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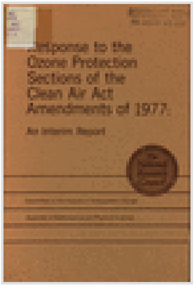

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Response to the Ozone Protection Sections of the Clean Air Act Amendments of 1977:

An Interim Report

Committee on the Impacts of Stratospheric Change
Assembly of Mathematical and Physical Sciences
National Research Council

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NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the Councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the Committee responsible for the report were chosen for their special competencies and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by the Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

Available from
Committee on the Impacts of Stratospheric Change
2101 Constitution Avenue
Washington, D.C. 20418

NATIONAL RESEARCH COUNCIL
ASSEMBLY OF MATHEMATICAL AND PHYSICAL SCIENCES
2101 Constitution Avenue Washington, D.C. 20418

Committee on
Impacts of Stratospheric Change

December 15, 1977

Dr. Philip Handler
President
National Academy of Sciences
Washington, D.C. 20418

Dear President Handler:

This interim report represents the Committee's short-term response to the desire expressed in the Clean Air Act Amendment of 1977 for reports covering knowledge and research progress in three areas related to influences on human health and welfare arising from human controlled impacts on the stratosphere. It summarizes either the situation or the changes since our previous report in two of these three areas.

This Committee plans to prepare a much more complete report in about a year's time as well as an update of its previous report on the impact of chlorofluoromethanes. This latter report will, we hope, be available in the summer of 1978.

While the scope of the present report has been enlarged, because of the wording of the Clean Air Act Amendments for 1977, to cover <u>all</u> stratospheric changes, changes connected with ozone depletion continue to appear to be the most important, followed by effects due to infrared absorption of traced gases. Both of these kinds of effects were addressed in our earlier report; we can now say somewhat more about each.

Very sincerely yours,

John W Tukey

J. W. Tukey
Chairman

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NATIONAL ACADEMY OF SCIENCES

OFFICE OF THE PRESIDENT
2101 CONSTITUTION AVENUE
WASHINGTON, D.C. 20418

December 19, 1977

The Administrator
Environmental Protection Agency
Washington, D. C. 20460

Dear Sir:

I am pleased to submit a report, prepared by our Committee on Impacts of Stratospheric Change, on the effects on human activities on the stratosphere and consequent implications for human health and welfare. The report is responsive to the provisions of Sec. 153(d) of the Clean Air Act, as amended, which states that: "The Administrator shall undertake to contract with the National Academy of Sciences to study the state of knowledge and the adequacy of research efforts to understand (A) the effects of all substances, practices, processes, and activities which may affect the stratosphere, especially ozone in the stratosphere; (B) the health and welfare effects of modifications of the stratosphere, especially ozone in the stratosphere; and (C) methods of control of such substances, practices, and activities including alternatives, costs, feasibility, and timing." The Act calls for a report on the findings of the Academy by January 1, 1978.

The present report covers those aspects of (A) and (B) above that the Committee believed it was reasonable to analyze and interpret within the limited time available. A more detailed report from the Committee will be provided well before the deadline for your next report to Congress as mandated by Section 153. The Committee also plans to prepare, by the fall of 1978, an updating of its report of September 1976, "Halocarbons: Environmental Effects of CFM Release."

The Committee did not believe that an adequate and useful response to (C) could be prepared by 1 January 1978. The issues are too diverse and complex for a hasty collection of incomplete information to be of any value. We will be prepared, however, to carry out a detailed study of the issues on the longer time scale.

In many respects, the problem of inadvertent modification of the stratosphere and the attendant consequences can serve as a model of human impacts on the environment. Fortunately, it is a problem that has been discovered well before it poses any significant threat to human welfare. It is also a matter of worldwide scope that will require international cooperation both to understand its full magnitude and to minimize its adverse consequences. Control of these consequences will have to be based upon a sustained program of research in a variety of disciplines as outlined in this and earlier committee reports. If we fail to support such a research program we will lose the opportunity to act intelligently before the consequences reach significant and possibly very damaging proportions.

Sincerely yours,

PHandler

Philip Handler
President

Enclosure

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INTRODUCTION

1A. PURPOSE

This is the first report from the National Academy of Sciences-National Research Council in response to the direction of the Congress that the Administrator of the Environmental Protection Agency endeavor to contract with the National Academy of Sciences "to study the state of knowledge and the adequacy of research efforts to understand (a) the effects of all substances, practices, processes, and activities which may affect the stratosphere, especially ozone in the stratosphere; (b) the health and welfare effects of modifications of the stratosphere, especially ozone in the stratosphere; and (c) methods of control of such substances, practices, and activities including alternatives, costs, feasibility, and timing." For the reasons outlined in the letter of transmittal from the president of the Academy this interim report is limited to aspects of (a) and (b).

The present chapter outlines the routes by which human activities affect or seem likely to affect those aspects of the stratosphere that then may affect human health and welfare. Sometimes these routes are simple chains; more often they involve networks of relationships.

The links in these networks of cause and effect are of varied strength. A few are relatively well understood and expressable in numbers about which there is only moderate uncertainty. Others are known with considerably less precision; while still others are only suggested as deserving of study.

We have not tried to include all the links that have been suggested. On the other hand, we have by no means restricted our outline to those that we believe should be matters of immediate public concern. Thus, in addition to the few already recognized as matters of public concern, we have included others whose existence is reasonably certain but whose importance is not clear at this time.

1B. FOUR BROAD ASPECTS OF THE STRATOSPHERE

As far as our present understanding goes, the aspects of the stratosphere through which human activities will influence human health and welfare fall into four parts:

1. The total amount of ozone present and its distribution in height.
2. The general temperature pattern and general circulation of the stratosphere.
3. The presence of CO_2 and trace gases that absorb infrared radiation. These are shared between stratosphere and troposphere and exert their effects* in all parts of the earth's atmosphere (trace-gas and CO_2 greenhouse effects). Somewhat similar effects come from the presence of fine particles.
4. The presence both of radioactive particles that will gradually become radioactive fallout and of radioactive gases. The latter is also shared with the lower atmosphere.

The first two of these are the principal subjects of Chapters 3 and 4.

In this report we have tried to share the responsibility for the description and assessment of the implications of substances (gases) and particles that spread through the whole atmosphere rather than the stratosphere alone; (1) leaving the important CO_2 greenhouse effects to other studies and reports†; (2) discussing briefly the trace-gas greenhouse effects; (3) leaving tropospheric aerosol

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*By increasing the absorption of infrared radiation and thus reducing the flow of heat from earth toward space for a fixed surface temperature.

†For example, Geophysics Study Committee, *Energy and Climate* (National Academy of Sciences, Washington, D.C., 1977).

effects to other studies and reports; (4) mentioning the human-related portion of the stratospheric aerosol effects; (5) leaving the shared radioactive gases, which do not seem to be a matter of appreciable concern, to other studies and reports; and (6) mentioning the status of radioactive particles in the stratosphere.

1C. ROUTES OF EFFECTS ON HUMAN HEALTH AND WELFARE

The total amount of ozone in the atmosphere, most of which is in the stratosphere, determines the absorption of most of the incoming radiation of the sun of those wavelengths (290 to 320 nm) that contribute to DUV (biologically damaging ultraviolet radiation). Thus changes in total ozone cause changes in the amount of DUV reaching the earth's surface, with consequent effects upon

1. *Humans*, where DUV, even without ozone reduction, is a major cause of skin cancer.
2. Other *biological systems*, where effects have not yet been adequately studied.
3. Urban atmospheric pollution in the form of *photochemical smog*, where DUV contributes to the production of photochemical smog.
4. Altering the concentration in the lower atmosphere of hydroxyl radicals, which are important in the destruction of other trace constituents such as methyl chloroform and some trace gases that have greenhouse effects, (a) reducing the amounts of these materials in the troposphere and (b) decreasing the amounts reaching the stratosphere ("tropospheric cleanup").

Both the general stratospheric circulation and temperature and the intensity of the trace-gas greenhouse effect can contribute to changes in the earth's short-term and long-term climate. The impact of such changes on human health and welfare would come mainly through effects on agriculture.

The amount of radioactive holdup present in the stratosphere in the form of particles, and its distribution in height, is responsible for the rate at which long-delayed radioactive fallout takes place.

1D. ROUTES OF EFFECTS OF HUMAN ACTIVITIES ON THE TOTAL STRATOSPHERIC OZONE--FINAL STEPS

The total amount and distribution in height of stratospheric ozone is affected by the amounts of various gases reaching--or formed in--the stratosphere, especially

1. "Oxides of hydrogen" (HO_x, mainly the free radicals HO and HO_2, the compound H_2O_2, and free H atoms), the hydrogen arising from methane or arriving as water vapor.
2. "Oxides of chlorine" (ClO_x, mainly ClO radicals, Cl atoms, and HCl and $ClONO_2$ molecules) collectively formed in the stratosphere from chlorine-containing gases of both natural and human origin.
3. "Oxides of nitrogen" (NO_x, mainly the compounds NO, NO_2, N_2O_5, and HNO_3) either (a) formed in the stratosphere from N_2O that reaches the stratosphere from plant life, soils, and the ocean, the N_2O arising either naturally or as modified by cultivation and human uses of fertilizer; or (b) formed in the stratosphere by particle radiation from the sun, the exhausts of high-flying aircraft, cosmic radiation, or nuclear explosions; or (c) from N_2O formed, probably in small quantities, by the combustion of fossil fuels.

And to a still lesser degree by the smaller amounts reaching the stratosphere of

4. "Oxides of bromine" (BrO_x; similar to ClO_x) reaching the stratosphere from natural or human sources.

A further mechanism has been suggested, involving

5. Traces of certain metals on fine aerosol particles, traces of which may or may not be catalytically active.

1E. ROUTES OF EFFECTS OF HUMAN ACTIVITIES ON THE TOTAL STRATOSPHERIC OZONE--EARLIER STEPS

The occurrence of oxides of chlorine (ClO_x) in the stratosphere is almost exclusively a consequence of the passage of relatively stable chlorine-containing compounds from the troposphere through the lower stratosphere to where ultraviolet radiation breaks up the otherwise stable molecules. This occurs for the man-made chlorofluoromethanes* as well as for a variety of other compounds from both natural sources and human activities. (Of those generated by humans, the chlorofluoromethanes are the most important.)

More reactive compounds are attacked and, to varying degrees, destroyed in the troposphere. The role of trace constituents such as OH mainly formed by reactions of excited ("singlet-D") oxygen atoms with water is crucial in this destruction. In the troposphere, these very active oxygen atoms are formed by absorption by *tropo*spheric ozone, of some of that fraction of DUV radiation that reaches the troposphere (having escaped absorption by *strato*spheric ozone). However, human production of CO and NO_x, as from automobile exhausts and fossil-fuel burning for heat and power, is also important in altering this process of tropospheric cleanup.

The effectiveness of tropospheric cleanup may be important (a) in affecting the fraction of gases of intermediate stability (e.g., hydrogen-containing halocarbons) reaching the stratosphere, and there contributing to ozone decrease (through ClO_x production) and (b) in removing other trace constituents. The occurrence of tropospheric cleanup can, however, also lead to reaction products that contribute to such undesirable results as photochemical smog.

1F. ROUTES OF HUMAN EFFECTS ON OTHER STRATOSPHERIC ASPECTS

The general stratospheric circulation and temperature is influenced by

1. The effect due to all infrared-absorbing gases (both as an average and as it varies from place to place and from one height to another);
2. The distribution of stratospheric ozone in space and time;
3. The amounts of water vapor present in the stratosphere at various locations and heights;
4. The amounts of NO_x present, from both natural and human sources, in the stratosphere;
5. The reflectivity (albedo) of the earth's surface,

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*Often known under one of their tradenames as Freons.

which might be changed by cultivation, forest removal, irrigation, and other human activities.

