

CHAPTER 3

ATMOSPHERIC CHEMISTRY, AEROSOLS AND SOLAR RADIATION

3.0 INTRODUCTION

The Earth atmosphere is the fluid that most directly connects the other components of the Earth System (the oceans, marine and terrestrial ecosystems, geosphere and cryosphere) and provides the medium in which these components interact. It is a chemically complex and dynamic mixture whose composition and structure are determined by competing processes. Changes in that balance, caused by natural phenomena or human action, can strongly influence life on Earth, either directly through change in atmospheric composition, or indirectly through chemical impacts on climate and the biosphere. Atmospheric change is the result of strongly interactive chemical and physical processes. Atmospheric radiation transfer and temperature depends on chemical composition and the distribution of atmospheric aerosols, while the nature and rates of chemical processes depend on temperature. As a result, chemistry plays a crucial role in determining the Earth climate, while the physics and dynamics of the atmosphere influence chemical processes and composition.

The goals are to measure and understand how and why the global atmospheric composition, aerosol distribution and properties, and solar radiation are changing in response to natural and anthropogenic forcings, and to enable accurate prediction of future changes in climate, ozone distribution, surface ultraviolet radiation, and global pollution.

3.1 MAIN SCIENCE QUESTIONS

- *How is stratospheric ozone changing as the abundance of ozone-destroying chemicals decreases?*

It has been established that the primary cause of the stratospheric ozone depletion observed over the last two decades is an increase in the concentrations of chlorofluorocarbons (CFCs) and other halogen-containing hydrocarbons of industrial origin. The depletion has been significant, ranging from a few percent per decade at mid-latitudes to greater than fifty percent seasonal losses at high latitudes, notably the annually recurring Antarctic ozone hole, as well as smaller but still large, winter/spring ozone losses observed recently in the Arctic. Reduction in atmospheric ozone amounts leads to increased flux of ultraviolet radiation at the surface, with harmful effects on plant and animal life, including human health.

In response to these findings, the nations of the world have ratified the *Montreal Protocol on Substances That Deplete the Ozone Layer* and agreed to phase out the production of several halogen-containing chemicals. Concentrations of many such compounds are now beginning to decrease in the lower atmosphere, and there is some evidence that ozone-depleting substances are approaching their anticipated peak levels in the stratosphere. All other things being equal, as the halogen burden falls in response to regulation, stratospheric ozone concentration should begin to recover. However, the future behavior of ozone will also be affected by the changing abundance of water vapor, methane, nitrous oxide, sulfate aerosols and changing temperature. Thus, for a given atmospheric halogen burden, stratospheric ozone amounts will not be the same in the future as it was in the past.

One of the principal challenges to atmospheric chemistry research in the coming decade are to identify the signs of ozone recovery and to verify that CFC substitutes and other industrially produced halogenated compounds are indeed decreasing as expected from the application of the Montreal Protocol. In addition to climate change, transient phenomena like volcanic eruptions and particle precipitation events, variations in solar irradiance and atmospheric circulation also affect stratospheric temperature, water vapor and aerosols and, in turn, influence stratospheric ozone. Systematic observation is crucial to understand the origins of observed ozone changes and their causes, natural or human. NASA's ozone observation and upper atmosphere research plans are consistent with the responsibility defined by federal legislation (NASA Authorization Act and the US Clean Air Act).

▪ ***What trends in atmospheric constituents and solar radiation are driving global climate?***

The increasing concentrations of long-lived trace constituents that absorb infrared radiation, such as carbon dioxide, nitrous oxide and methane, are primary forcing factors of global climate change. The growth rates of these constituents in the atmosphere show considerable variations, that are currently not well understood. Quantifying the sources and sinks of these constituents is the basic issue for establishing future greenhouse gas concentration scenarios. Transient variations and changes in the concentrations of shorter-lived greenhouse gases involve a more complex combination of dynamical, physical, and chemical processes. Ozone depletion in the lower stratosphere introduces a cooling tendency, while the increase in tropospheric ozone introduces a warming tendency. Water vapor in the upper troposphere and lower stratosphere likewise has a powerful greenhouse effect.

Stratospheric aerosols, such as that result from large volcanic eruptions (most recently Mt. Pinatubo in 1991) can significantly cool the Earth's climate, as has been demonstrated by several recent events. Tropospheric aerosols, on the other hand, can either cool or warm the atmosphere, depending upon their properties and the albedo of the underlying surface. Tropospheric aerosols also influence climate indirectly by modifying cloud microphysical properties and lifetime. The details of aerosol impacts on atmospheric radiation strongly depend on the nature of the aerosols. Thus, a major scientific challenge is understanding the processes that lead to aerosol formation, and the diversity of aerosol types and chemical compositions.

Solar radiation is the only climate forcing factor that is truly external to the Earth system. We know that the Sun's radiant energy output has increased materially since the distant past, apparently without any drastic change in Earth's climate – thus pointing to the existence of stabilizing feedback mechanisms operating over very long time scales. Over shorter time periods, however, any change in solar activity and radiation will affect the Earth's radiation budget and climate. Furthermore, solar radiation is considerably more variable in the ultraviolet part of the spectrum than at longer wavelengths (5% over a solar cycle in the spectral range involved in stratospheric ozone production). The extent to which resulting variations in stratospheric composition and thermal structure influence climate in the lower atmosphere remains to be quantified. Monitoring solar radiation (both total and spectrally resolved) is indispensable to account for solar-induced variations in atmospheric climate and chemistry.

▪ ***How do stratospheric trace constituents respond to climate change and chemical agents***

Changes in physical climate affect the distributions of trace constituents (e. g. ozone), and vice versa. As the already depleted ozone layer enters the period of its greatest vulnerability, recently detected stratospheric cooling – a predicted side effect of both ozone depletion and increased concentrations of other greenhouse gases – may delay the recovery of the ozone layer and enhance its susceptibility to natural or human-induced perturbations. For six out of the last nine years, unusually low late-winter to

early-spring ozone concentrations have been observed in the Arctic stratosphere, in conjunction with unusually cold and protracted stratospheric winters. In addition to these polar effects, increasing abundance of greenhouse gases can also affect the climate, chemistry and ozone content of the middle latitude stratosphere.

Chemical reactions that occur on the surface of stratospheric aerosol and/or cloud particles are temperature dependent; even a small decrease in temperature could cause a significant increase in the rates of these reactions. Changes in stratospheric water vapor, associated with changes in fluxes through the tropopause, could enhance the formation of aerosol and cloud particles that facilitate ozone destruction. Furthermore, a decrease in ozone amount would result in a reduction of solar UV absorption, further cooling, further enhancing ozone loss (a positive feedback). There is already strong evidence of cooling temperatures in the lower stratosphere, one of the largest climate signals measured in the atmosphere over the past 20 years.

Hydroxyl radicals (OH) formed largely from ozone and water vapor are the principal oxidizing agents in the atmosphere. Any changes in hydroxyl amounts, especially in the troposphere, will affect the lifetime and distribution of many trace constituents, including ozone, chlorofluorocarbon replacements, and even slower-reacting species like methane. Changes in hydroxyl will also affect the formation of aerosol. In addition to its direct impact on climate, the troposphere is the ultimate source and sink for stratospheric trace constituents; tropospheric interactions between meteorological, physical and chemical processes are still far from being adequately understood. The spatial and temporal variability of both long-lived and short-lived chemical constituents is a major impediment to scientific knowledge of global effects, such as radiative forcing of climate and long-range dispersion of pollutants.

Improving our understanding of this highly interactive system calls for detailed investigation of the relationships between the distributions of ozone, water vapor, aerosols, temperature, and relevant trace constituents, notably chlorine and bromine compounds and nitrogen oxides. In view of the high spatial variability of these constituents, good horizontal and vertical resolution will be needed, especially in the vicinity of the tropopause (upper troposphere and lower stratosphere).

- *What are the effects of regional pollution on the global atmosphere, and the effects of global chemical and climate changes on regional air quality?*

With the growth of population, economic activity and use of fossil fuels, particularly in Asia and Latin America, emissions of pollutant gases such as carbon monoxide, sulfur dioxide, and oxides of nitrogen will undoubtedly increase. There is now significant evidence that such gases can be transported over very large distances (e.g., across the Pacific or Atlantic oceans). Aerosol from biomass burning and effluents from fossil fuel combustion have been readily detected in the lower troposphere thousands of miles from their sources. The global effects of atmospheric pollution are poorly known because natural emissions of trace constituents, removal by wet and dry deposition and chemical reactions in the atmosphere or at the Earth's surface are not well characterized globally.

The combined effect of increased pollutant levels and changing temperatures, winds, and precipitation patterns must also be considered. Unlike the relatively calm stratospheric environment, the troposphere is a very active medium. Convection is a major element of complexity, effectively coupling the upper and lower troposphere. In particular, large scale atmospheric motions move pollutants around in ways that are greatly complicated by the significant geographical and seasonal variations in convection, which in turn plays a key role in establishing the vertical mixing of pollutants and, ultimately, their global distribution. Other complicating factors are the condensation, re-evaporation or precipitation of water, which affects the concentrations of soluble trace gases and aerosols, and lightning, which acts as a strong source of reactive constituents where it occurs. Aircraft traffic that results in the emission of ozone and aerosol

precursors, and other effluents directly in the upper troposphere and stratosphere may also be a factor. Further advances in the knowledge of tropospheric chemistry will require integrating chemical and meteorological knowledge in comprehensive interdisciplinary studies.

