1 Atmospheric Composition

Strategic Research Questions

3.1 What are the climate-relevant chemical, microphysical, and optical properties, and spatial and temporal distributions, of human-caused and naturally occurring aerosols?

3.2 What are the atmospheric sources and sinks of the greenhouse gases other than CO_2 and the implications for the Earth's energy balance?

3.3 What are the effects of regional pollution on the global atmosphere and the effects of global climate and chemical change on regional air quality and atmospheric chemical inputs to ecosystems?

3.4 What are the characteristics of the recovery of the stratospheric ozone layer in response to declining abundances of ozone-depleting gases and increasing abundances of greenhouse gases?

3.5 What are the couplings and feedback mechanisms among climate change, air pollution, and ozone layer depletion, and their relationship to the health of humans and ecosystems?

See Chapter 3 of the *Strategic Plan for the U.S. Climate Change Science Program* for detailed discussion of these research questions.

Human and ecosystem well-being are related to the composition of the Earth's atmosphere. Changes in global atmospheric conditions have caused changes in climate, air quality, and the Earth's protective layer of stratospheric ozone. CCSP seeks to create a research and observation framework for atmospheric composition that provides information on the interactions among changes in the atmosphere, climate, the environment, and human health (see Figure 1) needed for sound policy decisions.

Progress in research to date has informed decisions related to laws and international treaties that protect the national and global environment. In addition, research has revealed linkages between global change and ozone depletion, as well as local, regional, and global air quality degradation. It has become clear that these issues cannot be understood separately by scientists or policymakers. Better understanding of these connections will make possible the development and improvement of predictive models, and support more effective decisionmaking. CCSP-supported research in atmospheric composition for FY 2006 will continue to have a high-priority focus on reducing

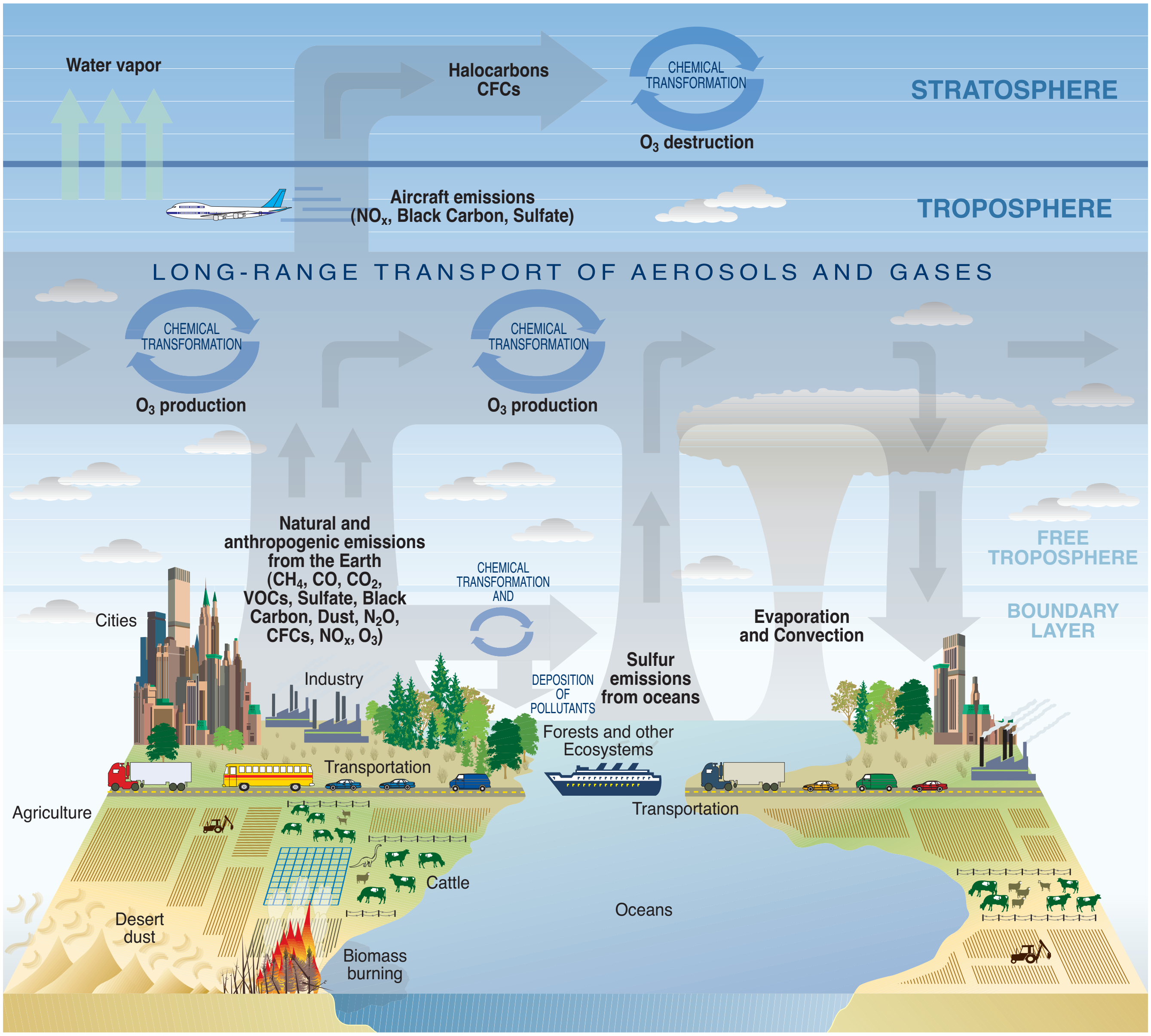
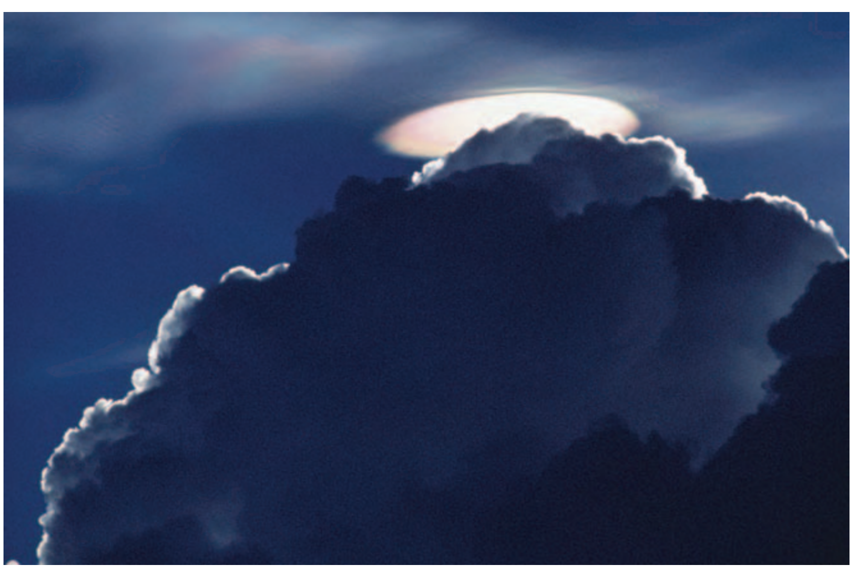