The additional effect from most infrared-absorbing trace gases extends throughout the whole atmosphere. The chlorofluoromethanes, F-11 ($CFCl_3$) and F-12 (CF_2Cl_2), are among the more effective of the trace gases thus far recognized as having relevant infrared absorption.

Radioactive particles in the stratosphere have arisen in large part as a consequence of nuclear tests but also from cosmic radiation.

2
GENERAL SUMMARY

2A. INTRODUCTION

The previous report from this committee, *Halocarbons: Environmental Effects of Chlorofluoromethane Release*, while focused on a more specific problem, actually discussed or mentioned most of the situations and problems outlined in the previous chapter. As a result, the present interim report covering all important recognized human interactions with the stratosphere can be almost entirely restricted to an update.

2B. NEW KNOWLEDGE

New knowledge has been gained in the year and three months since the completion of the earlier report. Some of the progress hoped for within two years has occurred, and we have learned some things that are surprising. New values of rate constants, most already reported in a NASA report,* have increased the calculated effectiveness of changing amounts of oxides of chlorine in reducing ozone but decreased the effects of changing amounts of oxides of nitrogen. This is a consequence of the recently recognized strong coupling between the chlorine and nitrogen reactions. As a further consequence, the sensitivity to changes of either depends markedly on the amount of the other present.

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*R. D. Hudson, ed., *Chlorofluoromethanes and the Stratosphere* (NASA, Washington, D.C., 1977), p. 192.

As a result, the estimated seriousness of release of chlorofluoromethanes measured by ozone reduction has been roughly doubled. (After this increase, the best single estimate remains within the limits of uncertainty stated in our previous report).*

The estimated impact of NO_x from the exhausts of SST's and other high-flying aircraft on stratospheric ozone is now quite small, almost certainly not a matter of immediate concern. The same can be said of the possible increases in N_2O releases associated with increasing uses of fertilizers over the next decade. However, the impact on stratospheric ozone of quite large increases of oxides of nitrogen is still calculated to be serious; thus we shall have to be keenly aware of the fertilizer-N_2O question for the more distant future.

Both new laboratory results and new measurements in the stratosphere have contributed to unresolved discrepancies between model calculations and measurements. A period of intercomparison and adjustment is needed before the modified results just outlined can be made more reliably quantitative. Increased attention has been given to the processes through which some compounds are attacked, largely by OH radicals, in the troposphere before reaching the stratosphere. Our concern with tropospheric cleanup has its paradoxes, as when the use of trichlorethylene for degreasing has been banned because it is rapidly attacked and its reaction products contribute to photochemical smog, while there is some concern that a substitute, methyl chloroform, which is not as readily attacked, may cause difficulty by reaching the stratosphere too easily and there producing ClO_x.

Potential climatic impacts of stratospheric change have been studied, and the overall picture seems slightly clearer. The largest concern remains the infrared absorption effect from increasing amounts of CO_2, spread throughout the atmosphere. Next after this comes infrared absorption effects of a variety of trace gases including chlorofluoromethanes and N_2O. Other climate effects involving the stratosphere more specifically (note that the two just mentioned involve the whole atmosphere) seem likely to be still less important, with the exception of

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*Our previous report estimated from 2 to 20 percent reduction in ozone as the ultimate result (after many decades) of continued release at 1973 rates (a conventional basis for comparison).

the possible effects of the redistribution of ozone in height associated with increases in such ozone antagonists as oxides of chlorine or oxides of nitrogen. (Such effects of ozone redistribution would have to involve modification of the general circulation and temperature patterns in the stratosphere.) We are still unable to assess the size of such climatic effects with adequate accuracy; work on climate modeling continues actively, but no breakthroughs are in sight.

The Environmental Protection Agency-funded short-term interagency program of research on the biological consequences of the sort of increases of ultraviolet radiation that comes from ozone reduction (natural or human influenced) has taught us some useful things, both about differences among the responses of different species and about the mechanisms by which some plants respond.

Knowledge of the relation between increased exposure to ozone-shielded ultraviolet light and human health has been somewhat strengthened. Estimates of increased malignant melanoma deaths from specified amounts of increased exposure are now somewhat lower, while evidence for the importance of intermittent exposure in producing melanoma is somewhat greater; the responsibility of human behavior changes for the great increase of malignant melanoma in recent decades is even clearer.

Nothing that has been learned recently about the impact of climatic changes (of plausible size) on the biological world has changed our estimates or uncertainties enough to deserve special mention here.

The present amount of radioactive particles resident in the stratosphere does not seem to be in any way a pressing concern.

We have found no important progress in assessing the impact of increased ultraviolet light on materials.

2C. ADEQUACY OF RESEARCH

Recommendations made in pages 21 to 27 of our previous report remain, we believe, as pressing and important as ever. (We add a few detailed recommendations in the body of this report.) Especially in the biological area, we still fail to find a long-term research program with adequate peer review and careful coupling of what is done to what is most needed. (Such coordination must, of course, be so planned and managed as to allow excellent novel ideas rapid entry into the program.) The short-term interagency program has been more useful than we might have expected. It is still, however, not a proper model for a longer-term program and is threatened with serious cuts. The issues that most need to be attacked are not those that will lead to regulations in 1978, 1979, or 1980. The solution of the important issues will contribute, instead, to a much deeper understanding of the problem.

Programs of stratospheric measurement and laboratory study of chemical reactions have been rewarding; they need to be pushed forward with vigor. The same is true of atmospheric modeling, particularly the development of higher-dimensional models.

Climate studies involving a wide variety of questions, including those mentioned in this report, are now being recognized as overdue. Efforts to improve our understanding of climate and climate change should be increased. In particular, while we have left the main responsibility for the response of climate to the CO_2 infrared absorption effect to others,* we dare not fail to emphasize the urgency of further work in this area.

Thought should be given to mechanisms that will accelerate our understanding of stratospheric circulation and structure.

Studies of the nitrogen cycle, including the relation of fertilizer application to N_2O production, deserve continuing emphasis.

Studies of the relation of the human health to ultraviolet exposure, particularly in relation to skin type, have begun to expand. This expansion should continue.

More details of some of these matters will be found in the summary sections of the following chapters.

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*For example, the Geophysics Study Committee document, *Energy and Climate*, cited earlier.

3

STRATOSPHERIC CHEMISTRY AND TRANSPORT

3A. INTRODUCTION

A nonspecialist, reading successive reports on the effects of human activities on the ozone shield, might wonder why the "best" projections of ozone depletion for individual pollutants have changed with time while the range of uncertainties have not narrowed appreciably. He would be wrong to conclude, however, that little progress has been made in our understanding of the stratosphere in general and ozone in particular. Significant advances have been made, but with them has come an increasing realization of the complexity of the stratosphere, as to both its motions and its chemistry, forcing the consideration of an increasing number of detailed questions.

Models used to describe the atmosphere have had to use approximations for both vertical and horizontal motions as well as for a number of other physical properties. These include the way the solar intensity changes with altitude and time, including such effects as multiple scattering from molecules and particles, and reflections from the surface and from clouds. Although considerable progress has been made in treating these problems theoretically, the approximations have yet to be directly tested by physical measurements in the stratosphere.

On the chemical side, it is worth remembering that, before the early 1960's, four chemical reactions were

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Based on draft prepared by the Committee's Panel on Stratospheric Chemistry and Transport: H. I. Schiff, *chairman*; J. Chang, R. Dickinson, J. P. Friend, F. Kaufman, R. A. Marcus, J. W. Tukey, C. Walling, and G. Watson.

thought sufficient to explain the major features of ozone in the stratosphere. The substantial efforts (started by CIAP*) in the early 1970's both in stratospheric and laboratory measurements have resulted in more than 100 chemical reactions being included in current stratospheric models. There has also been an increasing awareness of the close coupling between many of the chemical families (NO_x, HO_x, and ClO_x) that affect ozone. This means that injection of one substance changes not only one but all the catalytic cycles that determine the ozone concentration. It is, therefore, not surprising that new measurements, either in the laboratory or in the stratosphere, while answering certain questions, also reveal additional ones. The new measurements have, however, brought us closer to an adequate quantitative understanding of the stratosphere.

3B. SUMMARY

Several new measurements of chemical rate constants have resulted in a downward revision of the predictions of ozone reduction from SST's and from N_2O releases related to increased use of fertilizers. Current estimates for SST effects indicate that appreciable ozone reduction is unlikely for fleet sizes currently projected. Possible effects of increased fertilizer usage are much more difficult to assess since they involve the additional complexities of biological cycles. Fortunately, the time scales of the possible fertilizer effects are sufficiently long that the solution of some of these problems can be left to vigorous, orderly research.

New values for some of the key rate constants have had the effect of increasing the calculated ozone reduction corresponding to continuing releases of CFM's at the 1974 levels. The central figures (of 11 to 16 percent) given by the NASA report, which uses these rate constants, are about a factor of 2 larger than the corresponding value given in our earlier report (which used the older estimates)

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*CIAP, the Climatic Impact Assessment Program of the Department of Transportation, with which was associated the work of the National Research Council's Climatic Impact Committee.

but are still within the range of uncertainties of the latter.

Because the reaction with HO provides a relatively strong tropospheric sink for these compounds, the recovery of the ozone layer following any reduction in their release rate will occur in a much shorter time scale than is the case for F-11 and F-12.

At present, the major threat to the ozone layer is from the releases of F-11 and F-12. There are no new developments that suggest that the effects will be smaller than the ones outlined in our previous report; in fact, as noted above, the best available figure is about twice what the best figure was in 1976. Other halocarbons, particularly methyl chloroform in view of the continued exponential growth in its production, appear to pose a next greatest threat to stratospheric ozone. The N_2O-fertilizer threat is presently uncertain but, if important, would occur on a longer time scale.

The new values of the rate constants have produced some inconsistencies between the model calculations and a number of the stratospheric measurements. There are also some serious problems in explaining some of the recent ClO measurements and the latitudinal and temporal dependencies of NO_2. Until these problems are resolved, the quantitative predictions of the models including the allowances for recognized uncertainties (e.g., those in the NASA report) must still be viewed with reasonable caution.

The possibility of inactive removal of $CFCl_3$ (F-11) and CF_2Cl_2(F-12) in the troposphere by processes that prevent them from reaching the stratosphere is still an open question. Attempts to make direct overall tests for the existence of such sinks, by comparisons between the release rates of these CFM's into the atmosphere and the amounts measured in the atmosphere or their time dependence, remain inconclusive, largely because of recognized uncertainties in the measurements. The suggestion that surface photolysis in arid regions may be significant remains speculative.

The increasing production of other chlorine-containing compounds, particularly CH_3CCl_3 (methylchloroform) and CHF_2Cl (F-22) has gained attention. While these substances are subject to removal in the troposphere by reaction with OH,* the rates are slow enough that an appreciable fraction

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*We use "OH" here for emphasis and historical connection, although we shall use the chemically somewhat preferred "HO" hereafter.

of these substances can make their way into the stratosphere. The uncertainty in the OH concentration makes this fraction difficult to evaluate accurately.