Anthropogenic emissions transported on a global scale enhance the background concentrations of pollutants in populated regions far downwind from their source, increasing the severity and scale of pollution episodes in these regions. For example, it has been suggested that rising Asian emissions could adversely affect ozone air quality in the western U.S. It has also been suggested that long-range transport of African dust could contribute to degradation of visibility in the southeastern U.S. As air quality standards become more stringent to respond to increasing public expectations the influence of rising background concentrations becomes an increasingly important issue. Changes in atmospheric stability and circulation due to regional pollution is another mechanism by which perturbations in atmospheric composition could induce changes in regional air quality.

Understanding and predicting tropospheric transport, physics and chemistry will be a frontier area of atmospheric research for the next decade and probably beyond. Tropospheric chemistry poses an interdisciplinary challenge for the Earth science community. These are global scale problems that are particularly well suited to the use of space observations and correlative *in situ* measurements. Knowledge of these phenomena is needed to evaluate the effects of chemical changes on the global hydrological cycle, the cycling of nutrient compounds through the earth environment, the accumulation of greenhouse gases, the acidity of rain and snow, and the formation of ozone in the lower troposphere.

3.2 SCOPE AND NATURE OF THE PROBLEM

The main science questions map onto scientific problems that go well beyond the traditional discipline of atmospheric chemistry. Close collaboration between atmospheric chemists, radiation science specialists, meteorologists, and interdisciplinary scientists interested in biogeochemical cycles will be needed to successfully address these issues.

Stratospheric Ozone

How is the global distribution of ozone changing and how will it evolve in the future, given likely changes in both industrial activities and the underlying climate? Changes in the amount of ozone in the Earth's atmosphere that occurred over the last few decades are well documented by both ground- and space-based measurements. Statistically significant decreases in total column ozone amounts have been observed over most of the globe (with exception of the tropics), including depletion rates on the order of several percent per decade at mid-latitudes. Considerably larger losses have been observed at high latitudes, especially in the Antarctic spring where major ozone depletion (the "ozone hole") occurs systematically every year. The ozone loss corresponds to more than 50% of the original total column amount during Austral spring and temporarily reaches almost 100% at some lower stratospheric altitudes. In the Arctic, no such systematic ozone loss has taken place, but large decreases in total column amount have been observed in late winter and early spring, reaching 50% during one particular winter (1996-97).

Ground-, aircraft-, and space-based observations have shown that chemical reactions involving industrial halogen products, whose concentrations are affected by chemical reactions occurring at the surface of aerosol and cloud particles, are primarily responsible for ozone depletion in the lower stratosphere. The concentration of chlorine in the stratosphere, now above 3.5 parts per billion by volume, dramatically exceeds the level (~0.6 parts per billion) that would exist in the pristine stratosphere unaffected by industrial activity. Bromine is also found in the stratosphere at levels much greater than would occur in the absence of industrial activity. Chemical reactions occurring at the surface of aerosol and cloud particles are a necessary intermediate step to explain the magnitude of both polar ozone depletion (polar stratospheric clouds) and mid-latitude depletion in the lower stratosphere (sulfate aerosols). The amount of sulfate aerosols present can be greatly enhanced by oxidation of sulfur dioxide injected directly into the stratosphere by large volcanic eruptions. The conversion of SO_2 to H_2SO_4 takes place with a time constant of a month, with subsequent decay of the resulting aerosol over a period of 3-5 years. The large reduction in global ozone amount that occurred subsequently to the eruption of Mt. Pinatubo in June, 1991 was definitely shown to result from such aerosol-induced chemical reactions.

Global measurement by surface-based networks have shown that the implementation of the Montreal Protocol on Substances that Deplete the Ozone Layer and its subsequent amendments has achieved a significant reduction in the atmospheric concentrations of several chlorine-containing compounds (carbon tetrachloride, methyl chloroform). Growth in other source gases, such as trichlorofluoromethane (CFC 11), has essentially ceased. Satellite, balloon, and aircraft measurements have shown evidence of slower growth of chlorine levels in the stratosphere. As emission controls become more effective, stratospheric chlorine levels are expected to begin declining during the next decade. On the other hand, bromine-containing halons, also regulated under the Montreal Protocol, continue to increase at rates which exceed predictions and could delay the expected recovery of stratospheric ozone if the current trends continue into the next decade.

The sources and sinks of halogenated hydrocarbons, such as the hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs), which serve as replacements for the banned chlorofluorocarbons (CFCs), are not well understood. The concentrations of HCFCs and HFCs are now growing rapidly in the atmosphere. This lack of understanding is especially evident for methyl bromide, which has both

industrial sources and natural sources and sinks at the land surface and in the ocean. Methyl bromide is the primary source of bromine that can reach the Earth's stratosphere. Significant uncertainties exist in the global budget of methyl bromide and the strength of surface sources, even to the point where it is not known which regions are sources or sinks. This issue is complicated by the fact that the cycling of methyl bromide between the surface and the atmosphere may be quite rapid, and the net accumulation is a relatively small residual between larger production and removal terms. There is also increasing interest in understanding the sources of naturally-occurring chlorinated hydrocarbons, about which little is known, as well as the role of the oceans as a sink for chlorinated hydrocarbons, such as methyl chloroform and carbon tetrachloride. These unresolved questions highlight the need for improved understanding and continued monitoring of surface concentrations of the full range of industrially produced halocarbons.

Ozone recovery can be affected by climate-related changes. Stratospheric cooling, increase in the concentrations of stratospheric water vapor or other chemically-active molecules, and change in circulation patterns that affect stratosphere-troposphere exchanges or the stability and temperature of winter polar vortices, could all have significant impacts on future ozone levels. It is conceivable that climate change might be at least partly responsible for large Arctic ozone losses observed in recent winters. These findings indicate that verifying the recovery of stratospheric ozone will be complicated by the large interannual variability in the stratospheric circulation, which needs to be documented as well. In addition, the mechanisms coupling climate change, the flow of wave energy between the troposphere and the stratosphere, and the stability of the polar vortices need to be understood. Finally, the timing of ozone recovery will also be affected by changes in the amount and distribution of stratospheric aerosols (see below).

Aerosol Chemistry, Sources and Sinks

The role of aerosols in the global environment has received increased attention in recent years, notably because the global-mean radiative forcing associated with anthropogenic aerosols could be on the same order of magnitude (but with the opposite sign) as greenhouse gas forcing (IPCC, 1995). In addition, aerosols play an important catalytic role in atmospheric chemistry (notably ozone-destroying reactions) and contribute significantly to surface deposition of nutrients as well as particulate matter that may be harmful to human health. In particular, terrestrial and marine aerosol transported over long distances from their sources can provide critical nutrient subsidies, such as iodine transported by salt aerosols into the interior of continents or iron from mineral dust deposited in the oceans. The ocean ecosystem is critically dependent upon such iron inputs to sustain nitrogen fixation and photosynthesis by phytoplankton. As the mobilization of iron-rich mineral dust from desert regions is related to winds and aridity, a feedback process exists between the hydrological cycle and the rate of absorption of atmospheric CO₂ mediated by biological processes in the ocean.

The complexity of aerosol physics and chemistry, their space and time variability, and the diversity of aerosol compositions and particle sizes, add up to a daunting challenge for both experimental research and modeling. The primary cause of the extreme diversity of aerosol types and properties is the multiplicity of aerosol sources: sea spray, smoke from fires, volcanic ash and mineral dust from deserts and arid regions, smoke from fires, and atmospheric formation of sulfate particles from the oxidation of sulfur-containing compounds from natural or industrial sources. Biological production of dimethyl-sulfide (DMS), dimethyl-sulfonium propionate (DMSP) and, ultimately, sulfur-based aerosols by oceanic phytoplankton may constitute an important feedback mechanism between oceanic biological productivity and climate. Volcanic eruptions can inject enormous amounts of ash and SO₂ in the troposphere and stratosphere. The lofting of mineral dust under dry and windy meteorological conditions is partly controlled by human-induced changes in land use and agricultural practices. Biomass burning, particularly in the tropics, is a major source of tropospheric aerosol, and an important contributor to global budgets of radiatively and chemically active trace gases.

Stratospheric aerosols, linked to volcanic eruptions, are at their lowest level since the inception of global aerosol measurements some 20 years ago. Observations of these low values, coupled with better information on the concentrations of aerosol precursors and knowledge of aerosol microphysics, have called into question established ideas about the origins of background aerosols. The contribution of aviation to stratospheric aerosol loading is an open question. Quantifying the processes that govern the stratospheric aerosol background will be an important scientific goal, should injection by volcanic eruption remain small.

Atmospheric Processing and Removal of Trace Constituents and Aerosols

The global burden of atmospheric methane has more than doubled since pre-industrial times, accounting for about 20% of the estimated climate forcing by anthropogenic greenhouse gas emissions. Yet analysis of measurements for the period 1984-1996 reveals that the methane growth rate has varied considerably and recently slowed down; the origin of these variations is not understood. The cause of this decline in methane growth rate could be due to an increase in the atmospheric sink resulting from the increase in the concentration of tropospheric OH, although a decrease in methane emissions cannot be ruled out. If the current trend persists, the methane concentration in the atmosphere could stabilize at a level only 5% above those existing today. This new finding underscores the importance of acquiring consistent global methane concentration records, as well as improved understanding of the processes that control methane distribution. These include methane emission from both natural and anthropogenic sources and methane destruction dominated by reaction with the hydroxyl radical, the concentration of which may also be changing.

The observed increase in upper stratospheric water vapor goes well beyond that which could result from an increase in production from the oxidation of methane, suggesting larger water transport from the troposphere to the stratosphere. Such an increase could result from a relatively small warming of tropopause temperature (not substantiated by observation yet). Another difficulty in ascertaining trends in stratospheric water vapor is the high degree of variability observed in upper tropospheric water vapor. Better measurements of upper tropospheric water vapor and temperature will improve our understanding of this important global change parameter.

