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?

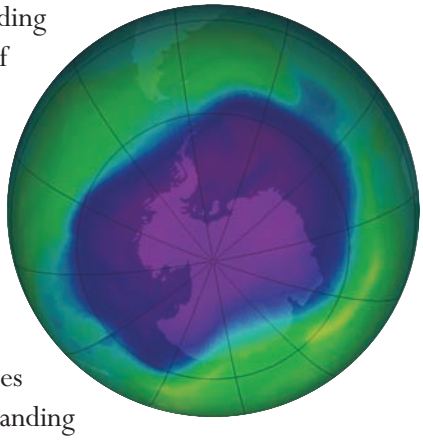
3.4 What are the characteristics of the recovery of the stratospheric ozone layer in response to declining abundances of ozone-depleting gases and increasing abundances of greenhouse gases?

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.

Earth's atmosphere is a complex mixture of gases and particles. Although most are present at trace amounts of one-millionth or less of the total, those very small abundances are sufficient to play a major role in regional and global issues that affect the well-being of humans and ecosystems. Indeed, changes in atmospheric composition are the drivers of major environment-related issues of today: climate change, depletion of the stratospheric ozone layer, and air quality at the Earth's surface. The composition of the atmosphere is changing due to both natural and anthropogenic influences. The changes have been significant in the approximately 250 years since the beginning of the Industrial Revolution. CCSP is developing a framework for research and observations that will elucidate the processes at work in our atmosphere and the way they influence

atmospheric composition and hence climate, the environment, and human health, with the ultimate aim of providing the scientific understanding needed to underpin decisions and planning in the 21st century.



Great strides have been made in the last two decades concerning our understanding of how atmospheric composition relates to the global environment. The cause of the Antarctic ozone hole, first observed in the 1980s and initially a geophysical mystery, has been conclusively linked to the trace amounts of human-made chlorine- and bromine-containing gases released to the atmosphere starting in the 1960s. The rise in greenhouse gases in the atmosphere has been meticulously documented for several decades. The interactions between natural emissions and emissions caused by human activity have been shown to lie at the heart of the quality of the air we breathe. Increasingly, researchers are taking up the challenge of understanding the complex interactions between issues such as climate change, ozone depletion, and air quality. Better scientific understanding holds the promise of enabling future decisions that will be effective across multiple—and interwoven—societal concerns.

CCSP-supported research in atmospheric composition for FY 2007 will have a high-priority focus on improving the predictive understanding of the role of natural and human-influenced aerosols (airborne fine particles) on the climate system. Aerosols interact with radiation directly (by absorbing or scattering radiation) as well as indirectly (e.g., by influencing the formation and properties of clouds). Unlike greenhouse gases, aerosols can either warm or cool the atmosphere through these interactions. Furthermore, the lifetime of aerosols in the atmosphere is shorter than that of many greenhouse gases, therefore the time frame for responding to their influence is correspondingly shorter than for the long-lived greenhouse gases. The understanding of how aerosols influence climate has been identified as one of the most uncertain scientific areas and a high priority for research. The interaction between aerosols, clouds, precipitation, and climate is a specific focus of FY 2007 research in the atmospheric composition component of CCSP. Research also focuses on understanding the role of non-CO_2 greenhouse gases (e.g., changes in water vapor distribution due to human influences) in climate, as well as investigating the long-range processes that transport and transform greenhouse gases in the atmosphere.

The overall research approach for understanding the role of atmospheric composition is an integrated application of long-term systematic observations, laboratory research, intensive field studies focused on gaining a process-level understanding of atmospheric phenomena, and diagnostic analyses and modeling that advance predictive capabilities. These endeavors are coupled with periodic assessments of understanding and development

of products that synthesize findings in ways that are useful to decisionmakers and planners. Research is highly collaborative and involves partnerships throughout the Nation and the world.

