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ATMOSPHERIC RADIATION MEASUREMENT: A PROGRAM FOR IMPROVING RADIATIVE FORCING AND FEEDBACK IN GENERAL CIRCULATION MODELS

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1. INTRODUCTION

The Atmospheric Radiation Measurement (ARM) Program is a key element of the Department of Energy's (DOE's) global change research strategy. ARM represents a long-term commitment to conduct comprehensive studies of the spectral atmospheric radiative energy balance profile for a wide range of cloud conditions and surface types, and to develop the knowledge necessary to improve parameterizations of radiative processes under various cloud regimes for use in general circulation models (GCMs) and related models.

The importance of the ARM program is apparent from the results of model assessments of the impact on global climate change. Recent studies suggest that radiatively active trace gas emissions caused by human activity can lead to a global warming of 1.5 to 4.5 degrees Celsius and to important changes in water availability during the next century (Cess, *et al.* 1989). These broad-scale changes can be even more significant at regional levels, where large shifts in temperature and precipitation patterns are shown to occur. However, these analyses also indicate that considerable uncertainty exists in these estimates, with the manner in which cloud radiative processes are parameterized among the most significant uncertainty. Thus, although the findings have significant policy implications in assessment of global and regional climate change, their uncertainties greatly influence the policy debate.

ARM's highly focused observational and analytical research is intended to accelerate improvements and reduce key uncertainties associated with the way in which GCMs treat cloud cover and cloud characteristics and the resulting radiative forcing. This paper summarizes the scientific context for ARM, ARM's experimental approach, and recent activities within the ARM program.

2. THE SCIENTIFIC OBJECTIVES OF ARM

The ARM is a ten-year program designed to reduce key uncertainties of cloud parameterizations in current GCMs and related models and to test new, improved cloud parameterizations. Specifically, the goals of ARM are 1) to improve the treatment of radiative transfer in GCMs for clear sky, general overcast, and broken cloud conditions and 2) to provide a test bed for cloud parameterizations used in GCMs. Thus, the scientific requirements for ARM are to:

- Develop a quantitative description of the spectral radiative energy balance profile under a wide range of meteorological conditions. These descriptions must be derived from measurements and must be quantified at a level consistent with climatologically significant energy fluxes of 1 to 2 W/m^2.
- Identify and investigate processes controlling the radiative and surface energy balance. Evaluation of our understanding of these processes must come from a direct and comprehensive comparison of field observations with detailed calculations of the radiation field and associated cloud and aerosol interactions, and surface energy flux.
- Develop necessary knowledge to improve parameterizations of radiative properties of the atmosphere for use in GCMs. This requires intensive measurements at a variety of temporal and physical scales, with a major emphasis of the role of clouds, including their distribution and microphysical processes.

To meet these scientific requirements, the ARM program is composed of three major activity areas, all coordinated by a Technical Management Team. These are 1) an experimental component known as Clouds and Radiation Testbed (CART), 2) a Science Team component, and 3) an instrumentation development component. More specific information on each of these areas, including some recent activities, is presented later in this paper following a general discussion on the complexities and key uncertainties in the cloud-climate system, which provides the scientific context for ARM.

3. THE CLOUD-CLIMATE SYSTEM

There is close yet complex coupling among clouds, temperature, and trace gases and aerosols. In the following sections we discuss the scientific basis of this coupled system in the contex of the ARM program, key sources of uncertainty in the cloud component, and the assumptions necessary to model the cloud component in GCMs.

3.1 Scientific and Observational Context

By reflecting and scattering incoming shortwave radiation, clouds act to prevent surface warming. The albedo and infrared absorption characteristics of the earth's surface are closely coupled to this system. Cloud formation can also be affected by influences on the temperature distribution of the atmosphere from radiative absorption of trace gases. The presence of aerosols changes the absorption properties of the atmosphere, thus affecting temperature fields. In addition the small soluble surfaces of many aerosols can act as condensation nuclei, which are important in the formation of clouds. Warmer temperatures allow the atmosphere to hold more moisture and thereby enhance the possibility of cloud formation.

Human activity introduces gases and aerosols into this system, perturbing the forcing and feedback mechanisms and resulting in a redistribution of temperature fields and clouds. Wigley (1989), for example, indicates that anthropogenic aerosols resulting from human activities may directly and indirectly influence changes in regional cloudiness in a way that largely offsets warming due to direct absorption of radiation by anthropogenic gases. A major purpose for the development of GCMs and related models is to describe this complex system, and to assess how anthropogenic emissions influence the redistribution of global temperature and precipitation.

3.2 Uncertainties in the Cloud Component

While there are many elements of uncertainty in describing this system, the cloud component is viewed as a major source of uncertainty in GCMs (Cess, et al., 1989). Other modeling studies indicate that the individual effects of longwave warming and shortwave cooling by clouds can be as much as a factor of ten times the forcing caused by a doubling of the atmospheric CO_2 concentrations arising from anthropogenic sources (Ramanathan, *et al.* 1989). Small uncertainties in the parameterization of cloud radiative forcing can therefore lead to large uncertainties in model results on global warming or cooling, relative to results generated from investigation of the effect of increases in trace gases alone.

