Global Air Quality: An Imperative for Long-Term Observational Strategies

Committee on Atmospheric Chemistry, Board on Atmospheric Sciences and Climate, National Research Council

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Committee on Atmospheric Chemistry
Board on Atmospheric Sciences and Climate
Division on Earth and Life Studies
National Research Council

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Preface

The National Research Council's Committee on Atmospheric Chemistry (NRC/CAC) was established to serve as a focal point for NRC activities on issues related to atmospheric chemical change and its impacts on air quality, climate, stratospheric ozone depletion, and other related issues. The committee consists of 12 members with expertise covering the areas of tropospheric and stratospheric chemistry; urban/regional air pollution; modeling of climate, chemistry, and atmospheric dynamics; *in situ* and remote sensing observational systems; and interfaces of science and public policy.

This CAC study was motivated by a concern that, in the coming decades, dramatic increases in global population and urbanization levels, as well as changes in global climate, may significantly affect air quality over large regions of the globe. The charge to the committee was to examine the linkages among regional/global changes in atmospheric composition, climate change, and air quality. In particular, their task was to:

- review projections of how future global industrialization, urbanization, and other human activities could alter regional and global atmospheric composition and chemistry over the next century;
- discuss how these global chemical changes could affect atmospheric radiative forcing and impact U.S. air quality;
- describe the gaps in scientific understanding that currently impede our ability to accurately assess such changes;
- identify the observational data that are most critical for detecting and documenting these chemical changes, characterize the capacity of current

observational systems to collect such data, and suggest ways that these systems can be enhanced or better integrated to provide the necessary observations;

- identify any other research needs related to this issue and, where appropriate, discuss what types of institutional arrangements (on both the national and international level) would be most effective for carrying out this work.

As the title of the report implies, the committee determined that a top priority in the study of global air quality change is the need to strengthen our capacity for sustaining long-term observations of atmospheric composition. However, the committee did not attempt to make highly detailed recommendations about specific observations that should be made (for instance, by suggesting locations and frequencies of measurements for particular chemical species). Determining these types of details is a research challenge requiring in-depth analyses that are beyond the committee's scope. Similarly, this report does not provide a comprehensive literature review on all the topics of relevance to this study; rather, the publications cited in the report were chosen only as useful examples or sources of further information for the reader.

In developing the goals of this study, and in formulating their conclusions and recommendations, the CAC members sought input from a wide variety of sources including members of the NRC Board on Atmospheric Sciences and Climate; representatives of federal agencies including the National Aeronautics and Space Administration, the U.S. Environmental Protection Agency, the National Oceanic and Atmospheric Administration, the National Science Foundation, and the Department of Energy; leaders of international scientific organizations including the Intergovernmental Panel on Climate Change, the International Global Atmospheric Chemistry project, the organization on Stratospheric Processes and their Role in Climate; and numerous individual scientists who are involved in research related to this topic. The committee extends its gratitude to all of the individuals who participated in the CAC meetings and who provided background information for this study. The committee is also grateful to the NRC staff who provided invaluable input for developing this study and preparing this report. This project was supported by the U.S. Global Change Research Program, the National Aeronautics and Space Administration, and the U.S. Environmental Protection Agency.

Contents

Executive Summary

Human industrial activities including the combustion of fossil fuels, and land-use activities including biomass burning and agriculture, lead to the emission of gases and particles that perturb atmospheric composition in numerous ways. One such perturbation is the build-up of long-lived greenhouse gases including carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and chlorofluorocarbons (CFCs). The scientific community has made considerable efforts to document the long-term atmospheric trends for these species and to assess how these changes will affect global climate in the coming years. Less well documented are changes in the atmospheric concentration and distribution of shorter-lived species including nitrogen oxides (NO_x), volatile organic compounds (VOCs), carbon monoxide (CO), sulfur dioxide (SO_2), ozone (O_3, which is formed in the troposphere from chemical reactions involving NO_x and VOCs) and airborne particulate matter (PM, which encompasses a diverse class of chemical species including sulfates, nitrates, soot, organics, and mineral dust).

Air pollution is generally studied in terms of immediate local concerns rather than as a long-term "global change" issue. In the coming decades, however, rapid population growth and urbanization in many regions of the world, as well as changing climatic conditions, may expand the scope of air quality concerns by significantly altering atmospheric composition over broad regional and even global scales. Ozone and PM are of particular concern because their atmospheric residence times are long enough to influence air quality in regions far from their sources and because they also contribute to climate change. Our ability to understand observed changes in global air quality and to accurately predict future changes will depend strongly on answering two important questions:

- *How can global air quality change affect, and in turn be affected by, global climate change?*

  Although air quality and climate are generally treated as separate issues, they are closely coupled through atmospheric chemical, radiative, and dynamical processes. The accumulation of pollutants in the atmosphere can affect climate through direct and indirect contributions to earth's radiative balance, and through chemical reactions that alter the lifetime of certain greenhouse gases. In turn, meteorological parameters such as temperature, humidity, and precipitation can affect the sources, chemical transformations, transport, and deposition of air pollutants. Our understanding of many of these climate-chemistry linkages is in its infancy. A better understanding is needed in order to make accurate estimates of future changes in climate and air quality and to evaluate options for mitigating harmful changes.

- *How is global air quality affected by the international and intercontinental transport of air pollutants?*

  Total global emissions of species including NO_x, VOCs, and CO may rise dramatically in the coming decades due to increasing population and industrialization, and in particular, the growth of "megacities" in many regions of the world. The transport of pollutants such as ozone and PM across national boundaries and between continents will increase in importance as total emissions rise. Such pollutant transport connects all the countries of the world to varying degrees and can raise "background" pollution levels over large regions of the globe. Quantifying this long-range transport is essential in order to understand what future changes may occur in U.S. air quality, to assess how U.S. pollutant emissions affect other regions of the world, and to develop realistic and effective air quality management plans for the coming decades.

Addressing these complex questions about global air quality change will require a comprehensive research strategy that integrates atmospheric observations covering a wide range of spatial and temporal scales together with diagnostic, global, and regional models. Other key elements in this research framework include inventories of pollutant emissions, meteorological data to describe atmospheric conditions and transport, laboratory measurements to characterize important chemical reactions, and process studies to provide detailed understanding of complex chemical and dynamical phenomena.

Some components of this research framework are in a more mature state than others. For instance, significant progress is being made in the development of regional/global chemical transport models and their integration with global climate models. Likewise, in recent years the atmospheric chemistry community has organized numerous field campaigns that combine model analyses with in-

tensive observations from a variety of platforms, which has enhanced our understanding of some complex chemical and radiative processes.

In contrast, we currently do not have the capacity to observe many important medium- and long-term changes (that is, changes occurring over the course of years to decades) in the chemistry and composition of the lower atmosphere. If these observational capabilities are not strengthened, this will greatly limit our ability to document the evolution of the atmosphere in the coming decades. This also limits the value of the developments cited above, since a strong observational base is needed to test and improve model predictions, and to provide a longer-term context for the observational "snapshots" obtained through intensive field campaigns. A range of observational platforms and techniques will be needed to provide measurements at the earth's surface and in the free troposphere several kilometers above the surface. Satellite measurements ultimately hold the greatest promise for comprehensive global observations in the lower atmosphere, but these observational techniques are still largely in the developmental stage. Obtaining global coverage through ground-based and other *in situ* observations will require that similar measurements be made by numerous international scientific groups, with careful calibration and intercomparison of different measurement systems.

The following are the committee's conclusions and recommendations:

Key findings:

Current observational systems are not adequate for characterizing many important medium- and long-term global air quality changes. Some particularly notable weaknesses in our current observational capabilities include the lack of (i) long-term measurements of reactive compounds and PM, (ii) methods for obtaining vertical profile data, and (iii) measurement sites that allow for a meaningful examination of long-range transport and trends in background concentrations.

The global air quality issues discussed in this report intersect with the concerns of several federal agencies, yet none of these agencies have a clear mandate to lead U.S. research efforts or maintain the long-term observational programs that are needed to address these issues.

Recommendations:

- Maintain and strengthen the existing measurement programs that are essential for detecting and understanding global air quality changes. High priority should be given to programs that aid in assessing long-term trends of background ozone and PM.

- Establish new capabilities to provide long-term measurements and vertical profiles of reactive compounds and PM that will allow meaningful examination of long-range transport and trends in background concentrations.

  These two recommendations will require providing support to:

  - develop uniform and traceable standards, on a global basis, for calibration of both gas-phase and aerosol measurements;

  - improve measurement technologies for use in current observational platforms (such as ground-based air quality monitoring networks, commercial aircraft, and balloons/sondes), and in new potential platforms such as "supersites" for measuring a comprehensive suite of compounds in remote locations and unmanned aerial vehicles for long-duration sampling of the atmosphere over a wide range of altitudes;

  - integrate measurements obtained from different observational programs and platforms, with a particular focus on integrating remotely-sensed satellite observations with *in situ* aircraft and ground-based observations;

  - promote observational programs specifically designed to address chemical and meteorological data requirements for the development and evaluation of models.

- Responsibility for carrying out this work should be clearly assigned to a U.S. federal agency (or interagency) research program, and the United States should play a leadership role in fostering international cooperative research and observational activities to enhance our understanding of global air quality changes.

1

Introduction

Human activities are changing the composition of the earth's atmosphere in numerous ways. For instance, since the beginning of the industrial era, the atmospheric abundances of the gases carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) have been increasing due to the combustion of fossil fuels and other industrial activities, and due to land-use activities such as biomass burning and agriculture. Once released, these gases remain in the atmosphere up to many decades, and as greenhouse gases (GHGs), their increasing abundances contribute to global climate change.

Significant increases in the emissions of some short-lived gases have also occurred in many regions. Important examples include nitrogen oxides (NO_x), volatile organic compounds (VOCs), carbon monoxide (CO), and sulfur dioxide (SO_2). The background concentration of tropospheric ozone (O_3), which is formed in the lower atmosphere from chemical reactions involving NO_x and VOCs, has approximately doubled over the past century. Human activities have also increased airborne particulate matter (PM), which encompasses a diverse class of chemical species including sulfates, nitrates, soot, organics, and mineral dust. These gases and particles are relatively short-lived, remaining in the atmosphere for only days to months near the surface. Ozone and PM are of particular concern because their atmospheric residence times are long enough to influence air quality in regions far from their sources and because they also contribute to climate change.