Global satellite observation have led to rapid advances in knowledge of the distribution of tropospheric aerosols, especially those resulting from long-range transport of mineral desert dust, smoke from biomass burning, sulfate aerosols, and volcanic ash. *In situ* measurements have shown the composition of tropospheric aerosols to be considerably more complex than previously thought, including the presence of large amounts of organic compounds in some cases. Once airborne, the fate of aerosol particles can vary enormously. In some regions, aerosol particles serve as cloud condensation nuclei and are thus removed from the atmosphere. At other locations, aerosols can be transported over long distances, e.g. the observed transatlantic transport of mineral dust from the Sahara desert or the near complete circuit of the Earth made in the mid-troposphere by aerosol particles originating from biomass burning. Hygroscopic aerosols are particularly susceptible to transformation in the moist troposphere; their particle size distributions "age" as a result of such transformation, with corresponding changes in sedimentation rate and optical properties. In the stratosphere, aerosol particles formed by oxidation of sulfur dioxide have an e-folding time of nearly a year and disperse over the whole Earth; their global distribution is a sensitive indicator of the effectiveness of large-scale atmospheric transport and mixing. In view of the complexity of the dynamical, physical and chemical processes involved, developing reliable representation of aerosols in atmospheric models is a daunting scientific challenge.

Solar Radiation Forcing of Atmospheric Chemistry and Climate

The Sun is a mildly variable star that exhibits cyclical variations in its internal activity and radiation output. The primary external forcing of the Earth climate is produced by variations in the Sun's total energy output, which are currently too small (less than 0.2% over an 11 year solar cycle) to induce significant changes in the lower atmosphere. The much larger variability in solar ultraviolet radiation does affect the chemistry and composition of the stratosphere and upper atmosphere. Variations are a few percent in the ultraviolet spectrum range involved in stratospheric ozone production, but as large as a factor 2 at the shorter wavelengths that affect photochemistry in the upper atmosphere (above the stratosphere). Changes induced by solar variability in the stratosphere could potentially affect the tropospheric general circulation and climate. Neither the mechanisms nor the extent of these interactions are definitely known at present.

Chemistry-Climate Connection

Climate change can affect atmospheric chemistry in several ways. Temperature affects reaction rates and the natural emission rates of source gases. Changes in atmospheric circulation can modify the transport of radiatively and chemically active trace constituents within the troposphere and stratosphere, as well as across the tropopause. Weather phenomena determine the location and duration of persistent meteorological conditions that enhance air pollution or cause its removal from the atmosphere. Aerosol formation and microphysics are affected by atmospheric temperature and moisture. Conversely, ozone, other chemically active trace constituents, and aerosols contribute significantly to the atmospheric greenhouse effect and can induce changes in the atmospheric circulation and climate. Understanding the linkages between atmospheric chemistry and climate change is a major multi-disciplinary challenge for Earth system science.

On the other hand, half the total increase in global greenhouse effect since the beginning of the industrial era is due to the combined radiative effect trace constituents other than CO₂. On a per molecule basis, compounds such as methane, nitrous oxide, chlorofluorocarbons, and ozone are significantly more effective greenhouse gases than CO₂. One explanation of the apparent discrepancy between climate model simulations and the global warming trend observed over the last 30-40 years is an underestimation of the (negative) radiative forcing due to anthropogenic aerosols. Knowledge of the time-dependent 3-dimensional distribution of aerosol will be needed in order to discriminate between natural and anthropogenic sources. The same information is also needed to exploit opportunities for natural "climate forcing experiments" offered by volcanic eruptions that inject large amounts of sulfur dioxide into the stratosphere and can enhance the stratospheric aerosol loading for periods of several years.

The optical properties of aerosol are complex functions of number density, particle size distribution, particle shape, and chemical composition of the material. The direct radiative forcing caused by aerosols depends upon all these properties and also, most importantly, upon masking by clouds and the underlying surface albedo. In addition, indirect radiative forcing can be induced by certain classes of aerosol acting as cloud condensation nuclei (See section 4.3.2 in the next chapter).

Global Pollution

There is growing evidence that increasing regional air pollution contributes significantly to planetary scale changes in atmospheric trace constituents, including precursors for ozone formation, other oxidants, and particulate matter. The development of Asian economies and the growing use of fossil fuels will lead to significant increases in air pollution in East Asia and to degradation of air quality in currently clean air regions of the Pacific Basin. A significantly increased pollution plume from Asia could potentially affect air quality on the West Coast of the United States. This is just one example of cross-boundary pollution issues that will assume increasing importance in the next few decades. Large plumes of pollutants are transported to the free troposphere through convection and travel great distances

downwind of their sources. Observations made over the tropical Pacific Ocean and off the west coast of North America show clear evidence of such transport into regions that would otherwise be free of pollutants. Effluents from African and South American biomass burning has been observed over the tropical Atlantic. African and South American plumes are seen over the central and Eastern South Pacific Ocean. Mineral dust from the Saharan desert crosses the Atlantic Ocean all the way to the Caribbean Sea and, in some instances, the southeastern US. The consequences of long-range transport of atmospheric pollutants need to be quantified so that non-local influences can be factored into local changes in atmospheric chemistry and pollution.

3.3 NASA PROGRAM ELEMENTS

The NASA research strategy in atmospheric chemistry involves four categories of activities: process studies, systematic observation of the distribution of relevant quantities, model simulations and retrospective analysis of past observation records, and model predictions of future atmospheric changes. Scientific breakthroughs in understanding the complex problems described in the previous section come from the synergy of these four research elements.

Process-level understanding is needed to form an accurate conceptual picture of how chemical constituents and aerosol particles form, interact, and are transported in the atmosphere. Laboratory experiments provide much of the detailed information on gas-phase reactions of trace constituents, the formation of aerosol particles from chemical precursors, fundamental chemical and physical properties of aerosol particles, and chemical reactions that occur at the surface of aerosol and cloud particles. Theoretical knowledge can also be applied to understand aerosol particle formation and growth. Information on transport processes comes from the interpretation of detailed atmospheric constituent distribution data, as well as knowledge of the atmospheric circulation.

Ground-, aircraft-, balloon-, and space-based measurements are used to determine the concentration of chemical constituents, the distribution and properties of aerosol particles, transport between different regions of the atmosphere, and atmospheric temperature and wind fields. Parameters that affect atmospheric processes, such as solar ultraviolet radiation and energetic particle fluxes, also need to be documented. Observations range over all spatial and temporal scales, from short-term, focused studies of a particular region of the atmosphere, to systematic global measurement by satellites and surface-based networks.

Detailed comparison of observations with the results of model simulations test both our understanding of processes as they occur in the atmosphere and our understanding of the past evolution of atmospheric composition in relation to changes in halogen abundance, variations in solar radiation, and effects of volcanic eruptions. Models range in complexity from relatively simple photochemical box models, that represent chemistry within each atmospheric volume assuming no atmospheric transport, to fully interactive 3-dimensional models that can simulate chemical, radiative, and transport processes.

Predictions of the future changes in atmospheric composition and physical properties usually require fully interactive multi-dimensional models representing atmospheric chemistry and transport. Much of this activity has been applied to international assessments, especially the Scientific Assessment of Ozone Depletion: 1998, by the World Meteorological Organization and the United Nations Environment Programme (WMO, 1998). Such models have been used, for example, to predict the response of the atmosphere to the different halogen loading scenarios that could result from the Montreal Protocol (e.g. speeded up phase-out of some compounds). Important assessment activity has also been carried out to support the NASA Office of Aero-Space Technology to estimate the atmospheric effects of aviation. Even more ambitious model developments attempt to simulate detailed interactions and feedback between meteorological, chemical, and radiative processes, such as gas and mixed phase chemical processes in the troposphere.

Particulate matter is another type of trace constituent with unique physical and chemical properties. A comprehensive aerosol research program must consider processes that cut across all disciplines of Earth system science, including: (i) aerosol production, transport, and scavenging (ii) impacts on radiation transfer, (iii) impacts on chemical reactions and, (iv) nucleation processes. Advances in global aerosol science so far have been based mainly on inferences from model simulations built on much theoretical knowledge and limited observational data. The strategy is to reverse this trend and:

- Augment systematic aerosol measurements that can be applied to test global aerosol models (AERONET, first EOS mission series).

- Develop *in situ* airborne measurement campaigns and global observation (PICASSO-CENA research satellite mission) to investigate aerosol properties and processes.

3.3.1 GLOBAL SYSTEMATIC MEASUREMENTS

Currently, the NASA global atmospheric chemistry research program is principally organized around a major global monitoring and discovery mission EOS Aura (2002-2007) as part of the Earth Observing System (EOS) program. EOS Aura will build on the heritage of measurements initiated by the still active UARS spacecraft launched in 1991. Instruments on the EOS Aura mission that are focused on stratospheric chemistry include:

- The High Resolution Dynamics Limb Sounder (HIRDLS) instrument, using infrared emission to measure ozone, water vapor, temperature, and a broad range of trace constituents, including long-lived source gases and most of the important nitrogen-containing species (N_2O , NO_2 , HNO_3 , N_2O_5 , ClONO_2) in the upper troposphere and stratosphere. The high vertical and horizontal resolution of HIRDLS measurements will make a unique contribution to knowledge of stratospheric constituents and the transport of trace gases between different regions of the stratosphere and troposphere. HIRDLS is a joint development of the US and United Kingdom.
- The Microwave Limb Sounder (MLS) using emission in the microwave spectrum to observe the distribution of ozone, water vapor, long-lived tracers like N_2O , halogen compounds and, for the first time, the OH, HO_2 , and BrO radicals. Simultaneous measurements of ClO and HCl will also be made for the first time from space. These observations will allow a critical test of current understanding of the partitioning of chlorine between possible reservoir species. MLS measurements will thus enable the first ever global characterization of catalytic ozone destruction rates through measurements of radicals in all key chemical families (HO_x , NO_x , ClO_x , and BrO_x).
- The Ozone Monitoring Instrument (OMI), supplied by the Netherlands, will measure total column amounts of ozone. The OMI total column ozone data will extend the record obtained from the Total Ozone Mapping Spectrometer (TOMS) and the European Global Ozone Monitoring Experiment (GOME) at higher spatial resolution. OMI will also measure total column amounts of several other trace gases, notably NO_2 , and information on ozone profiles as well.

