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 composition of the atmosphere—its gases and particles—plays a critical role in connecting human welfare with global and regional changes because the atmosphere links all of the principal components of the Earth system. The atmosphere interacts with the oceans, land, terrestrial and marine plants and animals, and the cryosphere (regions of ice and snow). Because of these linkages, the atmosphere is a conduit of change.

Emissions from natural sources and human activities enter the atmosphere at the surface and are transported to other geographical locations and often higher altitudes.

Some emissions undergo chemical transformation or removal while in the atmosphere or interact with cloud formation and precipitation. Some natural events and human activities that change atmospheric composition also change the Earth's radiative (energy) balance. Subsequent responses to changes in atmospheric composition by the stratospheric ozone layer, the climate system, and regional chemical composition (air quality) create multiple environmental effects that can influence human health and natural systems.

Changes in atmospheric composition are indicators of many potential environmental issues. Observations of trends in atmospheric composition are among the earliest harbingers of global changes. For example, the decline of the concentrations of ozone-depleting substances, such as the chlorofluorocarbons (CFCs), has been the first measure of the effectiveness of international agreements to end production and use of these compounds.

A principal feature of the atmosphere is that it acts as a long-term "reservoir" for certain trace gases that can cause global changes. The long removal times of some gases, such as CO_2 (more than 100 years) and perfluorocarbons (more than 1,000 years), imply that any associated global changes could persist over decades, centuries, and millennia—affecting all countries and populations.

The overall research approach for understanding the role of atmospheric composition is an integrated application of long-term systematic observations, laboratory and field studies, and modeling, with periodic assessments of understanding and significance to decisionmaking. Most of the activities related to atmospheric composition research are part of national and international partnerships. Such partnerships are necessitated by the breadth and complexity of current issues and because the atmosphere links all nations. Current research on atmospheric composition is based on the substantial body of knowledge and understanding available from the work of many scientists.

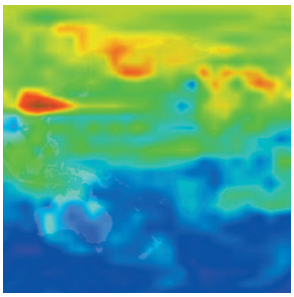
Highlights of Recent Research

Highlights of recent research supported by CCSP participating agencies follow.

Global atmospheric methane levels constant. Increases in methane are the second most important contribution to the radiative forcing of climate from human activities. Methane has a relatively short lifetime in the atmosphere. The globally averaged abundance of methane is monitored using an extensive network of surface sampling sites. Methane growth rates in the atmosphere have been steadily decreasing

since 1985. Recent analysis now shows that global methane abundances have been constant from 1999 through 2002. This lack of change strongly suggests that methane may have reached a steady state in the atmosphere determined by a balance between methane emissions and removal processes. The annual averages also show an abrupt drop in the early 1990s in the difference between methane values in the Northern and Southern Hemispheres. This change is attributed to reductions in emissions from the former Soviet Union, which might have accelerated the approach to steady state conditions. Future global measurements will continue to be of great importance to improve our understanding of the global methane budget.

Global aerosol measurements. Observational studies have identified chemical markers for distinct aerosol sources such as smoke from forest fires and African dust. These markers can be used to identify aerosol transported over continental and intercontinental scales. Ground-based measurement stations in California and in the Azores—two key aerosol observing sites in an emerging global network—have been of particular value in recent studies. Data from ground-based sites obtained during the Intercontinental Transport and Chemical Transformation (ITCT-2K2) field campaign suggest that the springtime background aerosol found in the western United States was Asian in origin.



The ability to characterize and understand the nature and origins of atmospheric aerosols is rapidly improving. In an intensive field campaign, detailed investigations were undertaken of the composition, size, shape, and mixing state of individual particles of mineral dust and urban aerosols originating from East Asia—a region that is the source of substantial emissions, dust storms, and pollution plumes. The optical properties (important for understanding the role of aerosols in climate change) varied significantly depending on the mixture of components, and a range of shapes of elemental carbon-containing aerosols was observed.

In addition, models for aqueous and gas-phase chemistry are being coupled with thermodynamic models for an improved description of aerosol-gas-cloud interactions.