3C. MODELS OF THE STRATOSPHERE

Coupled chemical-kinetics and transport models of the stratosphere consist of a set of mathematical equations describing the complex chemical and photochemical interactions among the relevant chemical species in the stratosphere and of the processes by which these species are transported. In the current generation of stratospheric models, although the coupling between chemistry and atmospheric transport is included, atmospheric chemistry clearly receives the major attention. Typical models include a hundred or so chemical interactions among 20-30 chemical species. The system of mutual interactions is so complex that the set of equations in the model can only be solved on large computers. Even with the largest computers it is not possible to represent the detailed three-dimensional motions in the atmosphere while including the detailed chemical interactions. Consequently, reduced models have been developed that represent either only the vertical direction (1-D models) or only the vertical and latitudinal directions (2-D models). Much of the current knowledge about the chemical structure and the coupling processes of the stratosphere has been derived with the help of 1-D models. Compared with the development of 1-D models, the progress in 2-D modeling has been slow. The recent direction in 2-D model development parallels the 1-D model development of several years ago, i.e., validation of individual subprocesses, analysis of model variability, intermodel comparison, and model sensitivity analysis.

In the meantime, 1-D models have been improved in various ways. Several deficiencies have been partially removed. Uncertainties due to omission of stratospheric temperature feedback and of totally self-consistent diurnal averaging have been studied and in many instances reduced. Systematic sensitivity analyses of model predictions to uncertainties in input variables are beginning. Along with these developments has come a heightened awareness of the complexity of the system, e.g., the coupling of stratospheric chemistry to radiative and transport processes.

It is now completely clear that all the major chemical catalytic cycles are closely coupled and cannot be studied separately. In fact, recent progress in stratospheric chemistry has come mainly from new laboratory data on chemical-kinetic reactions linking various basic catalytic cycles. As a consequence, the predictions of ozone perturbations due to various natural or man-made causes have been revised. Predicted ozone reductions from NO_x increases have become smaller or nonexistent, whether the source is SST or extended usage of fertilizers. This change is a result of the existence of canceling effects--ozone reduction in the upper stratosphere and ozone increase in the lower stratosphere--both coming from an increase in NO_x. The apparent extent of this cancellation is highly sensitive to the choice of transport-coefficient profile used in the model; within the context of 1-D models there is no basis for determining a best value. Further clarification of the issue requires a multidimensional model with more realistic representation of atmospheric motions.

One-dimensional models are not now expected to undergo major structural changes except in the modification of chemical-kinetic systems by the inclusion of new chemical species, new reactions, or improved reaction-rate coefficients. Because of the necessarily greater simplification in model chemistry and the increased coupling to detailed transport, we must be careful in the development of multidimensional models. The details of their chemistry must be carefully validated, and the details of their transport must be compared with atmospheric data. The 3-D models that are under development include very limited chemistry. (The extent of possible improvement is not clear at this time.) It is important that more research effort be devoted to the understanding of the relative--and most likely complimentary--roles of models involving different numbers of dimensions.

3D. INPUT TERMS FOR MODEL CALCULATIONS (SOURCES)

As the preceding section has indicated, a very large collection of physical and chemical parameters is required as input into the basic equation of 1-D models even though such models are, in themselves, crude approximations to the incredible complexity of the real atmosphere. These parameters fall into three groups: (a) strengths of sources of substances likely to affect stratospheric ozone; (b) rate coefficients that characterize the speed of chemical and photochemical reactions among all species that directly or indirectly affect the ozone budget; and (c) the 1-D transport (eddy-mixing) coefficient as function of altitude (the transport-profile). It is well to remember that, even in the present highly simplified description of the stratosphere, 20 to 30 different chemical species undergoing 100 to 150 chemical and photochemical reactions have to be considered and that the dependence on temperature and pressure of the corresponding reaction rates needs to be known. For solar photodissociation reactions, one also needs to know the absorption spectrum of the particular substance, its quantum yield (fraction of light that leads to decomposition) as a function of wavelength, and the concentrations and absorption coefficients of all other substances above any given altitude that may reduce the available solar flux. The present chapter and the two that follow will give a brief update covering information on these parameters not included in earlier, much more detailed reports.*

NO_x Sources

Although all calculations indicate that present and currently projected fleets of SST's would have an unimportant impact on stratospheric ozone, it should be pointed out that the large discrepancy between different experimental determinations of the NO_x emission index of SST jet engines (values of 16 to 18 g of NO_2 per kg of fuel versus values of 30 to 40 for this index) has not yet been resolved.

Concerning the fertilizer-N_2O problem, much experimental work on the fractional yield of N_2O in denitrification is currently under way. Estimates range from about 6 to 40 percent corresponding to long or short atmospheric lifetimes (~140 or ~20 years) of N_2O (Logan *et al.*, 1977). Projected human-controlled sources of N_2O due to fertilizer use and fossil-fuel combustion have been estimated to rise by about an order of magnitude over the next hundred years (Logan *et al.*, 1977).

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*For example, the Panel's last report, *Halocarbons: Effects on Stratospheric Ozone*, and the NASA report *Chlorofluoromethanes and the Stratosphere*.

CFM Sources

The Manufacturing Chemists Association (MCA) has continued to gather data on production of CFM's, and on the basis of this information, estimates of worldwide production and release have been made by the Dupont Company (McCarthy, 1977). Data are summarized in Table 1.

TABLE 1. Halocarbon Production

	(10^3 Metric Tons)			
	F-11	F-12	F-22[a]	Methyl Chloroform[b]
1973 production	368	441	77	351
1974 production	400	474	88	390
1975 production	357	416	74	375
1976 production	336	406	93	439
Cumulative	3784	5505	845	3320
Cumulative release	3235	4784	Not available	2981

[a] Reporting companies only.
[b] Neely and Plonka (1977).

World production of both F-11 and F-12 in 1976 was about 1 percent below figures for 1973, and a further small decline is anticipated in 1977. It is estimated that U.S. sales of aerosol propellants in 1977 will have decreased by 50 percent compared with 1976. The corresponding European decrease is estimated at 10 percent (McCarthy, 1977). In contrast, production of F-22, used almost entirely for refrigerants, continues to rise. MCA reports 93,000 metric tons produced in 1976 versus 77,000 metric tons in 1973 by reporting companies. (World production would be slightly higher.) Continued monitoring of production, use, and release of all these CFM's should certainly be maintained.

Other Halocarbon Sources

No new data are available on carbon tetrachloride (CCl_4). Since most is consumed in CFM manufacture, it is probable that production and release rates remain similar to our previous estimates for 1973. Continued monitoring of production, release, and tropospheric concentration is desirable.

New data are available on methyl chloroform, CH_3CCl_3. Worldwide production has been increasing at 17 percent per year since 1958 and is estimated at 439,000 metric tons in 1976 (Neely and Plonka, 1977). Since its chief use is as a solvent, most of this production escapes to the atmosphere. Measured tropospheric concentrations are approximately 90 ppt in the northern hemisphere and 76 ppt worldwide. Methyl chloroform ranks just after F-11 and F-12 as a major current human-controlled source of stratospheric halogen, and therefore continued monitoring is important.

Although CH_3Cl is the most abundant halocarbon in the troposphere at present, it is predominantly of natural origin. Its global distribution, sources, and atmospheric lifetime are of obvious interest and deserve further study.

In our previous report we concluded that organic bromine-containing compounds had human-controlled sources too small to be of serious concern at present. No important new data have appeared, but since bromine is probably more effective than chlorine in the reduction of stratospheric ozone, continued monitoring of the release of organic bromine compounds is desirable.

ClO_x from Space Shuttle Operations

Our last report concluded that introduction of chlorine into the stratosphere through Space Shuttle operations at currently contemplated levels would have an insignificant effect on stratospheric ozone. This calculated ozone reduction is now likely to be somewhat larger than earlier measurements as a consequence of newly measured rate constants. If contemplated levels of operation increase appreciably, the matter will need re-examination.

3E. RATE COEFFICIENTS FOR MODEL CALCULATIONS

Most of the important processes and their rate coefficients were recently reviewed in a NASA assessment report. On the whole, there has been substantial progress in the laboratory measurement of rate coefficients and a general narrowing of their uncertainty limits. This is clearly shown in comparing the cumulative uncertainty in projected ozone reduction calculated in our 1976 report (p. 157) due to seven important reaction rate constants with a similar calculation based on the more recently tabulated reliability estimates of the NASA report (Table 1, pp. 8-13). This cumulative uncertainty range is seen to have decreased from a factor of 5.1 in 1976 to a factor of 2.2 in 1977. Even when the NASA report widened its sensitivity analysis to include 35 chemical reactions (pp. 188-189) rather than the original 7, the cumulative uncertainty range was reported to be roughly a factor of 4.4 (all at the 95 percent confidence level).

This progress has led to a rather firm data base in which approximately 80 percent of the needed rate parameters are now known to better than about ±30 to 40 percent (at plus or minus a single standard deviation, corresponding to a confidence level of about 68 percent). Further experimental improvements, which must be implemented reaction by reaction, are able to reduce such uncertainties to the ±10 to 20 percent range, but this has so far had limited effect on the cumulative uncertainties that are controlled by those processes that are least well known. It is unfortunately true, however, that, accompanying very substantial overall progress, the recent development of our understanding of stratospheric chemistry has been dominated by major upheavals caused by the recognition of the importance of processes whose role either had not been properly appreciated (such as the addition of chlorine nitrate chemistry, which delayed the publication of the previous panel report*) or whose rate coefficient had been grossly misjudged (such as the rate constant for the important reaction $HO_2 + NO \rightarrow HO + NO_2$, whose recently established larger value has made chlorine catalysis of ozone much more effective than previously thought and NO_x catalysis less so). To say how many more major upheavals we should expect in the future is rather like trying to foresee the unforeseeable. Yet it is certainly true that there is less and less likelihood of future such major upheavals.

The areas of reaction rate measurements and photochemistry in which new data have recently become available or are now being collected will now be reviewed briefly.

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Some processes whose importance may require drastic revision, at least in terms of large possible changes in their rate parameters, will be identified.

The Panel report divided processes into those involving active or inactive removal of human-controlled materials. Active removal indicates involvement in catalytic "odd-oxygen" destruction processes, i.e., includes all catalytic O_x, NO_x, HO_x, ClO_x, and other cycles, their coupling steps, and all other major reactions of these stratospheric species. Inactive removal, in the kinetics/photochemistry sense, indicates interception by reaction or photolysis before the substances reach the stratosphere.

Active removal by the primary O_x, NO_x, HO_x, and ClO_x catalytic cycles is well understood, but a few of the key rate constants have not yet been measured with sufficient accuracy. This is particularly true of the HO_x cycle for which a number of HO_2 reactions are only now beginning to be characterized. The reactions of HO_2 with O_3, O, HO, HO_2, H, NO_2, Cl, and possibly ClO are in need of further study, and for two of these reactions (those with NO_2 and ClO giving the reaction products HO_2NO_2 and ClOH) have not yet been seriously considered as parts of the stratospheric scheme. Preliminary theoretical (Jaffee, 1977) and experimental (Timmons, 1977) work has indicated that solar photolysis of ClOH is less effective than previously believed. If a fast formation process exists for it, ClOH, like $ClONO_2$, may be a temporary, pseudo-inert reservoir species, although the likelihood of a major impact on the ClO_x cycle appears to be small.

For the O_x and NO_x steps, fewer uncertainties remain. The quantum yield of ozone and the formation of singlet D near the threshold wavelength of 310 nm is still somewhat uncertain. The formation and removal steps of N_2O_5 also need to be clarified.

The importance of the role of CO by virtue of its reaction with HO (followed by H + O_2, HO_2 + NO, and NO_2 photolysis) requires that our understanding of CO sources be increased.