Highlights of Recent Research

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

Upper Atmospheric Water Vapor Trends and Feedbacks.[2,12,13] Climate models predict that as ocean surfaces warm due to a warming climate, the increased evaporation will moisten all levels of the atmosphere, especially the upper atmosphere. Since water vapor is itself a greenhouse gas, this moistening further increases the warming. Models of Earth's climate suggest that this serves as a powerful positive feedback, more than doubling the sensitivity of the surface temperature to anthropogenic greenhouse gas forcing. Regional moistening trends in the lower atmosphere have been observed since the mid-1970s from balloon-borne measurements and are strongly linked to changes in surface temperature. Upper atmospheric trends are, however, difficult to assess from conventional observing systems, such as balloon-borne sensors, due to fundamental limitations of observing capabilities. New CCSP research has used data from the High Resolution Infrared Radiometer Sounder (HIRS) and Microwave Sounding Unit (MSU) satellite instruments to evaluate upper atmospheric water vapor and temperature trends since 1979. The observed trends are consistent with those obtained from global climate model simulations; both indicate moistening over this period. The observed results are starkly different from what would be expected in a "no moistening" scenario, which would have greatly reduced the magnitude of model estimates of future global warming. This study provides strong evidence supporting the capability of current climate models to simulate this important climate process, and adds to the credibility of climate model projections of future global warming.

In other CCSP research, simultaneous observations of water vapor in the upper atmosphere and cloud ice from the Microwave Limb Sounder (MLS) on the NASA Aura satellite have provided new evidence for another kind of positive feedback through convective cloud-induced enhancement of the greenhouse effect in the tropics. The work shows that when sea surface temperature exceeds 27°C (about 300 Kelvin, see Figure 1), water evaporated from the warm surface is carried to the upper atmosphere through the formation of towering cumulus clouds. Ice particles in the upper levels of these clouds eventually evaporate, leaving increased water vapor concentrations in the upper atmosphere. Analyses indicate that this cloud-induced