EOS Aura will also carry the totally new Tropospheric Emission Spectrometer (TES) instrument for exploratory survey of trace tropospheric constituents. The high spectral resolution ($\sim 0.025 \text{ cm}^{-1}$) of the TES instrument will allow vertically resolved measurements of ozone and other trace constituents, including the important nitrogen oxide family that plays a major role in ozone production in the lower atmosphere. Other key tropospheric molecules measured by TES include carbon monoxide and water vapor. TES will operate in both a nadir- and limb-viewing mode, and can be pointed so as to allow observation of interesting regions more than once per day.

The EOS Terra (AM-1) mission will carry another tropospheric instrument the Measurement of Pollution in the Troposphere (MOPITT). This sensor will determine the vertical structure of carbon monoxide (CO) in the troposphere, as well as the total column amounts of CO and methane (CH_4). Combined with other optical measurements on the same platform, MOPITT will determine the relationship between CO concentration, aerosol, smoke, and surface sources such as biomass burning. Partial information on smoke aerosol and the fires responsible will also be available from the Moderate Resolution Imaging Spectrometer (MODIS), another instrument on Terra.

Altogether, the EOS program will provide a vast amount of information on stratospheric chemistry and initiate a new research thrust with the exploration of large-scale chemical processes in the troposphere. From the analysis of EOS data combined with other observations (UARS, aircraft campaigns, and surface-based measurements), NASA expects major advances in stratospheric chemistry that will allow progressing beyond the need for fully comprehensive sets of chemical measurements. In this perspective,

future missions will achieve their scientific goals through the interpretation of a representative subset of EOS measurements. The feasibility of this long-term observing strategy hinges on a sufficiently good understanding of the processes which control the relationships between different constituents, under a wide range of geophysical conditions. *In situ* measurements will be essential in acquiring this basic information.

Different sensors, principally imaging radiometers, on several Earth Probes, EOS Terra, Aqua, follow-on operational precursor missions, and operational missions, will continue to provide the background information on the distribution, particle size distribution, and optical reflectance of global aerosols. Relatively thin surface-based networks (AERONET) provide complementary information on aerosol transmission and optical thickness. Solar occultation sensors and limb-scanning sensors (SAGE-IV and follow-on; HIRDLS and NPOESS/OMPS) sample the distribution stratospheric aerosols and provide profile information at relatively wide space- and time-intervals. However, none of these systems is expected to deliver the detailed measurements needed to quantify the radiative forcing of climate by changes in atmospheric aerosol loading. On the other hand, a robust space-based measurement strategy exists to document the changes in solar radiation output that are relevant to the Earth system science, with the required time-resolution, instantaneous precision, and long-term accuracy/consistency.

The next phase of atmospheric chemistry research will marry on-going modeling and intensive studies of specific physical/chemical processes with long-term systematic measurement of a limited range of critical stratospheric trace constituents, focusing on two principal objectives: (1) verifying the recovery of the ozone layer following the projected decrease in anthropogenic halogens and, (2) quantifying climate feedback associated with changes in upper tropospheric and lower stratospheric ozone and water vapor. Tropospheric chemistry, on the other hand, is still a research frontier and progress will require further exploratory global surveys and field studies. Thus, the NASA observing strategy will involve three distinct observing system components: a space-based global mapping component (implemented principally by operational environmental agencies), a space-based precision monitoring component specifically focused on NASA research objectives, and supporting surface-based measurements.

3.3.1.1 Total Column Ozone and Aerosol Mapping

Total ozone data have come principally from the series of Total Ozone Mapping Spectrometer (TOMS), and the Solar Backscatter Ultraviolet (SBUV) measurements on the NOAA polar-orbiting operational environmental satellite series. Both are based on solar ultraviolet backscatter measurements to determine ozone amounts. TOMS is a cross-track scanning instrument that provides daily global mapping of total column ozone with long-term accuracy on the order of 1% per decade. SBUV is a nadir-viewing instrument that provides total column and partial ozone profile data along the sunlit portion of the sub-satellite track, i. e. typically 16 orbits per day.

In addition to total column ozone, TOMS also provides unique data sets on the distribution of aerosol and particulate matter in the troposphere, including sulfate particles and UV-absorbing particles such as mineral dust, volcanic ash, smoke particles from fires and biomass burning. TOMS aerosol data are of particular interest because they currently constitute the only source of information on the large-scale aerosol loading of the atmosphere over land areas. Because they provide information on total column ozone, clouds, aerosols, and surface reflectivity, TOMS measurements are particularly suitable for inferring UV radiation fluxes at the surface of the Earth. A number of "residual techniques" have been developed to infer total tropospheric ozone from TOMS and other (stratospheric) ozone data, especially in the tropics. The method is being validated by an enhanced network of ozone sonde sites in the tropics, under the Southern Hemisphere Additional Ozone Sondes project (SHADOZ). These tropospheric

ozone and aerosol observations are helping to prepare for future space-based observation missions dedicated to tropospheric chemistry problems.

The TOMS instrument was flown on Nimbus 7, Meteor-3, ADEOS, and currently on Earth Probe TOMS since 1997. The next TOMS flight, QuikTOMS, is planned for a launch in 2000 aboard a small dedicated satellite. Space-based measurements by other agencies will be important in filling any ozone observation gap. The European Space Agency's GOME instrument has been operating since 1995 and the successor instrument SCIAMACHY on the European ENVISAT mission is expected to provide more comprehensive data, beginning in 2000. NASA investigators played a significant role in the exploitation of GOME data and the development of retrieval algorithms for both GOME and SCIAMACHY. This type of cooperative effort is expected to continue in the future.

Four SBUV/2 instruments are still planned for launch on the NOAA Polar-orbiting Operational Environmental Satellite series in 1999, 2001, 2004, and 2007. In the long term, the US National Polar-orbiting Operational Environmental Satellite System (NPOESS) will deploy an Ozone Mapping and Profiler Suite (OMPS) that encompasses the capabilities of TOMS and will maintain total column ozone measurements as well as ozone profile measurements, both based on observation of backscatter UV radiation. The UV limb scattering technique was demonstrated by the Shuttle Ozone Limb Sounding Experiment/Limb Ozone Retrieval Experiment (SOLCE/LORE) on the Space Shuttle; another space flight is scheduled in 2001 for further refinement of the measurement technique. Likewise, Japan's space agency NASDA is planning to develop a UV backscatter instrument (Ozone Dynamics Ultraviolet Spectrometer) for deployment of a new series of Global Change Observation Missions (GCOM-A) beginning in 2005. A similar ozone profiling sensor (GOME-2) will also be carried by the first two European METOP operational polar-orbiting environmental satellites.

Total Aerosol Loading

Currently, no existing observing technique or combination of techniques can document all relevant aspects of the very complex chemical, microphysical and meteorological processes governing the distribution and properties of atmospheric aerosols. The problem is particularly vexing in the troposphere, where aerosol lifetime is only a few days and their distribution vary widely. A Global Aerosol Climatology Project (GACP) has been established under NASA leadership to analyze available satellite radiance measurements and field observations, infer aerosol distribution and properties, and compile a 20-year global aerosol data set. Determining the global distribution of aerosols and its spatial and temporal variations is an essential element of NASA's climate change research strategy, particularly in order to test and improve aerosol chemistry, microphysics and transport models which will eventually be used to predict the climate impacts of various classes of aerosol. Systematic measurements conducted by NASA include:

- Spatial and temporal distribution of total aerosol loading in the middle and upper troposphere over land and ocean, inferred from the Total Ozone Mapping Spectrometer (TOMS) ultraviolet backscatter measurements (also provides an indication of the nature of the aerosol, e. g. mineral dust versus smoke, etc.).
- Distribution and optical properties of aerosol over land and ocean, inferred from solar reflectance measurements by the Moderate-Resolution Imaging Spectro-Radiometer (MODIS) on EOS Terra and Aqua, and the Multi-angle Imaging Spectro-Radiometer (MISR) on EOS Terra. Measurements of optical depth at several visible wavelengths can be used to derive the Angstrom coefficient which is determined by the aerosol size distribution.

It is anticipated that further progress will be made in satellite retrievals of aerosol distribution, physical properties and chemical composition using a combination of lidar, radiometric and photo-polarimetric techniques. Specific plans for implementation of a comprehensive space- and surface-based global aerosol observing system have not been formulated yet, although some steps have been made by the

GACP. In the short term, the main focus of NASA will be on experimenting with new aerosol observing techniques and exploratory satellite missions (see below).

3.3.1.2 Stratospheric Composition Monitoring

Ozone, Aerosol and Polar Stratospheric Cloud Profiles

The principal source of high resolution stratospheric ozone and aerosol profile data has been the Stratospheric Aerosol and Gas Experiment (SAGE) program, based on the principle of occultation of solar radiation by the limb of the atmosphere. The main limitation of SAGE measurements is spatial coverage, since solar occultations occur only in two latitudes bands for a given orbit. SAGE data have been very useful for determining long-term trends in ozone vertical profile, especially in the lower stratosphere, and SAGE data analysis and trend studies continue to be a research priority. NASA is also currently supporting the systematic analysis of high latitude solar occultation data acquired by the Polar Ozone Aerosol Monitor (POAM-3) instruments on the French SPOT-4 earth observation satellite series (in polar orbit).