Figure 1: Chemical and Transport Processes Related to Atmospheric Composition. These processes link the atmosphere with other components of the Earth system, including the oceans, land, and terrestrial and marine plants and animals. *Credit: CCSP Strategic Plan* (illustrated by P. Rekacewicz).

scientific uncertainties associated with atmospheric aerosols (particulate matter). Aerosols are similar to greenhouse gases in that their atmospheric concentrations have increased greatly since the Industrial Revolution, and because they impose changes on the energy balance of the planet. Unlike greenhouse gases, however, different aerosols can have either a warming or a cooling influence on the climate. They also have environmental effects beyond altering climate, for example affecting regional air quality as well as human and ecosystem health in both source regions and in regions distant from the source. Observational advances have yielded important information on the geographical and vertical distribution of atmospheric aerosols and their diverse warming and cooling influences on Earth's radiation budget and their indirect effects on climate and the hydrological cycle. Nevertheless, significant further study is required to more fully quantify aerosol evolution, composition, vertical distribution, and global and regional impacts so that climate and air quality models can be more effectively evaluated and improved.

During the past 2 decades, scientists have made significant progress in understanding the causes and implications of variations in atmospheric composition. However, many questions remain. For example, halogen chemistry is known to be largely responsible for stratospheric ozone depletion, but the roles of chemistry versus dynamics remain to be fully quantified. The connection between climate change and ozone chemistry has been recognized, but uncertainties remain about the effects of climate change on the timing and extent of ozone recovery. In the troposphere, scientists have observed varying trends in ozone; however, its spatial evolution and trends remain to be quantified. Similarly, the spatial and temporal variations in oxidizing capacity – the atmosphere's ability to cleanse itself and prevent the buildup of harmful emissions – require further characterization in order to quantify the changing abundances of trace gases and aerosols. Global observations have shown the transport of tropospheric ozone over large (hemispheric) distances. However, the extent to which regional pollution can be attributed to such long-range transport remains to be quantified. In the climate area, radiatively important changes in atmospheric water vapor have been observed, but these temporal variations are not sufficiently quantified so that future changes can be projected.



Highlights of Recent Research

The following are selected highlights of recent research supported by CCSP participating agencies.