New techniques for aerosol measurements. Rapid progress in new techniques for aerosol measurements promises to improve the ability to characterize aerosols in future field and laboratory studies. NOAA, NASA, and NSF have supported several new analytical techniques developed to probe aerosol composition that are proving effective in identifying aerosol sources and changes in composition. Examples include aerosol sampling with mass spectrometers, liquid surfaces, improved rotating drum impactors, and relative-humidity-controlled samplers, and an aerosol analytical method based on synchrotron spectroscopy.

Efforts at the National Institute of Standards and Technology (NIST) have also focused on improving the accuracy of thermal-optical analysis, a method widely used

for monitoring black carbon levels. In another NIST effort to study the effect of soot on radiative transport in the atmosphere, quantitative measurements were performed on the extinction coefficient of soot in the infrared region.

Scientific assessment of ozone layer depletion. In the 2002 *Scientific Assessment of Ozone Depletion*, published in 2003, the world's leading scientists defined the current understanding of the ozone layer and the phenomenon of stratospheric ozone depletion. This global, multiagency effort was led by NOAA, NASA, the European Commission, the UN Environment Programme, and the World Meteorological Organization. Scientists in several agencies and in academia played leading roles in authoring the report, as well as carrying out its review and final publication. The Ozone Assessment plays a particularly unique role as a "bridge" between the scientific community and decisionmakers and is designed to underpin future international decisions about ozone-depleting substances and the protection of the ozone layer.

Findings of the 2002 Ozone Assessment include an update on atmospheric processes underlying ozone abundance at the poles and globally, observations of ozone-depleting substances in the atmosphere, expectations for recovery of the ozone layer, and approaches to evaluating the ozone-layer impacts of very short-lived halogen-containing substances. Antarctic ozone depletion has been found to be large throughout the last decade. The size of the Antarctic ozone hole in September 2003 is the largest on record. It is not yet possible to say whether the annual peak in the area of the ozone hole has reached its maximum. The abundance of ozone-depleting compounds controlled by the Montreal Protocol continues to decline slowly in the lower atmosphere. The assessment concludes that the global ozone layer is expected to begin

Figure 4:
Covers of three 2002 Ozone Assessment reports.

recovery within the next decade or so, assuming continued international compliance with the Montreal Protocol. The ozone layer will remain particularly vulnerable during the next decade because halogen-containing gases in the stratosphere will be at their greatest values.

Splitting of the 2002 Antarctic ozone hole. The Antarctic ozone hole is a springtime fixture in southern high latitudes, occurring every year in the stratosphere as the result of high chlorine and bromine levels and low winter temperatures. In late September 2002, a major sudden warming occurred in the Southern Hemisphere stratosphere (see Figure 5). This unusual event split the polar vortex and the associated Antarctic ozone hole into two distinct parts. An unexpected major warming such as that observed in 2002 likely occurs only once a century and, hence, does not change the predictions of ozone recovery in the next decades as chlorine and bromine levels decline in the global stratosphere. Year-to-year changes in ozone hole area (or depth) are more likely to reflect changes in stratospheric "weather" conditions, rather than longer term changes in chlorine and bromine levels. NASA, NOAA, and NSF are supporting model evaluations of this unusual event to further our understanding of stratospheric transport and dynamics.

Observations and modeling of Asian pollution outflow. Emission inventories for atmospheric constituents are essential inputs to models used for simulating and projecting changes in air quality and climate. These inventories are constructed in what is known as the 'bottom up' method by using current knowledge of consumption