Two flight models of an improved SAGE III instrument have been fabricated for deployment on a Russian Meteor-3M spacecraft in the first quarter of 2000 (sun-synchronous polar orbit) and the International Space Station in 2003 (51.5° inclination orbit). Solar occultation from polar sun-synchronous orbits restricts observation to high latitudes only (the lunar occultation capability of SAGE III will help to “fill in” the tropics and mid-latitudes). An additional SAGE III instrument is under consideration, for another flight opportunity not yet determined. It is the intention that this program should merge with the NASA systematic measurement program (see below).

Stellar occultation is a promising technique for ozone profiling, as the multiplicity of stars provides many more occultation opportunities than either the Sun or the Moon and enables greatly expanded spatial coverage. In addition, the point-like nature of stellar sources simplifies the retrieval process. This approach has been successfully demonstrated with the UVISI instrument on the DOD MSX mission (with NASA support for data analysis). NASA has also undertaken the development, under the Instrument Incubator Program, of a more capable and lightweight stellar occultation instrument. Likewise, the Global Ozone Monitoring by Occultation of Stars (GOMOS) instrument on the European ENVISAT mission (launched in 2000) will make use of the stellar occultation technique.

Systematic Stratospheric Chemistry Measurement Program

Verification of ozone recovery and the attribution of observed variations to specific causal factors require accurate systematic measurement of stratospheric ozone, atmospheric temperature, aerosols and polar stratospheric clouds, water vapor, dominant species in the chloride, bromide and nitrogen oxide families, CH₄, N₂O and HF, and spectrally resolved solar ultraviolet radiation in the photochemically active spectral region (200-400nm). There are three basic latitude regions in the stratosphere that are relatively well mixed: the tropics, mid-latitudes and polar regions. Unequivocal verification of stratospheric ozone recovery will require time series of accurate measurements at high vertical resolution (1-2 km) in each of these regions. The centerpiece of NASA's stratospheric chemistry research program is high-precision systematic measurement of trace constituents, aiming for identification of potentially significant species, highest possible accuracy, and long-term consistency, with the minimum sampling density compatible with the science objective.

The nominal concept for the implementation of this program, aiming to maintain key measurements after the completion of EOS Aura, is a two-component observing system consisting of an payload on the International Space Station (ISS) and a series of polar-orbiting spacecraft. The scientific objective would be met by solar or stellar occultation measurements from one (ISS) in an inclined orbit and another platform in polar sun-synchronous orbit. The former would provide occultations in the low to mid-latitude range, the latter at high latitudes. Occultation measurements are essentially self-calibrating and have the potential for excellent long-term accuracy and consistency. A broad-gauge, survey-type

instrument (e. g., Fourier transform IR spectrometer) is envisaged to detect the presence and concentration of a wide range of current or potential active compounds. An emission spectrometer would add a very significant enhancement to the horizontal and temporal coverage of the system (See Box 3).

Box 3 **Stratospheric Chemistry Survey Mission**

The scientific objective of the mission is systematic monitoring of key stratospheric trace constituents, particulate matter, and relevant physical properties, as needed to understand the evolution, stabilization and eventual recovery of the stratospheric ozone layer, as expected from the application of the Montreal Protocol. The program consists of two components:

- International Space Station attached payload for occultation measurements in ultraviolet (SAGE III or SAGE-like instrument) and infrared spectrum (high-spectral resolution Fourier transform spectrometer instrument or equivalent emission sensor). Several instrument payloads will be needed for measurement continuity through the critical ozone stabilization and recovery period, and for payload refurbishment and periodic laboratory calibration.
- A polar-orbiting sun-synchronous satellite series, with the same measurement objectives. Due to orbital geometry, solar occultation from polar orbit provides only limited geographic coverage. The alternative stellar occultation technique would overcome this limitation and provide higher vertical resolution. An emission spectrometer may be added to complement occultation measurements. Emission spectrometry is potentially more informative, enabling the detection of free radicals and much enhanced sampling of the global atmosphere, but is less likely to provide the same vertical resolution and long-term calibration consistency.

3.3.1.3 Systematic Measurement of Total and Spectrally Resolved Solar Irradiance

NASA has taken the lead in long-term measurement of radiant energy received from the Sun. Nimbus-6 measurements, beginning in late 1975 and continuing until 1993, provided the longest data set currently available. Unambiguous evidence of variability in total solar irradiance (TSI) was first obtained from the highly precise Active Cavity Radiometer Irradiance Monitor (ACRIM) radiometer on the Solar Maximum Mission (SMM) in 1980. Monitoring variations in total solar irradiance on climate time scales is a challenging task, because accurate TSI measurements can only be made from space, free from any residual atmospheric absorption, and because changes in TSI are quite small, on the order of 0.1% over a 11 year solar cycle. There is theoretical evidence, however, that considerably larger TSI changes may have occurred in the past, particularly during the Maunder solar activity minimum (1645-1715). The validity of these theories has direct implication concerning the explanation of climate warming since the "Little Ice Age" period.

Identifying trends in total solar irradiance over several solar cycles requires an exacting calibration and/or intercomparison strategy in order to achieve the required long-term consistency with a succession of independent instruments. Relatively large absolute calibration differences may exist between successive instrumental records, certainly much larger than the small relative error margin allowed by the precision and stability of individual radiometers. The NASA strategy is based on:

- Best effort to ensure sufficient overlap between successive TSI measurement missions, so as to enable proper intercomparison between one instrumental record and the next;
- Best possible calibration of each flight instrument against absolute radiometric standards, both in air and in vacuum, and provision for on-board monitoring of instrument aging; and
- Periodic in-space calibration, based on intercomparison of current flight instruments with an identical fresh sensor carried on board the Space Shuttle for a 1-2 week calibration campaign, as insurance against the contingency of early termination before a replacement sensor is in place.

In the near future, two overlapping total solar irradiance measurement missions are planned: the ACRIM Earth Probe mission as part of the EOS program (launch in fall 1999), and the SORCE mission in July 2002 (see Box 4). In the long-term, the National Polar-orbiting Operational Environmental Satellite System (NPOESS) program is planning to continue total and spectrally resolved solar irradiance measurements to the required accuracy indefinitely in the future (beginning at the end of the next decade). An additional TSI monitoring mission is being envisaged to bridge the interval between SORCE and NPOESS, if necessary. A supporting research program will be pursued in order to keep the relevant scientific experts involved in maintaining this fundamental data record and encourage further scientific investigations of solar variability. This effort will be strengthened by coordination with similar international efforts, through the establishment of an international investigator group.

Box 4 Solar Irradiance Mission

The Solar Radiation and Climate Experiment (SORCE) mission, planned for a launch in July 2002, is a five-year mission with the following combination of instruments:

- Total Irradiance Monitor (TIM) measuring total (full spectrum) solar irradiance with absolute accuracy of 0.1% or better, and relative precision of 2×10^{-5} per year.
- Spectral Irradiance Monitor (SIM) measuring the solar radiation spectrum from 200nm (ultraviolet) to 2500nm (short-wave infrared), with spectral resolution of 1nm for short wavelengths to 25nm for long wavelengths, and absolute accuracy of 2% or better.
- The Solar Stellar Irradiance Comparison Experiment (SOLSTICE), an instrument developed for EOS that will measure the solar ultraviolet spectrum (from 5 to 440nm) with 3% absolute accuracy and 1nm resolution.

In the long term, the NPOESS program is planning to embark a combination of the TIM and SIM solar radiometers developed by NASA, also known as the Total Solar Irradiance Monitor (TSIM), and will continue this measurement indefinitely in the future from the end of the next decade onward. A bridging mission (not yet identified) may be required in

the 2005 to 2006 timeframe in order to maintain the continuity of TSIM measurements and ensure adequate overlap with NPOESS.

In relative terms, variability in the ultraviolet (UV) region of the solar spectrum is much larger, on the order of 5%. While variations in solar UV radiation contribute only a small amount to the variability in total solar energy absorbed by the Earth atmosphere, their impacts on stratospheric chemistry and temperature structure (static stability) are substantial and could constitute one of the pathways through which solar variability could induce changes in the Earth's climate. Stratospheric photochemical reaction rates are very dependent upon variable solar ultraviolet (UV) radiation in specific and relatively narrow spectral bands. Changes in UV radiation reaching the surface of the Earth is also likely to impact land and ocean ecosystems. Accurate knowledge of both total and spectrally resolved solar irradiance over several solar cycles is crucial to test the validity of these solar variability theories.

Precise knowledge of spectrally-resolved solar UV irradiance is essential for understanding and modeling these reactions. The two existing records of spectrally-resolved solar UV irradiance were obtained from the SBUV series of instruments on NOAA satellites and two instruments aboard UARS, the Solar Ultraviolet Spectral Irradiance Monitor (SUSIM) and the Solar-Stellar Irradiance Comparison Experiment (SOLSTICE). These measurements will be pursued by the SORCE solar radiation monitoring mission in 2002 (Box 4). As a contingency provision against a possible hiatus in spectrally resolved UV measurements, further research will be conducted to develop a more quantitative relationship between solar UV irradiance and proxy indicators of solar activity, such as the 10.7 cm radio-frequency flux.

3.3.1.4 Surface-Based Networks

Systematic measurements from surface-based stations are indispensable, at the current stage of knowledge and observing techniques to validate satellite-derived chemical data and provide otherwise unobtainable information about the concentration of critical trace constituents, especially long-lived chemical species that are well mixed in the troposphere. The objective is to achieve sufficient geographic coverage and sampling density to provide representative information on the spatial distribution and temporal variability of critical chemical species.