Evidence of this uncertainty is available from field observations. The Earth Radiation Budget Experiment (ERBE) has produced information on cloud forcing and feedback processes, showing that these effects over the globe are large (Ramanathan, *et al.* 1989). This study indicates that changes in these forcing functions induced by changes in cloudiness can modify the heat balance and the resulting climatic regime. Globally-averaged cloud forcing appears to produce net cooling (Gibson, *et al.*, 1990, Ramanathan, *et al.*, 1989), and the influence of cloud forcing varies across latitudes, with the strongest effect indicated in mid-latitude storms.

Surface characteristics of the earth, which also have an important influence on radiative forcing and feedback, also represent sources of uncertainty. Surface radiative properties change during the day and from season to season due to changes in soil moisture, vegetation, and solar angle.

3.3 Modeling the Cloud Component of Radiative Transfer

Cloud parameterizations in GCMs generally employ simplifying assumptions regarding scattering and absorption of short wave radiation and transmission and absorption in infrared radiation. The frequency dependence of a cloud is often assumed to be "grey" or black in the infrared spectrum, and clouds are characterized as having idealized shapes, or represented as plane-parallel models. When compared against more exact spectral line-by-line models of infrared and shortwave radiative transfer, the simpler algorithms can introduce important errors (*e.g.* Schwarzkopf and Ramaswamy, 1990), and therefore these algorithms represent a key uncertainty in the overall model.

Other simplifying assumptions, required in order to complete the radiative transfer calculations in GCM, also introduce important uncertainties. For example, simplifying assumptions about the albedo and infrared emittance of the earth's surface for each GCM grid cell must be made. This requires specification of a single land use type and surface radiative characteristic for a grid cell where in actuality considerable variability is likely to exist.

4. OVERVIEW OF THE ARM APPROACH

The ARM is designed to establish a technical base for improving parameterizations and reducing key uncertainties in the modeling of the cloud-climate system. A Technical Management Team, composed of representatives from the various DOE national laboratories, has responsibilities for the operational aspects of ARM, including identification of study sites, modeling support, coordination with other programs, and instrumentation and site operation requirements. The Technical Management Team also receives input from the Science Team (described below) concerning the scientific issues to be studied. An overview of the ARM approach is given here, along with some recent program developments.

4.1 Clouds and Radiation Testbed (CART)

The CART includes an experimental framework and a shared data system that draws together all experimental elements and provides ready access to major experimental results for the Science Team and other investigators.

The experimental approach will combine a central intensive radiation and cloud observation facility, conceptualized in Figure 1, with a three-dimensional mapping network, an extended observing network, and mobile, aircraft and satellite observing systems. The approach is designed to address cloud parameterizations for a single grid cell of a GCM. However, a single GCM grid cell is orders of magnitude larger (>500 km^2) than the spatial scale associated with important cloud processes, and a single GCM time step is considerably longer than the life cycle of most cloud processes. As a consequence, the ARM experimental approach is configured to allow for the development and testing of subgrid scale cloud parameterization, coupled with longer-term measurements over spatial scales comparable to a GCM grid. Only with this approach can the critical uncertainties associated with subgrid scale cloud inhomogeneities and surface albedo variations be reduced in climate models. The ARM experimental approach will gather data that can be used to characterize the statistics of clouds on a subgrid-scale.

Details of this experimental approach have been published by the Department of Energy (1990) and summarized in Patrinos *et al.* (1990). In brief, the conceptual approach utilizes a central measurement facility, augmented by a three-dimensional mapping network surrounding the facility. The major focus of this network is to reconstruct the cloud geometry surrounding the central facility. A more extended network, covering the spatial scale of a GCM grid cell, will surround this core network. This larger-scale network coverage is intended to provide a data base extending over time scales comparable to GCM time steps and of sufficient duration to capture important year-to-year meteorological variability. The exact number of sites to be chosen for installation of this measurement configuration will depend on a number of criteria relating to factors such as climatological and meteorological conditions, surface characteristics, logistical support, synergism with other research activities, and programmatic budgets. At present, approximately four to six sites are anticipated.

In addition to this permanently placed measurement network at each site, ARM will employ mobile observing systems and conduct intensive campaign studies. Direct sampling from aircraft to support the campaigns will be made. In addition, routine research aircraft missions will be conducted to support, for example, calibration of remote sensing systems and to profile the radiation field as a function of altitude. Finally, satellite data will be used to provide top-of-the-atmosphere radiative fluxes, and to augment atmospheric profile data.

The CART data environment, described in a parallel proceedings by Melton *et al.*, 1991, is intended to provide data streams of field measurements and model predictions that form a test bed for model performance. Testing model performance represents a "quasi real-time" requirement, where some models will be tested in real time, while others, which require longer-term runs, will be tested within a several-month mode. This design is intended to minimize periods when excessive data processing would be required if all model performance was to be done after the field measurements are collected.

