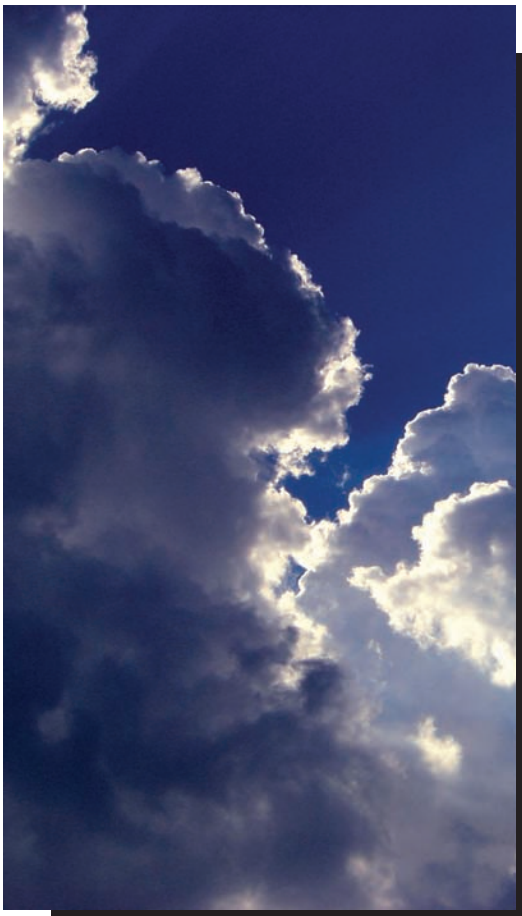


1 Atmospheric Composition

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?

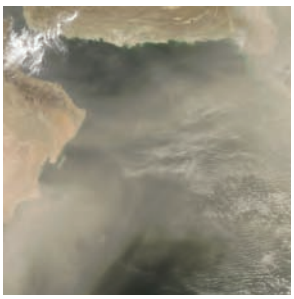
3.5 What are the couplings and feedback mechanisms among climate change, air pollution, and ozone layer depletion, and their relationship to the health of humans and ecosystems?

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

The Earth's atmosphere is composed of gases and particles that vary with spatial scale and with time, influencing climate, air quality, the stratospheric ozone layer, and weather. Interactions between these components have impacts on human health and the vitality of ecosystems and hence have high relevance to society. CCSP research on atmospheric composition focuses primarily on how human activities and natural processes affect atmospheric composition, and how these changes in turn relate to societal issues. The issues embrace multiple disciplines, cross many spatial scales, and are highly interrelated. Consequently, CCSP research is a highly coordinated endeavor that involves observational studies, laboratory investigations, and modeling analyses to provide the timely, accurate, and useful scientific information needed by decisionmakers nationally and internationally.

Progress has been made to date in understanding the role of atmospheric composition in Earth's climate. Efforts have been dedicated to the areas of largest uncertainty in understanding how atmospheric constituents other than carbon dioxide (CO_2) affect the forcing of climate. Atmospheric fine particles ("aerosols") can have either warming or cooling effects, depending on many factors. CCSP research has made progress in defining those factors, and has recently taken steps to address the next levels of complexity in the issue by looking at the interactions of aerosols with clouds. For FY 2009, CCSP's atmospheric composition research will focus on aerosols and aerosol/cloud interactions in the polar environment through analyses of measurements from satellites, aircraft, and the surface made during the International Polar Year (spring 2007 to summer 2008). Additional FY 2009 work focuses on linked air quality-climate modeling systems, future emission projections, and communicating research results to air quality decisionmakers.

Highlights of Recent Research



The following paragraphs provide selected highlights of recent research supported by CCSP-participating agencies.