Aura Mission Successfully Launched and Operating.[7,9] The Aura satellite was launched on 15 July 2004, from Vandenberg Air Force Base, California (see <aura.gsfc.nasa.gov/test/>). Aura is already providing the first-ever daily global measurements of tropospheric ozone and many other trace gases that affect air quality. The spacecraft is delivering these results, in addition to observations of the stratosphere, with unprecedented spatial resolution. Aura's global view of Earth's atmospheric composition provides needed information for research, and for supporting environmental management and policy development related to issues of local, regional, and global air quality, climate change, and stratospheric ozone depletion and recovery. Aura carries four instruments: the Ozone Monitoring Instrument (OMI), the Microwave Limb Sounder (MLS), the High Resolution Dynamics Limb Sounder (HIRDLS), and the Tropospheric Emission Spectrometer (TES). OMI was built by The Netherlands and Finland in collaboration with NASA. HIRDLS was built by the United Kingdom and the United States.

Aura will help scientists globally monitor pollution production and transport from city-to-city, region-to-region, and continent-to continent, on a day-by-day basis, for the first time. Aura's view from space enables us to understand the long-range pathways of pollutants, and early research results using MLS measurements of carbon monoxide

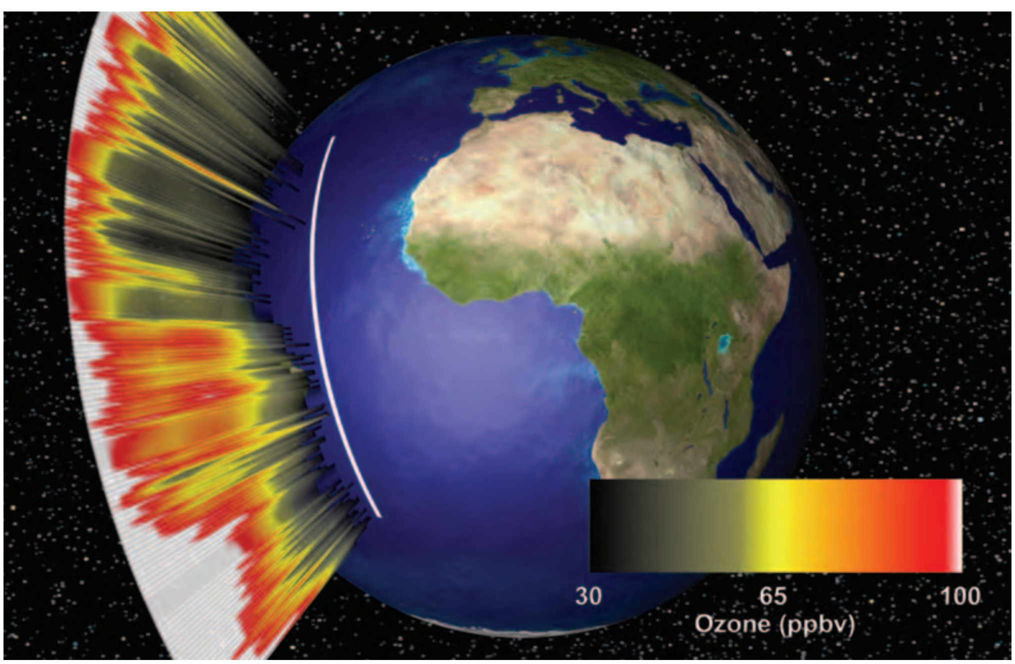
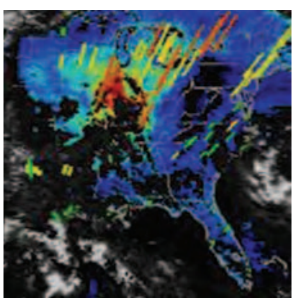


Figure 2: Aura TES Transect of Atmospheric Pollutants. The Tropospheric Emission Spectrometer (TES) on NASA's EOS Aura satellite is providing new observations of the vertical distribution of pollutants, including ozone, in the lowest part of the atmosphere. In this vertical slice taken in early September 2004 over the Atlantic, plumes of ozone that formed downwind of forest fires burning in both South America and Africa can be seen south of the equator from just above the surface to ~18 km. New measurements of pollutants and greenhouse gases such as ozone in the troposphere will allow scientists to estimate the impact of regional pollution events on global air quality and climate. *Credit: Jet Propulsion Laboratory.*

have begun to quantify the role of strong convective weather systems and long-range transport of pollution. Aura measurements also offer new insights into the processes that control the distribution of the trace gases important to climate change, and how climate changes influence the recovery of the protective stratospheric ozone layer. Analyses using measurements by the MLS instrument on Aura gave researchers the needed data to diagnose with unprecedented detail and spatial coverage polar ozone loss for the 2004 Antarctic ozone hole.