Chemical Monitoring Network

NASA surface-based observation activities are carried out in the framework of much larger international programs, notably networks operated by the NOAA Climate Monitoring and Diagnostics Laboratory (CMDL) and those coordinated by the Global Atmosphere Watch of the World Meteorological Organization (WMO). Meteorological data are obtained from the WMO World Weather Watch. Other surface data (such as atmospheric concentration of ozone and carbon monoxide, or nitrate and sulfate amounts in rain water) are obtained from pollution monitoring networks operated by regulatory environmental agencies. In this context, NASA's special contribution to ground-based measurement networks is the deployment of research-quality instruments for the determination of long-term trends in atmospheric trace constituent concentration. The main activities are the NASA-sponsored AGAGE (Advanced Global Atmospheric Gases Experiment) and participation in the international Network for the Detection of Stratospheric Change (NDSC). AGAGE provides systematic measurements of surface concentrations of trace constituents, and NDSC probes the chemical composition of the lower stratosphere.

The AGAGE network is based on gas chromatography coupled with mass spectrometry to measure halogen-containing species including hydrochlorofluorocarbons (HCFCs), and bromine-containing species (like halons and methyl bromide), as well as nitrous oxide and methane. There are currently five operating AGAGE stations (Mace Head, Ireland; Trinidad Head, California; Ragged Point, Barbados; American Samoa; and Cape Grim, Tasmania). NASA will continue operating these facilities and will

upgrade measurement capabilities and long-term calibration, but does not plan to expand its network. NASA will cooperate instead with other agencies that can maintain research-quality monitoring stations in order to better cover the northern hemisphere.

NDSC is an internationally sponsored program, supported by NASA and several foreign partners, to systematically operate, at a number of sites around the world, surface-based remote-sensing systems that meet mutually-agreed research-quality standards. The instruments deployed at NDSC "primary sites" include visible and infrared Fourier transform spectrometers, millimeter-wave emission spectrometers, visible/UV absorption spectrometers, lidar, and balloon sondes to characterize total column amounts and/or profiles of important species. A very high standard of calibration consistency is maintained between stations and over time through a continuing program of instrument intercomparison in the laboratory and in the field. NASA will continue to seek the cooperation of foreign partners to expand the network in under-represented areas (low latitude regions of Latin America, Africa, and South Asia).

Aerosol Monitoring Network

Systematic observations of total column aerosol content (derived from multi-spectral solar radiation absorption measurements) are carried out by a worldwide network of some 50 monitoring stations (the Aerosol Robotic Network or AERONET) installed by NASA with the cooperation of a number of local institutions. The primary purpose of AERONET is to acquire validation data for global aerosol products inferred from radiometric measurements on the Terra mission.

Ultraviolet Radiation Monitoring Network

The principal source of concern about the depletion of stratospheric ozone is the biological impact of the resulting increase in the flux of actinic (UV) solar radiation that reaches the Earth surface. Scientific knowledge of the solar UV flux at the Earth surface has been long stymied by the absence of suitably calibrated and stable instruments to measure such fluxes. NASA uses the data from a US network of some 60 UV measuring stations (operated by several agencies under the auspices of the US Global Change Research Program) to validate global estimates of surface UV fluxes derived from TOMS observations. The combination of the surface UV network and space-based measurements now provides a means for accurate long-term assessment of changes in surface UV radiation.

3.3.2 EXPLORATORY RESEARCH SATELLITE MISSIONS

The NASA global change research strategy for the next decade is expected to shift from comprehensive surveys of a wide range of chemical surveys (exemplified by UARS and EOS Aura) to more specific process-oriented research satellite missions. At the same time, the emphasis for process-oriented atmospheric chemistry research is expected to shift toward the lower stratosphere and troposphere. The discovery potential of tropospheric chemistry research can justify several experimental Earth Probe missions, each focused on specific process-related science questions.

Tropospheric Chemistry Research Satellites

Like in the stratosphere, large-scale chemical processes in the troposphere are dominated by ozone, which is the key oxidant species. Ozone photolysis is a principal source of the hydroxyl radical (OH) which is the most important agent in the degradation of hydrocarbons. Furthermore, because of its oxidant function, tropospheric ozone causes acute and chronic health problems in humans and attacks plant and animal populations. Ozone is also a greenhouse gas in its own right and contributes to climate radiative forcing, although the impacts are markedly different in the stratosphere and troposphere. The

distribution of ozone in the troposphere is influenced by transport from the stratosphere, by regional sources of precursor gases (e. g. biomass burning and urban pollution), and by variations in the atmospheric circulation. Key precursor species include nitrogen oxides, methane and other organic species of anthropogenic and biogenic origins. Both precursor families are subject to the same atmospheric transport which causes a high degree of spatial variability, on scales of hundreds of meters vertically to hundreds of kilometers horizontally.

Two specific challenges need to be addressed to effectively investigate tropospheric chemical processes: horizontal/vertical resolution and temporal sampling. Tropospheric ozone is expected to show considerable vertical stratification and fine horizontal structures that contain information on the origin and age of important species. Likewise, the altitude distribution of upper tropospheric ozone is a critical parameter in climate models. Variability is also important, including significant diurnal variations. On the balance, however, scientific priority was given to vertical resolution over temporal resolution. High vertical resolution will be achievable for ozone with active sounding techniques from low Earth orbit (e. g. differential absorption lidar or DIAL). High vertical resolution observations would be equally valuable for ozone precursors and tracer species, but will probably not be achievable within the time-frame of this plan. Even at relatively coarse vertical resolution, trace species measurements will still be informative in conjunction with high-resolution ozone distribution data for tropospheric chemistry process studies.

High frequency temporal sampling would be best achieved by observation from a geostationary platform. However, the sensitivity and, especially, the vertical resolution of passive radiometric or spectrometric measurement feasible from geostationary orbit is not yet well established and NASA plans to invest in the development of innovative observing technologies to overcome current limitations, in particular, through the New Millennium flight demonstration program.

The planned Triana mission will be the first Earth observing mission to be deployed at the first Lagrange point (L1) - the neutral gravity point between the Sun and the Earth. From L1, Triana will have a continuous view of the sunlit side of the planet. Triana at L1 will complement the current fleet of satellites in low-Earth orbit and geostationary orbit, and will demonstrate the scientific and practical value of the L1 observing location. The Triana Earth Polychromatic Imaging Camera (EPIC) will provide an important complement to Total Ozone Mapping Spectrometer (TOMS) observations and deliver unique data on changes in ozone, clouds, and aerosol from sunrise to sunset for climate science applications and estimation of the amount of UV radiation reaching the surface. Triana's advanced radiometers will measure the radiances of reflected and emitted energy from a unique range of viewing angles with unprecedented accuracy and will test the current understanding of radiative transfer processes in the atmosphere.

Aerosol Research Satellite Sensors and Missions

Currently, the primary aerosol properties inferred routinely from space measurements are total reflectance and a particle size parameter. The MISR and MODIS instruments on EOS Terra provide more accurate measurements, but still do not allow independent estimates of aerosol radiative forcing, due to the lack of information on aerosol vertical distribution, and optical properties.

The next step will be the PICASSO-CENA experimental satellite mission (in cooperation with the French space agency CNES). PICASSO-CENA will be launched in 2003 together with Cloudsat and fly in formation with Cloudsat and EOS Aqua. The primary sensor of the PICASSO-CENA mission is a dual-frequency, infrared and visible (green), dual polarization lidar profiler that will directly determine the vertical distribution of aerosols over both land and ocean. PICASSO-CENA data, complemented by simultaneous EOS Aqua (MODIS, CERES) measurements, will directly determine aerosol optical depth, number density, and optical properties; they will yield estimates of aerosol radiative forcing essentially

free from *a priori* assumptions. The 3-year PICASSO mission will provide a large representative set of seasonal conditions and geographic regions, but still a relatively discrete sample of the whole atmosphere. It is anticipated that lidar profile data will allow back-tracking the atmospheric trajectories of aerosol layers and help identify their sources.

Solar occultation and limb-viewing measurements also contribute to determining the vertical structure of the aerosols, albeit with poor horizontal resolution. The SAGE IV instrument will include a longer wavelength channel (1.5 μm), which affords a better probability of reaching the troposphere. The HIRDLS instrument on EOS Aura will provide spatially dense aerosol profile information derived from emitted atmospheric radiation, free from the geographic coverage limitations of SAGE.

3.3.3 LABORATORY STUDIES AND FIELD MEASUREMENT CAMPAIGNS

Laboratory measurements of basic physical and chemical properties and processes provide an essential fundamental underpinning for scientific interpretation of field measurements and global satellite-based surveys. First, laboratory measurements of kinetic rates of chemical reactions and photochemical processes, molecular cross sections and quantum yields, and information on heterogeneous processes are needed to understand atmospheric transformations and inform numerical models. Current results of laboratory measurements are critically evaluated in the biennial report on *Chemical Kinetics and Photochemical Data Evaluation*, published by NASA. Second, spectroscopic measurements and related photochemical phenomena are necessary to support NASA's remote sensing projects. Particularly important is the spectroscopy of polyatomic molecules at the temperature and pressure representative of stratospheric and upper tropospheric conditions. Finally, a new range of studies is needed to determine thermodynamic and chemical properties of atmospheric aerosol and their interaction with stratospheric cloud particles, notably the three-component water-nitric acid-sulfuric acid system under stratospheric conditions.