Climate-Relevant Properties of Aerosols

Aerosol Forcing Effects on Climate Change are Better Defined.[1,2] Aerosols (atmospheric fine particles such as pollution, smoke, and desert dust) in sunlight directly heat the atmosphere if they absorb light, and cool the surface by absorbing and scattering light. Evaluating the net radiative effect of aerosols has been a key uncertainty in past Intergovernmental Panel on Climate Change (IPCC) assessments, partly due to the highly variable horizontal and vertical distribution of aerosol particles of differing chemical composition, size, and shape. This in turn hampers modeling efforts to understand the total amount and vertical distribution of solar radiation indicated by satellite observations. A recent field campaign studied the radiative forcing of atmospheric brown clouds—large pollution plumes that increasingly cover large regions of dry season southern Asia (see Figure 3). Critical measurements of solar heating profiles above, within, and below these pollution-dominated plumes over the Indian Ocean in the Northern Maldives were made with small unmanned aerial vehicles (UAVs) instrumented with aerosol and radiation detectors and positioned to make vertically aligned measurements. Findings indicated that continental air masses with higher aerosol particle concentrations, and in particular those containing the carbonaceous component of soot, exhibited increased aerosol absorption and heating by as much as

Ganges Valley Brown Cloud

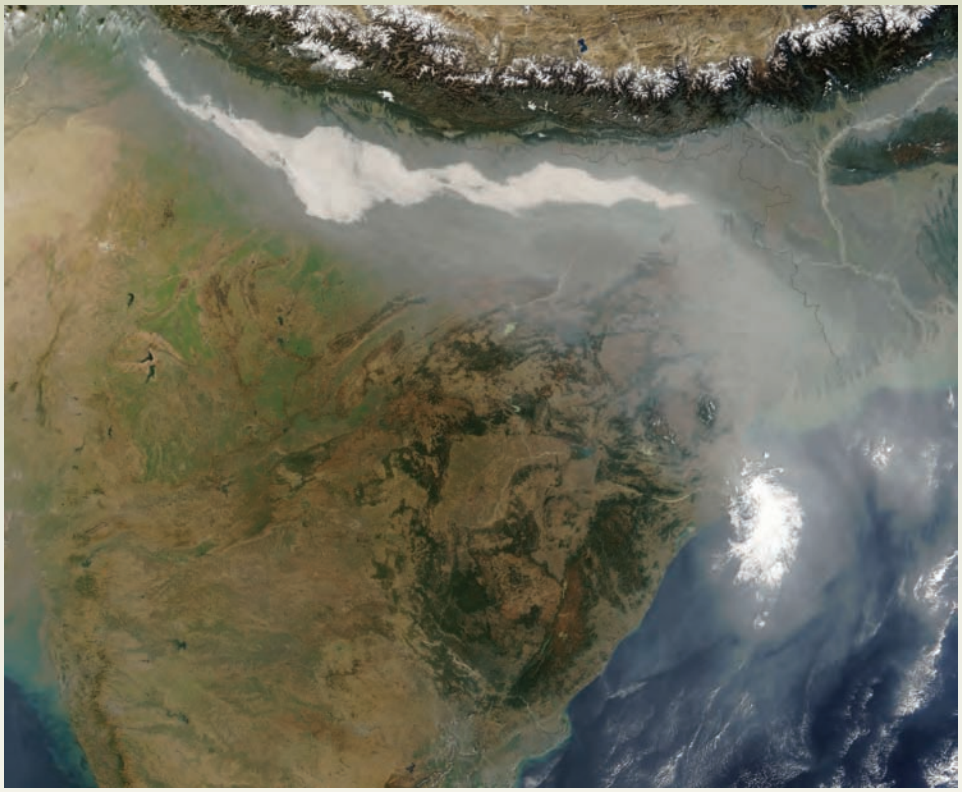


Figure 3: Ganges Valley Brown Cloud. Ganges Valley brown cloud plume drifts out over the Bay of Bengal and the Indian Ocean. *Credit: NASA / Goddard Space Flight Center*.