Changes in global air quality are determined by the release of key chemical compounds from source regions, and by the subsequent accumulation and interaction of these species throughout the earth's atmosphere. Our ability to under-

stand observed changes in global air quality and to predict future changes will depend strongly on answering two important questions:

- *How can global air quality change affect, and in turn be affected by, global climate change?*

  Although air quality and climate are generally treated as separate issues, they are closely coupled through atmospheric chemical, radiative, and dynamical processes. The accumulation of pollutants in the atmosphere can affect climate through direct and indirect contributions to earth's radiative balance, and through chemical reactions that alter the lifetime of certain greenhouse gases. In turn, meteorological parameters such as temperature, humidity, and precipitation can affect the sources, chemical transformations, transport, and deposition of air pollutants. Our understanding of many of these climate-chemistry linkages is in its infancy. A better understanding is needed in order to make accurate estimates of future changes in climate and air quality and to evaluate options for mitigating harmful changes.

- *How is global air quality affected by the international and intercontinental transport of air pollutants?*

  Total global emissions of species including NO_x, VOCs, and CO may rise dramatically in the coming decades due to increasing population and industrialization, and in particular, the growth of "megacities" in many regions of the world. The transport of pollutants such as ozone and PM across national boundaries and between continents will increase in importance as total emissions rise. Such pollutant transport interconnects all the countries of the world to varying degrees and can raise "background"[1] pollution levels over large regions of the globe. Quantifying this long-range transport is essential in order to understand what future changes may occur in U.S. air quality, to assess how U.S. pollutant emissions affect global air quality, and to develop realistic and effective air quality management plans for the coming decades.

Answering the questions posed above requires a research strategy that integrates atmospheric observations covering a wide range of parameters and spatial and temporal scales together with diagnostic, global, and regional models (see Figure 1-1). In particular, a renewed and comprehensive observational strategy with global cooperation is required in order to fill the significant gaps and shortfalls in our current observational capabilities. If we do not maintain existing

[1] "Background" is used here in a narrow sense as the concentration of a pollutant that would prevail in the absence of local anthropogenic emissions.

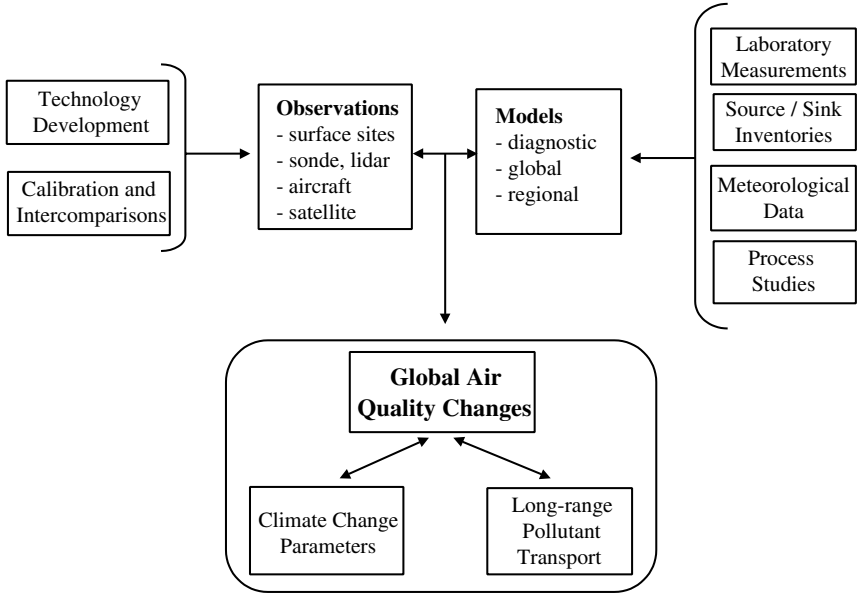

FIGURE 1-1 Schematic of the research framework needed to address questions about global air quality changes.

observational programs and develop strategies to enhance our observational capabilities, we will lose critical information about the state of the atmosphere as it evolves in this period of rapid population growth and industrialization. Predictions about air quality changes in the 21st century are highly uncertain, in part because of the limited observational record of changes in the atmospheric concentration of short-lived gases and aerosols since the pre-industrial era. Recent technical advances have greatly enhanced the possibilities for obtaining needed observations; the challenge now is to make a national commitment to developing and maintaining comprehensive observational programs that will utilize these technologies.

To address these global air quality issues, a range of observational platforms and techniques will likely be needed, including measurements at the earth's surface and in the free troposphere several kilometers above the surface (which can be accessed through platforms such as aircraft, balloons, and satellites). Satellite measurements ultimately hold the greatest promise for comprehensive global observations in the lower atmosphere, but these observational techniques are still largely in the developmental stage. Obtaining global coverage through ground-based observations will require that similar measurements be made by a wide variety of international scientific groups. Careful calibration and intercom-

Box 1-1
Global Air Pollution Concerns

As urbanization and industrialization have intensified during the last few decades, urban air quality has become an increasingly pressing public health concern throughout many regions of the world, particularly in developing countries. More than 3 billion people—about half the world's population—are now concentrated in urban areas, and by 2010 the global urban population is expected to swell to more than 4 billion (UNEP, 1999).

By one estimate, more than 1 billion people are currently exposed to harmful levels of air pollution (Schwele, 1995). Severe particulate air pollution is a chronic problem in much of Asia, primarily as a result of coal combustion in factories and power plants, and the use of coal and wood for cooking and home heating. Motor vehicles are an increasingly important contributor to air pollution in much of the world, with more than 600 million vehicles in use, a number that could double within the next 25 years (Dunn, 1996). Automobiles are the dominant source of air pollution in many Latin American cities, including Sao Paulo, Santiago, and Mexico City, where they have had to restrict automobile use in an effort to manage severe air pollution episodes (UNEP, 1999).

U.S. air quality, as measured at thousands of monitoring stations across the country, has shown steady improvement over the past 20 years due in part to the implementation of air quality regulatory programs and cleaner technologies for motor vehicles and stationary pollution sources. There are, however, still many areas of the country that are not in compliance with ambient air quality standards for pollutants such as ozone and PM (EPA, 1999).

parison of measurement systems will be required to ensure the value of these combined data sets.

Atmospheric models are also a key element in the research framework for addressing global air quality changes, as they represent the primary tool for forecasting the future state of the atmosphere. Critical sources of input for models include inventories of pollutant emissions, meteorological data to describe atmospheric conditions and transport, laboratory measurements to identify and describe important chemical reactions, and process studies that provide detailed understanding of specific aspects of the atmosphere's complex and interactive chemistry and transport. Observational data sets are necessary for judging whether model simulations are representative of current or past atmospheric conditions. Without this type of evaluation, our understanding will be incomplete and the value of model predictions will remain highly uncertain.

For all of these reasons, a commitment to long-term observational strategies emerges as imperative for understanding global air quality changes in the 21st century. The United States needs to play a leadership role in such a commitment because of our advanced technological capabilities and large contributions to global pollution emissions. Without such a commitment, we will critically limit

our understanding of the current state of the atmosphere and our ability to assess possible future changes. The remainder of this report focuses on issues related to the key research objectives discussed above, specifically:

- Chapter 2 contains a review of the interactions between climate change and atmospheric chemical change.
- Chapter 3 contains a review of observational and modeling studies of intercontinental pollution transport.
- Chapter 4 contains an analysis of our current capabilities for observing atmospheric chemical changes related to global air quality concerns.
- Chapter 5 contains the committee's key findings and recommendations.

Throughout these chapters, a series of text boxes are included to provide a broader context for the scientific concerns and imperatives discussed in the main body of the report.

2

Climate, Atmospheric Chemistry, and Global Air Quality

There are numerous linkages among the chemical and physical processes that affect climate change and air quality. Examples of climate/chemistry interactions that must be considered as part of any comprehensive assessment of future atmospheric evolution are discussed in the following list and summarized in Figure 2-1. Some of these linkages have been the subject of intensive research efforts, while others are still largely unexplored and highly uncertain.

Impacts of Climate Change on Tropospheric / Stratospheric Chemistry

- Changes in temperature and humidity patterns resulting from global climate change can directly affect the concentrations of many important tropospheric chemical species. For instance, increases in temperature accelerate the rates of reactions involved in the production of tropospheric ozone (e.g., Johnson et al., 1999). At the same time, increases in atmospheric water vapor can affect both the chemical production *and* the chemical destruction of tropospheric ozone (Stevenson et al., 2000), and thus the net impact of these concurrent changes is difficult to assess.

- The atmospheric accumulation of greenhouse gases has led to cooling of the stratosphere. This, in turn, may prolong the recovery of the stratospheric ozone layer, since colder winters and more stable polar vortices generally increase polar ozone losses (Shindell et al., 1998). This may also affect stratospheric circulation and alter the lifetime of greenhouse gases such as N_2O and CFCs, which are destroyed primarily in the strato-

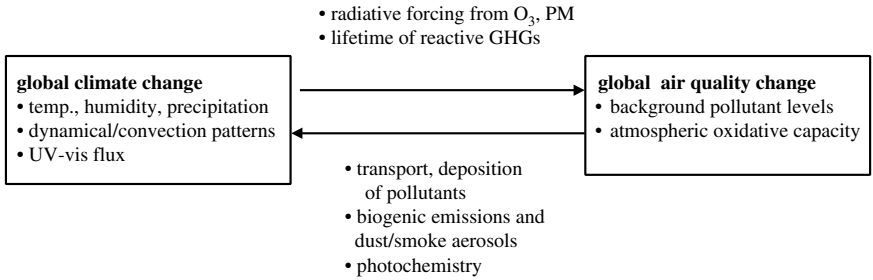

FIGURE 2-1 A schematic summary of the feedbacks between climate change and air quality change.

sphere. Depressed stratospheric ozone levels increase the flux of ultraviolet radiation to the troposphere; under conditions of high ambient NO_x levels, this can increase tropospheric OH/HO_2 and lead to increased tropospheric ozone production.