The CART data system requirements make conventional data system configurations inadequate for this program. The design and implementation of the CART Data System has followed a systematic approach, starting with a thorough definition of the required system functions, and leading

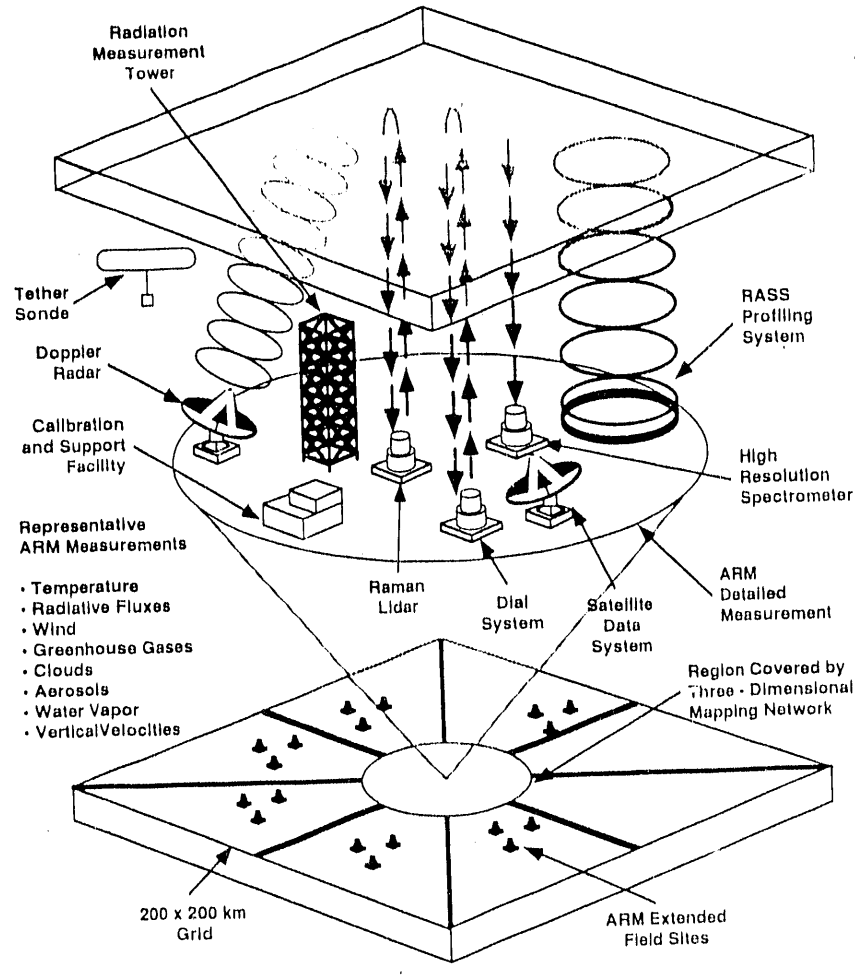

Fig. 1. Conceptual design for the central facility of the ARM experimental network.

iteratively to development of a system architecture and deployment of the system in the field. Currently the system architecture is designed to accept measurements from the site as well as external measurements (such as satellite data or data collected by other agencies). As the data streams enter the data system, they are converted to engineering units and subjected to various quality assurance checks. Information on the data is included in a "meta-data" stream. The site data system delivers the data stream associated with a specific experiment to the appropriate member of the Science Team (the Science Team is discussed in the next section). In addition, all raw and quality-assured engineering data are delivered to the CART Data Archive Center.

4.2 The Science Team

The Science Team will guide the development of the ARM program and conduct specific experiments relevant to improving parameterizations of clouds and radiative processes in GCMs and related models. This team will be organized into specific technical areas with functional responsibilities such as: GCMs and related models, cloud formation and parameterization, radiative modeling, remote sensing, radiometric measurements, and 4-D data assimilation. The Science Team is expected to work directly with the ARM Technical Management Team.

The Science Team is composed of principal investigators selected from responses to two programmatic requests for proposals (RFPs) released by DOE early in 1990. One RFP solicited research proposals as part of ARM from the university and non-DOE federal communities and the other solicited proposals specifically from DOE national laboratories. About 30 projects have been selected. The first joint meeting of the Science Team took place in November 1990.

4.3 Collaboration with Other Programs

Throughout the program, DOE will collaborate extensively with related research programs of other federal agencies and with the international research community. An important criterion for selection of the ARM sites and for conducting campaigns is the synergism with other agency and multinational programs. Examples of other studies where ARM campaigns could be conducted jointly include the National Aeronautic and Space Administration's (NASA's) FIRE programs planned for the midwest United States and for the Azores, and the international TOGA/COARE program planned for the western equatorial Pacific warm pool, and the GEWEX Continental Scale International Project. These programs have goals related to improving the understanding and predictability of climate change, and offer the possibility of extensive measurement and data collection systems that would be synergistic to meeting ARM's goals.

4.4 Recent Program Developments

Two developments have recently taken place that are important in the implementation of ARM field activities. These are the ARM experimental site selection process, and the establsihment of SPECTRE, funded by both NASA and DOE, as a pilot study for ARM during the FIRE CIRRUS experiment.

Selection of ARM Experimental Sites

A process is under way for selecting the ARM sites. Each site will be selected from a larger region, known as a "locale", defined as a relatively compact and contiguous geographical region that has generally homogeneous climatic and geographical properties. The selection process is proceeding at a schedule structured to accommodate the first site operations in 1992.

The locale selection process is one of the activities of the Technical Management Team, and began during the summer of 1990. Approximately five locales have been selected for use in determining the location of ARM sites. Since the locale selection process began in parallel with selection of the Science Team, it was recognized that the full complement of projects to be conducted at ARM sites is not yet known. Thus, the initial selection process attempted to provide a suite of locales which collectively meet the overall goals of ARM, with possible refinement of this ensemble to occur following review by the Science Team.