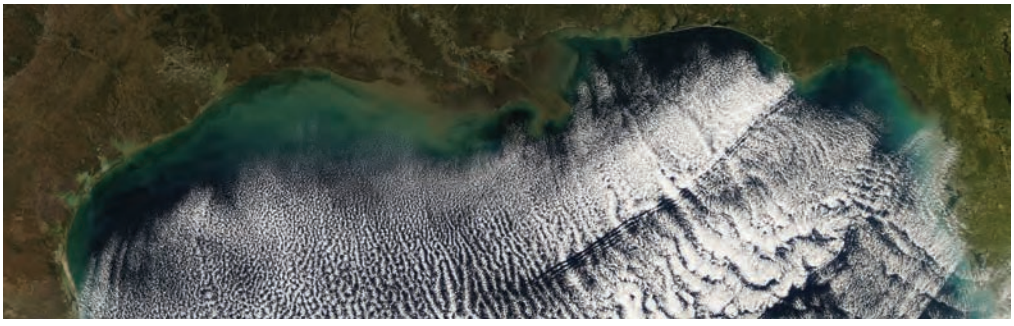
50% over background oceanic conditions. Other improvements in understanding have been made using the Aerosol Robotic NETwork (AERONET), a network of about 230 automated surface instruments that measure the optical properties of aerosols. Analysis of AERONET data collected in 2004 in the United Arab Emirates is leading to a better understanding of the dynamics of desert dust and pollution aerosols over a variety of environments in the Arabian Peninsula and over the Persian Gulf.

Current Trends in Arctic Haze and Implications for Climate Forcing.[3] The long-range transport of anthropogenic pollution from North America, Europe, and western Asia creates the aerosols associated with so-called Arctic haze. U.S., Finnish, and Canadian researchers have recently compiled long-term data to determine trends in and climate impacts of Arctic haze. The analysis confirmed previously reported results of a decreasing trend in Arctic haze between the early 1980s and the mid-1990s. In addition, the analysis revealed evidence of increasing levels of aerosol scattering, black carbon, and nitrate over the past decade. Calculations of direct radiative forcing by Arctic haze for a representative case during the haze maximum (mid-April) resulted in an estimated

2 to 3 Wm^{-2} of additional heating to the atmosphere and approximately 1 Wm^{-2} of cooling at the surface. CCSP researchers, as part of the International Polar Year, returned to the Arctic in 2008 using aircraft and ship platforms to better characterize the direct and indirect climate impacts of the Arctic haze.

Cloud-Aerosol-Climate Feedbacks and Interactions

Improved Understanding of Connections Between Aerosol Chemistry, Clouds, and Climate.[4,5,6,7]
The increasing levels of aerosol from human activities affect cloud properties, cloud lifetime, and precipitation processes, and hence climate. However, the relationships are not well understood and the aerosol/cloud processes are one of the largest uncertainties in current understanding of climate change. Aerosols affect cloud properties by serving as cloud condensation nuclei (CCN) and thereby activating the formation of cloud droplets. The process of cloud drop activation is a function of both the size and composition of the aerosol particles, which, in turn, depend on the source of the aerosol and transformations that occur downwind. CCSP scientists conducted airborne and shipboard measurements of the aerosol number size distribution, aerosol chemical composition, and CCN concentration during the 2006 Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS). Aircraft measurements showed that the high organic emissions of the Houston Ship Channel region lead to a high organic acid content in the aerosol, and further revealed details of how the organic acid content evolves with increasing altitude within and above clouds to produce a ubiquitous layer of organic aerosol above cloud. Analysis of the GoMACCS shipboard data showed that when the organic content of the aerosol increases, the aerosol is less likely to form CCN and hence less likely to activate the formation of cloud droplets. These studies have led to an improved understanding of the links between aerosol composition and cloud drop formation. The work has yielded a simplified means of representing the processes in climate models, ultimately contributing to the development of an improved predictive capability. In the continental United States, 8 years of surface measurements at the Atmospheric Radiation Monitoring (ARM) Southern Great Plains