Aura instruments measure five of the six "criteria pollutants" identified by EPA. The complexity of pollution transport makes it difficult to quantify the extent to which human activities affect local air quality. In addition, the presence of the stratospheric ozone layer between the satellite and the troposphere makes "seeing" tropospheric ozone very difficult. Aura's TES uses new technology to see through the stratospheric ozone layer to measure tropospheric ozone (see Figure 2).

Aura enables new insights into the physical and chemical processes that influence the stratospheric ozone layer and climate. It is producing the most complete suite of chemical measurements ever available to understand the ozone layer and its recovery. These include the first measurements of chemically reactive hydrogen-containing species involved in ozone destruction, and the first simultaneous measurements of key forms of chlorine and bromine, which are also important for ozone destruction.



Smoke Inhibition of Cloud Formation.[6] Urban air pollution and smoke from fires can have important effects on the climate system. The net effect of aerosols on the atmospheric radiation budget and climate is a key uncertainty in attempts to model and project climate change. Aerosols can counteract regional greenhouse warming by reflecting solar radiation to space or by enhancing cloud reflectance and lifetime. However, some aerosols can add to the warming by absorbing sunlight, which also may have the effect of slowing down the hydrological cycle and reducing cloud cover.

While most greenhouse gases have long lifetimes and a homogeneous distribution in the global atmosphere, aerosols, due to their short lifetimes, have a heterogeneous spatial and temporal distribution. Thus, daily satellite observations and continuous *in situ* measurements are needed to observe the emission and transport of dense aerosol plumes downwind of populated and polluted regions and regions with vegetation fires.

Remote-sensing observations are providing the first measurements of the effect of aerosols, including sunlight-absorbing black carbon, or soot, on inhibition of cloud formation (see Figure 3). Observations by the Moderate-Resolution Imaging Spectroradiometer (MODIS) instrument aboard the Aqua satellite over the Amazon region during the biomass burning season in 2003 showed that scattered cumulus cloud cover was reduced from 40% in clean conditions down to 0% in heavy smoke

Figure 3: Reduction in Amazon Cloud Cover due to Smoke. Aerosols can have both a warming and cooling influence on global climate through a variety of diverse regional effects. This figure shows the reduction of cloudiness in a geographical region due to smoke aerosols from biomass burning. The reduction in cloud cover causes the region to reflect less sunlight, thereby allowing the surface to become warmer by the increased absorption of direct sunlight. The two satellite images of regions of the Amazon show marked difference in cloud cover due to the presence of smoke from biomass burning. The panel to the left without smoke aerosols has 40% cloud cover while the panel to the right with smoke is virtually cloud free. The partially cloud-covered region reflects an average of 36 Wm^{-2} of the incident sunlight and the cloud-free area reflects a smaller 28 Wm^{-2}. The fraction of the sunlight that is not reflected is absorbed by the atmosphere and surface. These satellite images were acquired by the MODIS instrument aboard the Aqua satellite on 3 August 2003.
Credit: R. Simmon, J. Allen, and Y. Kaufman, NASA/Goddard Space Flight Center.

conditions. The reduction of clouds due to smoke aerosols leads to less sunlight being reflected and more sunlight being absorbed by the Earth, resulting in warming. This effect may be offsetting some of the cooling attributed to sulfate aerosols.

Multi-Platform Studies of Aerosol Properties and Radiative Effects.[5]
On five occasions spanning the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) field campaign in spring 2001, the Multiangle Imaging Spectroradiometer (MISR) instrument on the NASA Terra satellite took data coincident with high-quality observations by instruments on two or more surface and airborne platforms. The cases capture a range of clean, polluted, and dusty aerosol conditions. A wealth of data was collected through the joint support of NSF, NASA, NOAA, and the Office of Naval Research (ONR). Scientists synthesized the data from over 40 field instruments and satellite observations into layer-by-layer environmental snapshots

that summarize what is known about the atmospheric and surface states at key locations during each event. Aerosols within a few kilometers of the surface were composed primarily of Asian dust with mixtures of pollution added from Asian and non-Asian sources. Medium- and coarse-mode particle size distributions varied little among the events; however, the column aerosol optical depth varied considerably depending on the near-surface amounts of absorbing aerosols. The consistency of component particle microphysical properties among the five events, even in this relatively complex aerosol environment, suggests that global, satellite-derived maps of aerosol optical depth and aerosol mixture (air-mass type) extent, combined with targeted *in situ* component microphysical property measurements, can provide a detailed global picture of aerosol properties and distributions, enabling studies of aerosol impacts on climate.