The selection of potential locales was based on several criteria. From an initial list of candidate locales, a subset was derived by considering their ability to stress models in the area of climatology, surface properties, atmospheric processes, and surface energy flux. Other criteria include climatological homogeneity, surface property homogeneity, logistical impacts, and synergism with other programs. Ensembles of locales were developed from an evaluation of their attributes in context of the listed criteria, and in the context of meeting the ARM objectives. A list of potential locales was developed in time for review and evaluation at the full Science Team meeting held in November 1990.

SPECTRE: An ARM Pilot Study

An important activity within ARM is collaboration with other research programs that have synergistic objectives to ARM (see Section 4.3). One such program, FIRE CIRRUS, supported by NASA, will study the influence of cirrus clouds on radiative transfer over a relatively homogeneous region in the midwestern United States in the fall of 1991. A pilot study, funded jointly by DOE and NASA, has been established with FIRE CIRRUS to obtain improved measurements for use in calibrating spectral line-by-line models. These models are used to develop parameterizations of infrared radiative transfer in GCMs. This program, known as SPECTRE, is an outgrowth of the finding that few measurements are available to calibrate line-by-line models to within an accuracy of a few percent (see Luther, *et al.* 1988). During FIRE CIRRUS, accurate measurements of infrared radiance at high spectral resolution will be made simultaneously with measurements of profiles of the radiatively important atmospheric properties. Achieving accuracies of 1-2% in the parameterizations of radiative transfer in GCMs is important, since changes of radiation of energy of as little as 1% still have climatological significance.

6. SUMMARY

A critical focus of the ARM program is to reduce the uncertainties in GCMs associated with cloud radiative processes. Progress on ARM has been made in a number of areas. Of specific importance, a Science Team has been established to provide guidance in the development of the ARM program so that ARM can meet its objectives of improving and reducing the uncertainties of cloud parameterizations and cloud radiative forcing functions in GCMs. In addition, a process is in place that will lead to the selection of CART sites, with the goal of commencing field operations in 1992. A CART Data System is being designed to provide the modeled and measured data from a select set of sites having a variety of meteorological, climatological and surface characteristics; the intended use of the data system is to conduct improved parameterizations and testing on GCMs and their components related to cloud radiative forcing. A pilot study, SPECTRE, will be conducted in the fall of 1991 in conjunction with NASA's FIRE CIRRUS program to obtain improved measures of infrared radiative transfer for use in line-by-line models.

7. ACKNOWLEDGMENTS

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4.1

CHLOROFLUOROCARBONS (CFCs) IN THE ATMOSPHERE: STRATOSPHERIC OZONE DEPLETION AND THE GREENHOUSE EFFECT

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1. STRATOSPHERIC PHOTOLYSIS OF CHLOROFLUOROCARBONS

The relationship between the chlorofluorocarbon (CFC) gases and stratospheric ozone depletion was first outlined in a short paper in Nature [1], and then in a long review paper [2]. The CFC gases (the most important are CCl_2F_2, CCl_3F, CCl_2FCClF_2) are chemically inert in the lower atmosphere, with the consequence that they can survive for very long periods of time--even 100 years of more--without destruction. The usual tropospheric sinks--photolysis, rainout, oxidation--don't affect the CFCs because they are transparent, insoluble and non-reactive. However, eventually the individual molecules diffuse upward into the middle stratosphere and are decomposed by short wavelength solar ultraviolet radiation, as shown in (1) for CCl_2F_2 (CFC-12). These wavelengths of ultraviolet radiation do

$$CCl_2F_2 + \text{u.v.} \ (<220 \text{ nm}) \longrightarrow Cl + CClF_2 \qquad (1)$$

not penetrate down to the surface of the Earth because they are absorbed in the lower stratosphere either by molecular O_2 or by ozone, O_3. The Cl atom released in (1) reacts within a fraction of a second with O_3 by (2), with the formation of the free radical chlorine oxide ClO. The ClO radical can then react within minutes with atomic oxygen by (3), once again releasing Cl. The combination of (2) plus (3) constitutes the ClO_x free radical catalytic chain reaction and repeats itself over and

$$Cl + O_3 \longrightarrow ClO + O_2 \qquad (2)$$
$$ClO + O \longrightarrow Cl + O_2 \qquad (3)$$
$$O + O_3 \longrightarrow O_2 + O_2 \qquad (4)$$

over again before finally being stopped. The two reactions summed together as in (4) convert back to molecular oxygen one ozone molecule and one atom of oxygen which would otherwise have formed ozone, making this ClO_x chain an exceptionally efficient method for ozone destruction.

Usually the ClO_x chain is interrupted by reactions such as (5) with CH_4 or (6) with NO_2 which form temporary reservoir molecules such as HCl and chlorine nitrate (ClONO2). Chlorine can

$$Cl + CH_4 \longrightarrow HCl + CH_3 \qquad (5)$$
$$ClO + NO_2 + M \longrightarrow ClONO_2 + M \qquad (6)$$

remain chemically inactive in these reservoir molecules for a few hours or days, only to be released once more as Cl by photolysis of $ClONO_2$ or HO reaction with HCl. The average chain length of the ClO_x chain in the stratosphere before drifting back into the troposphere (usually as HCl) is about 100,000. It is the combination of this chain multiplication factor of 10^5 plus the release to the atmosphere every year of about one million tons of CFCs which has made stratospheric ozone depletion by chlorofluorocarbons a major global environmental problem.