The principal method developed by NASA to understand chemical and physical processes in the atmosphere relies on simultaneous *in situ* and/or remote sensing measurements of related parameters. Troposphere and stratosphere field campaigns are carried out from a variety of platforms, including stratospheric balloons, high altitude ER-2 and WB-57 aircraft, and the DC-8, and P-3 aircraft. Several factors are lessening the traditional specialization of field measurement campaigns for stratospheric or tropospheric studies. First, scientific questions are evolving toward the consideration of phenomena occurring just above and below the tropopause, such as troposphere-stratosphere exchanges. Second, increased flexibility in the operation of several aircraft, notably the ER-2 and uninhabited aerial vehicles (UAVs), now allows access to both the troposphere and the stratosphere with the same equipment. Finally, more capable sensors can be applied to chemical studies, notably measurement of the hydroxyl radical and many key precursor species in the lower stratosphere and upper troposphere. Balloon and aircraft campaigns will be needed in association with EOS Aura to provide detailed photochemical process studies of diurnally varying species that complement the global observations provided by EOS Aura, which will only sample a given latitude at a fixed time of day and to acquire higher precision and spatial resolution data for further process studies.

In the coming decade, growing demands for validation activities in support of space-based atmospheric chemistry measurements and other Earth science disciplines will place a heavy burden on NASA's aircraft and other non-space facilities. In addition, many important issues in atmospheric chemistry require measurement and analysis at spatial and temporal scales that are not accessible by satellite observation and can best be addressed through a combination of space and non-space measurements. Consultations are underway with the atmospheric science community for planning the Integration of Satellite Calibration/Validation and Research Oriented Field Missions in Atmospheric Chemistry in the Next Decade. An atmospheric chemistry research plan for integration of the research and analysis (R&A) field campaigns and the satellite missions in the next decade will be the end-product of these consultations.

Tropospheric Aircraft Campaigns

Field campaigns undertaken as part of the Global Troposphere Experiment (GTE) have relied on NASA DC-8 and P-3 aircraft for *in situ* and remote (lidar) measurement of ozone, other trace constituents, aerosols and meteorological parameters in remote regions of the world. The focus of these missions was to define the tropospheric background and to look at long-distance impacts of human activities on the troposphere at regional and global scales. The most recent GTE mission was the Pacific Exploratory Mission (PEM) TROPICS-B, implemented in March 1999. Given the relative paucity of process-scale

tropospheric chemistry data, further GTE campaigns are in principle scheduled approximately every other year. The focus of these campaigns in the next decade will be on the Pacific Basin, a relatively clean-air region of the world that is adjacent to and will be strongly affected by the very rapid population growth and economic development in Asia. Efforts will be continued during the during this period to enhance the chemically comprehensive suit of measurements on the DC-8 and P-3 through reductions of instrument size and weight, and improvements in sensitivity and temporal resolution. Following up on the GTE initial explorations of general process and chemistry in the major tropical source regions, South America and Southern Africa, light aircraft are being used by NASA and cooperating agencies to address recognized gaps in the understanding of the carbon and nitrogen cycles. These studies include the work of the LBA-Ecology and SAFARI-2000 programs (see Chapter 1).

Stratospheric Aircraft Campaigns

Stratospheric field campaigns usually involve both ER-2 and DC-8 aircraft. Over the past decade, the focus of attention was both northern and southern high latitudes regions where substantial ozone depletion was taking place. More recent campaigns have shifted attention toward mid-latitudes; the most recent Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) campaign, completed in 1997, was aimed at the study of photochemistry in the mid to high-latitude lower stratosphere from spring to late summer. The next major undertaking in the 1999- 2000 timeframe is SAGE III Ozone Loss and Validation Experiment (SOLVE) campaign, integrating space-based (SAGE III), airborne (ER-2 and DC-8) and high altitude balloon measurements.

The SOLVE project is being cited as a paradigm of the type of planning needed to foster scientifically productive flight and R&A programs in the coming decade. The objectives of the SOLVE campaign are to provide calibration and validation data for the SAGE III instrument on Meteor-3M and to evaluate the potential for enhanced ozone losses at high latitudes in the Arctic. The latter objective is driven by recently observed Arctic ozone losses on the same order of magnitude as Antarctic ozone losses. The substantial losses observed during the last decade over the Arctic are not reproduced by current atmospheric models. This inability to quantitatively explain present-day ozone losses undercuts the credibility of model predictions of future ozone losses and recovery under conditions of increased concentrations of greenhouse gases. The synergy demonstrated by SOLVE between space-based and *in situ* measurements, data analysis and modeling studies will guide the conception of atmospheric chemistry field campaigns in the coming decade to serve both satellite remote sensing validation and scientific discovery objectives.

Plans for further field measurement studies beyond SOLVE are open. Candidate projects could include additional aircraft missions focused on Arctic ozone (the first few years of the next century is the period when stratospheric chlorine loading has the greatest potential for ozone destruction) or ones upper tropospheric photochemistry, the properties and effects of gravity waves, the chemical and radiative effects of visible and sub-visible cirrus clouds in the tropics, or troposphere-stratosphere exchange. There is a strong likelihood that one or more aircraft campaigns will be organized to acquire validation and/or correlative data for EOS Aura measurements.

The current ER-2 payload has the capability for *in situ* measurement of nearly all important trace constituents in the lower stratosphere, including the important radical species OH, HO₂, NO, NO₂, ClO, and BrO. Together with measurements of source and reservoir gases (such as HCl and ClONO₂) and UV radiation, this payload is fully adequate to provide a critical test of photochemical models in the lower stratosphere. Uncrewed Aerial Vehicle (UAV) technology has progressed to the point of readiness for initial science mission demonstrations that will lead to these platforms becoming an important component of the airborne science field campaigns. Unique capabilities of UAVs include long flight duration at altitude (spanning a full diurnal cycle), higher subsonic flight altitudes, and the ability to

perform missions that would be hazardous or unsuitable for piloted aircraft. Future UAV capabilities could allow much expanded studies of the 22-25km region of the stratosphere (eventually up to 30km).

Balloon Campaigns

Balloon platforms are typically used to measure the vertical distribution of trace constituents above the ER-2 operational ceiling and have been essential elements of the calibration/validation program for space flight missions such as UARS. Large research balloons can reach altitudes up to 40 km, and carry both *in situ* sensors and remote sensing instruments. The Observations from the Middle Stratosphere (OMS) instrument payload was initially developed for comprehensive *in situ* stratospheric measurements, predominantly measurements of tracer concentrations. Since then, the addition of several remote-sensing instruments allows OMS to provide measurements of the complete suite of free radicals, source, reservoir, and tracer species needed for detailed process studies. It is foreseen that balloon flights will continue over the next few years, in conjunction with stratospheric aircraft campaigns. Development of new balloon capabilities (for example, ultra long-duration flights) may expand the range of high altitude balloon investigations. Balloon measurements will play a critical role in the SOLVE campaign.

In situ and Surface-based Aerosol Process Studies

Field campaigns, involving coordinated use of multiple aircraft platforms, surface- and satellite-based measurements, are an integral component of the program. Each field experiment is a coordinated effort with the activities sponsored by other US federal agencies and international institutions. For example, the Smoke, Clouds and Radiation (SCAR) experiment series was designed to obtain simultaneous *in situ* and remote measurements of the physical and chemical properties of aerosols produced by biomass burning and other human activities. The SCAR field studies also generated data for the evaluation of MODIS-compatible remote sensing algorithms. SCAR experiments were conducted in 1993 (effect of industrial pollution over the Atlantic seaboard), 1994 (biomass burning in California and Oregon), and 1995 (biomass burning in Brazil). Future plans for further field campaigns are yet to be determined, especially in relation to the validation of MODIS, MISR, and PICASSO-CENA aerosol products.

3.3.4 MODEL DEVELOPMENT AND STUDIES

Model development, validation and simulations are critical to nearly all aspects of atmospheric chemistry, as they provide the vehicle through which observations are interpreted and understanding of atmospheric processes can be tested. Models also constitute the principal tool for assessing future impacts on atmospheric composition. It is convenient to group model studies in three categories: process modeling, application to retrospective analysis, and model predictions.

Process Models

Process models are designed to provide the best possible representation of selected atmospheric processes. Various types of models are used for different classes of chemical processes, including (i) constrained photochemical models with the fullest possible representation of all constituents and chemical processes within a fixed air mass, (ii) aerosol microphysics models providing a detailed description of new particle formation, transformation and loss by gravitational settling or entrainment in clouds, (iii) parcel trajectory models that simulate the evolution of one or a number of air parcels as they move through the atmosphere, (iv) radiative transfer models for computation of radiation flux (especially ultraviolet and visible) in the atmospheric column, taking into account absorption and scattering by trace gases, aerosols, clouds, and the underlying surface, and (v) dynamical models that simulate the formation and propagation of waves created by various sources. Process models are typically constrained by comprehensive data sets from intensive field measurement campaigns. The ability to accurately

reproduce observed phenomena constitutes a critical test of our understanding of the particular processes represented in such models.

Retrospective Models

These models are designed to simulate the past evolution of trace constituent and/or aerosols given the strength of surface sources and sinks (or boundary conditions). The models may be utilized to reproduce changes in trace constituents in response to time-dependent meteorological forcing and sources, or simulate long-term equilibrium conditions as a function of changing boundary conditions (surface halogen abundance, solar input, etc.) or other constraints (e.g. sulfate aerosol distributions). Short simulations require that meteorological fields be specified for the time period of interest; this information is typically derived from meteorological data assimilation systems or forced dynamical models (e.g. stratospheric models forced with known tropospheric meteorology, or tropospheric mesoscale models forced by far-field conditions). Over longer time periods, the actual interannual variability may be ignored, although attempts have been made to take it explicitly into account. The requirement for boundary conditions or basic meteorological data ties atmospheric chemistry research to the global water and energy cycle component of the NASA Earth science program (See chapter 4). It is foreseen that interactive 3-dimensional transport-chemistry models will soon be widely utilized for extended simulations and/or predictions.