- Within the troposphere, changes in the vertical temperature structure may affect the evolution and dynamics of the boundary layer and associated mixing processes that influence the dispersion and transport of air pollutants. Changes in the thermal structure of the troposphere and stratosphere may also affect the location of the tropopause and cross-tropopause transport of ozone and its precursors (IPCC, 1996).[1]

Impacts of Climate Change on Emissions of Greenhouse Gases and Aerosol/Ozone Precursors

- Climate can affect the emissions of many biogenic compounds that play an influential role in tropospheric chemistry. For example, changes in temperature, soil moisture, and solar radiation can all lead to changes in the emission of ozone precursors such as isoprene and terpenes (Tingley et al., 1979; Lamb et al., 1985). Changes in temperature could also affect biological activity in the oceans and the resulting emissions of halogenated hydrocarbons such as methyl bromide, and sulfur compounds such as dimethyl sulfide.

[1]This reference is to the IPCC's second assessment report. The IPCC's third assessment report is in press and will be available by late 2001. The Summary for Policymakers is currently available online at www.ipcc.ch.

- Changes in temperature can affect the emission of methane from high-latitude wetlands. As an example, the temporary cooling of the troposphere after the eruption of Mount Pinatubo led to a dramatic decrease in the global methane growth rate, thought to be due in part to a decrease in natural methane emissions (WMO, 1999).

- Widespread climatic changes would likely alter the patterns of human activities such as agriculture, biomass burning, and energy consumption (e.g., demand for heating/air conditioning), and thus would affect the emissions of pollutant gases and particles that result from these activities. Likewise, changes in land-use activities and patterns of fires and droughts could influence the level of smoke and mineral dust aerosols in the atmosphere.

Impacts of Atmospheric Chemical Changes on Climate

- Increases in the atmospheric burden of greenhouse gases (including H_2O, O_3, CO_2, CH_4, N_2O, and CFCs) can change the earth's radiative balance and the temperature structure of the atmosphere. The potential climatic impacts of greenhouse gas radiative forcing are discussed extensively in international assessment reports such as IPCC (1996) and WMO (1999).

- Atmospheric aerosols affect climate by scattering and absorbing ultraviolet and visible (UV-Vis) radiation and by altering the formation processes, optical properties, and precipitation efficiency of clouds. Some studies find that pollution aerosols augment cloud albedo and thus exert a cooling influence (Albrecht, 1989; Pincus and Baker, 1994), while other studies find that the dark haze caused by pollution aerosols absorb solar radiation and reduce certain types of cloud coverage (Ackerman et al., 2000). The net impacts of these forcings are highly uncertain because they are dependent upon aerosol composition, and geographical and vertical distribution (IPCC, 1996; NRC, 1996).

- Large-scale air pollution emissions can significantly affect the concentration of the atmosphere's primary oxidizing agent, the hydroxyl radical (OH). Net changes are difficult to project, however, since increases in NO_x tend to elevate OH levels, while increases in CO tend to decrease OH. Large-scale changes in OH, in turn, affect the lifetime and thus the atmospheric abundance of reactive greenhouse gases such as CH_4, HFCs, and HCFCs.

The preceding list is not a comprehensive description of all the potentially important linkages between climate and atmospheric chemistry; it is only meant

Box 2-1
Policy Responses to Global Climate Change

The Kyoto Protocol to the United Nations Framework Convention on Climate Change, adopted in 1997, includes commitments that would limit the emissions of greenhouse gases from industrialized countries to an average of 95% of 1990 levels during the period 2008 through 2012. If the Protocol is ratified by enough countries to enter into force, it would control emissions of carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF_6). Each Party to the Protocol would have flexibility to select its own mix of control strategies and relative emphasis on each of the controlled greenhouse gases.

CO_2 is the dominant greenhouse gas emitted by most countries, and thus controlling CO_2 emissions is an essential component of any effective strategy for stabilizing radiative forcing. At the same time, non-CO_2 GHGs have much higher global warming potentials per unit mass and are also an important target for emission reductions. Tropospheric ozone and PM (in particular, black carbon aerosols) also contribute to global radiative forcing, but control of these species and their precursors is not currently included in the provisions of the Kyoto Protocol. Ozone and PM could be particularly effective targets for emission control efforts, however, as recently underscored by Hansen et al. (2000), since many countries already have domestic regulations that aim to control these species, and since reducing ozone and PM emissions can not only aid in efforts to control global climate, but also can improve local air quality, health, and agricultural productivity.

to demonstrate the fact that these linkages are complex and involve nonlinear couplings among numerous processes. For example, the atmospheric concentrations and distributions of two important greenhouse gases, O_3 and CH_4, are affected by anthropogenic and biogenic emissions of NO_x, VOCs, and CO, by the tropospheric oxidative capacity (in particular, OH concentrations), and by climatic parameters such as temperature, humidity, and UV flux. Because many of these parameters can change simultaneously, predicting associated atmospheric chemical changes is a complex and scientifically challenging task.

Improving our understanding of the interactions between climate and air quality will depend primarily on developing more sophisticated modeling tools; in particular, it will require the ability to couple local- and regional-scale air quality models (which cover spatial scales of a few hundred meters to hundreds of kilometers) with global-scale climate and chemistry models. In recent years there has been significant progress in developing these types of coupled models. However, one of the primary constraints in developing and testing such models is the lack of comprehensive observational databases, and thus strengthening our long-term observational capabilities emerges as an imperative.

3

The Effects of Changing Global Atmospheric Composition on Air Quality

It has long been recognized that pollution emissions can affect air quality beyond national borders. For instance, field studies have shown that the prevailing westerly winds typically carry ozone and its precursors from the eastern United States into Canada, the North Atlantic, and beyond (e.g., Prados et al., 1999); and in turn, it is known that air masses reaching the United States can carry pollution originating from many other parts of the world. The following is a discussion about some of the current and possible future effects of cross-border and intercontinental transport of air pollutants, with an emphasis on assessing the potential implications for U.S. air quality.

Observations of Current Impacts

Long-range transport of aerosols and trace gases from Asia is known to alter the composition of the remote Pacific troposphere (e.g., Uematsu et al., 1983; Xiao et al., 1997). In spring, when storm and frontal activity in Asia is most prevalent, outflows of continental pollution and dust are observed in both *in situ* observations and in satellite studies (e.g., Prospero and Savoie, 1989; Merrill et al., 1989; Herman et al., 1997; Wilkening et al., 2000).

There is growing observational evidence that the effects of pollutant outflow from Asia extend to North America. One study found evidence that Asian boundary layer air can be transported to the upper troposphere over California in two to four days (Kritz, 1990). Surface observations of CO coupled with air transport "back trajectories" were used to infer that anthropogenic emissions from Asia

were transported to the surface of North America during the spring of 1997 (Jaffe et al., 1999). In the spring of 1999, CO and ozone attributed to emissions originating in Asia were again detected off the coast of Washington State (Jaffe et al., 2001). Analysis of ozone data from U.S. air quality monitoring networks reveals that the low end of the ozone probability distribution has been increasing over the past two decades, which investigators attribute to rising levels of background pollution transported from outside the United States (Lin et al., 2000).

The long-range transport of dust aerosols to the United States from Africa has been well documented. For instance, Saharan dust transported to the southeastern U.S. during summer months has been observed to impact atmospheric PM concentrations in Miami (Prospero, 1999), and has also been detected in Great Smoky Mountains National Park and even further inland (Perry et al., 1997).

The long-range transport of gases and aerosols from biomass burning has also been detected. Emissions from large forest fires in Canada increased both CO and aerosol concentrations along the eastern seaboard of the United States in 1995 (Wotawa and Trainer, 2000). In May 1998, smoke from numerous fires burning in Mexico and Central America was transported into the southern U.S., prompting Texas and other states to issue public health advisories. Smoke from these fires was documented in Oklahoma (Peppler, 2000), and satellite and ground-based observations showed this smoke plume extending northeastward into the Smoky Mountains (Kreidenweis et al., 2001). Analyses of monitoring data from Big Bend National Park in Texas suggest that smoke transport to the park was an annual spring event (Gebhart et al., 2000).

Long-range pollution transport is known to affect even very remote regions of the earth. The wintertime phenomenon known as arctic haze has been extensively studied since its first identification in the 1950s. These studies revealed evidence that the observed haze was caused by mid-latitude emissions from fossil fuel combustion, smelting, and other industry, primarily from Eurasia (Barrie, 1986). Since then, a series of field studies have greatly improved our understanding of how pollution transported to the Arctic affects atmospheric chemistry and climate within this region (for instance, see Harriss et al., 1992; MacCracken et al., 1986; Blanchet, 1989).

Modeling Studies of Current/Future Impacts

A number of three-dimensional global chemical transport models (GCTMs) have been used to study the issue of long-range pollution transport. One modeling study predicted that future developments in Asia could lead to emissions that will significantly perturb free tropospheric ozone chemistry over the Pacific (Berntsen et al., 1996). More recently, it was calculated that Asian emissions have already raised mean background springtime levels of CO and O_3 by an average of 34 and 4 ppb, respectively, over the eastern North Pacific (Berntsen et

al.,1999).[2] Another study predicted that if Asian NO_x emissions were to triple between 1985 to 2010, this increase could enhance surface background ozone over the western United States by 2-6 ppb (Jacob et al., 1999).

While these simulations estimate monthly or seasonal mean changes, many observations suggest that Asian pollution outflow is highly episodic and may not be well represented as a uniform background enhancement. For instance, recent observations of CO at an observatory on the western coast of Washington State found strong synoptic-scale fluctuations with changes of 20 to 50 ppb over a few days (Jaffe et al., 2001). A model simulation by Yienger et al. (2000) predicted these types of large synoptic-scale fluctuations in CO. In this study it was also estimated that the current Asian contributions to episodic ozone events over the western United States are in the range of 3-10 ppb, and that future ozone episodes along the U.S. west coast could have Asian contribution as high as 40 ppb (based on a "worst case" emission scenario wherein Asian NO_x emissions increase by a factor of four from 1990 to 2020).

Long-Term Projections

Studies carried out for the Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) have extended estimates of future air quality changes to the year 2100. These studies are based on projections of future pollution emissions which, in turn, depend on changes in driving forces such as population, social and economic development, and technology. Attempts to predict changes in these driving forces and resulting emissions are highly uncertain when their time horizon extends as far as a hundred years into the future. Recognizing these uncertainties, however, one can examine the implications of a range of possible future emissions by developing a set of alternative scenarios.