2. TROPOSPHERIC SINKS FOR CHLOROFLUOROCARBONS?

An important aspect of the chlorofluorocarbon problem is further consideration of the question of possible tropospheric sinks--whether solar ultraviolet decomposition is the only important removal process for the CFC molecules. Can rainout, oxidation, tropospheric photolysis, some unknown process or a combination of these processes remove some of the CFCs? The most complete test of the inertness of the CFCs rests upon the determination in the atmosphere itself of the lifetime of the CFC molecules. If some important tropospheric removal process were to exist for the CFCs (e.g., freezing out on Antarctic snow), then a substantial fraction of the CFCs would already have disappeared from the atmosphere. However, experimental atmospheric measurements have shown that the concentrations of CCl_2F_2, CCl_3F (CFC-11) and CCl_2FCClF_2 (CFC-113) have each increased very rapidly over the past fifteen years. In fact, the total organochlorine concentration of the atmosphere in 1990 (Figure 1) is about 3.8 parts per billion by volume (ppbv), compared to about 1.8 ppbv in 1974 and 0.8 ppbv in 1950. The major contributions to Cl in the atmosphere now come from CCl_2F_2, 0.96 ppbv Cl (0.48 ppbv CCl_2F_2 x 2 Cl atoms per molecule); CCl_3F, 0.84 ppbv; CCl_2FCClF_2, 0.24 ppbv; CH_3CCl_3 (methylchloroform), 0.48 ppbv; CCl_4 (carbon tetrachloride), 0.56 ppbv; and CH_3Cl (methyl chloride), 0.60 ppbv. All of these compounds except methyl chloride are entirely manmade. Comparison of the amounts of the CFCs now in the atmosphere with the amounts already released confirm that most of the CFC molecules are still there, leading to estimated lifetimes of about 75 years for CFC-11, 100 years for CFC-113, and 120 years for CFC-12.

3. OBSERVATIONS OF OZONE LOSS OVER ANTARCTICA

A second major aspect of ozone depletion by chlorofluorocarbons has been the search for evidence of actual ozone loss. By 1984, ozone loss in the upper stratosphere--the most sensitive altitude for loss by the ClO_x chain of (2) plus (3)--had been established, but statistical searches for total ozone loss had not disclosed any significant decreases. This situation was changed by the decisive discovery with ground-based instruments of massive losses of ozone over Antarctica [3,4], followed quickly by confirmation with satellite instruments that the ozone losses had occurred over an area roughly the size of Antarctica itself. These losses appear each year in early spring, only to be diluted away as the Antarctic summer approaches. By October 1989, the springtime ozone concentration over Antarctica had fallen by 70% from its 1960s levels, and the ozone loss had been demonstrated to occur very rapidly as soon as sunlight ended the long polar darkness in the lower stratosphere. About 3% of the entire world supply of ozone was lost in one month over Antarctica in 1989.

Three expeditions to the southern polar region were carried out during 1986 and 1987, two on the ground in Antarctica and the third using aircraft based in Punta Arenas, Chile. These expeditions have successfully elucidated the chemical and physical processes involved in the formation of the Antarctic ozone hole each September. First, the winter meteorology of Antarctica is dominated by the polar vortex which keeps stratospheric air trapped in the darkness over Antarctica throughout the winter and well into the following spring. The temperatures in this air fall to -85°C and -90°C, cold enough for the formation of polar stratospheric clouds (PSCs). These PSC clouds occur in two forms: initially, crystals of nitric acid trihydrate ($HNO_3 \cdot 3H_2O$) are formed, and later when it is still cooler, water ice forms as well.

The abundant particles in these PSC clouds furnish surfaces on which important heterogeneous nitrogen and chlorine chemistry occurs very rapidly in the darkness:

(a) the nitrogen oxides (NO, NO_2, NO_3, N_2O_5) are all converted into nitric acid and trapped in the cloud particles, leaving a gaseous air mass in which nitrogen oxides are essentially absent;

(b) chlorine reservoir compounds such as HCl and chlorine nitrate react with each other or with H_2O to form reactive Cl_2 or HOCl, as shown in (7) and (8). These chlorine compounds escape into the atmosphere while the nitric acid remains in the clouds. The first

$$HCl + ClONO_2 \longrightarrow Cl_2 + HNO_3 \qquad (7)$$
$$H_2O + ClONO_2 \longrightarrow HOCl + HNO_3 \qquad (8)$$

sunlight of approaching spring breaks into the polar darkness and photodissociates HOCl to release Cl, or Cl_2 into two Cl atoms, each of which then attacks O_3 by (2) to form ClO. In the absence of NO_2, chlorine nitrate cannot form, the concentration of ClO increases to the ppbv level, and two ClO radicals can react to form the chlorine oxide dimer, ClOOCl in (9). This molecule is then broken apart by sunlight in (10), releasing both Cl atoms again. This

$$2ClO + M \longrightarrow ClOOCl + M \qquad (9)$$
$$ClOOCl + u.v. \longrightarrow Cl + ClO_2$$
$$\longrightarrow 2\,Cl + O_2 \qquad (10)$$

sequence represents a different ClO_x chain which sums to $2\,O_3 \longrightarrow 3\,O_2$, and which continues to operate effectively until the brighter sunlight of mid-spring causes the PSCs to evaporate. As the clouds disappear, HNO_3 is set free and soon reacts to release NO_2. This, in turn, can combine with ClO to form the reservoir molecule chlorine nitrate and put an end to the runaway chain reaction of September and early October. Concentrations of ClO as high as 1.3 ppbv have been measured over Antarctica and also over the Arctic during the winter of 1988-89--more chlorine as ClO than was present in all chemical forms in 1970, as shown in Figure 1. Detailed