It is also anticipated that more attention will be given to the application of data assimilation methods to atmospheric chemistry variables. Current efforts in this area include assimilation of long-lived stratospheric tracers (UARS data), ozone and tropospheric carbon monoxide concentration. A related application is "inverse modeling", i. e. the process of using chemical constituent observations with an atmospheric model to infer information about model processes that affect the distribution of these constituent. This technique has been applied to studies of chlorofluorocarbon sources and sinks. It is anticipated that the technique will soon be expanded to constituents with more complex sources and sinks distributions, such as carbon monoxide and methane.

Model evaluation is a strong activity in the NASA program, based on the comparison of model outputs with measurements. Such efforts include the "Models and Measurements" intercomparison completed in 1998. This effort, conducted jointly by the Office of Earth Science and the Office of Aero-Space Technology, was a critical test of the ability of various model to reproduce observed phenomena under prescribed conditions. Measurements of indicators of the stratospheric age of air parcels (CO_2 and SF_6) have proven valuable for diagnosing inadequacies in model transport. Observations of conservative tracers such as NO_y and Cl_y , along with the radical species, have been used to test the accuracy of component chemical mechanisms in models. Models have trouble simultaneously simulating the age of both the air and chemical tracers. Comparisons with measurements of species which are produced only in the stratosphere, such as the isotopes ^7Be and ^{10}Be , will help resolve this issues. Such efforts are expected to continue in the future, with a shift in priority toward tropospheric chemistry. NASA also expects to continue supporting the intercomparison studies undertaken under the aegis of the Global Integration and Model component of the International Global Atmospheric Chemistry (IGAC) program.

Predictive Models

Prognostic models are designed to simulate the future evolution of the atmosphere. In many cases, these models are substantially similar to those used for retrospective studies. In the prognostic mode, however, meteorological fields must either be assumed from some future atmospheric circulation scenario (typical method in two-dimensional models) or from a three-dimensional general circulation model (GCM). The use of GCMs will be particularly important in simulations of the combined response of the atmospheric circulation and chemistry to climate change, as the dynamical state of the atmosphere may change significantly. Such models will provide the means to assess the feedback mechanisms between

chemistry, radiation, transport, and eventually biospheric processes and feedback. Considerable work remains to be done to develop fast chemistry and microphysics algorithms for use in time-dependent models, as well as high performance numerical integration schemes for core dynamics (see section 2.3.4). Additional speedup will accrue from the use of massively-parallel computers (at non-trivial cost in software complexity and lack of flexibility).

Model-Based Assessments

The atmospheric chemistry community has a long history of supporting atmospheric environment impact assessments. The most widely known are the periodic Scientific Assessments of Ozone Depletion, carried out on behalf of the World Meteorological Organization (WMO) and the United Nations Environment Programme (UNEP) as well as the assessment of climate forcing factors on behalf of the Intergovernmental Panel on Climate Change (IPCC, 1995). Additional studies have been organized by the Office of Aero-Space Technology to assess the impact of projected supersonic and subsonic aircraft fleets on the atmosphere. A special assessment of the atmospheric effects of aviation is currently being carried out for the IPCC, on behalf of the International Civil Aviation Organization. The international research program on Stratospheric Processes and their Role in Climate (SPARC) under the World Climate Research Program also organized assessments of changes in the vertical distribution of ozone. SPARC produced an updated assessment of trends in stratospheric temperature, which was incorporated in the Trends in Stratospheric Temperatures section of the 1998 Scientific Assessment of Ozone Depletion (WMO, 1999). SPARC is likewise sponsoring an analysis of the variability and trends in stratospheric water vapor. Predictive and equilibrium atmospheric chemistry models are an essential tool for this purpose. NASA expects to continue supporting these important applications of knowledge acquired in the ESE atmospheric chemistry program.

Aerosol Impact Assessment Models

Given current state of knowledge, the most practical means to estimate the global radiative impact of anthropogenic aerosols (or a particular class of aerosols) and gauge the relative importance of anthropogenic and natural emissions in the global aerosol distribution, may be the numerical simulation of atmospheric transport, aerosol formation and evolution, and radiative transfer. The problem is to improve the degree of confidence in such models at the level of the representation of basic microphysical and chemical processes (local process studies), and at the level of global model simulations (global model product validation).

Unlike clouds, aerosols appear with a large diversity of chemical compositions, physical properties and derived radiative characteristics. Chemical, physical and radiative properties are closely linked to the specific sources of the aerosols. Complete aerosol emission inventories are a crucial input for the advancement of coupled climate-aerosol model simulations. Since radiative properties of aerosols are strongly linked to their size distribution, it is essential that the distribution of water vapor and the growth of aerosol particles as a function of relative humidity be properly characterized in the models. Satellite measurements can be used to constrain models of both the direct and indirect radiative forcing by anthropogenic aerosols. However, it is fair to say that a comprehensive strategy does not yet exist for systematic refinement, testing and validation of current global aerosol models.

3.4 LINKAGES

Linkages with other NASA programs

Understanding the long-distance transport and dispersion of trace species in the troposphere and stratosphere is dependent upon progress in knowledge of the atmospheric circulation (notably through 4-dimensional physical data assimilation) and the ability to realistically simulate atmospheric dynamics with general circulation models. Conversely, assimilation of tracer data is a promising avenue for progress in the analysis of the global atmospheric circulation. Another important linkage is the study of interlocking physical and chemical processes that control aerosol formation, transformation and removal. These scientific issues are being studied from the perspective of both atmospheric chemistry and physics (Chapter 3 and 4, respectively). Both aerosol and trace atmospheric gases have significant impact on atmospheric radiation transfer. The radiative impact of these atmospheric constituents represents a major linkage between atmospheric chemistry and global climate change, that must be factored in climate change prediction and assessments (Chapter 7).

Many important trace species have biological sources in the ocean or over land. Quantifying these emissions and understanding the terrestrial or marine processes that govern the release of trace constituents to the atmosphere is a scientific issue of interest to both atmospheric chemistry and the biogeochemistry of ecosystems (Chapter 2).

The Atmospheric Chemistry program also cooperates with the Aero-Space Technology Enterprise in the assessment of the potential effects of subsonic and supersonic aviation on climate and the global environment, including engine emission of effluents that affect the concentration of ozone, and particulate matter and aerosol precursors that affect the formation of condensation trails and clouds.

Linkages with other US agencies

The NASA Atmospheric Chemistry research program maintains a very active cooperation with the National Oceanic and Atmospheric Administration (NOAA) Environmental Research Laboratories (ERL) for the conduct of airborne campaigns and surface-based measurements. NASA and NOAA have developed complementary monitoring networks for accurate measurement of the surface concentration of long-lived trace constituents. The NOAA flask sampling network has a broad geographic coverage and collects weekly samples that are analyzed at a central facility in the US. The NASA Advanced Global Atmospheric Gases Experiment (AGAGE) network operates only a small number of stations distributed in latitudes, but makes much more frequent (hourly) measurements at the sites themselves. The two agencies organize detailed intercomparison between the two networks to ensure their consistency.

NASA and NOAA collaborate in the joint exploitation of ozone data obtained from operational SBUV and TOMS sensors. In the same context, NASA cooperates with DOD and NOAA for the development of the advanced Ozone Mapping and Profiler Suite (OMPS) of instruments that will fly on the National Polar-orbiting Operational Environmental Satellite System (see section 4.3.1.1). NASA also actively cooperates with several DOD research establishments, notably the Naval Research Laboratory (NRL) for the exploitation and scientific analysis of measurements obtained by the NRL Polar Ozone & Aerosol Monitor (POAM) embarked on a French SPOT Earth observation satellite.

International linkages

Atmospheric chemistry, especially in relation with the depletion of stratospheric ozone and changes in the concentration of greenhouse gases, has high international visibility. The NASA atmospheric chemistry program has been a major contributor to scientific advances in characterizing the causes of

ozone depletion, which led to the *Montreal Protocol on Substances that Deplete the Ozone Layer* and subsequent amendments. NASA maintains the observing capability to monitor stratospheric ozone and actively contributes to periodic international assessments of the state of the ozone layer commissioned by the World Meteorological Organization (WMO) and the United Nations Environment Program (UNEP). NASA also contributed to the Intergovernmental Panel on Climate Change (IPCC) assessment of the climatic impacts of ozone and other greenhouse gases, the IPCC/ICAO Special Report on Aviation and the Global Atmosphere. Likewise, NASA is a major partner in international cooperative research initiatives organized by the international World Climate Research Program, particularly the study of Stratospheric Processes and their Role in Climate (SPARC), and the International Geosphere-Biosphere Program, particularly the IGBP International Global Atmospheric Chemistry core-project.

Bilateral cooperative programs or projects have also been developed with many foreign agency partners. Original instruments developed by Germany (CRISTA), the Netherlands (OMI), Canada (MOPITT) or in cooperation with UK (HIRDLS) will fly on several EOS missions or the International Space Station. Furthermore, NASA has been actively cooperating with the European Space Agency in the joint analysis of data from ESA's Global Ozone Monitoring Experiment (GOME) sensor and supports US co-investigators selected by ESA for its ENVISAT mission. NASA also maintains similar scientific collaboration with Japan, notably through joint announcement of research opportunities for the ADEOS and ADEOS-2 missions. Finally, NASA plays a leadership role in the Network for Detection of Stratospheric Change (NDSC) that operates a variety of advanced remote sensing instruments for monitoring stratospheric ozone and other atmospheric constituents in partnership with NOAA, NSF and many foreign institutions (see section 4.3.1.4).

3.5 REFERENCES

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