Intercontinental Tracking of Pollution and Aerosols. An expanse from the western United States to the European continent was the setting in summer 2004 for more than 200 scientists who participated in the largest climate and air quality study to date, the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT; see <www.al.noaa.gov/ICARTT>). The research was aimed at developing a better understanding of the factors that are involved in the intercontinental transport of pollution and the radiation balance in North America and the North Atlantic. Several U.S. agencies (with scientists in NOAA and NASA as co-leads), academic institutions, and international partners from five other countries used satellite, aircraft, shipboard, and land-based observations to obtain unprecedented information about the composition and transformation of air masses as they crossed the United States, traversed the Atlantic Ocean, and arrived in western Europe (see

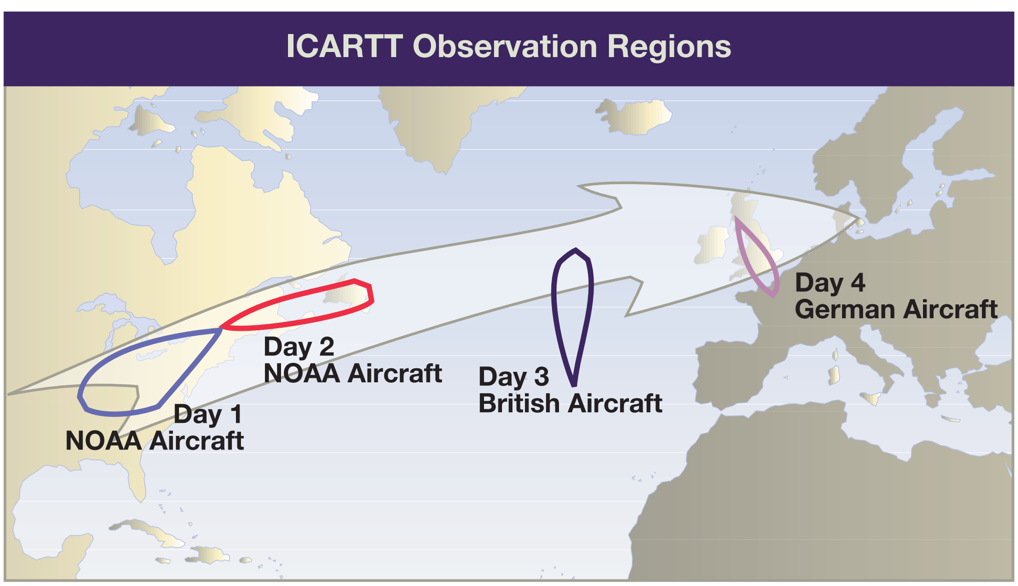
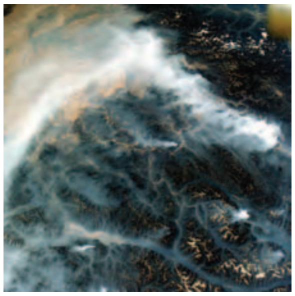


Figure 4: ICARTT Observation Regions. Observation regions used to study intercontinental transport and transformation of gases and aerosol particles during the Summer 2004 ICARTT experiment. *Credit: O. Cooper, NOAA/Aeronomy Laboratory.*

Figure 4). Natural and anthropogenic emissions react in the atmosphere to produce gases and aerosol particles that affect climate. Tracking the sources, atmospheric transformations, and intercontinental transport of these chemical species by ICARTT is advancing U.S. and international climate research.

Nighttime Chemistry More of a Factor than Previously Recognized.[2] A new technique has enabled measurement of previously hard-to-measure trace gases in the atmosphere. The observations have opened a new frontier area in studying the atmospheric chemistry that occurs at night. The approach uses an advanced spectroscopy technique to measure trace gases in the reactive nitrogen family, some of which occur primarily at night. The gases play important roles in the chemistry that produces ozone, a greenhouse gas and pollutant that has both climate and air quality implications. In atmospheric measurements off the coast of New England, researchers found that the nighttime chemistry involving nitrogen-containing trace gases can effectively remove these gases from the atmosphere, thus "short circuit" the chemistry that would have produced ozone the next day. Related work in 2004 was aimed at developing an aircraft-ready version of the instrument and applying it in the extensive ICARTT summer climate research field campaign.

Identifying which Atmospheric Aerosol Particles are Effective "Seeds" for Cloud Formation.[3] Among the least-understood but potentially important processes in atmospheric composition science is the relationship between small aerosol particles and ice-cloud formation. The process has important implications for the radiation balance of the atmosphere. Researchers have developed a novel technique to determine the chemical composition of those aerosols capable of forming atmospheric ice clouds, commonly termed "ice nuclei," both on a particle-by-particle basis and in real time. This technique was used in 2003 and 2004 field experiments to address the need for additional and more quantitative information on the chemical composition of atmospheric ice nuclei. Among the important results was the finding that the most efficient ice nuclei are not ubiquitous sulfate aerosols, but are instead rare mineral or fly-ash particles, some of anthropogenic origin. Aerosols rich in organic material were shown to be inefficient ice nuclei. These results have contributed to our understanding of the interaction of aerosol particles with clouds and will provide valuable information for global climate models.