A comprehensive set of long-term emission scenarios has recently been developed under the auspices of the IPCC Special Report on Emission Scenarios (Nakicenovic and Swart, 2000), which are based on a range of assumptions about the rate and direction of economic and technological change and the degree of globalization. These scenarios include projections for species that influence air quality, including SO_2, NO_x, CH_4, VOCs, and CO. The IPCC selected six "illustrative" scenarios (out of the 40 that were originally developed) to represent a range of assumptions and modeling approaches. None of these scenarios is identified as more probable than any other. The results of the six illustrative scenarios for emissions of SO_2 and NO_x over the next 100 years are displayed in Figure 3-1. Note that the total emissions of SO_2 generally decrease over time in these scenarios, due to an assumed widespread implementation of emission control technologies.

[2]For reference, Berntsen et al. estimated the average springtime concentrations of CO (in the marine boundary layer) to be 151 ppb, and the average concentrations of ozone to be 31 ppb.

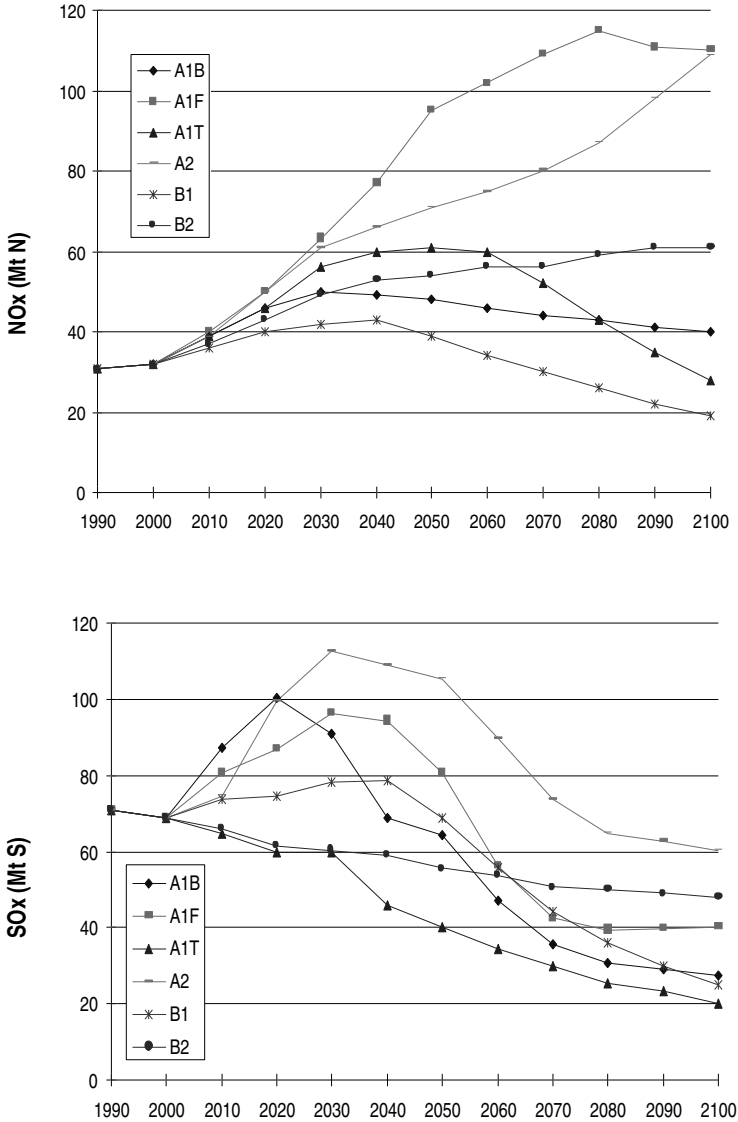

FIGURE 3-1 IPCC emission scenarios for NO_X and for SO_2. The "A" scenarios represent materially oriented development, whereas the "B" scenarios involve a transition to information and service-oriented economies. The "1" scenarios represent globally oriented development, and the "2" scenarios represent locally and regionally oriented development. Within the A1 family, the three variants are based on different energy sector developments: fossil fuel intensive (A1F); transition to non-fossil energy sources (A1T); and balance across all energy sources (A1B). See Nakcenovic and Swart (2000) for more details.

Box 3-1
Current Policy Approaches to Addressing Transboundary Air Pollution

The importance of air pollution transport across national boundaries has been recognized in some regions for more than twenty years. The original focus of attention was acid deposition in Europe, which led to the Convention on Long-Range Transboundary Air Pollution (LRTAP) signed in 1979. Since then the Convention has been extended by eight protocols, most recently the 1999 Protocol to Abate Acidification, Eutrophication, and Ground-level Ozone. This Protocol makes regional air quality a central focus of the LRTAP Convention and includes a schedule of specific emission limits on SO_X, NO_X, VOCs, and ammonia for countries throughout Europe. It also establishes air quality objectives for Europe similar to U.S. National Ambient Air Quality Standards.

The United States and Canada also participate in the air quality protocols of the LRTAP convention, although U.S. and Canadian obligations are simply designed to mirror pre-existing national laws. More pertinent in North America are the U.S.-Canada Air Quality Agreement, signed in 1991, and the Commission on Environmental Cooperation, established pursuant to the North America Free Trade Agreement. While the Air Quality Agreement initially addressed only acid deposition, the United States and Canada recently negotiated a new annex to the agreement in response to findings that there is significant cross-border transport of ozone in eastern North America (as discussed in the report *Ground-Level Ozone: Occurrence and Transport in Eastern North America* by the United States-Canada Air Quality Committee, 1999).

These scenarios were used with a variety of GCTMs to explore how global air quality and climate could be affected over the next century by the projected increases in pollution emissions. When using a "high end" emission scenario (A2, which assumes the largest emissions of CH_4 and ozone precursors of the six IPCC illustrative scenarios), all of the models projected large increases in global tropospheric ozone levels. Specifically, it was estimated that zonal mean surface ozone concentrations over northern midlatitudes for the month of July would increase from current values (estimated at about 40 ppb) to more than 70 ppb, and more than 80 ppb near the tropopause, by the year 2100 (Ehhalt and Prather, 2001). These projected changes, which result from the cumulative impact of all emissions, could have serious consequences for the air quality of most of the northern hemisphere.

It should be emphasized, however, that alternatives to this high-end emission scenario do not lead to such large-scale air quality changes. Moreover, although all models predicted significant responses of ozone to changes in precursor emissions, estimates of the overall magnitude and geographical distribution of these impacts varied widely. Also, the models used for these studies do not yet include some important processes such as the changing chemistry of the

stratosphere and the impacts of climate and land-use change on biogenic emissions of ozone precursors. Nevertheless, these studies illustrate that changes in air pollution emissions occurring over the next several decades could *potentially* have impacts far beyond the regions in which they are emitted, and that in the 21st century, a global perspective may be needed to meet regional air quality objectives.

4

Current Capabilities for Observing Global Air Quality Changes

As discussed in Chapter 1, improving our understanding of issues such as climate/chemistry interactions and intercontinental pollution transport will require a comprehensive research framework that integrates observations covering a wide range of temporal and spatial scales with modeling and process studies. Some components of this research framework are in a more mature state than others. For instance, significant progress is being made in the development of regional/global chemical transport models and their integration with global climate models. Likewise, in recent years the atmospheric chemistry community has organized many successful field campaigns that combine model analyses with intensive observations from a variety of platforms. These studies have enhanced our understanding of many complex chemical and radiative processes that play a role in global air quality change.

In contrast, current observational programs are not adequate to detect many of the important medium- and long-term atmospheric chemical changes discussed in the previous chapters. This weakness greatly limits our ability to document the evolution of the atmosphere in the coming decades. If improvements in observational capabilities are not made, this record could be lost forever. It also limits the value of the developments cited above, since a strong observational base is needed to test and improve model predictions, and to provide a longer-term context for the observational "snapshots" obtained through intensive field campaigns.

Current tropospheric observational programs include global and regional networks designed to measure background atmospheric composition at selected remote sites; regulatory monitoring networks that analyze day-to-day air quality

changes at numerous sites located primarily in urban areas; remote-sensing (satellite) instruments that provide global-scale observations of selected atmospheric species; and a variety of balloon- and aircraft-borne instruments used for *in situ* measurement campaigns. These different observational platforms vary widely in their scope and degree of analytical rigor. The following sections describe the capabilities and limitations of these different observational systems for addressing the global air quality issues discussed in this report. Gas-phase species and aerosols are discussed separately since each presents unique observational challenges.

Gas-phase Species: *In Situ* Observations

The temporal and spatial sampling requirements for observations of a gas-phase chemical species are closely linked to the atmospheric lifetime of that species: the required geographical density of sampling locations increases as the atmospheric lifetime of the species decreases. The necessary sampling density also increases with the need for higher levels of measurement precision, a requirement that is often related to the nature of the individual measurement sites. For instance, the more a sampling site is affected by local sources or local meteorology (e.g., upslope/downslope diurnal winds, passing fronts, land/sea breezes), the more frequent the sampling must be in order to resolve these effects. At many sites, hourly or more frequent sampling is often required to resolve measurable changes from background values for some species.

There are a few networks of surface observational sites that are far removed from local sources and sinks and thus can be used to obtain a regionally representative "baseline" of atmospheric composition. This includes U.S.-supported networks operated by the National Oceanic and Atmospheric Administration's Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) and by the Advanced Global Atmospheric Gases Experiment (AGAGE), as well as a number of non-U.S. national and regional efforts (e.g., in Australia, the European Community, Japan, and New Zealand). These networks employ *in situ* automated measurements as well as flask sampling, which permits centralized analyses of samples from different sites and helps verify the relative calibrations of the *in situ* measurements.