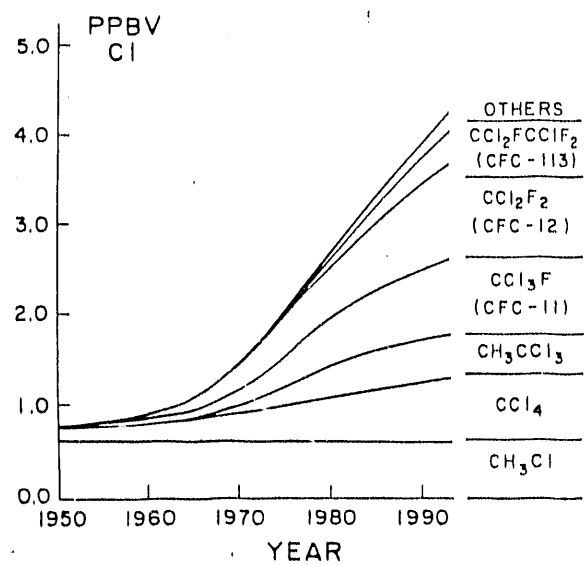

FIGURE 1. Organochlorine Concentrations in the Atmosphere, 1950-1990.

study of the experimental data conclusively shows that the ClO_x chains are the cause of the ozone depletion over Antarctica and that most of the chlorine in the ClO_x chains was put into the at-

mosphere by mankind. The most convincing single experimental data set is shown in Figure 2 which displays the

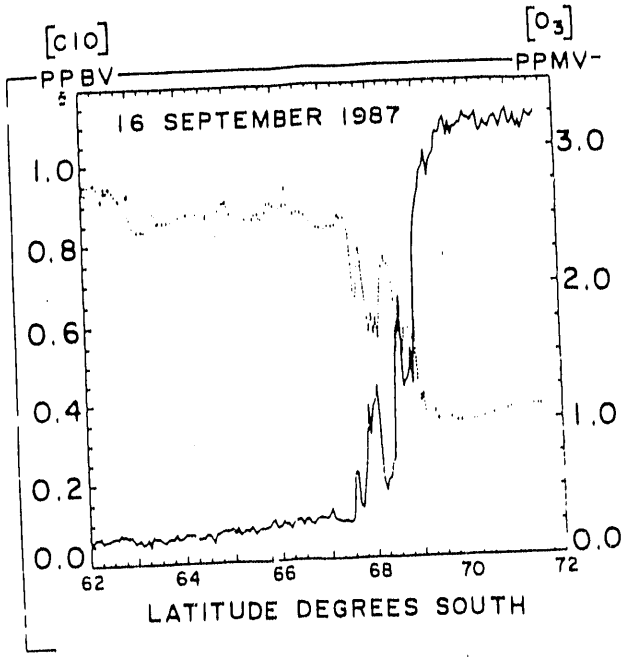

FIGURE 2. Chlorine Oxide and Ozone Concentrations over Antarctica at 18 Kilometer Altitude, September 16, 1990, as Measured on ER-2 Aircraft
———— ClO
-------- O_3

relationship between ClO and O_3 on the September 16 flight of the ER-2 aircraft [5]. On this day, the polar vortex had an irregular shape and ClO and O_3 went up and down several times in a strongly anti-correlated manner.

4. GLOBAL OZONE LOSSES

During 1986-1988, the historical ground-based records of total ozone were re-examined by the NASA Ozone Trends Panel and were found to show distinct seasonal differences not allowed for in the previous statistical analyses [6,7,8]. The revised statistical procedures emphasized a search for possible ozone variations on a month-by-month basis (e.g. change in January and in July might be different). With this approach, the data showed that ozone losses had already occurred in the broad latitude bands from 30°N to 64°N, with losses of about 2% since 1970 averaged over the entire year (Figure 3). The losses were shown to have been heaviest in the five-month period from November through March, and the probability is high that much of this ozone loss has originated through chemical mechanisms

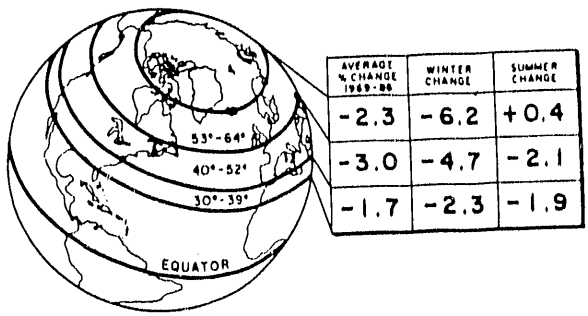

FIGURE 3. Measured Changes in Ozone Concentrations from 1969 to 1986 for Three Latitude Bonds in the Northern Hemisphere.
Winter = December through March
Summer = June, July, August

similar to those operating over Antarctica. These statistical procedures also made allowance for effects on ozone concentrations at each station attributable to the 11-year solar cycle, the QBO, and atmospheric testing of nuclear weapons three decades ago. An insufficient number of ground stations exists with 25 years or more of high quality ozone measurements to allow estimates of the ozone changes in other latitude bands in the tropics or in the southern hemisphere. However, the satellite instruments can be calibrated as they pass over the ground stations in the northern hemisphere and then used to monitor changes since 1978 in these regions. The satellite data indicate losses of ozone at essentially all latitudes over the past decade.