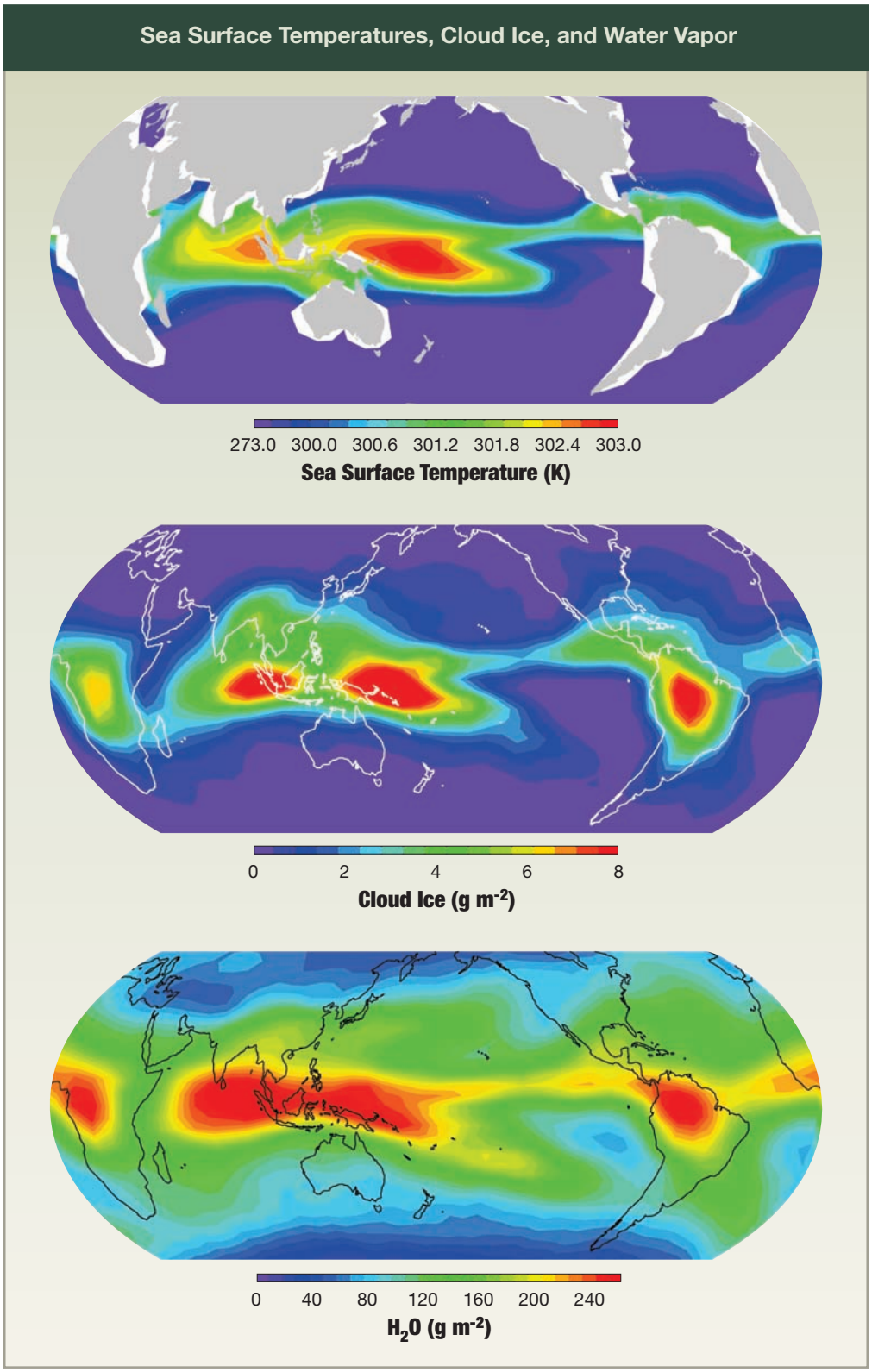


Figure 1: Sea Surface Temperatures, Cloud Ice, and Water Vapor. Observations of sea surface temperature [from the National Center for Environmental Prediction (top)], cloud ice [from Aura Microwave Limb Sounder (MLS, middle)], and upper tropospheric water vapor [from Aura MLS (bottom)] averaged for August 2004 through July 2005. These products illustrate increasing sea surface temperature associated with increasing convective clouds and increasing upper tropospheric water vapor. This convective cloud-induced moistening of the upper troposphere leads to an enhanced greenhouse effect due to water vapor—three times larger than without convection—exacerbating the impact of global warming. *Credit: H. Su, California Institute of Technology/ Jet Propulsion Laboratory.*

moistening of the tropical upper troposphere leads to an enhanced positive water vapor feedback that is about three times larger than what would be expected in the absence of the clouds. Figure 1 illustrate these relationships. These results indicate that convection is an important, if not dominant, process in the previously identified tropical "super greenhouse effect."

A 350-Year Atmospheric History of a Climate-Related Trace Gas.[5] CCSP scientists have published a 350-year atmospheric history of the most abundant sulfur-containing gas in the lower atmosphere, carbonyl sulfide (COS). The research provides the longest atmospheric record of COS to date and shows how the atmospheric abundances of this gas have increased and decreased with industrial sulfur emissions (see Figure 2). Carbonyl sulfide is a relatively long-lived sulfur trace gas with both natural and anthropogenic sources (the latter arising from a variety of industrial processes). Sulfur in COS accounts for about 50% of the sulfate aerosol layer, the atmosphere's layer of sulfate-containing fine particles, during non-volcanic periods. This layer affects depletion of the ozone layer as well as heating and cooling of the atmosphere and hence climate. CCSP researchers analyzed Antarctic air trapped in ice cores and consolidated snow that provided a record of COS concentrations back to about the year 1650. These data suggest that atmospheric amounts of COS increased substantially during the Industrial Revolution, but decreased by about 10% in recent years as global industrial emissions of sulfur declined. Given that a close relationship exists between the atmospheric history of COS and global industrial sulfur emissions,