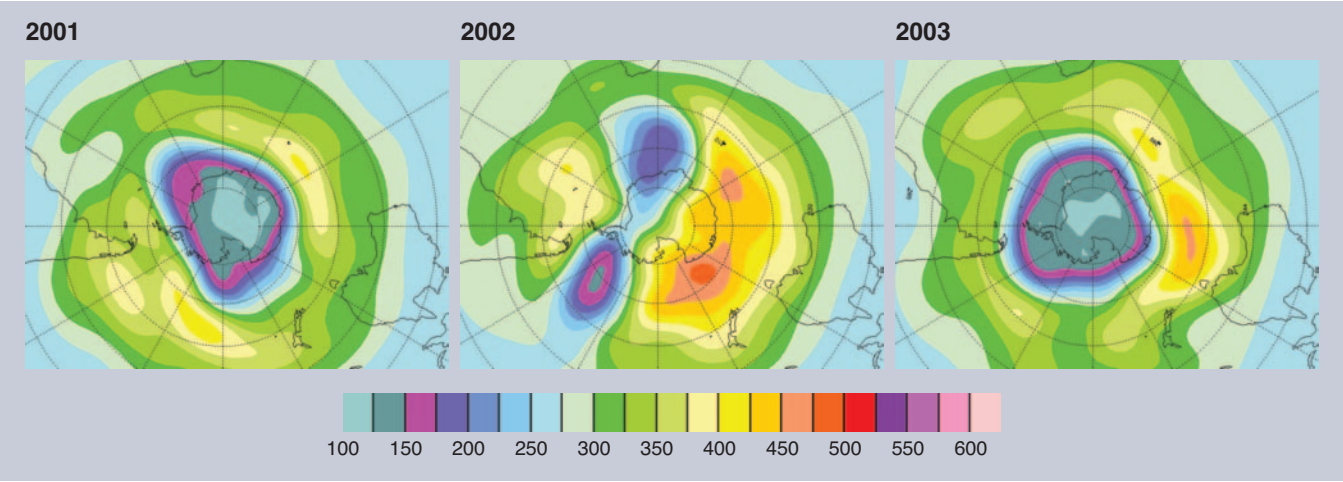
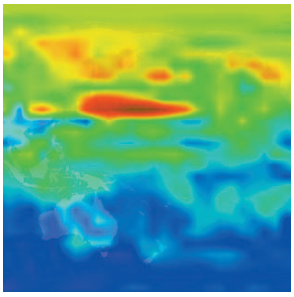
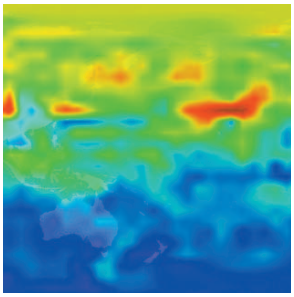


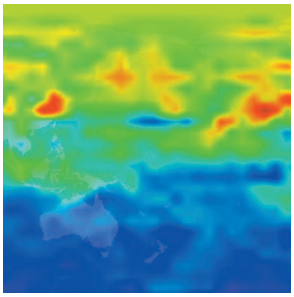
Figure 5: Satellite maps of total ozone over Antarctica on 24 September 2001, 2002, and 2003. The color scale shows the amount of ozone in Dobson units, indicating the depth of the hole. The images are based on multiple satellite records and analyses.
*Credit: Susan Solomon, "The hole truth." Nature, **427**, 22 January 2004. World Ozone and Ultraviolet Radiation Data Centre, Toronto, Canada.*

and practices in various nations, regions, and economic sectors. Recently, much progress has been made in demonstrating the value of global satellite and airborne observations for improving knowledge of emissions inventories, through combining the bottom-up derived information with 'top-down' data derived from observations. NASA's Transport and Chemical Evolution over the Pacific (TRACE-P) mission integrated aircraft and satellite data with chemical transport models (CTM) to analyze Asian carbon monoxide (CO) sources. A central objective of TRACE-P was to improve knowledge of Asian emission inventories through the addition of top-down constraints derived from observations in airmass outflow from Asia. Asian emissions for the period of the TRACE-P mission, based on the best bottom-up data sets, were generated prior to the mission. These emission estimates were used by five different CTMs to forecast the location of pollution plumes and to direct the observing aircraft toward this Asian outflow, thereby optimizing the testing of the model forecasts. Validation of the satellite-based CO observations was conducted during the mission to provide a seamless aircraft-satellite CO data set. These TRACE-P CO data were then assimilated into the models and used to derive the emission inventories with a method known as inverse modeling. Major conclusions were that the bottom-up estimates of anthropogenic emissions from China were 50% too low, biomass burning emissions from southeast Asia were 60% too high, and Japan and Korean emissions were roughly correct. The advantage of the combined bottom-up and top-down approach has been demonstrated for other important atmospheric constituents, such as tropospheric ozone precursors and also for absorbing aerosols.





Effects of regional pollution on the global atmosphere. Substantial progress is being made in describing the fate of anthropogenic emissions in the global atmosphere. New measurement techniques and observational studies addressing regional pollution were supported by NOAA, NSF, and other agencies. Rapid airborne measurements of formaldehyde (CH_2O) provided new insight into the reactive intermediate products of volatile organic carbon (VOC) oxidation in petrochemical plumes. Since VOC oxidation drives ozone production in the polluted boundary layer, this is a valuable new tool to evaluate the effect of regional pollution on the global atmosphere. A new chemical ionization mass spectrometry technique is now available for organic nitrogen species, which will improve understanding of how nitrogen oxide emissions are chemically transformed and transported globally. Regional- and global-scale models have been linked to capture intercontinental transport of regional emissions and the results are being compared with observations from multiple ground-based sites.