5. INCREASING CONCENTRATIONS OF TRACE GASES AND THE GREENHOUSE EFFECT

The chlorofluorocarbons are not the only trace gases whose concentrations are increasing in the atmosphere. Carbon dioxide has been regularly measured for more than 30 years, and the concentration pattern exhibits a yearly cycle superimposed on a steady growth, as shown in Figure 4. The amount of carbon dioxide decreases every spring and summer in the Northern hemisphere as it photosynthesized into growing green plants, only to return to a higher concentration the following autumn and winter. This 10% increase in 30 years in the total amount of carbon dioxide in the atmosphere is chiefly the result of the burning of the carbon-based fossil fuels, that is, coal, oil, and natural gas. Trace impurities such as SO_2 are also released during the burning of fossil fuels, but even "clean" fuels always give carbon dioxide. Additional

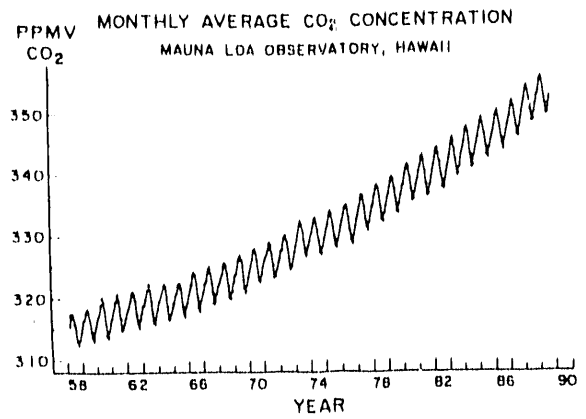

FIGURE 4. Measured CO2 Concentrations at Mauna Loa, Hawaii.

carbon dioxide is released during the burning of tropical forests.

The atmospheric concentration of methane is also growing. Our measurements since 1978 show in Figure 5 that

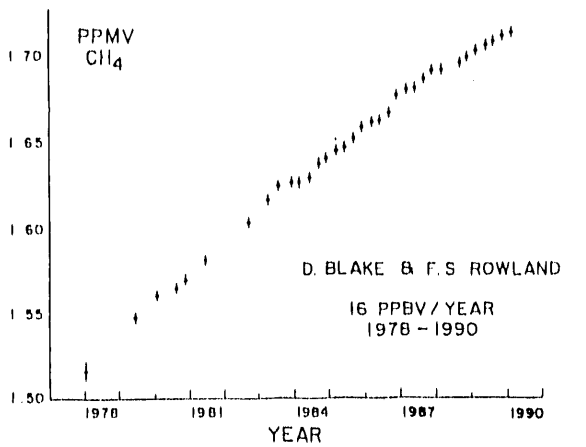

FIGURE 5. Globally-averaged Tropospheric Concentrations of Methane.

the amount of methane in the atmosphere has risen steadily from 1.52 to 1.71 parts per million by volume, 13% more in 12 years. The most abundant sources for atmospheric methane involve anaerobic biology--microbial action in the absence of oxygen or air. For example, methane is emitted from swamps and flooded rice paddies, and the stomachs of cattle give it off in large quantities. However, some CH_4 is also given off from fossil fuel applications--"natural gas" is itself primarily methane. Furthermore, nitrous oxide (N_2O) is increasing 0.2% per year, and ozone near the Earth's surface is also rising as well.

The major atmospheric problem caused by these increases in carbon dioxide, methane, the CFCs, and other gases is now described as the "greenhouse effect." Incoming solar energy arrives primarily as visible radiation, and an equivalent amount of energy must escape from the atmosphere in order to keep the earth in overall heat balance. The peak intensities for visible radiation occur at wavelengths around 500 nanometers (nm), in response to the solar surface temperature of about 5600K. With an average surface temperature near 288K, the peak intensity of the escaping radiation will be emitted at wavelengths around 20 times (i.e., 5600/288) longer than 500 nm, or 10,000 nm. These wavelengths are in the infrared (IR) region and are usually expressed as 10 microns rather than 10^4 nm. The internal vibrational motion of triatomic molecules have frequencies corresponding to these wavelengths with the consequence that naturally occurring molecules such as H_2O, O_3 and CO_2 are capable of absorbing this outgoing IR radiation, re-emitting it in all directions. With such IR escape to space partially hindered, Earth warms up sufficiently to force additional IR out through the transparent wavelengths. Estimates have been made that Earth's natural atmosphere is warmer by about 35°C than a hypothetical atmosphere free of such IR absorbing molecules.

Added molecules such as carbon dioxide and methane and the CFCs are also able to absorb this infrared radiation, preventing still more from making its way directly into space. The atmosphere will respond to this increased IR forcing by the accumulating trace gases, but the quantitative response is difficult to estimate. Most climatologists expect an increase in the average global temperature as one consequence of this trace gas perturbation, with estimated increases of 3.0±1.5°C by the year 2050. A major uncertainty in these estimates is connected with possible changes in cloudiness. An increase in horizontal area obscured by clouds would increase the visible albedo and reduce the intensity at the surface. Such a change would reduce both the amount of energy going into subsequent IR emission and the amount of sunlight available for photosynthesis. Actual measurements have shown that the average temperature of the earth's atmosphere over the last 100 years has risen by more than 0.5°C.