Satellite Images of Closed- and Open-Celled Clouds

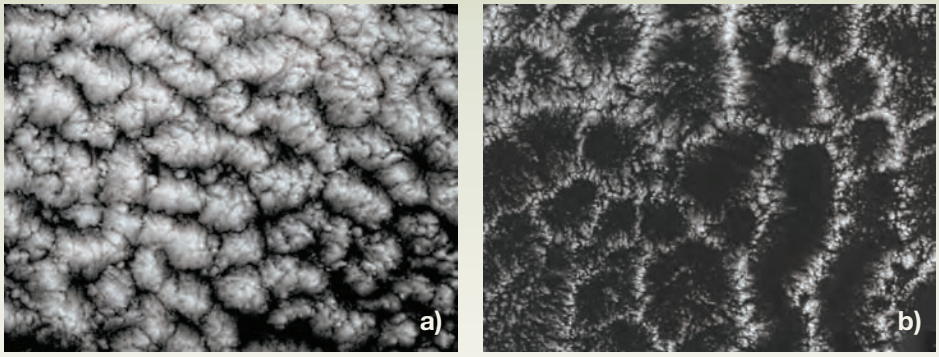
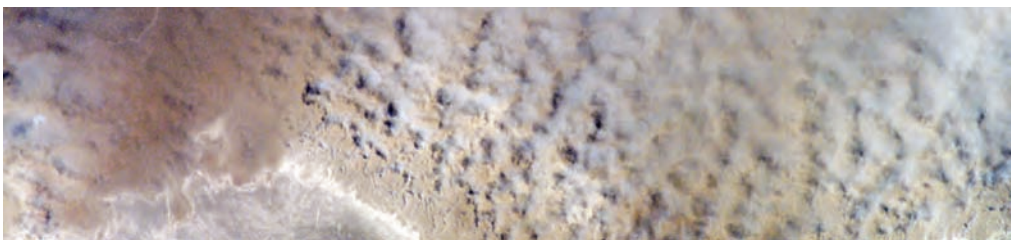


Figure 4: Satellite Images of Closed- and Open-Celled Clouds.[9] The width of the closed-cell (a) and open-cell (b) clouds images is approximately 200 km and 280 km, respectively. In closed cells, moist air rises in the center to form the cloud, then air descends at the edges to form the clearing. In open cells, the opposite happens. *Credit: M.J. Garay, UCLA (reproduced from the* **Bulletin of the American Meteorological Society** *with permission from the American Meteorological Society)*.

site in Oklahoma have advanced understanding of how clouds, water vapor, and climate interact. The data have clearly shown that higher cloud fractions are the key factor in reducing the amount of solar radiation at the Earth's surface, whereas the amount of precipitable water vapor in the clouds has a much greater impact on the cloud's absorption of outgoing infrared radiation.

Atmospheric Aerosol Pollution Affects Structure and Reflectivity of Clouds.[8] Viewed from space, stratocumulus clouds can have either an "open cell" or a "closed cell" appearance to their fine-scale structure (see Figure 4). The structure greatly affects the degree to which the clouds reflect light and, hence, the climate-relevant properties of the clouds. Recently it was hypothesized that precipitation may trigger the transition from closed to open cellular structure. Precipitation tends to occur in clean regions lacking in aerosol, thereby providing a potential link between the composition of the atmosphere and the organization of clouds. Polluted, non-precipitating clouds were shown to exhibit a closed cellular structure, whereas in clean conditions, open cells formed in response to drizzle. CCSP researchers have modeled the processes and confirmed the hypothesis. The transition from closed to open cells has dramatic implications for radiative forcing, essentially representing the transition from a highly reflective cloud to one of much lower reflectance.



Atmospheric Constituents other than CO_2, including Water Vapor, and Implications for Earth's Energy Balance

Lightning and Pollution Combine to Cause Ozone Enhancements in the Summer Upper Troposphere.[10,11,12,13,14] In the upper troposphere, ozone acts as a greenhouse gas and hence is relevant to climate. Analyzing data from a summer 2004 study, CCSP researchers found unexpectedly high levels of ozone in the upper troposphere 10 to 11 km above eastern North America during summer that were not attributable to either the high amounts of ozone pollution at Earth's surface or the higher ozone levels in the stratosphere. The researchers investigated the cause of the increased ozone, which can nearly double the amount of upper-tropospheric ozone above the region. It was found that a natural factor—the emission of nitrogen oxides from lightning—acts in concert with the generally higher background levels of ozone precursor compounds in today's polluted atmosphere to produce much of the upper-tropospheric ozone enhancement (see Figure 5). This ozone enhancement with a strong natural component contributes

