generally successful at achieving closure to within our measurement uncertainties between in situ measurements of aerosol optical properties and the column-integrated radiative impacts of aerosols, which suggests that our tools were adequate for the job in most cases. Through coordination of our in situ measurements with a variety of satellites, lidars, and Sun photometers we have also improved our ability to interpret remote sensing data in this complex environment. It is clear, however, that almost-fanatical attention to inlet- and plumbing-efficiencies is necessary to achieve this kind of result in the presence of supermicron aerosols like mineral dust, for which sampling artifacts are almost unavoidable.

[109] The ACE-Asia observations were integrated with chemical transport models from the start, so it is not surprising that these models have already been improved by comparison with our data. Important new dust source regions are now included and the (extremely difficult to predict) residential combustion sources of BC, OC, and CO are being improved. Yet our lack of either profile observations near source regions or observations of aerosol aging means that we cannot constrain important elements of these models, such as differentiating between primary and secondary sources of sulfate and nitrate.

[110] Measurements of radiative fluxes and optical depth have been used to infer the direct, clear-sky radiative forcing by east Asian aerosols, which is large and negative (on the order of -30 W m^{-2} in April). A wide variety of radiative transport models are also being used with our data to apportion this climatic impact among the many aerosol components and to make estimates of forcing in the presence of clouds. Similarly rigorous indirect effect experiments are needed.

[111] D. Fahey (personal communication, 2002) recently noted that anytime you observe something unexpected in the atmosphere, you have to determine whether the error is in the measurement or in the community's expectations (models). The implication of this is that our most interesting and valuable observations will be ones that disagree with models and are therefore subject to intense scrutiny and challenge. Only those observations that can be defended even when they produce seemingly unrealistic numbers will change our fundamental understanding of how the atmosphere works. Do we know each instrument's accuracy and precision well enough to correctly separate instrumental uncertainty from real geophysical variability in our data sets? Future experiments must spend even more of their effort on intercalibrations, laboratory testing with various aerosol types, and plumbing passing efficiency tests if they hope to add new knowledge about how aerosols affect the climate system.

[112] Redundancy is a powerful method for making data defensible, especially if some measurements are based on totally different physical principles. Agreement on unexpected values then has real power. Since new methods are constantly being introduced and since the accuracy of many measurements varies from platform to platform and group to

group, we have to make intercomparisons an integral part of experiments like ACE-Asia. It is so easy to make misleading aerosol measurements that future experiments on aerosol radiative forcing need to be as much a study of instrumentation testing and comparison as of geophysics. Otherwise they won't get the geophysics right.

[113] Perhaps the success of an endeavor like ACE-Asia should be judged by the amount of debate it produces. We only learn that our expectations were wrong when people can argue about whether the data are good enough to support the claim that we found something new. There needs to be far more instrument development and testing to resolve issues like the emissions of BC from Asian households and their radiative impact. As with any program, ACE-Asia has generated new questions that can only be answered using improved measurements and better strategies.

[114] The ACE-Asia science team has assembled a rich trove of aerosol data, especially for aerosol mixtures that include dust. Of course there are measurements we are more and less confident of. There are other and better ways of making the observations. These will be the stuff of future experiments.

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