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UNITED STATES DEPARTMENT OF COMMERCE The Under Secretary

National Oceanic and Atmospheric Administration Washington, D.C. 20230

AUG | 7 1987

MEMORANDUM FOR:

Domestic Policy Council Working Group

on Energy, Environment and Natural Resources

FROM:

Anthony J. Calio

SUBJECT:

Stratospheric Ozone: The State of the Science and NOAA's Current and Future Research

(July 1987)

Attached is a report, prepared by Dr. Daniel Albritton, Director of the NOAA Aeronomy Laboratory, and other NOAA scientists that summarizes the current scientific understanding of the potential for depletion of stratospheric ozone by man-made halogenated compounds. The report addresses both global and Antarctic ozone. In addition to describing the status of the science, the report also briefly summarizes NOAA's current and future research in these areas.

The document is constructed along the lines of the science questions that have come up frequently at the numerous domestic and international ozone-protocol meetings over the last several months. Specifically, the ozone/chlorine science is grouped by the following questions:

- What future ozone responses does theory predict for a few illustrative emission scenarios?
- o How good are these predictions?
- Do observations reveal any current chlorine-induced ozone depletions?

As an aid to judging the significance of a possible ozone loss, each of the predicted responses is compared to the observed natural variability, insofar as it has been established. For



example, at pages 19-20, the report discusses the conclusion that the "unexplained" component of the currently observed decrease in global ozone is within the range of natural variation and hence alone cannot be considered as conclusive evidence of an anthropogenic influence.

I wanted to send you a copy of this summary for your use in preparation for the Diplomatic Conference on the Protocol on Chlorofluorocarbons to the Vienna Convention for the Protection of the Ozone Layer, scheduled for this September in Montreal.

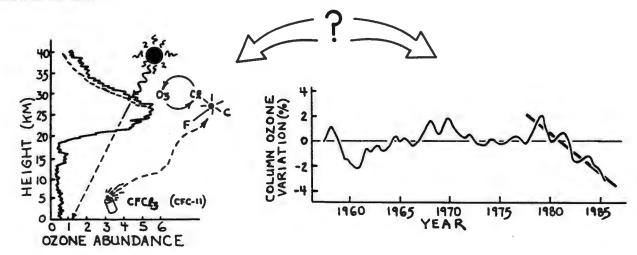
Comments are, of course welcomed and should be directed to Dr. Albritton at (303) 320-3218.

Attachment

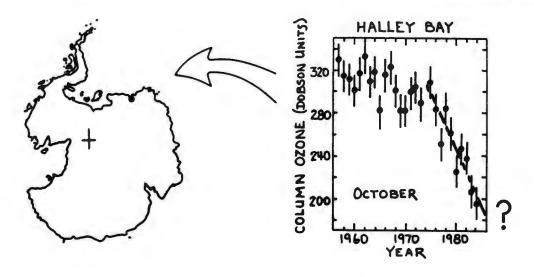
STRATOSPHERIC OZONE:

THE STATE OF THE SCIENCE AND NOAA'S CURRENT AND FUTURE RESEARCH July 1987

Global



Antarctica





COVER

GLOBAL OZONE

Emissions of man-made chlorine-containing chemicals are predicted to be depleting the earth's stratospheric ozone layer, which shields life on the surface from harmful solar ultraviolet radiation. This theory predicts that losses in the global-average overhead column of ozone are less than a percent at present, but would grow to several percent in the next century if emissions were to continue their current growth rates.

The 25-year monitoring record of global ozone shows that column ozone increased a few percent in the 1960's, remained relatively invariant through the 1970's, and decreased afterwards. Attention has focused on the recent decrease because it has occurred during the period of increasing stratospheric chlorine abundance. However, the powerful El Chichon volcanic eruption and the unusually strong 1982-83 El Nino also occurred during this period. Both could have influenced stratospheric ozone.

Is the recent global ozone decrease due to man-made chlorine chemistry, natural influences, or both?

ANTARCTIC OZONE

Observations have discovered growing ozone decreases over Antarctica in the austral spring season that have now reached 40 to 50%. Such decreases are the largest ozone changes ever observed. This trend was totally unanticipated.

Several theories have been proposed to explain this unusual phenomenon: man-made chlorine chemistry, sunspot-induced chemistry, and climatologically changed ozone circulation patterns. Recent measurements have shown that both the chemical and meteorological parameters in Antarctica have changed.