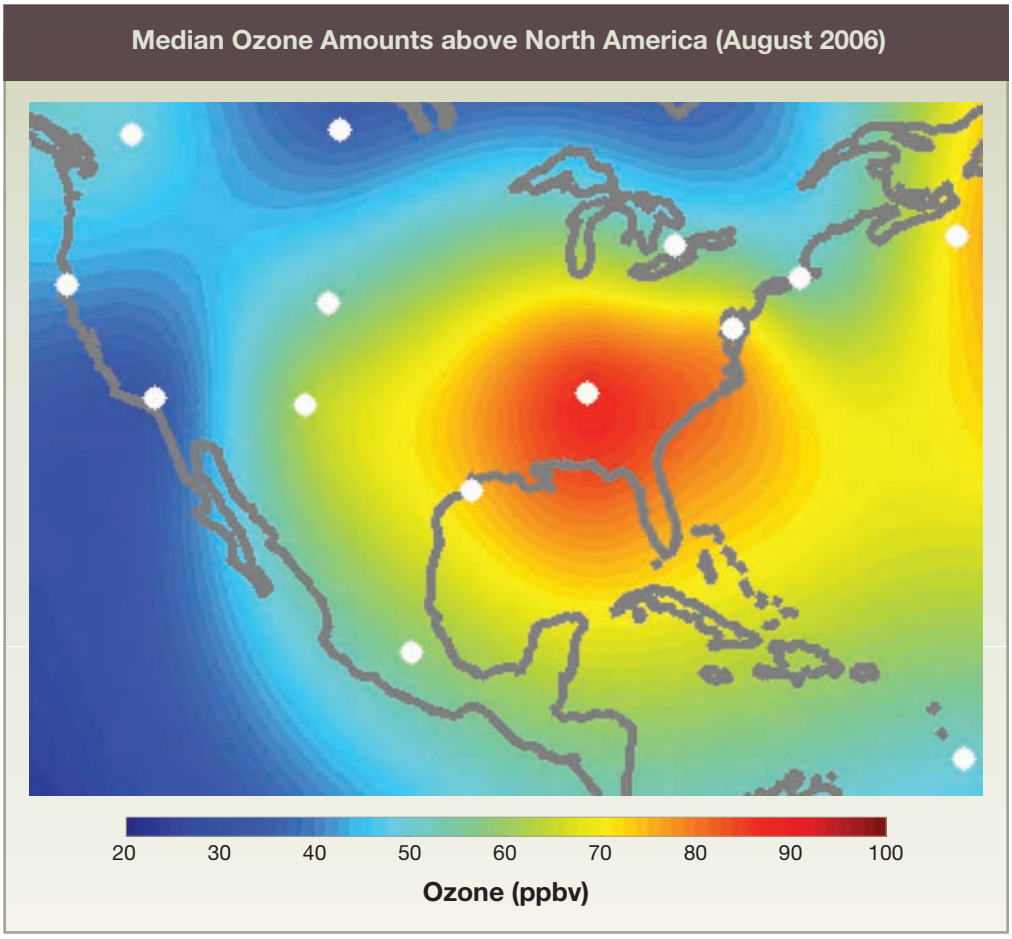


Figure 5: Median Ozone Amounts above North America (August 2006). Median ozone amounts above North America, in parts per billion, at 10 to 11 km during August 2006, after stratospheric ozone contribution was removed. The ozone enhancement is mainly due to nitrogen oxides emitted by lightning into the upper troposphere, followed by reactions with carbon monoxide, methane, and volatile organic compounds of both anthropogenic and natural origins. The white dots show the locations of observing stations. *Credit: O.R. Cooper, CIRES and NOAA / Earth System Research Laboratory (reproduced from* **Journal of Geophysical Research** *with permission from the American Geophysical Union).*

to the radiation budget at the regional scale. Other data gathered in the study demonstrate that the influence of lightning and convection on upper-tropospheric ozone has been previously underestimated. Lightning may have been underestimated by a factor of four, and faster convection rates may be needed to accurately model this region of the atmosphere. Large differences remain between observed and modeled levels of free radicals in the upper troposphere, an indicator that a major uncertainty remains in the understanding of how lightning-produced nitrogen oxides affect ozone in the upper troposphere.