These types of networks have generally proven successful in characterizing the overall distribution and temporal trends of long-lived gases including CO_2, CO, CH_4, N_2O, and CFCs[1] . In contrast, the atmospheric concentrations of shorter-lived (i.e., more reactive) species including NO_x, VOCs, O_3, and PM exhibit large spatial and temporal variations, and hence measurements at select baseline sites around the globe are generally far too sparse to characterize their overall

[1]Quantifying regional sources/sinks of these compounds requires more intensive measurement strategies, but this issue is beyond the scope of this report.

distribution and temporal trends. Integrating the observations from different networks could provide fairly dense coverage in some regions, but this would require substantially improved cooperation in matters such as calibration. Independent calibration standards are made by only a few research laboratories (e.g., AGAGE/SIO, NOAA/CMDL, Tohoku/Nippon Sanso), and comparisons of the calibration standards among these different laboratories have revealed significant disagreements for several important chemical species. This issue needs to be addressed in order to make optimal use of existing networks.

In the United States and many other countries, air quality monitoring networks provide extensive ambient measurements of pollutant species.[2] However, there are at present serious limitations to the use of these data in the study of large-scale atmospheric chemical changes. Most air quality monitoring sites are focused on heavily populated areas, and are designed simply to determine whether or not the area is in compliance with current air quality standards. There are very few air quality monitoring sites that allow meaningful examination of long-range transport and trends in background concentrations. Ozone trends in particular are difficult to assess because changes due to anthropogenic forcing are often confounded by natural variations. Finally, techniques used to monitor ozone precursors, NO_x and VOCs, do not yet have sufficient sensitivity or accuracy to contribute to trend analysis (Demerjian, 2000; NARSTO, 2000; Fiore, 1998; Wolff et al., 2001; Porter et al., 2001).

These types of urban and regional air quality monitoring networks rely entirely upon ground-based sites that sample within the boundary layer (the lowermost atmosphere). Addressing the global air quality issues highlighted in this report, however, will require observations that extend over a larger altitude range. For instance, the transport of pollution between Asia and the United States occurs primarily through the middle and upper troposphere, and because of the highly episodic nature of this transport, there can be significant inhomogeneity in the air masses reaching the continental United States. Thus, ground-based networks that only sample air masses within the boundary layer would not allow a quantitative determination of long-range pollutant fluxes. Likewise, the radiative forcing exerted by ozone and aerosols is highly sensitive to altitude, and thus it is important to have the capability to observe changes in chemistry and climate over a broad altitude range.

There are a variety of techniques to measure vertical profiles of atmospheric composition. Ozone profiles are commonly measured with ozonesondes, lightweight instruments carried aloft on balloons (the most common form is an elec-

[2]A report entitled "The Role of Monitoring Networks in the Management of the Nation's Air Quality" by the U.S. National Science and Technology Council provides a detailed summary of the different air quality and deposition monitoring networks operating in the United States. This report is available at: *http://www.nnic.noaa.gov/CENR/cenr.html*

Box 4-1
Meteorological Data Needs

Meteorological and dynamical processes operating over a wide range of spatial scales play a central role in the air quality changes discussed in this report. As described earlier, the emissions, chemical transformations, and deposition of air pollutants are affected by parameters such as temperature, cloud cover, humidity, and wind speed and direction. In addition, dynamical forces related to vertical temperature profiles control the dispersion of pollution within the urban/local boundary layer and the release of pollutants from the boundary layer into the free troposphere. Finally, the long-range transport of pollutants is influenced by atmospheric high- and low-pressure systems occurring on scales of hundreds to thousands of kilometers. Thus, the meteorological context of atmospheric chemical measurements must be established in order to accurately assess implications for air quality and to develop the models used for studying long-range pollution transport and climate-air quality linkages

trochemical concentration cell). In a comprehensive analysis of the various ozonesonde observations carried out worldwide, it was found that these data can be valuable for developing a global climatology of tropospheric ozone and for testing 3-D tropospheric chemistry models. However, it was also found that there are currently insufficient data for large regions of the earth, particularly in the tropics and subtropics (Logan, 1999). Ozone profile measurements can also be made with differential absorption lidar, a technology that can potentially be implemented on ground-based, airborne, and spaceborne platforms. Lidar provides high-resolution vertical profiles and thus may be particularly useful for identification of pollution plumes aloft. Lidar systems are also being developed for measurements of water vapor, aerosol, and cloud profiles.

Finally, aircraft provide a valuable platform for studying atmospheric chemical composition over a wide range of altitudes. The use of aircraft equipped with a large array of analytical instruments are becoming common in intensive field campaigns for measuring a comprehensive suite of trace gases and reactive species in the upper troposphere and lower stratosphere. More routine aircraft-based sampling is being carried out by the European MOZAIC program (Measurement of OZone by Airbus In-service airCraft), where measurements of ozone, water vapor, and temperature are collected by automated instruments mounted on passenger airliners (Marenco et al., 1998; Thouret et al., 2000). This approach of using sensors onboard commercial aircraft holds great potential as a tool for obtaining vertical profiles near airports and frequent measurements of atmospheric composition at flight altitudes. In order to play a major role in studies of global air quality change, however, it will be necessary to expand the geographic and temporal coverage of these types of measurements. Also, as

with the surface-based observations, developing more uniform standards for calibration and quality control will greatly facilitate efforts to integrate the observations obtained from different measurement programs.

Gas-phase Species: Remote Sensing (Satellite[3]) Observations

A considerable effort has been mounted to develop satellite remote sensing techniques for making global measurements in the troposphere. There are now about 20 instruments in orbit or under development that have substantial tropospheric monitoring capabilities. Table 4-1 lists some of these instruments, with an emphasis on nearer-term measurements. Measurements of tropospheric O_3, H_2O, NO_2, CO, CH_4, and HCHO have been demonstrated for potential future use, and there have also been significant efforts to extract tropospheric information from instruments currently in operation, for instance, extracting tropospheric ozone information from the Total Ozone Mapping Spectrometer (TOMS).

Information on the chemical state of the troposphere from satellite measurements is currently very limited because tropospheric measurements from space are inherently difficult. For example, nadir-looking measurements must be able to discriminate between stratospheric and tropospheric contributions of the species of interest. Limb-sounding measurements are often susceptible to interference from clouds and aerosols, and in some cases from H_2O absorption or emission. Another limitation is that most current and planned instruments are on polar orbiting platforms, which can only observe any particular location once every few days. In contrast, geostationary satellites allow instruments to observe one location for an extended period of time and thus would generally be more useful for studying pollution transport patterns.

Satellite instruments that could potentially be used for determination of long-term trends in tropospheric chemistry include the Global Ozone Monitoring Experiment (GOME) instruments on the European Space Agency's ERS-2 satellite and the Eumetsat Metop satellites (ca. 1995-2012 time series); the Infrared Atmospheric Sounding Interferometer (IASI) instruments on the Metop satellites (ca. 2006-2012 time series); the Ozone Mapping and Profiler Suite (OMPS) instruments on the U.S. NPOESS satellites (ca. 2008-2017 time series); and potentially, the U.S. TOMS instruments (1978 and continuing time series). Although these instruments are not optimized for tropospheric measurements, it is worthwhile to plan strategies for optimizing the interpretation of the collected tropospheric data and integrating these data with shorter-term satellite measurement programs (e.g., Earth Observing System).

[3]Most all of the current and planned remote sensing measurement programs reviewed here are satellite-based. It should be noted however, that the potential exists for using other remote sensing platforms such as the space shuttle.

Although much development work is still needed, the possibilities for using satellite-based measurements to obtain trend-quality tropospheric measurements of species including O_3, NO_2, and HCHO should eventually be realized. However, major challenges will need to be addressed as tropospheric measurements from satellites move from initial characterization to trend determination. Monitoring tropospheric trends using multiple instruments (often operated by different countries) will require tying together measurement records with intensive radiative transfer modeling, photochemical and transport modeling, and instrument characterization. Extensive ground-based and other *in situ* measurements will continue to be a critical component of remote sensing observational systems, as they are needed for evaluating and interpreting satellite data.

Aerosols: *In Situ* Observations

Particulate matter presents a "multi-dimensional" observational challenge, because it is important to characterize not only total PM concentration, but also size, composition, mass, and phase; and these parameters can evolve as the air mass containing the aerosol undergoes changes in humidity and temperature and exposure to new condensable emissions. Real-time measurements of particle size are possible but routine measurements of aerosol mass and chemical composition rely primarily on the accumulation of particles over extended sampling times with subsequent laboratory analyses (although techniques for real-time chemical analysis of aerosols are rapidly developing[4]).

In situ measurements can provide useful information about which aerosol types predominate in a given region, but there are serious gaps in existing aerosol observational programs. For example, the NOAA/CMDL sites are gathering useful aerosol optical data, but these measurements are very limited in time and space. Urban areas in the United States are monitored as part of the EPA's air quality regulatory framework (Demerjian, 2000), but large spatial gaps exist in many rural areas, where background aerosol plays a major role in determining local air quality. The IMPROVE network (Interagency Monitoring of Protected Visual Environments; Malm et al., 1994), collaboratively operated by several U.S. federal agencies, obtains information on aerosols in national parks and wilderness areas, and has recently been expanding to fill some of the observational gaps in sparsely populated areas. These data, however, cannot yet yield a picture of global or even regional aerosol distributions. There are opportunities for obtaining more complete information through better integration of existing measurement programs. For example, aerosol optical depths are measured by sun photometers at a number of sites (Aerosol Robotic Network-AERONET;

[4]A special issue of the journal Aerosol Science and Technology (v33, 2000) provides a detailed overview of real-time single-particle analysis techniques.

TABLE 4-1 Major Space-based Tropospheric Chemistry Platforms. SOURCE: adapted from Singh and Jacob (2000)

Sensors	**TOMS**[a]	**GOME**	**MOPITT**	**MODIS**	**SCIAMACHY**	**MIPAS**
Launch	**1978**	**1995**	**1999**	**1999**	**2001**	**2001**
O_3	column	column+			column+ Δz=3-4km limb	z>5 km, limb
H_2O				column	column+	z>5 km, limb
CO			3-4 levels		column+ Δz=3-4km limb	z>5 km, limb
NO						
NO_2		column			column+	
HNO_3						UT
CH_4			column		column+ Δz=3-4km limb	
HCHO		column			column+ Δz=3-4km limb	
SO_2	column	column			column	
BrO		column			column+ Δz=3-4km limb	
Aerosol	column	column		column	column/profiles	

[a]TOMS has been in operation since 1978. Last launch was in 1996 and data continues to be collected at this time. OMI will take over some TOMS functions in year 2003. [b]A number of additional derived chemical products such as acetone, methanol, H_2O_2, HCN, NH_3, HNO_4, SO_2, and PAN are possible.