Is this "ozone hole" the first indication of human-induced ozone destruction, a newly discovered natural phenomenon, or both?

STRATOSPHERIC OZONE:

THE STATE OF THE SCIENCE AND NOAA'S CURRENT AND FUTURE RESEARCH

31 July 1987

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SUMMARY

Atmospheric ozone is central to the chemistry, climate, and habitability of the planet. Hence, NOAA has committed a substantial effort to observing, understanding, and predicting ozone's atmospheric roles and effects as they relate to current environmental issues. The status of the science of stratospheric ozone and NOAA's research can be summarized briefly as follows.

GLOBAL STRATOSPHERIC OZONE DEPLETION

Background:

- According to a hypothesis proposed in 1974, man-made halogen-containing chemicals [including chlorofluorocarbons (CFC's), which contain chlorine, and Halons, which contain bromine] may be depleting stratospheric ozone globally.
- The ozone layer, which lies between 10 and 50 km, shields the earth's surface from harmful solar ultraviolet radiation and establishes the temperature structure (hence circulation patterns) of the stratosphere.
- Man-made emission rates of chlorine have greatly exceeded those of natural emissions.
- While the restrictions in the 1970's on the use of CFC's in aerosol sprays halted the rapidly increasing emission rates of the preceding decade, global emissions have begun increasing again in recent years.
- National policy discussions are focusing on the need for further emission regulations on the halogenated compounds, and international negotiations are also underway in that regard.

Theory:

- From a scientific perspective, any proposed regulatory policy should consider all of the fully halogenated compounds, which are characterized by high ozone depleting potentials and long atmospheric lifetimes, as a group for the purposes of regulation.
- Several chemical compounds that have low ozone depleting potentials may have significant value as possible future substitutes.
- The current understanding of stratospheric chemistry and circulation predicts that the halogenated compounds will alter the
 - total column amount of ozone overhead,
 - variation of this column ozone with latitude, and
 - vertical distribution of ozone.

- The fully halogenated compounds have long atmospheric lifetimes; therefore, their stratospheric abundance would continue to grow for many decades, even if their emissions were to plateau or decrease moderately.
- Hence, from the standpoint of limiting the maximum concentrations of chlorine and bromine in the stratosphere, emission-rate restrictions done earlier need not be as severe as those done much later.
- The response of stratospheric ozone to emissions of halogenated compounds is sensitive to the atmospheric growth rates of other trace gases, such as carbon dioxide, methane, and nitrous oxide, which are emitted from man-made and natural sources.

Predictions:

Policy has not set a "tolerable" stratospheric ozone loss. Therefore, as an aid to judging the <u>significance</u> of a possible ozone loss, each of the following predictions are compared to observed natural variability.

Global-average column ozone

- Model predictions for all scenarios that include continued growth of the emissions of the halogenated compounds at current rates or greater show global-average column ozone losses in the next century that are larger than or equal to observed natural ozone decadal variability, which is about 3 to 4%.
- Hence, a true global freeze of the sum of the chlorine and bromine reaching the stratosphere to current rates is consistent with limiting predicted long-term perturbations in the global-average column ozone to magnitudes less than the decadal natural variation.
- This prediction (as well as the ones below) assumes continual current percentage growth in carbon dioxide and methane. If less growth actually occurs, then more column ozone depletion is predicted; if more growth occurs, less column ozone depletion is predicted.

High-latitude column ozone

- The current understanding of the interaction of stratospheric circulation patterns and ozone-loss chemistry predicts that more column ozone loss will occur at higher latitudes than at the equator.
- Hence, a <u>significant decrease</u> in total chlorine and bromine emission rates is consistent with limiting predicted long-term perturbations in the column ozone at high latitudes to magnitudes less than decadal natural variation.

Upper-stratospheric ozone

- Even with a global freeze, current theory predicts substantial longterm losses of ozone at 40 km, which may alter stratospheric circulation patterns because of the accompanying cooling.
- Hence, a <u>significant decrease</u> in total chlorine and bromine emission rates is consistent with limiting predicted long-term losses in the ozone at 40 km and associated decreases in local temperatures to magnitudes less than the current (limited) observations of decadal natural variation, which is about 1 to 2°C.
- However, the couplings and feedbacks of chemistry, radiation, and circulation in the upper stratosphere have yet to be represented fully in three-dimensional models. Furthermore, the possible effects of the predicted upper-stratospheric cooling on life at the earth's surface are unknown at present.