Studies of Transcontinental Aerosol Plumes.[1,8] Transport and transformation processes in aerosol plumes were studied in field experiments off the east coasts of Asia (2001) and North America (2002), with the aim of evaluating and refining models of chemical transport and radiative transfer. This research has shown that dust transported out of Asia over the Pacific Ocean includes not only dust, but also anthropogenic gases and particles adsorbed/reacted onto the dust. The transport of dust-pollution plumes out of Asia and across the Pacific has important implications for air quality. In addition, recent satellite data suggest that these pollution plumes are having a regional effect on the Earth's reflectivity. Results from studies in eastern North America show that plumes originating in the United States can be as intense as those downwind of India and northern Asia. The direct radiative effect of the aerosols in these plumes is thus an important factor in climate. Forthcoming data analyses will allow researchers to quantify the radiative forcing due to aerosols and to apportion this forcing based on aerosol composition and, by inference, aerosol source. The information will provide the scientific basis for informing the development of effective strategies to reduce emissions and mitigate climate impacts.

Determining Aerosol Properties from Ground-Based Measurements. Since 1979, satellite retrievals have provided excellent spatial coverage of atmospheric column ozone, certain characteristics of aerosols, and ultraviolet (UV) radiation. Satellite data from Total Ozone Monitoring Spectrometer (TOMS) instruments (<toms.gsfc.nasa.gov>) have been essential for deriving global trends in UV radiation levels and resolving critical questions about the impacts of increased UV radiation due to stratospheric ozone depletion and changes in aerosols and clouds. Continuation and improvement of the TOMS UV data record is a goal of the new Ozone Monitoring Instrument launched on the Earth Observing System (EOS) Aura satellite in 2004.

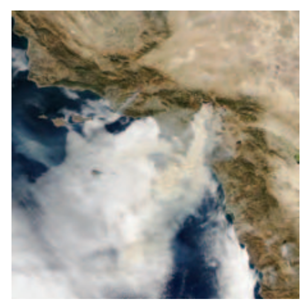
Currently, ground-based retrievals provide accurate measures of ozone needed for the validation of satellite data, as well as finer temporal resolution of these quantities necessary to determine diurnal variations and understand the observed long-term trends. An excellent example of ground-based support has been provided by the nine years of radiometer data from the CCSP-sponsored UV-B Monitoring and Research Program's (UVMRP) observational network, which has been used to assess the geographic distribution, trends, and year-to-year variability of UV-B radiation in the United States (<uvb.nrel.colostate.edu/UVB/home_page.html>).

Since 2002, CCSP's TOMS, UVMRP, and Aerosol Robotic Network (AERONET) programs have shared equipment, personnel, and analysis tools among member agencies such as USDA, NASA, and NOAA to quantify aerosol absorption using ground-based radiation measurements. Recently, UVMRP data have been used to extend measurements of aerosol properties into UV wavelengths when combined with data obtained from AERONET sun photometers (<aeronet.gsfc.nasa.gov/>).

The new analysis techniques applied to the ground-based radiation measurements provide aerosol scattering and absorption characteristics essential for determining the amount of UV radiation reaching the Earth's surface and for understanding the potential implications for climate change. Determining the spatial and temporal distribution of aerosol properties is critical for projecting climate change, understanding tropospheric chemistry, and making accurate satellite measurements of UV radiation, ozone, and aerosols.

Emissions of Ozone-Depleting Substances in Russia.[4] The emissions of six ozone-depleting substances (ODS) were estimated from measurements taken by scientists in a 17,000-km journey along the Russian trans-Siberian railway. The measurements are of global interest because Russian sources of ODS are thought to be a significant fraction of the total ODS production worldwide, and because estimates of global emissions are not in good agreement with observed atmospheric abundances over the last ~10 years. The research, based on a 2001 study, has thus far indicated that the modern emissions of ODS in Russia are too small to cause the large, contemporary shortfalls in global emission estimates. A follow-on study in spring 2004 was undertaken to examine the expected reduction of ODS concentrations since the earlier 2001 study. Preliminary results show that the abundances are indeed smaller, suggesting that the international ozone layer protection agreements of the Montreal Protocol are having their intended effect.

Highlights of Plans for FY 2006



CCSP will continue to gather and analyze information through measurement, modeling, and assessment studies to enhance understanding of atmospheric composition and of the processes affecting atmospheric chemistry. Key research plans for FY 2006 follow.