Holben et al., 1998), but few are co-located with chemical composition measurements.

Measuring vertical aerosol distributions is also critically important. Models that can reproduce surface aerosol observations quite well often differ significantly in their estimates of total column burden, and therefore the transport, of aerosols. Sporadic data from airborne field campaigns have been used to help understand the vertical distribution of aerosols, and balloon-borne measurement programs have provided valuable information on mid- and upper-tropospheric

TES[b]	HRDLS	OMI[a]	MLS	SAGE III	TRIANA
2002	**2002**	**2003**	**2002**	**2003**	**2002**
Δz=2/4km limb/nadir	Δz=1km (UT)	column	UT	Δz=1km (UT)	column
Δz=2/4km limb/nadir			UT	Δz=1km (UT)	
Δz=2/4km limb/nadir			UT		
tropical UT Δz=2km					
		column			Δz=1km (UT)
Δz=2km (UT)	Δz=1km (UT)				
column					
		column			
column		column			
		column			
		column		Δz=1km	column

Definitions: UT = upper troposphere; Δz is to the instrument vertical resolution; 'column' is the vertically integrated abundance of a species; 'column +' means that some information on vertical distribution is provided in addition to total column abundance; 'limb' and 'nadir' refer to observations made (respectively) along horizontal and vertical viewing paths.

aerosol size distributions (Hofmann, 1998). Lidars and lidar-in-space technology also offer new opportunities for filling this important observational need. At present, however, there is no clear plan for routine observations of aerosol vertical distributions.

As with gas-phase observations, obtaining accurate and precise aerosol measurements on a global basis requires the development of uniform, primary standards for calibration. Neither primary standards nor standard operating procedures are readily available for many research instruments. This lack of

consistency in operational measures presents a major obstacle to integrating the observations from different programs.

There are few long-term records of tropospheric aerosols, making it difficult to identify trends and to relate trends to anthropogenic activities. Even though some monitoring programs have been established over the last several decades, it can be difficult to interpret trends at individual sites when changes in the background air reaching that location are not characterized. For example, sulfate levels near coastal sites can be affected by ocean fluxes of reduced sulfur compounds, transport of natural precursors and aerosols, and anthropogenic emissions; and these different sources cannot be readily distinguished or quantified.

Surface observations of sulfate aerosols in remote regions are currently very difficult to reproduce in models. The agreement of model predictions with observations is much worse for other aerosol types, such as black carbon and organic species, for which we have fewer observations and less fully characterized atmospheric transformation and removal processes. Progress in improving these models is hindered by the lack of vertically-resolved aerosol data and by significant inaccuracies and poor spatial resolution in emissions inventories, particularly of biogenic and other organic emissions that condense in the atmosphere to form PM.

Aerosols: Remote Sensing (Satellite) Observations

Current satellite instruments (pre-1999) were not designed specifically to detect tropospheric aerosols, however they can provide a qualitative picture of the atmospheric distribution of some aerosol types. For instance, data from the GOES-8 satellite have been used for detection of smoke/haze from fires (e.g., Prins et al., 1998). Maps of "aerosol optical thickness" (AOT) over the oceans, showing widespread plumes attributed to dust, biomass burning, and pollution aerosols, have been produced using data from the polar-orbiting AVHRR instrument (e.g., Husar et al., 1997). The TOMS instrument has been shown to be sensitive to absorbing aerosols such as dust and smoke (Herman et al., 1997) and has been used to investigate long-range transport of African dust. By combining these data with observations from ground-based monitoring networks, much has been learned about global sources and transport of some important aerosol types. However, it is difficult to obtain quantitative information on aerosol type and mass flux from such images, largely because of the complex and poorly characterized radiative properties of most aerosols.

Recognition of the important role played by aerosols in climate forcing and in air quality has led to significant international support for satellite missions aimed at improving current observational capabilities. Newly-launched and proposed instruments (e.g., MODIS, MISR, and PICASSO-CENA) have been specifically designed for detection of aerosols and clouds (King et al., 2000). Nevertheless, it remains difficult to retrieve aerosol properties over land, particularly

for regions with low AOT; and unfortunately, these regions are of particular interest because they are most susceptible to anthropogenically-driven changes in aerosol loadings. Tropospheric aerosol determination is also limited by our incomplete understanding of aerosol light scattering physics.

Validation plans for MODIS and MISR include intercomparison with ground-based monitoring networks and intensive campaigns involving measurements from aircraft and other platforms. The international AERONET network of sunphotometers (Holben et al., 1998) plays a prominent role in evaluating the accuracy of AOT data. The vertical profiles of aerosol properties produced by the PICASSO-CENA lidar measurements will also need to be validated against ground-based and airborne *in situ* measurements. The value to these satellite missions of data from a long-term, ground-based network measuring aerosol chemical composition and physical properties is apparent. The network data help place the satellite measurements in context by defining characteristics such as typical aerosol loadings for particular locations. Ground-based observations can also provide information that cannot yet be obtained from space-borne instruments, such as the chemical composition (and hence the sources) of particulate matter. And finally, since these aerosol-specific satellite missions have a relatively short lifetime and thus limited applicability for trend analyses, continuous ground-based monitoring data are required.

Careful consideration needs to be given to making optimal use of the aerosol data being provided by current and planned satellite missions. Aerosol optical thickness, a common satellite data product, has limited utility as input to models for validation purposes; it is likely that combining satellite information with routine and intensive surface observations will be more useful. Continued investigations into coordination of efforts—for example, co-locating satellite validation monitoring sites with existing observational network sites, and ensuring continuity of the longer-term network data—should be undertaken and supported.

An Integrated Research Strategy

It is clear that our understanding of global air quality would be greatly aided by strengthening the infrastructure for obtaining high-quality atmospheric chemical measurements from ground-based stations and platforms such as balloon sondes and aircraft. However, *in situ* observations alone cannot provide sufficient spatial coverage to assess the global distribution and trends of short-lived gases and aerosols. Ideally, remote sensing instruments could provide global coverage, but the capability of satellites to provide accurate measurements of global atmospheric composition and long-term trends for tropospheric species lies years in the future. Even when comprehensive satellite observations do become available, correlative *in situ* measurements will still be needed to test the remote sensor inversion algorithms. The most feasible strategy then, is to use

Box 4-2
Key Factors in Maintaining High Quality Observations

Over the past several decades, beginning with the pioneering studies of atmospheric CO_2, a great deal has been learned about what is required to create and sustain effective, reliable networks for observing the chemical state of the atmosphere. Some of the key factors include the following:

- *Maintaining support for continuity of data sets.* Maintaining high-quality, long-term measurement programs for atmospheric trace gases and aerosols requires continuity of operational support. While all measurement programs should be subjected to rigorous scientific and technical evaluation, the continuity of the project itself should not be put in jeopardy by temporary shifts in funding priorities. There is a need to ensure that the support of observations is adequate to prevent breaks in the data record. Similarly, mechanisms are needed to provide support for development and validation of new instrumentation and its deployment in the field, which can often take many years.
- *Calibration.* Inadequate calibration of instruments in a measurement program limits the value of observations for understanding global atmospheric changes. Absolute calibration is critical for calculating the budgets and lifetimes of different chemical species. Relative calibration is essential in applications that require the use of observations from multiple measurement stations or networks. The goal should be to calibrate each species with an absolute accuracy approaching the analytical precision of the measurement technique.
- *Appropriate checks on data quality.* If a quality assurance/quality control structure has not been established or is inadequately funded for a particular measurement program, then the uncertainties associated with the measurements may render them unusable for trend analysis and model evaluation, and thus the collection efforts are wasted. Standardized quality assurance criteria are especially important in efforts to integrate data from multiple observational programs.
- *Collaboration among researchers with different missions but similar data needs.* Each type of measurement has limitations and is most useful when it can be meaningfully combined with other types of data. Effective coordination among programs with related observational needs can avoid redundant data collection efforts or data gaps that occur when individual programs lack the resources to adequately support continued observational efforts. For example, coordination of aerosol column measurements with surface aerosol observations can aid both the air quality and climate change research communities.
- *Personnel requirements.* Experience has shown that maintaining and advancing long-term observational science in atmospheric chemistry depends primarily on highly qualified and dedicated individuals. Attracting such individuals requires strong educational programs and promising career opportunities. Experience has also shown that having more than one laboratory striving for the same goal is the most effective way to assure that the highest quality observations will be made (i.e., all key species should be measured by more than one research group).

the limited available remote sensing data in combination with measurements from the range of available *in situ* platforms.

These integrated measurement programs form a core component of the overall research framework needed for the study of global air quality change. Models comprise another core component of this research framework, since they are needed to fulfill at least three important roles: (i) helping to determine the optimal locations for long-term observational sites and short-term process studies, in order to maximize the usefulness of the data collected; (ii) assimilating the observations acquired from different platforms and helping to place isolated measurements in a larger context; and (iii) providing a prognostic capability for predicting future air quality trends, for estimating transboundary pollution fluxes, and for assessing the impacts of coupling between climate and air quality changes. Fulfilling all of these roles will require careful integration of various types of modeling tools. For instance, process models can be embedded in regional models to provide detailed representation of certain critical processes; and likewise, regional models can be embedded in global models to expand the spatial and temporal resolution in areas of particular interest.

Coordinating measurements among various remote sensing and *in situ* platforms, and integrating these measurements with detailed modeling studies presents an immensely complex research challenge. This approach is being developed in focused process studies such as the Southern Hemisphere ADditional OZonesondes project (SHADOZ, which coordinates a network of balloon-borne ozonesondes with TOMS remote sensing measurements), and the INDOEX, TRACE-P, and ACE-Asia field campaigns (studies aimed at understanding the impacts of pollution outflow from the Asian continent). This level of integration, however, does not yet exist in an ongoing, operational sense or extend over the range of scales needed to accomplish the research objectives highlighted in this report.