Greenhouse effect

- In addition to their relation to stratospheric ozone, many of the halogenated compounds absorb infrared radiation and hence contribute to the "greenhouse" effect, which elevates surface temperatures.
- A <u>decrease</u> in the total emission rates of halogenated compounds appears to be consistent with limiting their predicted commitment to a long-term "greenhouse" warming such that it is less than the decadal natural variation, which is about 0.5°C.

Uncertainties:

- All of the above ozone responses are predictions made by atmospheric models that are based on the current understanding of the relevant atmospheric processes.
- While these models indeed simulate much of the present atmosphere very well, they are not perfect, which places a factor of two to three uncertainty on their predictive abilities.
- Since stratospheric ozone is sensitive to changes in other trace gases, an additional uncertainty is the difficulty in predicting their future abundances, particularly those of carbon dioxide and methane.

Nevertheless, most atmospheric scientists believe that if the present growth rates of chlorinated and brominated compounds continue unabated indefinitely, then it is highly likely that substantial global ozone depletions will occur in the next century, particularly at the higher altitudes and latitudes.

Ozone Trends and Interpretations:

- The ground-based "Dobson" spectrophotometer network provides a 25-year record of global-average column ozone, which increased a few percent in the 1960's, remained relatively invariant in the 1970's, and decreased afterwards. Attention has focused on this recent 4-5% decrease because (a) it is the most-rapid sustained change ever observed in global column ozone and (b) it has occurred during the period of increasing stratospheric chlorine abundances.
- The causes of the decrease are not yet known. Solar variability, volcanic aerosols, and general circulation changes are possible contributing natural mechanisms. Indeed, analyses of solar activity find no fully convincing statistical evidence of a human-caused depletion in global column ozone in recent years, which is consistent with theoretical predictions that current losses should be less than a percent.
- The "Umkehr" technique, which uses the Dobson instruments, yields an 18-year record of the vertical distribution of ozone. Over the past decade, the data show ozone decreases at the higher altitudes (and only there) that are remarkably close to model-predicted depletions. However, questions about artifacts due to volcanic dust, particularly since the 1982 eruption of El Chichon, prevent a definitive statement regarding the significance of the trend.
- Unpublished results from the NASA SBUV-1 instrument on the NIMBUS 7 research satellite apparently show distinct ozone decreases over the past eight years, both in the global column and in the upper-strato-spheric abundances, that are substantially larger than those of the Dobson and Umkehr data or the theoretical predictions. However, this analysis and the calibrations are undergoing intensive review, and the results are not yet accepted by the scientific community.
- Inconsistencies between parts of the data sets and the possibility of instrumental artifacts have divided the atmospheric science community, and there is presently no universal agreement on whether there is evidence of a chlorine-induced depletion in global ozone.

Future NOAA Research:

- NOAA will continue to focus on reducing the uncertainties in the theoretical predictions of stratospheric ozone perturbations.

 Specifically, the near-term efforts will be to:
 - measure, in collaboration with NASA, the abundance of stratospheric reactive nitrogen, which is an important parameter in ozone chemistry,
 - monitor the atmospheric concentrations of the fast-growing new chlorinated and brominated compounds,
 - construct theoretical models that accurately simulate the three-dimensional transport and chemistry of ozone,

- develop an improved theoretical treatment of radiative transfer in the atmosphere and test it against vertical-distribution radiation measurements, and
- develop improved model capability for evaluating the climatic responses to atmospheric ozone changes.
- NOAA will continue to focus on improving the monitoring record. Specifically, it will:
 - continue participation in the Dobson and Umkehr networks and improvement and assessment of these methods,
 - collaborate in a reassessment of the entire ozone data set, examine the significance of trends and the meaning of the inconsistences, and plan to report the results by the end of 1987,
 - continue to collaborate in the analysis of the trends in ozone, temperature, and other data and in the assessment of the agreement between observations and theory,
 - continue the recently begun series of measurements with the SBUV-2 instrument on the TIROS satellite series and plan to implement an improved measurement system on future satellite series, and
 - participate in the planning of a state-of-the-art ground-based remote-sensing network that will detect stratospheric change, serve as a test of stratospheric theories, and provide a complementary absolute calibration, backup, and extension of the satellite monitoring systems.