Improve Estimates of Aerosol Direct Radiative Forcing via Satellite Data Analysis. Much progress has been made in using global observations from space to assess aerosol radiative forcing, leading to the efforts planned for FY 2006. One major success is the similarity of several independent estimates of top-of-atmosphere aerosol direct radiative forcing over the ocean using satellite measurements of reflected sunlight. This success is traced to improved remote-sensing instrument calibration, spatial resolution, aerosol optical thickness evaluations, and improved algorithms for data retrieval in partially cloudy scenes.

Bottom-of-atmosphere aerosol radiative forcing is more uncertain, in part due to poorly constrained measurements of aerosol absorption and vertical distribution. Satellite observations are not able to differentiate aerosols due to anthropogenic sources from natural aerosols. However, satellite-retrieved measurements of aerosol optical thickness, using several methods, are generally greater than model predictions, suggesting that these measurements have information to contribute to model improvement.

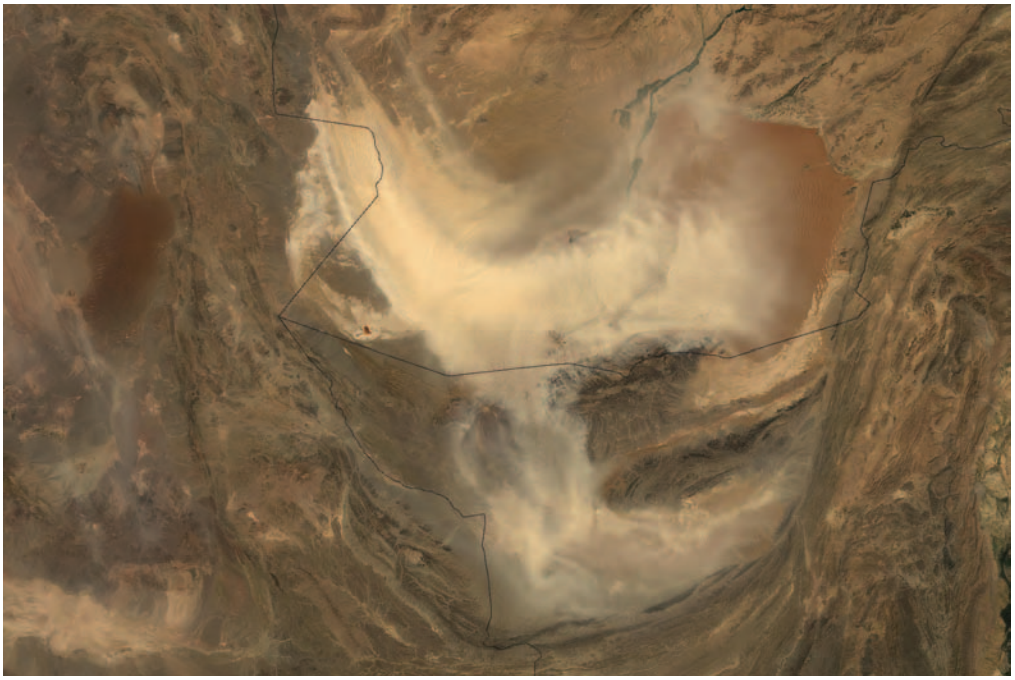
The lidar instrument aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite, scheduled for launch in fall 2005, promises to revolutionize knowledge of aerosol vertical distribution. Efforts are being made to develop methods of integrating global satellite observations with *in situ* and surface data to fill in major gaps in observations of aerosol particle properties such as single-scattering albedo, and to help separate anthropogenic from natural components. Also, further advances in the near term are expected from multi-angle, UV, and polarization-based techniques for deriving aerosol optical thickness over land.

These activities will address Question 3.1 of the CCSP Strategic Plan.

Intercontinental Chemical Transport Experiment – North America (INTEX-NA). The INTEX-NA is an integrated series of atmospheric measurements in support of achieving the Aura science goals and validation needs through field activities to be performed over and around North America. It seeks to understand the transport and transformation of trace gases and aerosols on transcontinental/intercontinental scales and their impact on air quality and climate. A particular focus in this study is to quantify and characterize the inflow and outflow of pollution over North America. The main constituents of interest are ozone and precursors, aerosols and precursors, and the long-lived greenhouse gases. INTEX-NA goals are greatly facilitated and enhanced by a number of concurrent and coordinated national and international field campaigns and satellite observations. Especially important to the success of INTEX-NA are carefully conducted coincident validation and science activities associated with ongoing satellite measurement programs, such as Terra, Aura, and Envisat. Synthesis of the ensemble of observations from surface, airborne, and space platforms, with the help of a hierarchy of models, is a necessary and important step toward achieving science objectives (see <cloud1.arc.nasa.gov/intex-na/>).