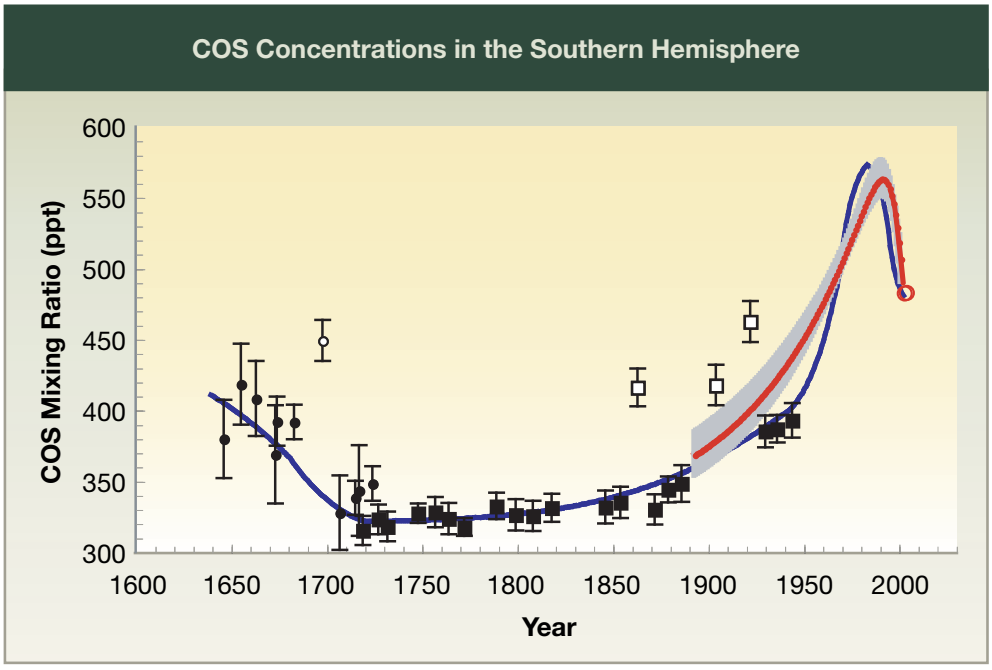


Figure 2: COS Concentrations in the Southern Hemisphere. The blue line represents a history of COS concentrations in the Southern Hemisphere. Note the rather significant increase in concentration coincident with the onset of global industrialization, and the decrease more recently as global sulfur emissions have declined. This history was derived from measurements of COS trapped in ice at Siple Dome, Antarctica (black points) and of COS trapped in the snowpack at the South Pole (red line with grey error bounds). Modern-day measurements from the South Pole and Tasmania are indicated with red circles. Open circle and squares were not used in the calculation because meaningful dates could not be assigned. *Credit: S.A. Montzka, NOAA/ Earth System Research Laboratory.*

the CCSP research suggests that human activities may have contributed to long-term changes in the atmosphere's sulfate aerosol layer.

Assessing and Improving the Inventories of Trace Gas Emissions from North America.[7,8] The CCSP community has made substantial progress in appraising the understanding of North American emissions of trace gases that are precursors for the formation of aerosols and ozone, both of which have implications for climate as well as for air quality in the lower atmosphere. The North American Research Strategy for Tropospheric Ozone (NARSTO) Emission Inventory Assessment, published in FY 2005, is a detailed examination of current approaches to measuring, inventorying, and modeling emissions of trace gases and an assessment of potential improvements to those approaches. Findings of particular note regarding the U.S. National Emission Inventory for mobile sources included: (i) carbon monoxide emissions are about half as large as earlier estimates suggested; and (ii) nitrogen oxide emissions increased throughout the 1990s, contrary to previous assumptions. Accurate estimates of emissions are needed to understand the quantity of aerosols, ozone, and their precursors that are exported from North America to the global atmosphere. Related CCSP research has examined the chemistry of the atmosphere in the New England region, especially with respect to pollutant emissions that affect ozone and aerosols. Findings show a different mix of volatile organic compounds than had been expected; the improved information will lead to a more accurate picture of the chemistry of continental air masses. The New England field research also documented the transport and transformation of aerosols and their associated radiative heating effects as they were carried downwind from their North American sources. This work will enable scientists to reduce the uncertainty in estimates of the effects of aerosols on climate.

Remote Sensing Measurement-based Assessments of Aerosol Direct Radiative Forcing.[14] Greenhouse gases and aerosol particles influence the climate system. While increasing greenhouse gases warm the climate system, aerosol particles have a diverse set of both cooling and warming influences on climate. Current scientific assessment finds that the net effect of aerosols is mostly to cool the climate system, temporarily reducing the impact of greenhouse gases on warming. Even though the greenhouse-gas warming is larger than the aerosol cooling, the uncertainty in the aerosol effect is five times larger than the uncertainty in the greenhouse effect. Reducing these uncertainties is critical to understanding of the climate system and its future change.