5

Findings and Recommendations

Climate change and intercontinental transport of air pollutants have the potential to significantly affect global air quality in the coming decades. Establishing a suitable infrastructure for reliable, long-term observations of the chemical state of the atmosphere is a critical component of the research framework needed to characterize global air quality and to help develop and evaluate the models used for projecting future changes. The following is a summary of the committee's priority finding and recommendations for addressing these issues:

Key findings:

Current observational systems are not adequate for characterizing many important medium- and long-term global air quality changes. Some particularly notable weaknesses in our current observational capabilities include the lack of (i) long-term measurements of reactive compounds and PM, (ii) methods for obtaining vertical profile data, and (iii) measurement sites that allow for a meaningful examination of long range transport and trends in background concentrations.

The global air quality issues discussed in this report intersect with the concerns of several federal agencies, yet none of these agencies have a clear mandate to lead U.S. research efforts or maintain the long-term observational programs that are needed to address these issues.

Recommendations:

- Maintain and strengthen the existing measurement programs that are essential for detecting and understanding global air quality changes. High

priority should be given to programs which aid in assessing long-term trends of background ozone and PM.

- Establish new capabilities to provide long-term measurements and vertical profiles of reactive compounds and PM that will allow meaningful examination of long-range transport and trends in background concentrations.

  These two recommendations will require providing support to:

  - develop uniform and traceable standards on a global basis for calibration of both gas-phase and aerosol measurements;

  - improve measurement technologies for use in current observational platforms (such as ground-based air quality monitoring networks, commercial aircraft, and balloons/sondes), and in new potential platforms such as "supersites" for measuring a comprehensive suite of compounds in remote locations, and unmanned aerial vehicles for long-duration sampling of the atmosphere over a wide range of altitudes;

  - integrate measurements obtained from different observational programs and platforms with a particular focus on integrating remotely sensed satellite observations with *in situ* aircraft and ground-based measurements;

  - promote observational programs specifically designed to address chemical and meteorological data requirements for the improvement and validation of models.

- Responsibility for carrying out this work should be clearly assigned to a U.S. federal agency (or interagency) research program, and the U.S. should play a leadership role in fostering international cooperative research and observational activities to enhance our understanding of global air quality changes.

In conclusion, the committee wishes to emphasize that developing global-scale research and observational programs inherently requires substantial international cooperation. In recent years, the atmospheric chemistry research community has successfully coordinated multinational programs to study issues such as stratospheric ozone depletion and the atmospheric effects of aviation. Studies of global air quality change should build upon these successes, and yet will likely require even deeper levels of international cooperation. For instance, quantifying long-range transport of air pollutants would be greatly aided by an

Box 5-1
Implications for U.S. Air Quality Management

To what extent will the United States be in control of its own air quality in the coming decades? It is not possible to provide a simple answer to this question given the vast uncertainties about future global economic development, technological advances, and policy choices. As discussed in this report, however, evidence does point to the plausibility of a scenario in which non-U.S. emissions and changing climatic conditions could significantly affect the air quality in some regions of the United States.

This concern will become even more pressing with the application of the national ambient air quality standards (NAAQS) that were issued by EPA in 1997. The ozone NAAQS was changed from a 1-hour, 0.12 parts per million (ppm) ambient concentration to an 8-hour, 0.08 ppm concentration.[1] The longer averaging time and lower concentration of the new standard means that background ozone will play an increasingly important role in determining whether or not an area is in compliance with the NAAQS. As background levels rise, local emission control strategies would become less effective at maintaining compliance with mandated standards. Because the observational record for PM is much less extensive than that for ozone, it is more difficult to assess likely future changes. There are similar concerns, though, that increases in background PM levels could greatly hinder the ability of some areas to comply with the PM NAAQS.[2]

Currently, air quality management plans focus primarily on controlling emissions at state and local levels. In recent years, the EPA has begun considering regional air quality management strategies, and an assessment of the North American Research Strategy for Tropospheric Ozone program (NARSTO, 2000) emphasized the importance of viewing ozone pollution from a continental perspective. It seems likely that this consideration will ultimately need to be broadened even further, to encompass a hemispheric-scale perspective. Likewise, current studies of the meteorological impacts on air quality will likely need to be expanded to include studies of how air quality is affected by long-term climatic changes. Meeting these new air quality management challenges requires a stronger scientific base which, in turn, will require strengthening the observational infrastructure for tropospheric chemistry.

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[1] The 0.08 ppm ozone NAAQS is defined as the 3-year average of the annual 4th-highest daily maximum 8-hour ozone concentrations.

[2] In 1997 EPA revised the PM NAAQS by adding a new annual $PM_{2.5}$ standard set at 15 micrograms per cubic meter ($\mu g/m^3$), and a new 24-hour $PM_{2.5}$ standard set at 65 $\mu g/m^3$. EPA retained the current annual PM_{10} standard of 50 $\mu g/m^3$, but changed the form of the PM_{10} 24-hour standard (150 $\mu g/m^3$) by replacing the 1-expected-exceedance form with a 99th percentile form, averaged over 3 years.

open exchange among countries of data from national emission inventories and air quality monitoring networks.

Programs such as the World Meteorological Organization's Global Atmosphere Watch have made substantial efforts to establish data centers and quality control programs to enhance integration of air quality measurements from different national and regional networks, and to establish observational sites in undersampled, remote regions around the world. Similarly, the International Global Atmospheric Chemistry project (of the International Geosphere-Biosphere Programme) has strongly endorsed the need for international exchange of calibration standards and has helped coordinate multinational field campaigns to address a variety of important issues related to global air quality. Maintaining these worthwhile activities depends upon continued support from the United States and other countries. The committee urges the U.S. federal agencies (working, where appropriate, with interested and qualified parties in academia, the private sector, etc.) to play a leadership role in fostering these types of cooperative efforts and developing new research and observational activities that will enhance our understanding of global air quality change.

References

Ackerman, A.S., O.B. Toon, D.E. Stevens, A.J. Heymsfield, V. Ramanathan, and E.J. Welton. 2000. Reduction of tropical cloudiness by soot. *Science*. 288:1042-1047.

Albrecht, B.A. 1989. Aerosols, cloud microphysics, and fractional cloudiness. *Science*. 245:1227-1228.

Barrie, L.A. 1986. Arctic air pollution: An overview of current knowledge. *Atmos. Environ*. 20:643-663.

Berntsen T., I.S.A Isaksen, W.C. Wang, and X.-Z. Liang. 1996. Impacts of increased anthropogenic emissions in Asia on tropospheric ozone and climate. A global 3-D model study. *Tellus*. 48B:13-32.

Berntsen, T.K., S. Karlsdottir, and D.A. Jaffe. 1999. Influence of Asian emissions on the composition of air reaching the North Western United States. *Geophys. Res. Lett*. 26:2171-2174.

Blanchet, J-P. 1989. Toward estimation of climatic effects due to Arctic aerosols. *Atmos. Environ*. 23:2609-2624.

Demerjian, K.L. 2000. A review of national monitoring networks in North America. *Atmos. Environ*. 34:1861-1188.

Dunn, S. 1996. Green Auto Racing. Natural Resources Defense Council, Washington.

Ehhalt D., and M. Prather (Coordinating Lead Authors). 2001. Atmospheric Chemistry and Greenhouse Gases. Chapter 4 in *Climate Change 2001:The Scientific Basis. Third Assessment Report of the Intergovernmental Panel on Climate Change (IPCC).* Cambridge University Press. *(in press).*

Environmental Protection Agency (EPA). 1999. National air quality and emissions trends report. EPA report #454/R-00-003.

Fiore, A.M., D.J. Jacob, J.A. Logan, and J.H. Yin. 1998. Long term trends in ground level ozone over the contiguous United States, 1980-1995. *J. Geophy. Res*. 103:1471-1480.

Gebhart, K.A., W.C. Malm, and M. Flores. 2000. A preliminary look at source-receptor relationships in the Texas-Mexico border area. *J. Air and Waste Management Assoc*. 50:858-868.

Hansen, J., M. Sato, R. Ruedy, A. Lacis, and V. Oinas. 2000. Global warming in the twenty-first century: an alternative scenario. *Proc. Natl. Academy of Sciences*. 97:9875-9880.

Harriss, R.C., S.C Wofsy, D.S Bartlett, M.C. Shipham, D.J. Jacob, J.M. Hoell, R.J. Bendura, J.W. Drewry, R.J. McNeal, R.L. Navarro, R.N. Gidge, and V.E. Rabine. 1992. The Arctic Boundary Layer Expedition (ABLE 3A): July-August 1988. *J. Geophy. Res*. 97:16383-16394.

Herman, J.R., P.K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier. 1997. Global distribution of UV-absorbing aerosol from Nimbus-7/TOMS data. *J. Geophys. Res*. 102:16911-16922.

Hofmann, D.J., R.S. Stone, M.E. Wood, T. Deshler, and J.M. Harris. 1998. An analysis of 25 years of balloon borne aerosol data in search of a signature of the subsonic commercial aircraft fleet. *Geophy. Res. Lett*. 25: 2433-2436.

Holben B.N., T.F. Eck, I. Slutsker, D. Tanre, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan, Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov. 1998. AERONET - A federated instrument network and data archive for aerosol characterization. *Rem. Sens. Environ*. 66:1-16.

Husar, R.B., J.M. Prospero, and L.L. Stowe. 1997. Characterization of tropospheric aerosols over the oceans with the NOAA Advanced Very High Resolution Radiometer Optical Thickness Operational Product. *J. Geophys. Res*. 102:16889-16909.

IPCC. 1996. Climate Change 1995. The Science of Climate Change, Intergovernmental Panel on Climate Change, J.T. Houghton, L.G. Meira Filho, B.A. Callander, N. Harris, A. Kattenberg, and K. Maskell, eds., Cambridge University Press, 572 pp.

Jacob, D.J., J.A. Logan, and P.P. Murti. 1999. Effect of rising Asian emissions on surface ozone in the United States. *Geophys. Res. Lett.* 26:2175-2178.

Jaffe, D.A., T. Anderson, D. Covert, R. Kotchenruther, B. Trost, J. Danielson, W. Simpson, T. Berntsen, S. Karlsdottir, D. Blake, J. Harris, and G. Carmichael. 1999. Transport of Asian air pollution to North America. *Geophys. Res. Lett*. 26:711-714.

Jaffe, D., T. Anderson, D. Covert, B. Trost, J. Danielson, W. Simpson, D. Blake, J. Harris, and D. Streets. 2001. Observations of Ozone and Related Species n the Northeast Pacific During the PHOBEA Campaigns: 1. Ground based observations at Cheeka Peak. *J. Geophys.Res*., 106:7749.