We must emphasize that laboratory techniques are now available to carry out virtually all of the required work by direct experimental methods with no need to depend on indirect inferences involving complex reaction mechanisms.

We must now ask the difficult question of which of these rate constants need be known with relatively great accuracy, i.e., how much the results of model calculations depend on the values of the rate coefficients. Work on this important problem is in its infancy as was indicated in the Panel and NASA reports. The problem is quite complex, since the desired sensitivity factors also depend on the structure of the model and on the specific questions asked. Such sensitivity analysis is likely to give rather nonlinear results in view of the multiple interactive effects among the reaction steps. Yet, work is very badly needed there to focus laboratory effort on those reactions where it is most needed.

Earlier guidelines, such as partitioning the total "odd-oxygen" destruction rates among the O_x, NO_x, HO_x, and ClO_x cycles by reducing them to ultrasimple terms (twice the rates of O with O_3, NO_2, ClO, etc.) are now considered to be unreliable. In fact, the direct NO_x contribution to total O_3 removal, which had been thought to be a major one (50 to 70 percent), is now seen to be much smaller; NO_x is calculated to decrease ozone only above 30 km and to increase it below. This does not mean that NO_x is any less important than had formerly been thought but that its major importance lies in the coupling reactions with ClO_x (which now turns out to be a major O_3 remover, along with HO_x) and with HO_x species.

The chemical interception processes (inactive removal) are highly dependent on the particular human-controlled perturbation. For aircraft or Space Shuttle injections, there are no known physical or chemical interceptors. For halocarbons, there is a long list of potential inactive removal processes, but most of them have been found to be inefficient, at least for fully halogenated methanes. Tropospheric reaction of F-11 and F-12 with HO was shown to be negligible (Chang and Kaufman, 1977a) and ion-molecule reactions may be similarly dismissed (Fehsenfeld *et al.*, 1976, 1977). Hydrogen-containing halocarbons react with HO in the troposphere at appreciable rates. Rate coefficients are known, but uncertainties in the tropospheric HO concentrations make the fraction of compounds--F-22 (CHF_2Cl) and methyl chloroform (CH_3CCl_3)--that are destroyed before reaching the stratosphere uncertain. Modeling of the measured north-south hemispheric distribution of CH_3CCl_3 points to a 10-year tropospheric removal time (Chang and Penner, 1977; McConnell and Schiff, 1977) and laboratory studies for F-22 suggest even slower removal (Chang and Kaufman, 1977b). The CH_3CCl_3 studies strongly suggest that the tropospheric HO concentration (in unpolluted areas) is in the range of 5×10^5 molecules per cm^3, considerably lower than measurements and theoretical models have indicated.

Unsaturated halocarbons react very rapidly with HO, and their interception is sufficiently effective to remove them from the list of potential threats, even at the lowest suggested values of HO.

The fertilizer-N_2O problem is still under active study but hinges mainly on the questions of ocean, freshwater, and soil sources and sinks of N_2O, i.e., on agricultural and biological chemistry. Biologically produced N_2O is believed to be the major source of stratospheric NO_x.

As noted in our previous report, several groups have suggested that increased use of nitrogenous fertilizers, acted on in turn by bacteria, could significantly increase N_2O production. Tropospheric concentrations of N_2O are well established.

The atmospheric chemistry of N_2O is reasonably well characterized. The process responsible for NO_x formation in the stratosphere is the reaction of N_2O with excited O atoms. This reaction has been studied with good success, as have the major steps that form and destroy excited O. The potential tropospheric interception of N_2O by reaction with HO has now been shown to be negligibly slow (Chang and Kaufman, 1977a).

The major uncertainties concerning N_2O are its tropospheric life, the strength and nature of natural sources, and the possible existence of tropospheric or biological sinks. The new higher value for the rate coefficient of the reaction $HO_2 + NO \rightarrow NO_2 + HO$ as determined by Howard and Evenson (1977) diminishes the calculated estimated ozone reduction attributable to modest N_2O increases by a substantial factor. All of these need further study before any firm judgment can be made as to the point at which a serious problem could be anticipated.

3F. TRANSPORT PARAMETERIZATION

The representation of atmospheric transport in current 1-D models of the stratosphere, by necessity, has been highly empirical. As was discussed in the NAS report and then in the NASA report, all estimates of the net vertical transport coefficients depend on adjusting the model so that the behavior of selected tracers agrees with the atmospheric measurements. In a detailed study* on the

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effect of variations in parameterized transport coefficients on model predictions, significant negative feedbacks were observed, which minimized the impact of large variations in assumed transport coefficients. This feedback, however, could not be quantified, and so was not included in the analysis. The somewhat subjective estimate of a factor of 2 uncertainty in the 1-D model transport inferred was then stretched to a factor of 3 to take into account the additional uncertainties in the time scales of CFM transport introduced by multidimensional effects in the real world. At this time there is no new information available to require a revision of this estimate.

3G. MODEL CHECKING BY ATMOSPHERIC MEASUREMENTS

The recommendation made in the NAS report that action to limit releases of CFM's should be delayed for a period up to 2 years was made, in part, because a number of atmospheric measurements were expected to be made during that period, which, it was believed, would decrease the uncertainties in the predictions of the effects of CFM's on stratospheric ozone. A number of these measurements have indeed been made. While the results of these measurements do confirm many aspects of the model, they have also raised some new, unanswered questions. Some ambivalence in interpretation of isolated measurements is not too surprising in a system as complex as the earth's atmosphere.

ClO_x, HO_x, and NO_x

Perhaps the most significant measurements in connection with the CFM problem are those of Cl and ClO (Anderson *et al.*, 1977), some of which are reviewed in the NASA report. The presence of these species in the stratosphere confirm their involvement in ozone chemistry, while the ratios of their concentrations are consistent with the catalytic chain mechanism proposed for ozone destruction. However, there remain some unexplained features of these measurements. In addition to a rather high variability, the height profiles of the concentrations of ClO have shapes that are not in complete agreement with model calculations and with measured HCl profiles. This is not too surprising, since these profiles will depend on the temperature and the concentrations of other constituents such as CH_4, O_3, and HO, none of which were measured simultaneously with Cl and ClO.

The most disturbing aspect of the ClO measurements is that a number of them show concentrations higher than can be accounted for if all known chlorine-containing compounds in the stratosphere were completely converted to ClO. It is difficult, if not impossible, to account for this by invoking unusual air motions. We are thus confronted with a fundamental difficulty that cannot be blamed on any inadequacies of the models. There appear to be only two possible explanations, errors in the measurements or the existence of some unidentified source of chlorine in the stratosphere.

The unexpected inertness of $ClONO_2$ to photodissociation and the recently measured large rate coefficient of the reaction of NO_2 with HO_2 means that there is strong coupling between reactions involving ClO_x, HO_x, and NO_x. Relatively few stratospheric measurements have been made of HO and HO_2, and the question of the variability of HO has yet to be established. While the general features of NO and NO_2 height profiles are consistent with the models, a number of observations still require explanation. These include Noxon's (1976) observation of a large decrease in NO_2 that occurs abruptly north of 50° latitude in the winter, the strange profile in NO at high latitude in the winter (Ridley and Schiff, 1977), and the maximum in NO_2 at noon observed by Kerr and McElroy (1976). Measurements of N_2O_5 have yet to be made and are important for understanding NO_x chemistry.

There is an obvious need for increased numbers of measurement of all of these reactive species and for increased accuracy. High on the list of priorities are simultaneous measurements of temperature, O_3, and the photochemically related ClO_x, HO_x, and NO_x species. Measurements using more than one technique should be compared at the same location and time. Particular attention should be given to calibration of instruments. Monitoring stations should be established to determine the degree of variability of the constituents and to establish seasonal and latitudinal trends.

Measurements of N_2O

The past year has also produced a large number of measurements of N_2O both in the stratosphere and in the troposphere over a fairly wide latitude range. The stratospheric measurements are useful in providing information about transport processes. The tropospheric measurements are of interest in connection with the fertilizer problem. One of the important questions to be answered concerns the lifetimes of N_2O in the troposphere. The longer this lifetime, the more potentially serious are the consequences of releasing a given additional amount of N_2O. Earlier estimates indicating a short lifetime were based mainly on the large fluctuation in concentrations with time and location. The recent measurements, however, show the N_2O in the troposphere to be remarkably constant over both northern and southern hemispheres. Although these new measurements indicate a longer lifetime for N_2O, this method of evaluating lifetime is not very sensitive, and the need remains for a fuller understanding of the sources and sinks for this compound. Measurements of N_2O in surface air and in ocean waters with sufficient geographic coverage to assess the role played by oceans are needed. Measurements in different types of air masses are also required. Some of these questions will, we hope, be discussed in greater detail in a forthcoming NRC report on nitrates.

Measurements of CFM's

During the past 2 years, there has been a large number of measurements of F-11 and F-12 both in the troposphere and in the stratosphere. These measurements are important for a number of reasons. They provide essential input data for the models used to describe the present atmosphere. The height profiles, i.e., the way in which the concentrations of these CFM's change with altitude in the stratosphere serve as a test for these models. The measurements are consistent with the photochemical aspects of the models, viz., that photolysis is the main loss process in the stratosphere. Since 1-D models treat transport only as an average taken over a wide range of space and time, they are incapable of explaining the observed variability of height profiles with time and location.

3H. INACTIVE TROPOSPHERIC REMOVAL?

Another important reason for measuring CFM's is to enable a comparison of the total amount released with the amount present in the atmosphere, the so-called global budget. In the NAS report it was pointed out that a major assumption in the models is that photodissociation in the stratosphere is the only removal process of CFM's in the atmosphere, i.e., that no removal processes occur in the troposphere. Since stratospheric photodissociation is a relatively slow removal process, even relatively inefficient tropospheric removal processes could be important. The most conclusive way to test for the existence of such processes would be by a global budget calculation. If the models are correct, then the amount released should equal the amount present in the atmosphere plus the amount that has been removed by photodissociation in the stratosphere. Any missing CFM's would be a measure of the amount removed by other processes that do not lead to ozone destruction.

At the time of writing of both the NAS and the NASA reports the combined uncertainties in the release rates and the global atmospheric amounts were too large to determine whether a significant tropospheric sink exists. Since then, considerable improvement has been achieved in the estimate of global release rates (McCarthy, 1977). Unfortunately, although there have also been advances in both the number and accuracy of the atmospheric measurements, the remaining uncertainties are still too large to allow definitive conclusions.

These uncertainties have several components. Since most of the CFM's are released at midlatitudes in the northern hemisphere, there will be latitude gradients that must be known to calculate the global amounts. There are inconsistencies between measurements of the latitudinal measurements by several groups (Lovelock *et al.*, 1973; Rassmussen *et al.*, 1976). The reason for these discrepancies appears to be, at least partially, due to a second problem--absolute calibration of the instruments. Although there has been considerable improvement in the precision of the measurements achieved by individual workers, there are still large differences between the analyses of the same air sample made by different groups. Reliable calibration standards are still not available. Therefore, the uncertainty in the absolute global amount is probably not less than 30 percent. The detection of a 50-year tropospheric sink would require an accuracy better than 15 percent. Finally, the amount that is estimated to be lost by photodissociation in the stratosphere is sensitive to the uncertainty in time scales for transport to the stratosphere, which, as has been reported earlier, has been assigned a value of 3.