Aerosols have both direct and indirect cooling effects on climate. The direct cooling effect is due to the aerosol particles' reflection of sunlight to space. Indirect effects include aerosol-induced changes in cloud properties, cloud distribution, and precipitation that can cool the Earth by enhancing cloud reflectivity. Past assessments of the direct

effect of aerosols on climate were conducted through modeling efforts. Now, CCSP research has assessed the direct impact of aerosols on climate through global measurements. New satellite sensors introduced in the last decade, as well as the Aerosol Robotic Network (AERONET), a global network of ground-based remote-sensing instruments, have provided the opportunity to make a measurement-based assessment of the aerosol direct effect on climate. A major scientific synthesis and assessment of the aerosol direct effect on climate has been completed, published, and served as a major resource for Working Group I of the Intergovernmental Panel on Climate Change (IPCC) in the preparation of its Fourth Assessment Report. The paper reviews recent progress in characterizing aerosols and assessing the aerosol direct effect, focusing on measurements. High-accuracy satellite measurements of aerosol properties have made it feasible to obtain observational constraints for the aerosol direct forcing, especially over the global ocean. Measurements show that aerosols in total reflect about 10% of incoming sunlight to space, much more than the models had indicated. The human contribution to the global aerosol concentrations suggested by the models agrees with the measurement assessment to within measurement accuracy and is on order of approximately 20%.

This review also identifies several issues that require significant future research efforts. Estimates of aerosol forcing over land are less well constrained than over ocean. Uncertainties in estimates of aerosol forcing are also larger on regional scales than on a global scale. The aerosol forcing under cloudy conditions remains relatively unexplored and quite uncertain. In addition, knowledge of the much more complex and probably more important aerosol indirect effects that modify cloud properties and abundance is much less certain and near-term interagency plans reflect the high priority placed on improved characterization of these aerosol-cloud interactions.

Improved Estimates of Organic Aerosols in the Lower Atmosphere.[4] Atmospheric aerosols are involved in issues ranging from air quality to climate change. Recent airborne measurements indicate that computer model simulations of aerosol composition severely underestimate the actual concentrations of organic aerosols in the free troposphere. The Asia-Pacific Regional Aerosol Characterization Experiment (ACE-Asia) was conducted in April and May of 2001. This large field campaign included two aircraft focused on sampling Asian aerosols transported over the Northwest Pacific Ocean. Three different teams of scientists made measurements of organic aerosols in the troposphere, from the surface to around 6.5 km. Results from the groups were very similar and showed that above 2 km, the concentrations of organic carbon were 10 to 100 times higher than the computer models predicted. However, the models were able to replicate adequately the concentrations of sulfate and elemental carbon, other aerosols that were also measured from the aircraft. The scientists believe that the underestimation of organic carbon by the models is due to

secondary organic aerosols, which are formed in the troposphere from the oxidation of volatile organic compounds. They are long-lived and can therefore play a large role in intercontinental pollution transport and radiative forcing of climate.

Relative Humidity and the Influence of Atmospheric Particles on Climate.[1,9,10] CCSP researchers have conducted several studies of how atmospheric humidity affects aerosols and the implications for the climate system. The warming and cooling effects of aerosol particles are among the most uncertain of the influences on climate. Many factors affect how aerosol particles interact directly with light, including the relative humidity of the atmosphere, which can affect the size of the particles. CCSP researchers have conducted several studies that show that the chemical composition of the aerosol is the overriding factor in how size changes with relative humidity. Aerosols that are largely inorganic in composition, such as those containing sulfate or sea salt, tend to take up water and grow larger in more humid atmospheres. Consequently, they scatter more light and hence have a larger climate cooling influence (shown schematically in Figure 3). On the other hand, aerosols that are primarily organic are more hydrophobic, and their interaction with light does not change much as the relative humidity of the atmosphere varies. CCSP research spanned a full range of approaches, including fundamental laboratory studies, analysis of extensive field measurements of aerosols in regions of Asia and North America, and modeling investigations. The work has clearly demonstrated that not all aerosols are alike when it comes to their effects on climate. Because aerosol composition was shown to be such a major factor, one implication is that the climate effects of aerosols will change as the aerosol ages and its chemical composition changes. Climate effects therefore