Johnson, C.E., W.J. Collins, D.S. Stevenson, and R.G. Derwent. 1999. The relative roles of climate and emissions changes on future oxidant concentrations. *J. Geophys. Res*. 104:18631-18645.

King, M.D., Y.J. Kaufman, D. Taure, and T. Nakajima. 2000. Remote sensing of tropospheric aerosols from space: Past, present and future. *Bull. Amer. Meteor. Soc*. 80:2229-2259.

Kreidenweis, M., L.A. Remer, R. Bruintjes, and O. Dubovik. 2001. Observations of smoke aerosol from biomass burning in Mexico. *Journal of Geophysical Research*. 106:3569-3590.

Kritz, M. 1990. The China Clipper-fast advective transport of radon rich air from the Asian boundary layer to the upper troposhere near California. *Tellus*. 42B:46-61.

Lamb, B., H. Westberg, G. Allwine, and T. Quarles. 1985. Biogenic hydrocarbon emissions from deciduous and coniferous trees in the United States. *J. Geophys. Res*. 90:2380-2390.

Lin C.-Y.C. , D.J. Jacob, J.W. Munger, and A.M. Fiore. 2000. Increasing background ozone in surface air over the United States. *Geophys. Res. Lett*. 27:3465-3468.

Logan, J.A. 1999. An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models, and development of a gridded climatology for tropospheric ozone. *J. Geophys. Res*. 104:16115-16149.

MacCracken, M.C., R.D. Cess, and G.L. Potter. 1986. Climatic effects of anthropogenic Arctic aerosols: An illustration of climate feedback mechanisms with one- and two-dimensional climate models. *J. Geophys Res*. 91:14445-14450.

Malm, W.C., J. Sisler, D. Huffman, R. Eldred, and T. Cahill. 1994. Spatial and seasonal trends in particle concentration and optical extinction in the U.S. *J. Geophys. Res*. 99:1347-1370.

Marenco, A., V. Thouret, P. Nedelec, H. Smit, M., Helten, D. Kley, F. Karcher, P. Simon, K. Law, J. Pyle, G. Poschmann, R. Van Wrede, C. Hume, and T. Cook. 1998. Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, An overview. *J. Geophys. Res*. 103:25631-25642.

Merrill, J.T., M. Uematsu, and R. Bleck. 1989. Meteorological analysis of long-range transport of mineral aerosols over the North Pacific. *J. Geophys. Res*. 94:8584-8598.

Nakicenovic, N. and R. Swart (eds.). 2000. Special Report on Emissions Scenarios. Report of Working Group III of the Intergovernmental Panel on Climate Change. Cambridge University Press. 599 pp.

NRC (National Research Council). 1996. Aerosol radiative forcing and climate change. National Academy Press, Washington D.C., 161pp.

NARSTO (North American Research Strategy for Tropospheric Ozone). 2000. An Assessment of Tropospheric Ozone Pollution: A North American Perspective. Available online at: www.cgenv.com/Narsto/.

Peppler, R.A. 2000. ARM Southern Great Plains Site Observations of the Smoke Pall Associated with the 1998 Central American Fires. *Bull. Am. Met. Soc*. 81:2563-2591.

Perry, S., T. Cahill, R. Eldred, D. Dutcher, and T. Gill, 1997. Long-range transport of Saharan dust into the eastern United States. *J. Geophys. Res*. 102:11225-11238.

Pincus, R. and M.B. Baker. 1994. Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer. *Nature*. 372:250-252.

Porter, P.S., S.T. Rao, I.G. Zuebenko, A.M. Dunker, and G.T. Wolff. 2001. Ozone air quality over North America: Part II An Analysis of trend detection and attribution techniques. *J. Air and Waste Mgmt. Assoc*. 51:283-306.

Prados, A.I., R.R. Dickerson, and J.T. Merrill. 1999. Transport of ozone and pollutants from North America to the North Atlantic Ocean during the 1996 Atmosphere/Ocean Chemistry Experiment (AEROCE) intensive. *J. Geophys. Res*. 104:26219-26233.

Prins, E.M., J.M. Feltz, and D.E. Ward. 1998. An overview of GOES-8 diurnal fire and smoke results for SCAR-B and 1995 fire season in South America. *J. Geophys. Res*. 103:31821-31835.

Prospero, J.M. 1999. Long-term measurements of the transport of African mineral dust to the southeastern United State: Implications for regional air quality. *J. Geophys. Res*. 104:15917-15927.

Prospero, J.M., and D.L. Savoie. 1989. Effects of continental sources on nitrate concentrations over the Pacific Ocean. *Nature*. 339:687-689.

Schwele, D. 1995. Public Health Implications of Urban Air Pollution in Developing Countries. Proceedings of the 10th World Clean Air Congress, Espoo Finland, 28 May - 2 June.

Shindell, D.T., D. Rind, D., and P. Lonergan. 1998. Increased polar stratospheric ozone losses and delayed eventual recovery owing to increasing greenhouse-gas concentrations. *Nature*. 392:589-592.

Singh, H.B., and D.J. Jacob. 2000. Future directions: Satellite observations of tropospheric chemistry. *Atmospheric Environment*. 34:4399-4401

Stevenson, D.S., C.E. Johnson, W.J. Collins, R.G. Derwent, and J.M. Edwards. 2000. Future estimates of tropospheric ozone radiative forcing and methane turnover – the impact of climate change. *Geophys. Res. Lett*. 27:2073-2076.

Tingey, D.T., M. Manning, L.C. Gorthaus, and W.F. Burng. 1979. The influence of light and temperature on isoprene emission rates from live oak. *Physiol. Plant*. 47:112-118.

Thouret, V. J., Y.N. Cho, R. Newell, A. Marenco, and H.G.J. Smit. 2000. General characteristics of tropospheric trace constituent layers observed in the MOZAIC program. *J. Geophys. Res*. 105: 17379-17392.

Uematsu, M., R.A. Duce, J.M. Prospero, L. Chen, J.T. Merrill, and R.L. McDonald. 1983. Transport of mineral aerosol from Asia over the North Pacific Ocean. *J. Geophys. Res*. 88:5343-5352.

UNEP (United Nations Environment Program). 1999. Global Environment Outlook. Earthscan publications Ltd., London. 398 pp.

Wilkening, K.E., L.A. Barrie, and M. Engle. 2000. Trans-Pacific Air Pollution. *Science*. 290:65-67.

Wolff, G.T., A.M. Dunker, S.T. Rao, P.S. Porter, and I.G. Zurbenko. 2001. Ozone air quality over North America: Part I - A review of reported trends. *J. Air and Waste Mgmt. Assoc*.51:273-282.

Wotawa, G., and M. Trainer. 2000. The influence of Canadian forest fires on pollutant concentrations in the United States. *Science*. 288:324-328.

WMO (World Meteorological Organization). 1999. Scientific Assessment of Ozone Depletion: 1998. Global Ozone Research and Monitoring Project - Report No. 44. Geneva, Switzerland. 732 pp.

Xiao, H., G.R. Carmicheal, J. Durchenwald, D. Thornton, and A. Bandy. 1997. Long-range transport of SO_X and dust in East Asia during the PEM-B Experiment. *J. Geophys. Res*. 102:28598-28612.

Yienger J.J., M.Galanter, T.A. Holloway, M.J. Phadnis, S.K. Guttikunda, G.R. Carmichael, W.J. Moxim, and H.L. Levy II. 2000. The episodic nature of air pollution transport from Asia to North America. *J. Geophy. Res*. 105:26931-26945.

Acronyms/Abbreviations

ACE	Aerosol Characterization Experiments
AERONET	Aerosol Robotic Network
AGAGE	Advanced Global Atmospheric Gases Experiment
AOT	Aerosol Optical Thickness
AVHRR	Advanced Very High Resolution Radar
CAC	Committee on Atmospheric Chemistry
CFC	Chlorofluorocarbon
CMDL	Climate Monitoring and Diagnostics Laboratory
EOS	Earth Observing System
GHG	Greenhouse gas
GOES	Geostationary Operational Environmental Satellites
GOME	Global Ozone Monitoring Experiment
GTCM	Global chemical transport model
HALOE	Halogen Occultation Experiment
HCFC	Hydrochlorofluorocarbon
HFC	Hydrofluorocarbon
HRDLS	High Resolution Dynamics Limb Sounder
IASI	Infrared Atmospheric Sounding Interferometer
ILAS II	Improved Limb Atmospheric Spectrometer-II
IMPROVE	Interagency Monitoring of Protected Visual Environments
INDOEX	INDian Ocean EXperiment
IPCC	Intergovernmental Panel on Climate Change
LRTAP	Long-Range Transboundary Air Pollution (Convention)

MIPAS	Michelson Interferometer for Passive Atmospheric Sounding
MISR	Multi-Angle Imaging Spectro-Radiometer Project
MLS	Microwave Limb Sounder
MODIS	Moderate-Resolution Imaging Spectrometer
MOPITT	Measurement of Pollution in the Troposphere
MOZAIC	Measurement of OZone by Airbus In-service airCraft
NAAQS	National Ambient Air Quality Standards
NARSTO	North American Research Strategy for Tropospheric Ozone
NOAA	National Oceanic and Atmospheric Administration
NPOESS	National Polar-orbiting Operational Environmental Satellite System
NRC	National Research Council
OMI	Ozone Monitoring Instrument
OMPS	Ozone Mapping and Profiler Suite
PICASSO-CENA	Pathfinder Instruments for Cloud and Aerosol Spacebourne Observations - Climatologie Etendue des Nuages et des Aerosols
PM	Particulate matter
POAM III	Polar Ozone and Aerosol Measurement III
PPB	Parts Per Billion
PPM	Parts Per Million
SAGE III	Stratospheric Aerosol and Gas Experiment
SCIAMACHY	Scanning Imaging Absorption SpectroMeter for Atmospheric Chartography
SHADOZ	Southern Hemisphere ADditional OZonesondes
SIO	Scripps Institution of Oceanography
TES	Tropospheric Emission Spectrometer
TOMS	Total Ozone Mapping Spectrometer
TRACE-P	TRAnsport and Chemical Evolution over the Pacific