Field Mission Investigates Atmospheric Composition, Clouds, and Climate in the Tropical Atmosphere. CCSP researchers carried out a field mission that gathered chemical and meteorological data for the cold, dry conditions of the upper tropical tropopause (an important transition region between the troposphere and stratosphere). This region of the Earth's atmosphere between 14 and 18 km plays a key role in both climate change science and depletion of the stratospheric ozone layer. The Tropical Composition, Clouds, and Climate Coupling (TC4) experiment, based in San Jose, Costa Rica, involved dozens of scientists from U.S. agencies and academia and used a unique combination of coordinated observations from satellites, ground stations in the inter-tropical convergence zone, and instrumented aircraft during July and August 2007 (see <www.espo.nasa.gov/tc4/>). One of the specific goals of TC4 was to study the composition, formation, and radiative properties of clouds (cirrus and sub-visible cirrus) in this region, thereby assessing the contributions of such clouds, aerosols, and water vapor to climate forcing. Other aspects of the campaign focused on understanding the convective processes that control the transport of trace gases and aerosols from the lower atmosphere into the tropical tropopause and thence into the stratosphere, where they can influence stratospheric ozone. This mission gathered data that CCSP researchers are using to expand the scientific understanding of climate-cloud-chemistry interactions in the highly active and climate-relevant region of the tropical tropopause. The measurements also will be used to test retrieval algorithms for several instruments on the Aura satellite that measure trace gases (High Resolution Dynamic Limb Sounder, Microwave Limb Sounder, and Thermal Emission Spectrometer).

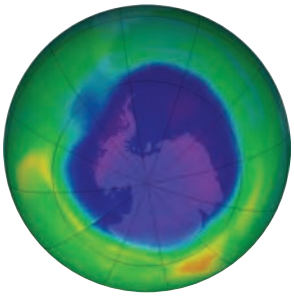
Global Transport of Pollution from Satellite Observations of Carbon Monoxide.[15,16] Carbon monoxide (CO) in the Earth's atmosphere is formed by the incomplete combustion of fossil fuels and biomass burning, and is primarily emitted at the surface. It can be lifted into the atmosphere by convection and transported around the globe by the prevailing winds. Its relatively long lifetime enables CO to be a good tracer of transport processes, such as the trans-Pacific transport of Asian pollutants to North America. Two years of observations from the Microwave Limb Sounder on the Aura satellite have provided the spatial distribution, temporal variation, and long-range transport of atmospheric CO and have shown the close relationship of concentrations of this gas to

surface emissions, deep convection, and horizontal winds. The transport of CO from Southeast Asia across the Pacific to North America occurs most frequently during the Northern Hemisphere summer when deep convection associated with the Asian monsoon is clustered over the strong anthropogenic emission regions. Measurements of the global distribution of CO over time provide a strong indicator of the connections between changes in air quality due to increased industrialization and climate.

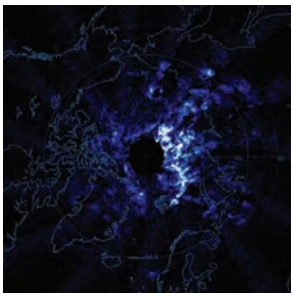
Regional Pollution and Global Climate Change

Impact of Global Change on U.S. Regional Air Quality.[17,18,19,20] Emerging study results show that climate change has the potential to increase ground-level ozone concentrations in many areas of the United States and to lengthen the season of elevated ozone events. These increases may be beyond the envelope of natural interannual variability. Planned and future emissions controls will lower U.S. ozone concentrations, but the impacts of global change may necessitate further reductions to meet national air quality standards.