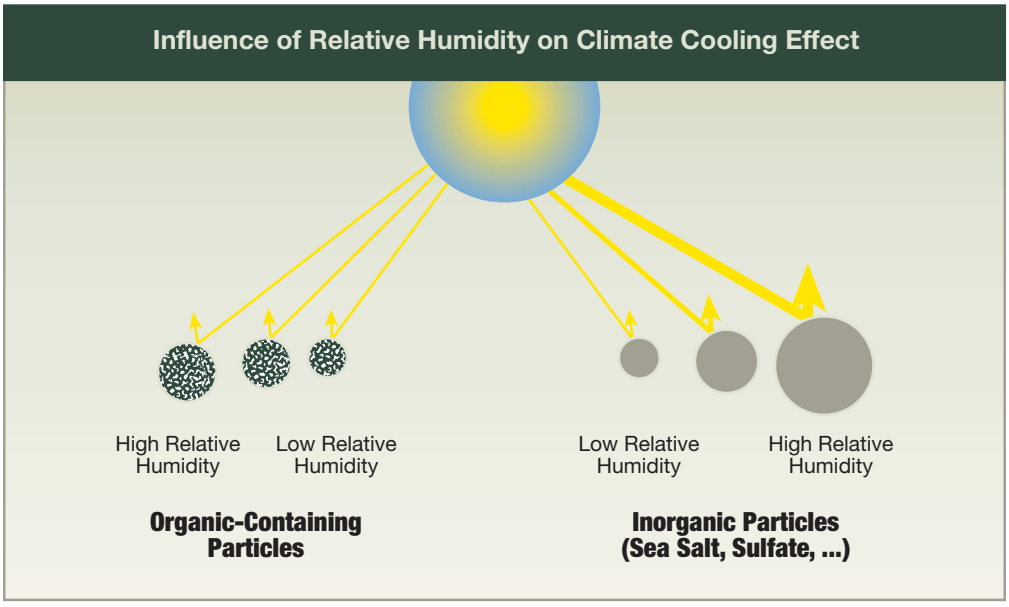


Figure 3: Influence of Relative Humidity on Climate Cooling Effect. Schematic contrasting how relative humidity influences the climate cooling effect of inorganic versus organic aerosol particles. *Credit: A.R. Ravishankara, NOAA/Earth System Research Laboratory.*

evolve as aerosol particles are transported from their original source. The research has led to more accurate representations of aerosol interactions in climate radiative calculations and will thereby improve the accuracy of estimates of the direct climate forcing by aerosols.

Cloud- and Aerosol-Related Fundamentals Elucidated.[6] The interactions between water vapor and condensed forms of water in the atmosphere underlie many of the physical processes that govern cloud formation. Cirrus clouds, polar stratospheric clouds, and the large volume of the atmosphere that has a temperature below freezing are key climate-relevant phenomena that depend on the fundamental physical properties of water vapor, liquid water, and ice. Furthermore, the formation of aerosols and the chemical processes within them are affected by heretofore poorly understood processes specific to water that is supercooled below the freezing point. CCSP research has resulted in a comprehensive review of the fundamental physical processes at the vapor/water/ice interface and an advancement of the parameterization of those processes that are most relevant to atmospheric composition and climate. The work provides a first look at the full range of temperature conditions relevant to the atmosphere, yielding new perspectives on processes pertinent to colder regions, such as the Antarctic stratosphere, that had not been previously considered. Various structural forms of liquid water and solid ice are shown to behave differently, thus research has highlighted the need to represent these aspects of real-world behavior in models of atmospheric chemistry and climate. In addition, the work has implications for methodologies used to calculate relative humidity in laboratory applications and in the real atmosphere.

Smoke Suppression of Clouds: A Climate-Related Effect of Aerosol Particles.[3] Improved modeling capabilities have enabled CCSP researchers to simulate smoke-cloud-surface interactions in biomass-burning regions and to study the mechanisms by which smoke suppresses cloud formation.