Several alternative methods for estimating tropospheric lifetimes have been proposed that are not so sensitive to the choice of time scale for transport to the stratosphere and do not require absolute measurements. One involves following the increase in the global amounts with time. Unfortunately, this method is not a very sensitive test for the existence of tropospheric sinks. It is somewhat ironic that the most sensitive test is provided by the rate at which the CFM's in the atmosphere would decrease following a complete cessation of their release.

In summary then, the quality of the measurement data is at present insufficient to allow conclusions about the existence of significant tropospheric sinks. Any of the methods outlined above would require the provision of reliable calibration standards and a number of monitoring stations distributed about the globe in sufficient numbers to permit reliable global averaging.

Individual Processes

Both the NAS and the NASA reports considered a wide variety of possible tropospheric removal processes and concluded that none were likely to be of comparable magnitude to the stratospheric removal process that leads to ozone destruction. Subsequent to those reports only one other process has been suggested.

A study by Ausloos *et al.* (1977) has prompted the idea that photodecomposition by sunlight of CFM's (and possibly also N_2O) adsorbed in desert surface sands and possibly on other mineral surfaces might constitute a significant sink. Laboratory studies by Ausloos *et al.* (1977) and by Gab *et al.* (1977) have shown that such photodecomposition does occur for some compounds including F-11 and F-12 adsorbed on silica gel and quartz grains. The rates of adsorption and the amounts of the compounds that can be adsorbed on natural mineral and sand grain surfaces remain to be determined before any assessment of the corresponding sink strength (or lifetime) can be made.

3I. STRATOSPHERIC AEROSOLS

Nature and Sources

Stratospheric aerosols are believed to consist mainly of solid or aqueous solutions of sulfuric acids and ammonium salts. The chemical processes responsible for the formation of stratospheric aerosols are not well established; it is likely that they are initiated by oxidation of sulfur dioxide (SO_2) in the gas phase by HO or, possibly, on the surfaces of other particles.

The main source of stratospheric sulfate aerosols appears to be large explosive eruptions of volcanos. Combustion of fossil fuels releases SO_2 into the troposphere, most of which is removed by rainout or adsorption on surfaces in the troposphere. Doubling or trebling SO_2 emissions in the troposphere is unlikely to change the background density of stratospheric aerosols to any significant degree except, perhaps, during long periods relatively free from volcanic explosions. (It should be pointed out that we do not have a clear idea of the long-term distribution of volcanic eruptions.)

Crutzen (1976) has recently suggested that carbonyl sulfide (OCS) may be an important source of stratospheric sulfur. This compound is relatively inert in the troposphere but will be photodissociated and oxidized to SO_2 after it has been transported into the midstratosphere. On the basis of model calculations and recent measurements of OCS in the troposphere (Maroulis, *et al.*, 1977), it appears possible that OCS may be a major precursor of sulfate aerosols in the atmosphere. Volcanic gases have been shown to contain OCS (Stoiber, *et al.*, 1971). However, the sources and sinks of OCS have not generally been determined, so that the impact of future human activities such as coal gasification, which is known to produce some OCS (Gibson *et al.*, 1974), cannot be reasonably assessed. As suggested by Crutzen (1976), it is important to establish the sources and sinks of OCS and to provide critical appraisal of the consequences on the stratosphere of future industrial developments, especially those pertaining to fuel conversion. It is also important to note that the determination of the global distribution of atmospheric OCS is a necessary part of the study of sources and sinks.

Effects of Stratospheric Aerosols

An increase in the stratospheric sulfate aerosols may have an effect on the climate and/or on stratospheric ozone. Possible climatic effects are discussed in Chapter 4.

The earlier NAS panel report concluded that sulfuric acid and the sulfates are unlikely to provide efficient catalytic surfaces for O_3 destruction. However, Johnston in CIAP Monograph 1 (1975) and in a private communication (1977) pointed out that the catalytic activity might be higher for sulfate aerosols containing metals such as manganese and vanadium, although there is no direct experimental evidence to support this suggestion. We would recommend research activities on these and other heterogeneous chemical reactions that might occur in the stratosphere.

Some Research Needs

At present, it appears that the major effects of human activities on the stratosphere, in the near future, are likely to be caused by release of halocarbons into the ambient air. Nevertheless, because of the close coupling between ClO_x, NO_x, and HO_x, continued studies of the processes that affect their amounts are indicated. In this interim report we will not attempt to provide a comprehensive list of these processes nor even to make a selection with an indicated priority for action. This will be left to the full report. We now summarize several research needs that have been mentioned earlier in the chapter.

Simultaneous measurements, in the stratosphere, of trace constituents that are photochemically related should be made using more than one technique, if possible, for each constituent. Diurnal, seasonal, and latitudinal dependences of the concentrations of these constituents should be established. The efficiency of the ClO_x catalytic cycle in destroying ozone is determined by the ratio of the concentration of ClO to the concentrations of all compounds containing chlorine. Therefore development of a method for determining the sum of the concentrations of all chlorine-containing compounds in the stratosphere would be of considerable interest. Measurements to test the effect of multiple scattering and albedo are desirable.

Measurements should be undertaken of those tropospheric constituents that may act as precursors or interceptors of stratospherically important substances. These include, for example, HO, CO, N_2O, and the halocarbons. Tropospheric measurements of F-11 and F-12 designed to test for possible tropospheric sinks for these substances should be pursued and a system for intercalibration of instruments established.

Laboratory work should be continued to improve the accuracy with which rate coefficients are known for various reactions--particularly those involving HO and HO_2 and chlorine compounds, including $HOCl_2$ among others.

Fuller understanding of the role played by atmospheric motions is required along with an improved data base for treating these motions in the model.

Improvement in the models used to describe the atmosphere and to make predictions on the effects of future human activities is desirable, but there must be careful evaluation of the advantages and limitations of each development particularly as higher-dimensional models are introduced.

Work should also continue on improving the assessment of the uncertainties of model predictions and their sensitivities to the various inputs.

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4

EFFECT OF STRATOSPHERIC CHANGES ON CLIMATE

4A. INTRODUCTION

The possible effects of stratospheric change on climate have recently received much attention. The field is changing so rapidly that it has seemed important to review and update the 1976 reports of the CISC and its Panel on Atmospheric Chemistry. Therefore, this chapter seeks to identify (a) recent research results that significantly alter the conclusions of the earlier report concerning effects on climate and (b) new questions and viewpoints that may influence the direction of future research. The Panel has relied heavily on unpublished papers prepared for this purpose by V. Ramanathan and J. D. Mahlman. The chapter consists of three sections: possible climatic effects of changing stratospheric aerosols, changing stratospheric ozone, and changing concentrations of other trace gases.

Research on climatic effects of changes in stratospheric ozone is carried out through experimentation with mathematical models of the atmosphere that incorporate some of the physical processes occurring in the atmosphere. Models differ in degree of complexity and in the processes chosen for investigation. Results may depend on the physical insight and perhaps the luck of the investigator in designing the model or in interpreting the results of calculation. These sensitive aspects of mathematical modeling have been well known, but they have taken on increased

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Based on a draft prepared by the Committee's Panel on Effects on Climate: R. G. Fleagle, J. Mahlman, J. M. Mitchell, H. A. Panofsky, and V. Ramanathan.

importance as the questions relating to the effects of ozone on climate have become sharpened.

It is recognized that a change in total ozone affects radiation reaching the surface in two competing ways and that the net effect on surface temperature therefore represents the small difference between two larger effects. On the one hand, a decrease of ozone causes increased warming by transmission of visible and ultraviolet light to the surface. On the other hand, the stratosphere cools, causing less infrared radiation to reach the ground. Consequently, other limitations of the models, such as incomplete treatment of feedback mechanisms and one-dimensional treatment of the problems, may be responsible for errors whose magnitudes are comparable with the direct effect of radiation. Therefore, even the direction of surface-temperature changes resulting from ozone changes remains in doubt.

Discussion of effects of ozone depletion have concentrated largely on possible effects on climate at the surface of the earth, especially on surface temperature. Changes of other parts of the atmosphere may be as important as those occurring at the surface, and the magnitudes may be considerably larger. For example, cooling of the lower stratosphere and upper troposphere through decrease in ozone concentration or rise in the height of maximum ozone concentration is likely to be much larger than cooling (or warming) at the surface. Such cooling will affect stratospheric water vapor and hence may affect cloud cover and therefore surface climate in an additional way. These effects are not included in current models. Consideration may also have to be given to effects of stratospheric structure on the propagation of long tropospheric waves associated with weather systems.

4B. SUMMARY AND CONCLUSIONS

Changes in stratospheric concentrations of ozone, nitrogen dioxide, or particles (aerosols) can affect the temperature both close to the ground and at higher levels. Once the temperature changes, changes in patterns of circulation and precipitation are also likely.

Changes in the concentration of trace gases, such as chlorofluoromethanes, which contribute to the greenhouse effect while distributed throughout the atmosphere, can also change temperatures at all levels, and hence patterns of circulation and precipitation.

Tentatively, it appears that with present trends:

1. Climate changes due to the CO_2 greenhouse effect, mainly from burning of fossil fuels, will be largest (of these three);
2. Climate changes due to widely diffused trace gases, particularly CFM's, will be of intermediate size;
3. Climate changes due to specifically stratospheric changes will be smallest (with the possible exception of ozone redistribution and indirect climate effects due to ozone reduction discussed in Section 4E).

The most important new results since the Committee's previous report apply to the total radiative and thermal effects resulting from a reduction in or redistribution of ozone (Ramanathan *et al.*, 1976; Fels, 1977). These suggest that considerable compensation occurs and that we need to be concerned about the CO_2 greenhouse effects even more than about climatic changes due to ozone reduction.

Although changes in radiative effects can be calculated quite accurately, the present extreme uncertainty in the calculated changes of temperature, wind, and precipitation is, at least in part, due to our current inability to give proper representation to the complexities of the atmosphere and the earth's geography in a computer-manageable model. No rapid improvement in this situation can be foreseen.

The previous report of the Committee made recommendations about needs for research in these areas, which we still recommend without change.

4C. RELATION BETWEEN OZONE CHANGE AND CLIMATE CHANGE

Ozone absorbs solar radiation in the ultraviolet and visible parts of the solar spectrum and also absorbs and emits infrared radiation in the 9.6-μm region. Solar ultraviolet absorption by ozone is the major source of heating within the stratosphere. In the 9.6-μm region, ozone absorbs the radiation emitted by the surface and emits at the much cooler stratospheric temperatures. The 9.6-μm absorption tends to heat the stratosphere, and the emission tends to cool the stratosphere. The net effect of the 9.6-μm ozone absorption and emission is one of heating in the lower stratosphere (i.e., between 12 and 25 km) and one of cooling the stratosphere above 25 km.

Reduction of stratospheric ozone has two competing effects on the surface temperature:

(i) More solar radiation reaches the troposphere and surface (because of the reduction in the stratospheric absorption of solar radiation, both in the ultraviolet and visible ranges), which tends to warm the surface;

(ii) The reduction in the solar and 9.6-μm absorption (due to the reduction in ozone) in the stratosphere cools the stratosphere, and hence less infrared radiation is emitted to the troposphere by the stratosphere, and this effect would tend to cool the surface.

As will be discussed, these two effects are of the same general size.

4D. EFFECTS OF OZONE CHANGES ON SURFACE TEMPERATURE

With our present knowledge of the radiative properties of ozone and its horizontal and vertical distribution, it is possible to calculate, to a useful accuracy, the perturbation in the distribution of radiative fluxes due to a reduction in ozone. However, we face serious difficulties when we attempt to estimate the corresponding change in the temperature distribution. This is due to the lack of a satisfactory three-dimensional model.