Anthropogenic emissions in megacities. The growth of pollution sources in the world's megacites has reduced air quality for millions of inhabitants. A broad-based initiative has been developed for the Mexico City Metropolitan Area to address the

Figure 6:
Aerial photo taken during Mexico City Metropolitan Area (MCMA) 2003 Field Measurement Campaign.
Credit: MCMA 2003 Field Measurement Campaign.

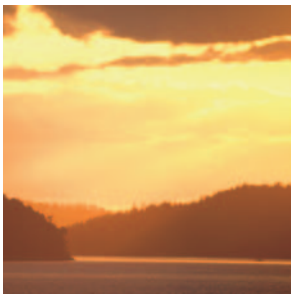
causes and possible solutions of the local and regional pollution that affects the area's large population. NSF and DOE supported deployment of state-of-the-art instruments near Mexico City in an April 2003 study in order to improve understanding of the space- and time-dependent nature of emissions and atmospheric composition in a developing megacity. Enhanced understanding of these emissions will lead to better assessment of the role of megacities in influencing regional and global air quality (see Figure 6).

Regional hydrocarbon pollution in the southwestern United States. Research in the southwestern United States has revealed significant quantities of light alkane hydrocarbons in the near-surface atmosphere of Texas, Oklahoma, and Kansas during both autumn and spring seasons. The levels are attributed to direct emissions from the oil and natural gas industries in the southwestern states. Observed alkyl nitrate mixing ratios in north-central Texas (maximum 34 ppbv ethane, 20 ppbv propane, and 13 ppbv n-butane) were comparable to urban smog values, with abundant formation of tropospheric ozone. Ozone production adversely affects human health and plant growth, and contributes to climate forcing. An estimated 4-6 teragrams of methane are released annually within the region, representing a significant fraction of the estimated total U.S. emissions. This result suggests that total U.S. natural gas emissions may have been previously underestimated. These measurements will lead to a more comprehensive understanding of U.S. greenhouse gas emissions and the role of fossil fuel hydrocarbon emissions in regional air quality.

Cirrus cloud study. A unique data set of properties of cirrus clouds was obtained in 2002 from a field campaign conducted in southern Florida. The Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment (CRYSTAL-FACE) was an interagency effort involving NASA, NOAA, NSF, DOE, and the Naval Research Laboratory. More than 60 *in situ* and remote measurements operated onboard six aircraft. Aircraft flights were coordinated with overpasses of instruments on the Terra and Aqua satellites in order to validate cloud and aerosol retrieval algorithms. Scientists are analyzing the multi-instrument data set to enhance understanding of the properties of cirrus clouds and their role in the climate system. CRYSTAL-FACE results published to date address the role of Saharan dust in cloud systems, the use of water isotope measurements to constrain the stratospheric dehydration process, the composition of cloud condensation nuclei, and the role of nitric acid in affecting relative humidity in low-temperature cirrus clouds.

Highlights of Plans for FY 2004 and FY 2005

The 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 and tropospheric chemistry. Key research plans for FY 2004 and FY 2005 are described below.



Tropospheric aerosols and climate. Atmospheric aerosols play a significant role in modifying the amounts of solar radiation absorbed at the surface and in the atmosphere. Changes in absorbed radiation can influence climate change, which can, in turn, affect the concentration of aerosols, especially on regional scales. The role of aerosols in climate processes will be examined as part of an international, interagency intensive field campaign based in the eastern United States in mid-2004. A joint NASA, NOAA, and DOE mission, using multiple instrumented aircraft, will make measurements focused on the properties and distribution of aerosols; processes affecting distribution; source-strength estimates; and radiative impact, with specific emphasis on regional and continental scales. Study objectives include using observations to diagnose the indirect effect of anthropogenic aerosols on cloudiness. Coordination with the NASA Aura and Terra satellites and the European Envisat satellite will facilitate connection with global data sets and provide validation to several satellite instruments. In separate studies, regular vertical profiling of aerosol parameters will be made from light aircraft. Data from these campaigns will be used to improve aerosol chemistry and transport models, and will address the CCSP aerosol transport/transformation and radiative impact priorities, as well as provide information that is common to a better understanding of both climate and air quality processes.