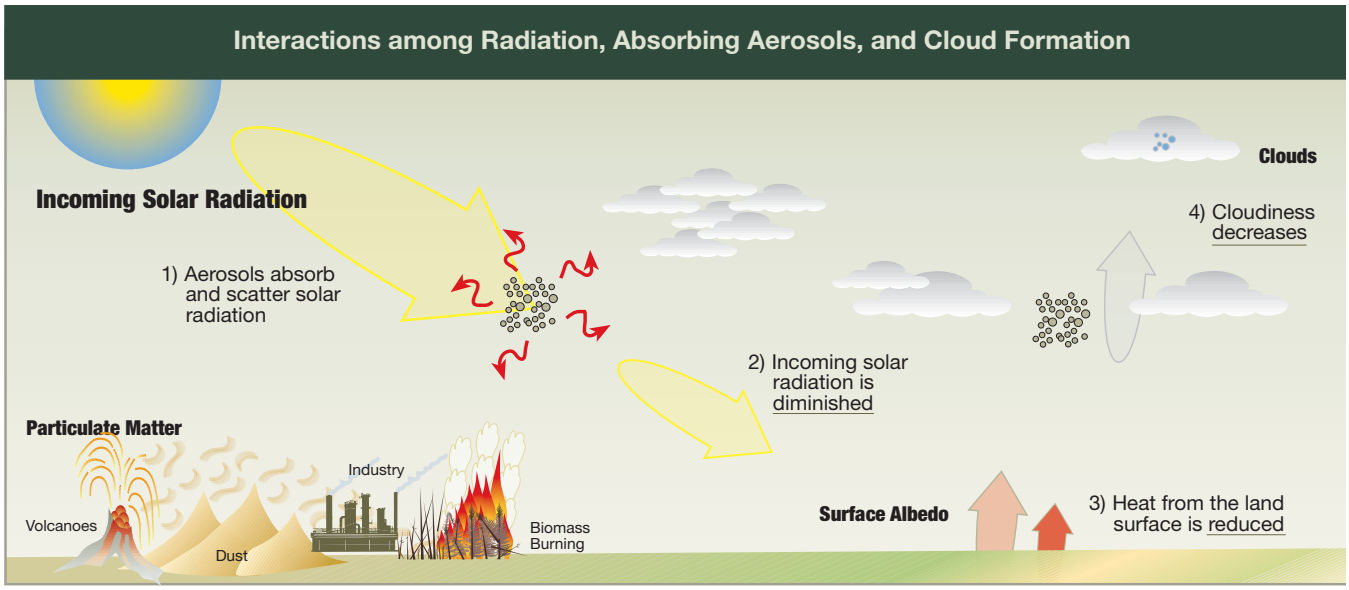


Figure 4: Interactions among Radiation, Absorbing Aerosols, and Cloud Formation. Schematic showing the interactions among radiation, absorbing aerosol particles in smoke, and cloud formation. *Credit: G. Feingold, NOAA/Earth System Research Laboratory.*

The model includes unprecedented treatment of coupled smoke aerosol-cloud-radiative feedbacks in three-dimensional simulations of biomass-burning regions of Amazonia. Absorbing aerosol particles, such as smoke particles, can affect cloudiness by modifying heating rates and atmospheric stability, by modifying how much cloud droplets are heated, or by modifying heat flows from the land surface. CCSP research has evaluated the relative importance of these processes in continental areas, demonstrating that the primary reason for smoke suppression of clouds is the reduction of surface heat flow caused by the presence of absorbing smoke particles (shown schematically in Figure 4). Further, the change in surface heat flow may cause changes in climate-relevant cloud properties, such as cloud amounts. However, the ultimate effect on cloudiness is case-specific and is highly dependent upon cloud type, smoke quantity and properties, location of the smoke within the atmosphere and relative to the clouds, and the degree to which surface heat flows are reduced. The research has helped to quantify a poorly understood aspect of the influence of atmospheric aerosol particles on the climate-related phenomena of cloudiness and surface heating.

Ozone Layer Shows Signs of Recovery.[11] New CCSP research has found that the atmosphere's protective ozone layer is no longer decreasing over much of the globe. The work is based on several different satellite records and surface monitoring instruments. Previous studies have shown that ozone in the topmost layer of the atmosphere may have stopped declining. However, very little ozone is in these top layers. The latest work focuses on the thickness of the entire ozone column above the Earth's surface,

and therefore has relevance to the amount of harmful ultraviolet radiation reaching the surface of the Earth. Using satellite-derived estimates of ozone levels, as well as ground-station data from North America, Europe, Hawaii, Australia, and New Zealand, researchers found that total column ozone amounts over the southern mid-latitudes have stopped declining and leveled off, while amounts over the mid- and high latitudes of the Northern Hemisphere have increased since 1996. While the study indicates the beginning stages of improvement in the ozone layer, ozone amounts are still lower than those observed 25 years ago and full recovery lies decades in the future.

Highlights of Plans for FY 2007

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 2007 follow.