The only published studies of the ozone-climate question rely on one-dimensional models of the climate system in which the entire three-dimensional earth-atmosphere system is averaged over the earth's surface and considered as a single column. The model imposes the condition that the net incoming solar radiation at the top of the atmosphere is in balance with the infrared radiation emitted by the earth-atmosphere system, so that the net energy flow vanishes. The dependent variable is the vertical temperature profile, and this is computed by prescribing globally averaged values of (1) incoming solar radiation, (2) vertical distributions of the optically active gases, and (3) clouds. A more detailed description of the 1-D model can be found in Manabe and Wetherald (1967) and

Ramanathan *et al.* (1976). Such a model also contains a limited number of climatic feedbacks.

Several 1-D studies (Manabe and Wetherald, 1967; Coakley *et al.*, 1976; Wang *et al.*, 1976; Fels, 1977) have estimated the change in average surface temperature due to a reduction in ozone. All of these studies indicate that removing ozone cools the surface. Figure 1 shows the results of Ramanathan *et al.* for uniform reduction in ozone (that is, ozone is everywhere reduced between 12 and 40 km by the same percentage). The results suggest that it is likely that infrared cooling dominates solar heating. However, the study indicates that the individual cooling and warming effects are larger than the net effect by a factor of 2 to 3, so that the net effect is quite uncertain and depends on model characteristics. Thus, for example, the figure shows that a 10 percent ozone reduction would be associated with a surface cooling of about 0.1°C. But the uncertainties are such that anything from a cooling of 0.3°C to a warming of 0.1°C is possible. Since a human-generated decrease of O_3 as large as 10 percent does not seem likely to be tolerated for reasons of human health and biological impact, the resulting

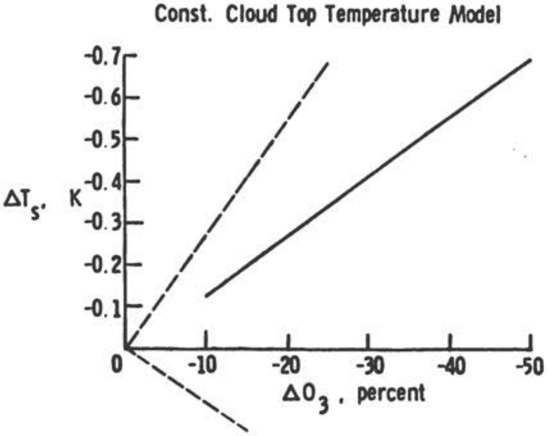

FIGURE 1 Change in the surface temperature as a function of ΔO_3. ΔO_3 is the uniform reduction in stratospheric O_3. ΔT_S is the computed surface temperature. 1-D model results of Ramanathan *et al.* (1976) shown by solid line. Dashed lines represent subjective estimated of uncertainty, based on some unpublished sensitivity studies by J. D. Mahlman *et al.*

temperature changes are not likely to be larger than those just mentioned. These numbers are relatively small compared with other temperature changes caused by human activities, especially the temperature increase expected early in the next century due to an increase of CO_2 in the atmosphere.

Ramanathan *et al.*'s study also showed that the surface temperature is sensitive to the vertical distribution of ozone. Figure 2 shows various ozone distributions, and Figure 3, the corresponding temperature distribution for the same total ozone mass. The surface cools significantly with an increase in the height of the peak ozone concentration. The 9.6-μm ozone absorption decreases with an increase in the altitude of the ozone maximum (because of the pressure-broadening effects), and hence the stratosphere cools. The cooling of the stratosphere leads to a decrease of the infrared radiation emitted downward, and hence the surface cools. Comparison of Figures 1 and 3 shows that a possible modification of the vertical distribution of ozone can have a much larger effect on surface temperature than a 10 percent ozone reduction. However, no strict comparison is possible because ozone decrease and redistributuion are not independent of each other; a redistribution without change of total ozone is unlikely.

All of these studies concentrate on surface temperature changes. Other variables, such as static atmospheric stability near the ground and precipitation are also likely to depend on ozone changes, but such effects have not been investigated.

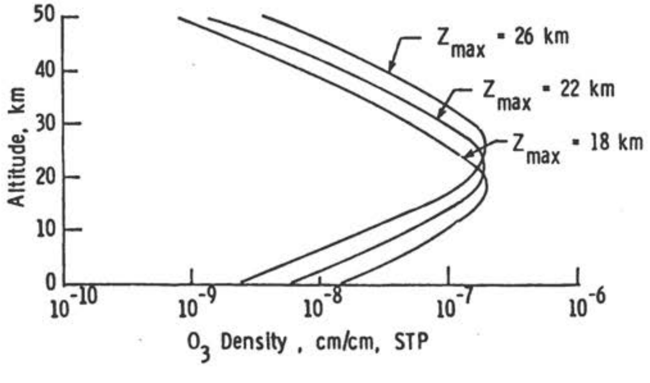

FIGURE 2 Ozone concentration profiles. Z_{max} is the altitude at which the density is maximum.

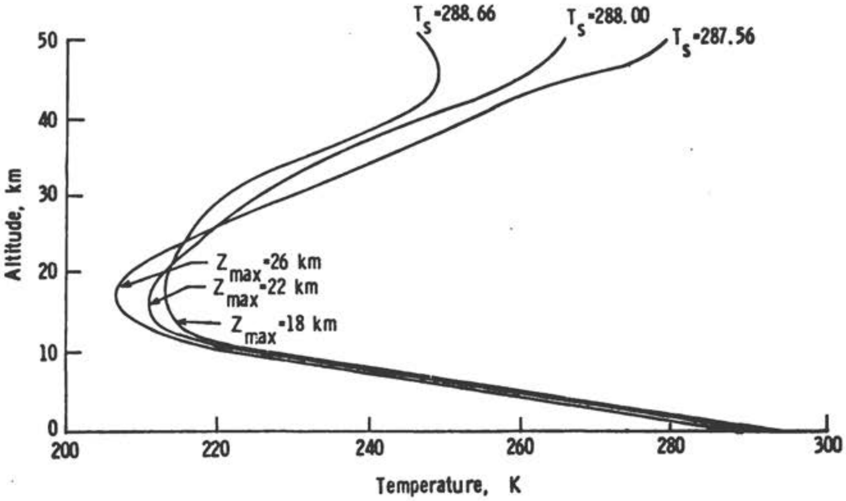

FIGURE 3 Radiative-convective model temperature profiles for the three O_3 distributions in Figure 2. Source: Ramanathan *et al.* (1976). T_s is the computed surface temperature here. z_{max} is the altitude at which the density is maximum.

The vertical distribution of ozone and the total ozone amount are strongly dependent on latitude and season. Thus, we should study the effects of ozone changes with models having two or three dimensions. As a first step in this direction, various investigators are attempting to estimate the effect of the present-day ozone distribution on the seasonal latitudinal energy budget of the troposphere and surface. Part of this work has been completed (Ramanathan and Dickinson, 1977), and Figure 4 shows the estimates of the energy input into the troposphere by the 9.6-μm band of ozone. This figure clearly reveals the seasonal and latitudinal dependence of the problem.

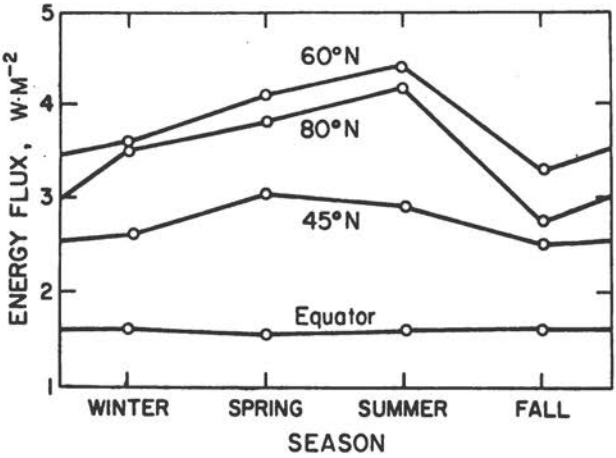

FIGURE 4 Tropospheric energy input by O_3 9.6-μm band (season values).

4E. INDIRECT CLIMATE EFFECTS DUE TO OZONE REDUCTION

So far, we have discussed the climate effects of ozone modification through its effects on radiative fluxes. There is yet another potential climatic effect of ozone modification through its influence on the stratospheric circulation. The stratosphere cools because of a O_3 reduction. The vertical distribution of the cooling is shown in Figure 5 for a uniform 50 percent reduction. As suggested by Dickinson (1974) and Bates (1977), changes of stratospheric temperatures could have significant impact on the troposphere through modification of dynamic coupling mechanisms. Bates, in fact, concludes from a linear model without energy dissipation that the equator-to-pole heat flux in the troposphere depends strongly on certain characteristics of the stratosphere such as the wind profile and the static stability. It is important to test these results by studying more realistic models.

Another possible climatic effect of ozone has been suggested by Ramanathan *et al.* (1976) and by Liu *et al.* (1977). From Figure 5, we see that the temperature near the tropopause drops considerably because of a reduction in ozone. This suggests the possibility of an effect on the H_2O concentration in the stratosphere. Mastenbrook

(1971) found that the H_2O mixing ratio in the stratosphere varies with variations in the temperature of the tropical tropopause. If this is generally true, the H_2O mixing ratio in the stratosphere will decrease with a reduction in the tropopause temperature. The reduction in H_2O mixing ratio will cause a decrease in the infrared emission by stratospheric H_2O, which will tend to cool the surface and troposphere.

Decreasing water vapor could also decrease cloudiness in the stratosphere. On the other hand, cooling could also lead to increased ice-crystal formations near the tropopause, with a consequent increase in solar energy reflected into space. Some of these factors may be nonzonal and might therefore systematically modify the distribution of eddy potential energy associated with planetary and synoptic-scale waves.

In conclusion, the work done to date on the ozone-climate problem with the aid of simple climate models has enabled us to identify various aspects of the problem. Detailed recommendations for quantitatively examining these aspects have been made in the recent EPA report (1977) and hence need not be repeated here.

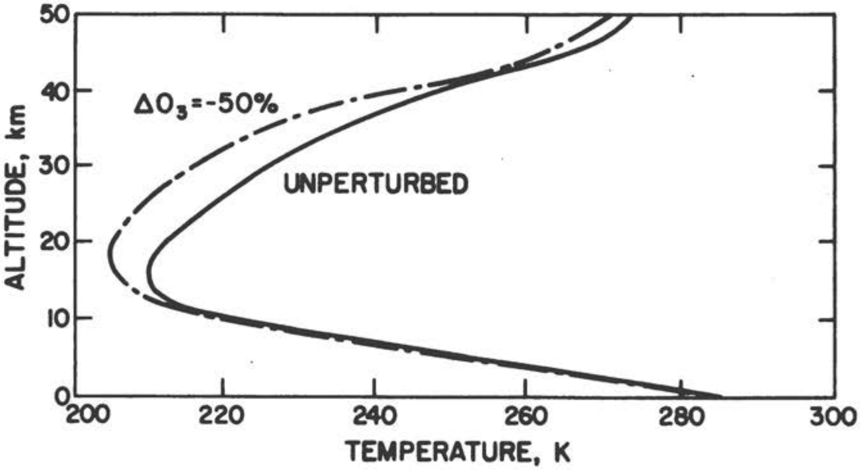

FIGURE 5 Effect of 50 percent reduction of stratospheric O_3 on temperature. Results obtained from a 1-D radiative-convective model. Source: Ramanathan *et al.* (1976).