ANTARCTIC STRATOSPHERIC OZONE DEPLETION

Background

- Dobson, satellite, and balloon-borne instruments have revealed growing, major (50%) ozone decreases over Antarctica in the austral spring season. The existence of this "ozone hole" is established beyond doubt.
- Halogen-chemical, sunspot-induced chemical, and dynamical/climatechange theories have been postulated. The first evokes man-made perturbations; the latter two evoke natural processes.
- The paucity of chemical and physical data at the time that these theories were proposed prevented assessment of their correctness.

1986 Ground-based Expedition

 Last summer, NSF, NASA, NOAA, and the CMA supported a 20-investigator expedition to McMurdo Base, Antarctic, led by a NOAA scientist, to provide additional data on the "ozone hole".

- The balloon-borne and ground-based ozone results show a 40% loss, mostly in the <u>lower</u> stratosphere, which makes the sunspot-induced chemical theory a <u>less-likely</u> explanation.
- Other measurements, also made by two different techniques, reveal that the abundance of the reactive chlorine compounds is highly elevated compared to other regions of the globe. Remaining uncertainties, however, prevent unequivocal identification of the man-made halogenated compounds as the cause of the ozone loss.
- Until the cause is established, the question as to the implication of the Antarctic decreases for global ozone cannot be answered.
- Because the current theories of the "ozone hole" require the unique physical features of Antarctica, the failure of the "standard" global ozone-depletion theories to predict its occurrence does not necessarily imply that these theories have major shortcomings regarding their predictions in other regions.

Plans for the 1987 Ground-based and Airborne Expeditions

- It is now established that chlorine chemistry and meteorological features are both enhanced in the Antarctic stratosphere, compatible with chemical and dynamical theories, respectively. Thus, it is mandatory to understand their respective roles in the "ozone hole" in much more detail. Consequently, NOAA and other agencies plan a greatly expanded effort during 1987.
- First, NOAA scientists will take an improved instrument back to NSF's McMurdo Base and will also provide year-round ozonesonde balloon launches from the South Pole during 1987. Scientists from other agencies and institutions will make up an expanded ground-based research effort this summer.
- In addition, NOAA will join NASA, NSF, CMA, and university investigators in an aircraft expedition to probe the stratosphere at the altitudes where the ozone decrease is occurring. The flights will be based out of Punta Arenas, Chile.
- NOAA scientists will have five chemical instruments on board NASA's ER-2 and DC-8 aircraft, and one of these investigators will serve as mission scientist. The full instrument payload will address very many of the key chemical constituents and physical parameters that are needed for testing the current chemical and dynamical theories.
- NOAA will help provide satellite ozone data that will locate the "ozone hole" and define its shape for the flight planning.

ASSESSMENTS AND SCIENTIFIC INPUT TO POLICY

• Scientific discoveries continue to be made (e.g., the "ozone hole") and understanding improves (e.g., the forthcoming trends re-evaluation). Hence, regular assessment of the state of knowledge is a key

part of policy refinement. NOAA will continue to contribute to such reviews and assessments.

• In addition, NOAA will continue its participation in the current international negotiations of a CFC/Halon protocol and will strive to help provide the scientific advice needed to base these negotiations on the most-complete current understanding of atmospheric circulation and chemistry.

STRATOSPHERIC OZONE:

THE STATE OF THE SCIENCE AND NOAA'S CURRENT AND FUTURE RESEARCH

I. INTRODUCTION

A. RATIONALE

The National Oceanic and Atmospheric Administration (NOAA) is focusing a substantial fraction of its environmental research on atmospheric ozone (03). The justification can be stated simply: ozone is the single most important chemically active trace gas in the earth's atmosphere. The reasons for this strong assertion are manyfold. The two that are the most relevant to this stratospheric-ozone summary are:

- The stratospheric ozone layer (between 10 to 50 km) is an ultraviolet shield that is essential to life on the planet.
- The temperature structure and hence the circulation patterns of the stratosphere depend on the distribution of ozone in this region.

B. ENVIRONMENTAL ISSUES

Around these key atmospheric roles of stratospheric ozone revolve two current environmental issues and their associated scientific questions:

- (1) Releases of man-made chlorine- and bromine-containing compounds into the atmosphere are predicted to be <u>depleting</u> the stratospheric ozone layer.
 - How scientifically sound is this hypothesis?
 - What are the future ozone depletions predicted for continued emissions of these compounds?
 - If these depletions were to occur, what would be the atmospheric and climatic consequences?
 - Is there any unequivocal observational evidence that stratospheric ozone is indeed being perturbed at present on a global scale by human activities?
- (2) Very recently, it was discovered that substantial year-to-year ozone decreases are occurring in the <u>Antarctic stratosphere</u> during late austral winter and early spring.
 - What is the cause of this "ozone hole", i.e., natural or man-made?