The current yearly CO_2 concentration increase of 1.5 parts per million by volume is approximately 100 times larger than for CH_4 at 16 parts per billion by volume, and 75,000 times greater than CCl_2F_2 at about 20 parts per trillion by volume. However, these incremental CO_2 molecules absorb precisely the same wavelengths absorbed by all of the existing CO_2, greatly reducing the incremental CO_2 IR absorption efficiency. Because the vibrations corresponding to the stretching of C-Cl and C-F

bonds fall in transparent wavelength bands, incremental additions of molecules such as CCl_2F_2 and CCl_3F are about 15,000 times as efficient in IR absorption as CO_2. Incremental methane is about 30 times more efficient than CO_2. For this reason, the accumulations of the other trace gases had an incremental greenhouse forcing effect in the 1980s approximately equivalent to that from CO_2.

This greenhouse effect is eventually expected to cause a general rise in the sea level around the world, partly from thermal expansion during the warming of the oceans and partly from added liquid water in the oceans after the melting of glacier and polar ice presently accumulated on land. Changes can also be expected in rainfall patterns, so that some regions now accustomed to plentiful rain might perhaps have less rain in the future. While the average global temperature is predicted to go up as the result of the greenhouse effect, regional temperature changes are expected often to be substantially different from the average, and can even include cooling in some locations. Future regional temperature and rainfall patterns are very hard to predict with current atmospheric models which usually operate on a grid scale encompassing 4 or 5 degrees of both latitude and longitude. Accurate model predictions on a much finer grid scale are not primarily limited by computer power, but by absence of sufficient experimental atmospheric knowledge for calibrating and validating the models.

6. CONSEQUENCES OF OZONE DEPLETION

The primary consequences of ozone depletion can be described under the categories of climatic and biological. The actual existence of the stratosphere, with temperatures increasing with increasing altitude, is caused by the absorption of solar ultraviolet radiation in the ozone layer. With less ozone in the upper stratosphere, as already observed, the temperature in the 40-50 km altitude range can be expected to decrease and, indeed, has dropped by 1.7±1.0°C in the past decade. With altered stratospheric temperatures and gradients, wind patterns could also eventually be effected.

Ultraviolet wavelengths between 293 nm and 320 nm are only partially absorbed by stratospheric ozone, and some of it penetrates to the surface of the Earth where it can be absorbed by the various biological species. With humans, the major consequences of this UV-B radiation are increased incidence of skin cancer, increased numbers of eye cataracts, and probably partial suppression of the immune system. Many other biological species are also susceptible to UV-B radiation, but the effects are well-studied and well-documented with only a few non-human biological species.

7. INTERNATIONAL REGULATION OF CFC EMISSIONS

An international protocol to control the emissions of CFCs to the atmosphere was agreed upon in Montreal in September 1987. Under its terms, the total global emissions of CFCs were to be reduced by 20% below the 1986 emission levels by 1994 and reduced an additional 30% by 1999. This agreement has now been strengthened at the meeting in London in June 1990, and calls for an essentially complete phaseout by the year 2000. Many of the major CFC producing countries are independently committed to an even faster phaseout schedule for the CFCs. Nevertheless, the amounts of CFCs emitted to the atmosphere during the five years from 1985 through 1989 exceeded the emissions in any preceding five-year period, and measurements of the CFCs in the atmosphere do not yet show any slackening in the rate of increase in their atmospheric concentrations.

8. CFC SUBSTITUTES

Replacements for CFCs in most of its uses are being developed very rapidly. The major uses in 1988 included aerosol propellant (CFC-11 plus CFC-12), plastic foam blowing (either CFC-11 or CFC-12), cleaning of electronics (CFC-113), refrigerant (CFC-12), etc. Some of the replacements can be described as "not-in-kind", as with hydrocarbons for CFCs or a manual air pump for an aerosol. Conservation is another important opportunity for saving CFCs. Finally, several new compounds from the HCFC and HFC classes are being produced. The difference is that these molecules have C-H bonds which can react with HO in the troposphere, as shown with $CHClF_2$ (HCFC-22) in equation (11). This compound is already in wide usage as the refrigerant

$$HO + CHClF_2 \longrightarrow H_2O + CClF_2 \qquad (11)$$

in home air conditioners. Other new compounds which will soon be available are HCFC-123 ($CHCl_2CF_3$) for CFC-11 and HFC-134A (CH_2FCF_3) for CFC-12 in some applications.

9. LONG LIFE FOR CFCs AND OZONE DEPLETION

Because about 10% of the CFCs are employed in usages which involve slow emission to the atmosphere, the maximum concentration of CFCs in the troposphere can be expected about a decade after the completion of the phaseout process

(i.e., about the year 2010 under the terms of the London revision of the Montreal Protocol). The maximum stratospheric effects can be anticipated another 5 or 10 years after that because of the delay time from release to decomposition in the upper stratosphere, or about the years 2015 to 2020. With the very long observed atmospheric lifetimes for the CFCs, stratospheric ozone depletion from the CFCs and the effects from this ozone loss will be felt on a global scale throughout the 21st century.

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