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

CCRI PRIORITY - AEROSOLS

The Climate Change Research Initiative (CCRI) will accelerate research on aerosols and leverage existing USGCRP research to address major gaps in understanding climate change. Uncertainties related to the effects of aerosols on climate are large, with both warming and cooling effects possible depending on the nature and distribution of the aerosol.

Research has demonstrated that atmospheric particles (aerosols) can cause a net cooling or warming tendency within the climate system, depending upon their physical and chemical characteristics. Sulfate-based aerosols, for example, tend to cool, whereas black carbon (soot) tends to warm the system. In addition to these direct effects, aerosols can also have indirect effects on radiative forcing (e.g., changes in cloud properties). When climate models include the effects of sulfate aerosol, the simulation of global mean surface temperatures is improved. One of the largest uncertainties about the net impact of aerosols on climate is the diverse warming and cooling influences of the very complex mixture of aerosol types and their spatial distributions. Further, the poorly understood impact of aerosols on the formation of both water droplets and ice crystals in clouds also results in large uncertainties in the ability to project climate changes. More detail is needed globally to describe the scattering and absorbing optical properties of aerosols from regional sources and how these aerosols impact other regions of the globe.

The CCRI will advance the understanding of the distribution of all major types of aerosols and their variability through time, the different contributions of aerosols from human activities, and the processes by which the different contributions are linked to global distributions of aerosols. The CCRI will support research to improve understanding of the processes by which trace gases and aerosols are transformed and transported in the atmosphere. Studies of how atmospheric chemistry, composition, and climate are linked will be emphasized, including those processes that control the abundance of constituents that affect the Earth's radiation budget, such as tropospheric methane, ozone, and aerosols.

The global distributions of a limited number of atmospheric parameters (including climatically relevant parameters such as ozone and aerosols) and their variabilities will be obtained from satellite observations over long periods of time along with more comprehensive suites of observations over briefer time periods. Satellite data recently obtained and to become available for the first time for methane, tropospheric ozone, and tropospheric aerosols will be analyzed and interpreted in the context of global models and assimilation systems.

The studies will provide an observational- and model-based evaluation of the radiative forcings associated with aerosol direct and indirect effects. These forcing results will contribute to the CCSP climate projections for research and assessment.

Following the 2004 field mission, a major activity within NOAA, DOE, and NASA will be to analyze and interpret the field data set, with an emphasis on modeling. These efforts are expected to yield substantial information on the fate and transport of pollutants emitted from North America and how their chemical and radiative properties evolve as they move across the Atlantic Ocean. The data interpretation will demonstrate how combining information from multiple sampling platforms (e.g., aircraft and satellites) allows a more complete description of the transport and transformation processes. Innovative approaches to studying the radiative effects of aerosols will also have been part of the 2004 mission, with the results expected to decrease the uncertainty in understanding of aerosol climate effects. The results will make substantial contributions to national (CCSP 2006 products) and international (IPCC Fourth Assessment Report) assessments. Tangible results in the form of reports and peer-reviewed publications are expected to begin appearing by the end of 2005.

These activities will address Questions 3.1 and 3.3 of the CCSP Strategic Plan.

Investigations of aerosol-cloud-climate interactions will continue beyond 2004, with novel instruments, techniques, and approaches developed to study aerosol indirect effects. Studies already planned include, for example, using an aerosol-gas-cloud chemical model, newly coupled with a thermodynamic model, to investigate the effects of organic aerosols on cloud microphysical properties.

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

In 2005, regular vertical profiling of aerosols also will continue, with the ultimate goal of developing an aerosol 'climatology' of North America. This continuous information will not only give scientists a comprehensive understanding of U.S. aerosol emissions, fate, and transport, but also will establish a baseline against which effects of policy measures to reduce human-induced aerosol emissions may be measured.

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

Aura satellite launch. Researchers will begin examining data from the new Aura satellite scheduled to be launched in mid-2004. The satellite will carry four instruments designed to measure trace gases and aerosols in the troposphere and stratosphere. Aura measurements made in the troposphere using infrared emissions will provide an unprecedented data set from space on tropospheric composition. Using the measurements of tropospheric ozone, carbon monoxide, nitric acid, water vapor, and other trace gases, the transport and chemical transformation of anthropogenic pollution can be investigated from the local to global scale. Also measurements of stratospheric ozone, ozone-depleting gases, and other stratospheric gases will improve detection and attribution of ozone layer recovery in the coming years.