4F. RADIATIVE EFFECTS OF TRACE GASES ON CLIMATE

Trace gases can affect climate in two major ways: (1) by absorbing solar radiation and (2) by absorbing and emitting infrared radiation, particularly in the regions of the spectrum that are normally quite transparent, that is, in the atmospheric window between 7 and 14 μm. The two gases that are important in this connection are ozone (which has been discussed in Section 4C) and nitrogen dioxide (NO_2). NO_2 could be added to the stratosphere, for example, if large fleets of SST's were built or as a result of increased use of fertilizers.

It was shown in an earlier NAS report (Climatic Impact Committee, 1975) that the absorption of visible light by NO_2 cannot be neglected.

Wang *et al.* (1976) have studied a number of largely man-made trace gases that have absorption lines in the window. Most of these gases are well distributed through the atmosphere, so that only perhaps 14 percent exist in the stratosphere. Of particular interest here are the fluorocarbons, the influence of which was already discussed in the NAS report *Halocarbons: Environmental Effects of Chlorofluoromethane Release*; the conclusion was that surface warming in the early twenty-first century (unless CFM production is curtailed) due to infrared absorption by the CFM's will be a significant fraction of the warming due to the increased CO_2 greenhouse effect. Additional computations of the CFM effect have confirmed these conclusions (Ramanathan, 1977). Other trace gases causing pronounced greenhouse effects are, according to Wang *et al.*, N_2O and CH_4.

A second point, previously not noted, involves a possible feedback of change in stratospheric ozone on the tropospheric greenhouse effect. Changing ozone can change OH in the troposphere, which, in turn, reacts with some of the gases listed by Wang *et al.* (1976), e.g., compounds containing hydrogen. Thus, changes in stratospheric ozone can modify the greenhouse effect.

4G. CLIMATIC IMPACT OF STRATOSPHERIC PARTICLES

A layer of variable particle density exists in the stratosphere centered about 20-km height, consisting mostly of sulfates. Most of these particles are produced by volcanoes, but human activities may well add to the total particle load (see Section 3I). If this is indeed the case, man also affects climate in this manner. In order to estimate the effect of changes in particle loading, the optical properties of these particles must be known. Several groups have given typical values of optical parameters and size distributions.

Cadle and Grams (1975) have presented a thorough review of the optical properties of stratospheric aerosols, while Toon and Pollack (1976) have proposed a globally averaged model for both stratospheric and tropospheric aerosols.

Utilizing various aerosol models characteristic of volcanic injections into the lower stratosphere, several investigators have made calculations of the theoretical impact on the global albedo and surface equilibrium temperature. For sulfate and sulfuric acid aerosols, the general indications are that the increase in global albedo due to increased reflection of sunlight is partially (~30 percent) offset by the increased greenhouse effect due to retained terrestrial radiation. It is generally concluded that absorption of infrared radiation can cause significant warming of the stratosphere in the vicinity of the aerosol layer (Harshvardhan and Cess, 1976).

In an empirical study, Oliver (1976) compared the chronology of estimated stratospheric particle injections by volcanoes since 1881 with the chronology of global-scale mean annual surface temperature anomalies in the same period. He related the two chronologies through a simple climate model that included thermal storage effects in the global climate system and calibrated the model to the 1883 eruption of Krakatoa. His results, indicating a fairly systematic transient surface cooling effect of volcanic ejecta, agreed quite well with theoretical results. The net effect of the largest volcanic eruptions is a surface cooling of less than 1 K (Coakley and Grams, 1976; Pollack *et al.*, 1976; Herman *et al.*, 1976; Luther, 1976).

There is good reason to believe that episodic injections of volcanic effluents into the stratosphere are a principal factor in the modulation of stratospheric particle loading and of stratospheric chemistry as well. However, more needs to be done to establish (1) specifically the composition of gases and particles that reach the stratosphere from volcanic eruptions, and in what amounts, and (2) what are the chemical and physical changes in the ejected materials while they are in residence in the stratosphere.

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5

BIOLOGICAL EFFECTS

5A. INTRODUCTION

The biological world is a balanced, dynamic system of both competing and cooperating species, each suited to live in a certain environmental range. This environment is provided by the physical and chemical features of the surroundings (greatly modified by activities of the other organisms present) plus the neighboring organisms themselves. Although the dynamic balance of this system undergoes fluctuations in the exact numbers and relationships of species, it constitutes over the long run the "natural" world with which we are familiar. Selection during biological evolution has presumably produced species that are well adapted to their particular niche within it.

Man grossly manipulates his environment in efforts to accomplish deliberate ends. These purposeful perturbations (as, for example, in permitting only selected plants and large animals to live in farmed areas, removing large masses of certain organisms in lumbering and fishing, changing natural water flows) rely for their expected benefits on the rest of the environment remaining "satisfactory." Perturbations that excessively disturb the biological world would change the framework within which man must live and thus force major readjustments of his activities.

Concern over the potential consequences to the biological world of stratospheric change revolves around (1) effects of increased solar DUV radiation and (2) the implications of changes in other climatic variables. It is clear that some potential consequences of such changes, such as effects on agricultural productivity, could be detrimental to human welfare. Others, however, are less easily characterized, because of the difficulty in defining optimum conditions for the biological world. The presumed effectiveness of past evolutionary adaptation, and the human need for a stable framework within which to operate, make it reasonable to believe that widespread changes are more likely harmful than helpful, particularly when these occur rapidly compared with usual evolutionary time scales. Thus, where a prospective man-made change cannot be proven unequivocally beneficial, it should be viewed with some suspicion.

Assessment of the probable impacts of solar uv and climatic changes on the biological world is a very substantial task. Because little has been learned about the implications of small climatic changes since the Committee's report of last year, this report concentrates on the small advances made in understanding the biological responses to increased solar uv flux.

5B. SUMMARY AND CONCLUSIONS

The previous report made specific recommendations for a broadly based biological research program, supported by adequate financial resources, and continued over a period of more than five years. In the year since its release, a short-term interagency program was initiated under the coordination of the Environmental Protection Agency (EPA). The overall uncertainties about potential biological effects of enhanced solar uv have not been materially lessened, but some results--emanating in part from the EPA program--provide additional insight into the dimensions of the problem.

Unfortunately, the financial resources initially allocated to this program were insufficient to address properly the comprehensive biological questions confronting us. Recent suggestions that the financial support of this program is to be significantly reduced, rather than increased, are particularly disconcerting. Without adequate and sustained financial support, it will not be possible to assess threats from stratospheric modification to public health and welfare (which necessarily includes the stability of the biological world).

Mechanisms of uv radiation action, and the differing sensitivity of different organisms, have been further outlined. These studies represent progress toward identifying species that might be most adversely affected by solar DUV intensification and the environmental settings in which the problem might be most acute. There is evidence that some organisms may normally live near their tolerance limits for solar uv, while others do not. It now appears, however, that the experiments that can be most readily performed in the controlled conditions of the laboratory--although useful--may not be able to provide direct answers about ultimate effects in the outdoor environment.

Reduction of the uncertainties surrounding these questions will ultimately require several years of study as emphasized in previous reports of the National Academy of Sciences. The short-term EPA research program, if adequately supported, is welcomed as a beginning, but commitment to a sustained program with substantial funding, rather than a series of short-term efforts of indeterminate length and continuity, is essential. Furthermore, subjecting such efforts, in advance, to intensive scientific peer scrutiny is necessary, if we are to ensure that financial resources are wisely committed.

5C. LAND VEGETATION

Higher plants have obvious importance both in agricultural systems and as the basis of primary production in natural land ecosystems, which makes their response to atmospheric ozone reduction (and the attendant increase in solar uv radiation) particularly significant. The fact that plants have evolved to maximize exposure of their living cells to sunlight, and thus facilitate solar energy capture in photosynthesis, increases our concern over the possible effects of intensified solar uv on them. Ultraviolet radiation sensitivity screening is under way for a comprehensive variety of agricultural plants in laboratories of U.S. Department of Agriculture at Beltsville, Maryland, and the University of Florida.

Comparison of preliminary results of different research projects under the EPA research program further corroborates earlier findings (Biggs *et al.* 1975) that uv sensitivity of plants appears to be greater in the artificial illumination of plant environmental growth chambers than in the open field. Whether this is due to the lower light levels in growth chambers or to some other aspects of the chamber environment clearly needs to be learned--although this is a complicated problem, not easily undertaken in a short-term research effort. Unfortunately, this finding means that those experiments that provide the most completely controlled conditions are not by themselves adequate to evaluate the consequences of increased solar DUV on plants in nature.

The first results of the screening programs support the earlier findings summarized in the report of last year, which emphasize differential sensitivity of higher plant species. In some cases, variations in uv sensitivity occur among different cultivars of even the same agricultural species. These variations may explain why certain varieties that thrive in the greenhouse fail to do well in the field. Such screening programs are useful to focus research and assessment efforts on varieties that might be most affected if stratospheric ozone were depleted, as well as to provide information for possible plant breeding to improve uv resistance. Yet basic understanding of the physiological and structural properties of plant species that affect uv radiation sensitivity is also needed. This is particularly important in the ultimate assessment of uv radiation impact on nonagricultural ecosystems, where systematic screening of the multitude of plant species is hardly conceivable. Such an endeavor would necessarily depend on understanding the characteristics and mechanisms of uv injury and tolerance in higher plants. Some progress in this area has been made over the past year.

5D. CHARACTERISTICS OF ULTRAVIOLET INJURY

The growth and productivity of higher plants appear to be altered by DUV radiation in at least three different ways. Ultraviolet radiation (1) reduces photosynthetic rates, (2) increases respiratory carbon consumption in the dark, and (3) impairs leaf expansion.

The amount of photosynthesis occurring under standard light conditions in a sensitive plant species decreases with increasing total DUV radiation doses given to the plants (Sisson and Caldwell, 1977). Even uv doses less than those currently being received in the natural environment caused some reduction compared with plants given no uv. Unlike some other uv radiation effects--such as the reduced survival of an aquatic alga (Nachtwey, 1976)--the effect of a given total dose was the same over a rather broad range of dose rates. Thus, it would appear that even a moderate enhancement of uv radiation in nature might have significant effects on productivity of plant leaves.

In a survey of a very limited number of crop species, Van and Garrard (1976) reported that only plants possessing the most common biochemical pathway of photosynthesis (the "C_3 pathway") were sensitive to uv radiation, while those possessing a physiologically more elaborate photosynthetic system known as "C_4" photosynthesis were not. The C_4 photosynthetic pathway is particularly prominent in plants native to environments of the earth's surface having naturally more intense solar uv radiation. There appears to be no immediate theoretical reason to suggest why the C_4 photosynthetic pathway should be less sensitive to uv radiation, and the very limited number of species examined would caution against generalization at this point. Nevertheless, if such a generalization ultimately proves to be true, the alternative photosynthetic pathways may represent an important plant adaptation to uv radiation.

5E. MECHANISMS OF ULTRAVIOLET RADIATION ACTION

Recent studies indicate that impairment of photosynthesis may involve either direct effects on chemicals involved in the photochemical reactions or disruption of chloroplast membranes or both (Okada *et al.*, 1976; Brandle *et al.*, 1977). Such damage does not, therefore, seem to concern cellular nucleic acids, and one of the major protective processes of biological organisms--the radiation repair mechanisms discussed in last year's Committee report--may not be operative in this case.