Recovery of the Stratospheric Ozone Layer



A New Formulation for Gauging the Effects of Ozone-Depleting Substances on the Ozone Layer.[21] Equivalent effective stratospheric chlorine (EESC) is a convenient parameter to quantify the effects of halogens (chlorine and bromine) on the depletion of the stratospheric ozone layer. EESC has been extensively used to evaluate future scenarios of ozone-depleting substances (ODS) in the stratosphere. CCSP research has led to a new formulation of EESC that provides revised estimates of ozone layer recovery. The work shows that ODS levels will recover to 1980 levels in the year 2041 in the mid-latitudes, and 2067 over Antarctica, assuming adherence to international agreements that regulate the use of ODS. The researchers also assessed the uncertainties in the estimated recovery times. The 95% confidence interval associated with the mid-latitude recovery is the time period from 2028 to 2049, while it is from 2056 to 2078 for Antarctic ODS recovery.



Highlights of Plans for FY 2009

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. The general emphasis will continue to be on quantifying the effects of aerosols and non-CO_2 greenhouse gases on climate. In FY 2009, the following research activities will be emphasized to meet the overall priority.

Analysis of International PolarYear Data Sets. Research across several agencies will focus on analyzing data sets gathered during the 2008 spring and summer experiments to study Arctic aerosols and their connections to clouds, radiation, and ice melting. Measurements from satellites, aircraft, and the surface were made to assess the influences of long-range pollution transported to the Arctic environment, the so-called Arctic haze. Specific areas of scientific investigation include the effects of Arctic haze on ozone chemistry and ice melting; studies of the influence of boreal forest fires on the Arctic climate; and an evaluation of aerosol/cloud influences in the Arctic.

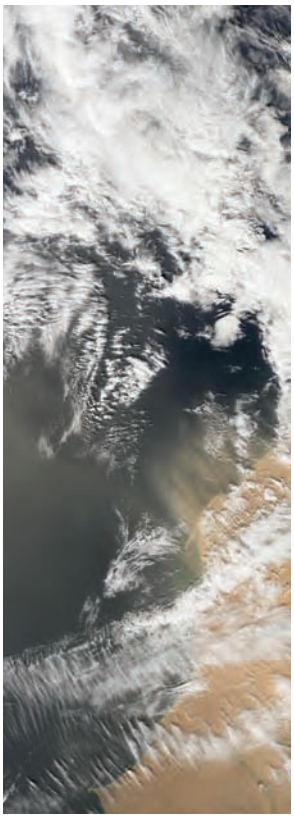
This activity will address Questions 3.1 and 3.3 of the CCSP Strategic Plan.

Utilization of Ground-Based Measurement Networks for Climate and Ozone Studies. Retrievals of atmospheric particulate absorption from AERONET will continue to be utilized in climate forcing studies and in the validation of current and future satellite missions, such as the Glory mission (2009 launch) that will measure sunlight absorption by aerosols. Network expansion will continue, with a focus on inadequately sampled regions that are important for understanding global climate change, such as China (both the polluted eastern regions and the western deserts that are a source of dust storms). An experimental effort is underway to investigate the sensitivity of ground-based instruments for nighttime measurements using moonlight. Studies using the Micro Pulse Lidar Network data will focus on the influence of polar stratospheric clouds on ozone destruction over the Antarctic and on the impacts of Arctic haze on polar climate.

This activity will address Questions 3.1 and 3.3 of the CCSP Strategic Plan.

Continue Regional Aerosol Study in China. In 2008, the ARM Mobile Facility (AMF) was deployed in China to identify and quantify the climatic effects of aerosols. The AMF was deployed at Lake Taihu (April-December) near Shanghai, China, and the ARM Ancillary Facility was also deployed from Linze (February-June) and Xianghe (July-December). Data will be used to improve rain remote sensing and understanding the roles of aerosols in affecting regional climate and atmospheric circulation.

This activity will address Questions 3.1, 3.2, and 3.3 of the CCSP Strategic Plan.