In 2005, the Aura satellite will be operational, producing large global data sets of trace gases and aerosols in the troposphere and stratosphere. The subsequent analysis and interpretation of these data sets by NASA, NOAA, NSF, and other agencies will be used to address a wide variety of research questions as noted above.

These activities will address Questions 3.2, 3.3, and 3.4 of the CCSP Strategic Plan.

Global transport of atmospheric pollutants. A 2004 study supported by NOAA and NASA will explore the processes governing the formation, transport, and fate of tropospheric ozone over northeastern North America and the North Atlantic Ocean. The instrumented aircraft and ship platforms involved in this study will be able to diagnose the sources of pollutants that increase regional ozone levels and follow the trajectories of emissions as they move eastward over the continental United States and ultimately offshore into the North Atlantic. With participation of European airborne platforms, the tracking of polluted air masses will continue across the Atlantic Ocean. This field research will address the need to quantify North American outflow of reactive

and long-lived gases and aerosols and to understand the balance between long-range transport and transformation of pollutants.

In 2005, a major activity within NOAA and NASA will be to analyze and interpret the field data sets obtained in the 2004 field mission. The results of these activities will yield insight into the effects of regional pollution on the global scale. The results will also form part of a detailed global survey of tropospheric ozone and its precursor species and provide valuable data for employing atmospheric chemistry models coupled with general circulation models to improve understanding of the feedbacks between regional air pollution and global climate change.

These activities will address Questions 3.2 and 3.3 of the CCSP Strategic Plan.

Megacity modeling studies. Data acquired in the Mexico City measurement-intensive study in April 2003 will be used to address the fate and impact of anthropogenic emissions from a megacity population. The study provided an unprecedented data set of emissions from the most polluted metropolitan area in the world. With NSF support, model studies will use the observations to constrain estimates of local and regional pollution transport and transformation. Of interest is the production of ozone and the formation of aerosols in the urban center and the export of ozone precursors and aerosols to the regional and global scale. Models are now able to incorporate emission sources on the subgrid scale in regional and global analysis.

These activities will address Questions 3.1, 3.2, and 3.3 of the CCSP Strategic Plan.

Consequences of climate change for U.S. air quality. A scenario-based assessment of the potential consequences of global change for regional U.S. air quality is being conducted, focusing on fine particles and ozone. The climate, emissions, and underlying socioeconomic scenarios are intended to provide a plausible depiction of future conditions, rather than predictions of what actually will happen. Research and assessment activities will focus on developing regional-scale inputs for air quality simulations, using the Community Multiscale Air Quality modeling system. The impacts of potential increases in transported pollutants due to further global industrialization, population growth, and increases in economic activity are also being assessed. The continuing research in FY 2004 and FY 2005 will build toward a 2007 assessment of changes in U.S. air quality due to climate change, which includes direct meteorological impacts on atmospheric chemistry and transport, and the effect of changes in temperature, cloud cover, and precipitation on air pollution emissions. Further research will result in a 2010 assessment that adds the emission impacts from technology, land use, and demographic changes to construct plausible scenarios of U.S. air quality 50 years into the future.

These activities will address Questions 3.3 and 3.5 of the CCSP Strategic Plan.

Figure 7:
The High-performance Instrumented Airborne Platform (HIAPER) aircraft.
Credit: University Corporation for Atmospheric Research.

Instrumenting the HIAPER research aircraft. In late FY 2004, a new platform for atmospheric research will be delivered. This new aircraft, a Gulfstream V (G-V), is designated as the High-performance Instrumented Airborne Platform (HIAPER, see Figure 7). The G-V's combination of payload capacity, altitude, and range make it an important new national resource for the study of meteorology and chemistry in the troposphere and lower stratosphere. Modifications specified by a scientific users group that will make the aircraft suitable for atmospheric research will be completed before delivery. In FY 2004, NSF will begin supporting the development and construction of new instrumentation for HIAPER. In FY 2005, the HIAPER aircraft will be fully flight tested, facility instrumentation will be evaluated, and one or more progressive science missions will be flown in preparation of the aircraft becoming formally available to the scientific community by the end of FY 2005.

The development of this platform and its instruments will potentially support a wide range of activities needed to address the Atmospheric Composition research element of the CCSP Strategic Plan.