Diminished leaf expansion after uv exposure seems due to inhibition of cell division rather than to reduced cell expansion (Dickson, 1977), and there is apparently no direct involvement of the plant phytochrome system (Lindoo and Caldwell, 1977). The reduction in growth is more than that expected from simply the reduced energy supply accompanying inhibited photosynthesis (Sisson and Caldwell, 1976). The resulting reduced display of foliage combines with the reduced photosynthetic rates to constrict further the plant's ability to fix carbon. Those higher plant species in which the regions where new cells arise are naturally protected against sunlight should be less sensitive to radiation. This may explain in part the lower uv sensitivity observed in the past for some grasses, where new cells are added at the bottom of the blade.

Although there is not likely to be a single determinant of plant susceptibility to increased solar uv radiation, these mechanistic studies should help to identify which groups of higher plant species would be most likely to be sensitive. Such information would also suggest the environmental conditions under which enhanced solar uv radiation might have a greater or lesser effect on plant processes. This, of course, is germane not only to the immediate concerns of agricultural productivity but to estimating the ultimate balance and stability of natural ecosystems as well. In these systems, the effects of intensified solar uv radiation may be manifested more in alterations of the competitive balance of species rather than reduced ecosystem productivity *per se*. Experimental evidence does suggest that supplemented DUV radiation can shift the competitive balance of higher plant species under field conditions (Fox, 1977).

Recent preliminary information suggests that some higher plants may be able to adapt to some extent to intensified solar uv radiation by altering the transmittance of this radiation through the outer epidermal cell layer of leaves (Robberecht, 1977), just as humans develop pigments (tan) in the sun. Certainly, the adaptive capacity of higher plants must also be included in an overall assessment of the potential impacts of increased DUV on the biological world.

5F. AQUATIC ORGANISMS

Many aquatic organisms of both freshwater and marine ecosystems have been shown to be quite sensitive to uv radiation (Calkins and Nachtwey, 1975). Studies of the past year further corroborate these earlier findings, suggesting that, in addition to phytoplankton and invertebrates, certain life stages of vertebrate organisms such as the embryonic forms of mackerel and anchovy are sensitive at intensities currently experienced very near the surface of ocean waters (J. Hunter, National Marine Fisheries, La Jolla, Calif., with J. Taylor and G. Moser, unpublished data). Changes in productivity, manifest as depressed chlorophyll *a* concentrations and reduced biomass, decreased community species diversity, and altered community structure have been recently demonstrated as a result of simulated intensification of solar uv radiation in experimental estuarine microecosystems (Worrest *et al.*, 1977). Some of these organisms may normally live near the limit of their uv tolerance, balancing this factor against the need to live in regions of high photosynthetic activity, where phytoplankton, which serve as food supplies for other marine organisms, are plentiful. The ultimate importance of this high uv sensitivity compared with terrestrial plants and animals depends, of course, on the depth at which such organisms normally exist during the daytime and on the depth of solar uv penetration. Studies currently under way may help to clear up this matter.

5G. OTHER SPECIES

Studies of uv effects on insects and agricultural animals have been initiated but require much experimentation. The latter may be important as attempts are made to promote modern farming in tropical latitudes with breeds developed in temperate zones. A study of squamous-cell carcinoma of the eye in Hereford cattle, suspected to be linked with uv radiation exposure, was initiated with the goal of establishing its determinants. At present, we are not aware of any studies involving uv sensitivity of nonagricultural animal species.

Studies of the effects of increased solar uv radiation on the interactions of higher plants with agents causing plant disease were begun during the past year.

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6
HUMAN HEALTH

6A. INTRODUCTION

The possible routes by which stratospheric changes can effect human health appear restricted to (1) changes in DUV as a consequence of changes in stratospheric ozone; (2) changes in climate from changes in infrared absorption (involving CO_2, ozone, and/or human-released trace gases); (3) conceivably, changes in minor constituents of ground level air. We know of no evidence that indicates that (3) is of any significant importance. Our interim judgment, to be reexamined in a later report, is that direct climatic effects on human health are so relatively small, compared with indirect influences through climatic effects on food production, as not to deserve separate discussion. This leaves decreased ozone, with consequent increased DUV acting on white human skin, with the greatest potential for human health effects.

6B. SUMMARY AND CONCLUSIONS

The most serious health consequence of possible stratospheric change still seems to be possible or probable increases in death from malignant melanoma (a type of skin cancer). Although likely increases in melanoma from

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Based on a draft prepared by the Committee's Panel on Effects on Human Health: John W. Tukey, chairman; T. B. Fitzpatrick, M. Henderson, and J. Lee.

small ozone changes, while perhaps as much as 4 times as large a percentage as the ozone changes, would still have to be small compared with the consequences of exposure to sunlight with unchanged ozone, solar exposure is a serious public health problem whether or not ozone changes occur. Some useful federal actions have been taken that may help with the broad problem.

Continuing collection of detail about anatomical locations of primary melanomas has only strengthened the conclusion drawn in *Halocarbons: Environmental Effects of Chlorofluoromethane Release* about the role of sunlight, specifically DUV, in malignant melanoma. Recent work (see Section 6F) suggests somewhat lower predictions for melanoma deaths consequent to specified increases in DUV than our last report suggested.

The evidence implicating DUV in the other, much more common but very rarely fatal kinds of skin cancer remains as before.

In addition to the needs for research pointed out in the Committee's previous report, we call attention to the importance of the following:

1. Developing ultraviolet sources and observing instruments that can express the sunburn erythema and tanning responses of individual's skins to measured amounts of DUV. This could lead to a more precise and sensitive classification of individual suspectability to the development of skin cancer.
2. Developing an experimental-animal model of UV-B induction of melanoma and using this model to study the importance of intermittent exposure.

6C. THE PREVIOUS REPORT

The impact of increased DUV on human health was summarized in *Halocarbons: Environmental Effects of Chlorofluoromethane Release*, and the general conclusions reached were as follows:

1. Increased DUV *could* involve an "increased incidence of malignant melanoma...and an increase in mortality from this cause."
2. Increased DUV *would* increase the incidence of basal- and squamous-cell carcinomas, a less serious, rarely fatal,

but much more prevalent form of DUV-induced skin cancer. This increased incidence would result in serious disfigurement and an increased economic burden.

3. Antirickets effects of conversion, in the skin, of 7-dehydrocholesterol to vitamin D, while an important benefit of DUV, would not be appreciately affected by small DUV changes.

This report recognized the susceptibility of certain persons for the development of DUV-induced skin cancer based on their sun-reactive skin type and in this regard made the following recommendations:

1. Education of the public regarding the damaging effects of DUV, and
2. Use of topical chemical preparations (sun screens) that are absorbed in the outer layers of the skin and then absorb the DUV that impinges on it.

6D. RECENT ACTIONS

Several recommendations for research have been implemented in 1977. The National Institutes of Health has funded educational programs for both the public and the medical profession regarding malignent melanoma. The Environmental Protection Agency has funded a survey of the relative incidence of the various sun-reactive skin types in the United States in order to identify the most susceptible population that could be affected by increased DUV.

The Food and Drug Administration has announced a program of efficacy labeling of sunscreens that should both enhance the effectiveness of what is sold and make individual choice of truly efficacious products possible.

6E. ONGOING STUDIES

Further inquiry into the available survey data on skin cancer is being pursued,* and a new survey has been

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*By the NRC Panel on Statistics on Skin Cancer and by its chairman, Elizabeth Scott of the University of California, Berkeley.

undertaken* by the National Cancer Institute to acquire more complete data on incidence and with some regard to skin color and sunburn sensitivity.

6F. EFFECT OF CHANGING DEATH RATES

Unpublished cohort studies (Lee *et al.*, 1977) indicate that death rates have been behaving for a quarter of a century in a pattern that is consistent with a continuing fall in the percentage of those persons developing melanoma who die from the disease. The anticipated improvement in survival for melanoma patients born in more recent years will reduce previous estimates of increased numbers of deaths that would otherwise be associated with the predicted increases in incidence. As a result, forecast total melanoma deaths as well as forecast extra deaths due to a specified increase in DUV may well be somewhat lower than the rates discussed in *Halocarbons: Environmental Effects of Chlorofluoromethane Release*. (We hope to convert this qualitative comment into quantitative estimates in a later report.)

6G. LOCALIZATION OF MALIGNANT MELANOMA IN RELATION TO SUNLIGHT

In 1970, the late James McDonald (1970), an atmospheric physicist, first suggested the sequence of stratospheric perturbation → decreased stratospheric ozone → increased DUV → increased skin cancer. At that time, the problem of skin cancer was considered a minor hazard and dismissed as "easily cured."

Our earlier report (*Halocarbons: Environmental Effects of Chlorofluoromethane Release*) pictured for the first time how malignant melanoma occurs not only in areas of the skin directly exposed to sunlight but even more frequently in areas that are only partially protected (trunk, limbs of females) and spares only those absolutely covered areas (anogenital areas and buttocks). In a more recent compilation of the data collected in the Melanoma Clinical

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*By J. Scotto of the National Cancer Institute.

Cooperative Group (Sober *et al.*, 1977) the precise anatomical locations for 761 patients were depicted on human models. A nonuniform distribution was observed, with the highest area of density occurring on the head, neck, and back in males and females and the lower legs in females. Spared areas were noted on the buttocks and on the breast and anterios torso of females. This distribution suggests that areas that are rarely, if ever, exposed to sunlight infrequently develop melanoma.

Arguments against the role of sunlight in explaining the striking increased incidence of malignant melanomas in the past two decades have been put forth by Urbach (1977), who has the opinion that solar exposure has only been implicated in one type of melanoma (lentigo maligna melanoma), which occurs principally on the face and which has not been increasing in incidence. Urbach postulated that if solar exposure has a role in the etiology of the other two types of melanoma, sunlight must act in conjunction with some other unknown factor.

Recent data by Fears (1977) would suggest that the occurrence of melanoma is more closely related to intermittent solar exposure than to total cumulative exposure. His data showed that the incidence of melanoma does not have a strong relation to age; while nonmelanoma skin cancer increases rapidly with age. These findings agree with the large-scale analysis of mortality from malignant melanoma made in England and Wales for the years 1949 to 1953 and 1959 to 1963 (cited by Lee *et al.*, 1977). In this analysis, a higher mortality was concentrated in younger professional and managerial workers, which is consistent with higher efficacy of intermittent exposure patterns associated with recreation, as contrasted with exposure from outdoor occupations.

In 1976, Magnus (1977) of Oslo reported on the analysis of 2541 melanoma patients observed in Norway during the period 1955-1974. Analysis supports an exogenic cause of melanoma in this series, exposure to sunlight being the important factor. Both the tripling of the incidence during this period and a steep increase during adolescence are attributed to increased recreational sunlight exposure as a result of (1) increasing leisure coincident with economic progress, industrialization, and urbanization and (2) change in clothing habits with a trend away from total concealment. Northern Norway (72°) has only one half the incidence rate of southern Norway (58°). This is attributed to greater exposure associated with the warmer climate and a higher DUV intensity in southern Norway.

As in other reported series, there were striking differences in the anatomical site of the primary cancer in males and females--males showing higher incidence in the neck-trunk sites and females showing a three times higher incidence on the lower limbs. It is to be noted that there was no change in incidence on face or feet during the entire observation period (1955-1974), supporting the hypothesis that change in behavioral pattern is an important factor in the striking increase in incidence of malignant melanoma since World War II.

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MATERIAL DAMAGE

GENERAL

A rough assessment of the likely damage to paints, plastics, and other surfaces due to increases of DUV was given in the Committee's last report. New studies have been initiated to assess the probable importance of DUV increase on stability of agricultural pesticides.

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