These activities will address Questions 3.1 and 3.2 of the CCSP Strategic Plan.

Development of an Instrument Package for an Airborne Aerosol Observatory. To date, observations of *in situ* aerosol properties have been primarily limited to intensive field campaigns, which provide a "snapshot" in time and space. A new instrumentation package for a light airplane will be completed and fielded; this deployment will permit more extensive *in situ* observations of the vertical and horizontal distributions of aerosol chemical, microphysical, and radiative properties over clean and polluted regions of the United States. The new approach will yield aerosol properties at differing locations and times, thus leading to a better characterization of aerosol

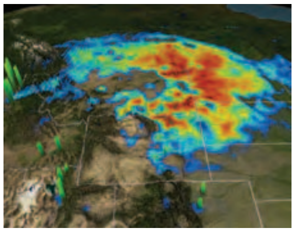


radiative properties above the surface, where few systematic measurements have been taken. They will also help validate satellite observations.

These activities will address Question 3.1 of the CCSP Strategic Plan.

Field and Laboratory Studies of Factors that Influence the Radiative Effect of Aerosols. The optical properties of aerosols are crucial parameters for calculating their radiative forcing. Laboratory studies will quantify how relative humidity affects the optical properties of aerosols of different composition. In conjunction with the laboratory studies, field measurements will examine water uptake by real atmospheric aerosols. The combination of these two measurements will better quantify how aerosols grow and how their radiative properties change under differing atmospheric relative humidity conditions. Incorporation of these real-world data in radiative transfer models, which calculate the radiative forcing of the climate system by aerosols, will reduce the uncertainty in the radiative forcing by aerosols.

These activities will address Question 3.1 of the CCSP Strategic Plan.



Potential Consequences of Climate Variability and Change for U.S. Air Quality. Researchers are conducting scenario-based analyses of the potential consequences of global change on regional U.S. air quality, focusing on fine particles and ozone. The climate, emissions, and underlying socioeconomic-based scenarios are intended to provide plausible projections of future conditions, rather than predictions of what actually will happen. Research activities focus on developing regional-scale inputs for air quality simulations using the Community Multiscale Air Quality modeling system. The potential impacts of increases in transported pollutants due to increases in global industrialization, population, and economic activity are also being analyzed. Research from FY 2004 onward is building toward a 2007 completion of an analysis of potential changes in U.S. air quality due to climate variability and change, including direct meteorological impacts on atmospheric chemistry and transport and the effect of temperature changes on emissions of air pollutants. Further research will result in a 2010 completion of an analysis that incorporates projected emission impacts from technology, land-use, and demographic changes to construct plausible scenarios of U.S. air quality for up to 50 years into the future.

These activities will address Question 3.3 of the CCSP Strategic Plan.

Mexico City Campaign in Spring 2006. Megacities are significant sources of aerosols influencing regional and global scale climate parameters. The Mexico City Metropolitan Area (MCMA), with 18 million people, is the world's second largest megacity after Tokyo. The recent MCMA-2003 field measurement campaign, which was part of the Integrated Program on Urban, Regional, and Global Air Pollution (see <www-eaps.mit.edu/megacities>), was designed to improve knowledge of the chemistry, dispersion, and transport processes of pollutants emitted to the MCMA atmosphere. In addition to being an example of a megacity that exports aerosols to the global environment, the Mexico City area provides an opportunity to study aspects of aerosol life cycles in a unique environment characterized by very high concentrations of soot, and secondary aerosols and precursors, that can be transported from the megacity into the surrounding region, thereby affecting regional radiative balance. DOE plans to conduct a 4-week field campaign during March 2006, using a G-1 aircraft and two instrumented surface sites. The planned study will examine and characterize changes in aerosol composition, size distribution, light scattering coefficient, absorption coefficient, optical depth, soot specific absorption, and radiative fluxes at the surface in the urban plume. The timing of the field experiment will allow close collaboration between the DOE Megacity Aerosol Experiment in Mexico City (MAX-Mex; see <www.asp.bnl.gov/MAX-Mex.html>), the NSF Megacity Impact on Regional and Global Environments (MIRAGE; see <www.ucar.edu/communications/quarterly/spring99/MIRAGE.html>) experiment, and the second phase of INTEX-NA (see <cloud1.arc.nasa.gov/intex-na>). The collaboration between the Mexican-, DOE-, NSF-, and NASA-supported scientists is part of a larger effort to understand the air pollution impacts of megacities, and is referred to as the Megacity Initiative: Local and Global Research Observations.

These activities will address Question 3.3 of the CCSP Strategic Plan.

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