Climate and Ozone Layer Assessments. CCSP research and leadership are playing a substantial role in international efforts to assess the state of scientific understanding with regard to climate and the ozone layer. Major preparation stages of the IPCC's Fourth Assessment Report are occurring during 2006, with many CCSP scientists serving as authors and reviewers; the United States provides the co-chair of IPCC's Working Group I and hosts its Technical Support Unit. In addition, the international assessment of ozone layer depletion was drafted and is being reviewed during 2006. Publication of both the climate and ozone layer assessments will occur in FY 2007.

These activities will address Questions 3.1, 3.2, 3.4, and 3.5 of the CCSP Strategic Plan.

Climate Field Study in the Texas/Gulf of Mexico Region. In August and September of 2006, CCSP researchers in several agencies, institutions, and academia led and participated in a field program that investigated important scientific questions that are common to both climate and air quality. The Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS) component will focus on providing a better understanding of the sources and atmospheric processes responsible for the formation and distribution of ozone and aerosols in the atmosphere and the influence that these species have on regional and global climate. Analyses will continue in FY 2007.

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

Evaluating the North American Contributions to "Greenhouse" Ozone. CCSP research will analyze ozone profiles measured across mid-latitude North America during a 2004 climate field study, with the aim of estimating how much

ozone forms over the continent in the summer. Further studies will elucidate the export of ozone from North America to the North Atlantic Ocean and Europe. Ozone in the lower atmosphere is a greenhouse gas that affects climate because it absorbs infrared radiation.

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

Analysis of Ozone and Ozone-Depleting Substances Data Sets for Trends and Ozone-Layer Recovery Detection. Decisions over the last 2 decades with regard to the protection of the stratospheric ozone layer have resulted in declining use of ozone-depleting substances (ODS). An important focus now is documenting and understanding how the atmosphere is responding to those decisions, both with regard to ODS abundances (which in general are in decline) and the ozone layer itself. Satellite and ground-based data sets have established a multi-decadal record that will be increasingly scrutinized for signs of recovery of the ozone layer via analyses of ozone and ODS.

These activities will address Question 3.4 of the CCSP Strategic Plan.

Oklahoma City Field Study. A field campaign to study North American aerosols involving CCSP scientists from multiple agencies is planned for the summer of 2007. Anthropogenic aerosols are relatively short-lived in the atmosphere and therefore have a high degree of variability over space and time. For this reason, a large contrast in aerosol properties is expected to be found between the upwind and downwind regions of their urban source area. The campaign will investigate the role of such differences for a mid-size, mid-latitude metropolitan area in North America and the consequences for the warming and cooling effects of aerosols. In addition, the campaign will evaluate changes to aerosols as they move through relatively simple fair-weather cumulus systems found over much of mid-latitude North America during the summer months.

This activity will address Question 3.1 of the CCSP Strategic Plan.

Megacity Initiative: Local And Global Research Observations (MILAGRO). The goal of the MILAGRO project is to characterize emissions, chemical transformations, and aerosol and pollutant outflow from Mexico City, the world's second largest megacity. In 2007, CCSP scientists supported by NSF, DOE, and NASA will analyze and evaluate data collected during the 2006 campaign. Field measurements involved numerous ground sites and research aircraft, which along with satellite observations provide information at multiple spatial scales, ranging from the urban Mexico City "super site" to larger areas extending downwind into the Gulf of Mexico. U.S. scientists together with university and government scientists from Mexico will review MILAGRO data and analysis plans at a workshop in Boulder, Colorado, in FY 2007. The data will enable characterization of the evolution of the Mexico City plume, including changes in reactive trace gas concentrations; the evolution of chemical, physical, optical, and cloud nucleating properties of aerosols; and the interactions between gases and aerosols. These measurements and analyses should prove valuable for developing and testing models of aerosol evolution.

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

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. The lidar instrument aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite, launched 28 April 2006, promises to revolutionize knowledge of aerosol vertical distribution and greatly expand knowledge of the effects of aerosols in the lowermost atmosphere. 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 (e.g., how they scatter versus absorb light), and to help separate anthropogenic from natural components. Further advances in the near term are also expected from multi-angle, ultraviolet, and polarization-based techniques for measuring the amount of aerosols in the atmosphere over land surfaces.

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

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