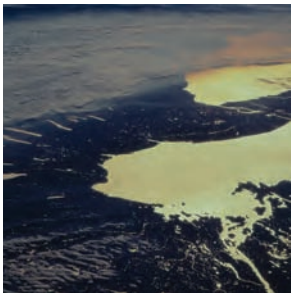


Cloud/Aerosols Field Study. Extensive and persistent layers of stratus clouds occur off the subtropical west coasts of Africa and of North and South America. These cloud decks have a significant impact on Earth's radiation budget. Aerosols, arising from natural processes and from human activity, have important influences on the brightness and persistence of these clouds. The Variability of the American Monsoon System (VAMOS) Ocean-Cloud-Atmosphere-Land Study (VOCALS) field mission will study the stratus deck off the Pacific coast of Chile and Peru, using *in situ* and remote aircraft observations, along with satellite and ship-based measurements. Natural and human sources of particles will be observed, as well as the roles these particles play in the determining the brightness and lifetimes of stratus clouds. The VOCALS planning and site research occurred early in 2008, and the field mission will occur in October/November 2009.

This activity will address Questions 3.1, 3.2, and 3.3 of the CCSP Strategic Plan.

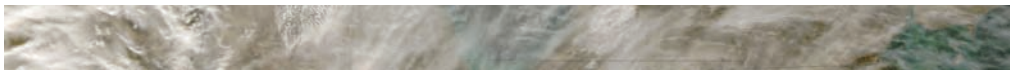
Examination and Intercomparison of Water Vapor Measurements from Aircraft, Balloons, and Satellites. Understanding changes in the distribution of water vapor, whether due to natural or anthropogenic causes, is essential to understanding the potential for climate change. Shortfalls in knowledge of the processes affecting water vapor concentrations near the interface between the troposphere and the stratosphere result primarily from the difficulties in making accurate water vapor measurements at these altitudes where concentrations are quite small. A number of research efforts will be continued or initiated to help resolve observed discrepancies in *in situ* water vapor observations. These activities are being conducted jointly by two CCSP agencies with the involvement of U.S. and international investigators from a wide range of government and academic institutions. The planned efforts include (1) single instrument laboratory studies designed to better characterize and understand instrument performance and calibration under a variety of atmospheric conditions; (2) the possible selection and use of a water vapor calibration standard to establish and/or confirm measurement accuracy and precision; and (3) multiple-instrument intercomparisons in the laboratory and field involving an independent referee to coordinate and present the results of each formal laboratory and flight intercomparison that includes instruments from different research groups.

This activity will address Questions 3.2 and 3.5 of the CCSP Strategic Plan.



Interactions of Climate Change and Air Quality. Understanding the combined effect of climate change and air quality is a key research question. Continuing work in FY 2009 will focus on (1) reducing the uncertainty for ground-level ozone; (2) assessing the impact of climate change on particulate matter; and (3) preliminary research to enable assessment of interactions with mercury. The FY 2009 work focuses on linked air quality-climate modeling systems, future emission projections, and communicating research results to air quality decisionmakers.

This activity will address Questions 3.3 and 3.5 of the CCSP Strategic Plan.



Aviation Impacts on the Upper Atmosphere and Climate Change. CCSP agencies concerned with understanding the impacts of aviation on climate change will address key uncertainties and research gaps through the Aviation-Climate Change Research Initiative (ACCRI). ACCRI will use a structured, sequential four-step approach, simultaneously applying the latest scientific knowledge and modeling/analysis capability to quantify the climate impacts of aviation. The main objective of ACCRI is to inform policymaking decisions for the U.S. Next Generation Air Transportation System and the International Civil Aviation Organization Committee on Aviation Environmental Protection based on the latest research results. A key goal of ACCRI is to identify, develop, and evaluate the metric(s) that can capture aviation-induced climate impacts at all relevant spatial and temporal scales and assist in tradeoff analysis whenever possible.

This activity will address Questions 3.3 and 3.5 of the CCSP Strategic Plan.

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