- What are the implications for global ozone?

These are challenging scientific and environmental questions to which policy makers rightly demand the best up-to-date, objective, scientific analyses that accurately reflect both the knowns and the unknowns of the situation. This summary describes (a) NOAA's current contributions to providing answers to these questions, (b) the future research plans in that regard, and (c) the current state of the science as it relates to public policy.

II. GLOBAL STRATOSPHERIC OZONE DEPLETION

A. THE PROBLEM

In 1974, two chemists at the University of California at Irvine hypothesized that man-made chlorofluorocarbons (CFC's) may be depleting the stratospheric ozone layer. Figure 1 schematically shows this hypothesis superimposed on a typical observed vertical-distribution profile of ozone in the atmosphere. The main features of the ozone-depletion hypothesis are noted and are as follows:

- (1) CFC's are being emitted at the earth's surface in what generally have been increasing quantities.
- (2) The only significant loss of these compounds is a slow (relative to the past emission rates) photodissociation in the stratosphere, the result of which has been a long-term buildup of stratospheric chlorine.
- (3) The chlorine fragments catalytically destroy many ozone molecules, i.e., the chlorine atom is not consumed in the total process.
- (4) A thinner ozone layer would absorb less solar ultraviolet light, resulting in a cooling of the upper stratosphere.

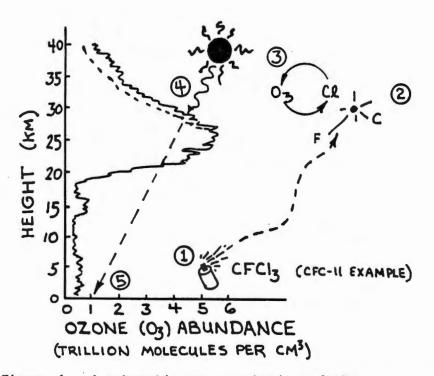


Figure 1. A schematic representation of the elements of the ozone-depletion hypothesis (Molina and Rowland, 1974). (Solid curve: ozone profile at 41°N, 106°W on 11 February, 1983. NOAA ultraviolet photometer data.)

(5) The increased ultraviolet radiation at the surface would cause elevated incidences of skin cancer, cataracts, and other biological effects, including disruption of some terrestrial and aquatic ecosystems.

The first four elements of this hypothesis relate to atmospheric responses. The last element relates to biological responses, which, while a crucially important part of the hypothesis, are outside the scope of this summary document and of NOAA research.

Over the decade since this hypothesis was proposed, the atmospheric scientific community has focused a substantial research effort along two general lines to test the hypothesis' correctness. The first is the development of a theoretical understanding of the response of the stratosphere to the emission of ozone-altering compounds, accompanied by process-oriented experimental observations that provide input to this model and appraisals of its validity. The second is a global, long-term monitoring effort designed not only to assist in the above development and testing, but also to search for direct observational evidence of the predicted human-caused perturbations of stratospheric ozone. The present status of these two endeavors are summarized separately in Sections II.B and II.C below.

B. GLOBAL OZONE DEPLETION THEORY

1. NOAA Current Research

NOAA has contributed to the development of this theory, as well as to the experimental appraisal of its validity. The NOAA theoretical work has taken two general directions. The first is a better understanding of the natural processes that form, distribute, and remove ozone, which give rise to the long-term natural background against which any human-caused trend will have to be identified. Sporadic and episodic events, such as bursts of solar particles, have provided natural short-term perturbations that have been used to test the theoretically predicted ozone responses. The second thrust is the development of improved models of the coupling of meteorology and chemistry, the results of which sounded an alert that ozone depletions are likely to be larger at higher latitudes than at the equator.

Shortly after the ozone-depletion hypothesis was proposed, NOAA's balloon-borne measurements demonstrated that the CFC's were indeed reaching the stratosphere in the quantities predicted by the theory. Laboratory studies revealed that some key reactions of the ozone-depleting chemistry were different than originally thought, thereby changing some aspects of the ozone-loss predictions.

Furthermore, long-term measurements of the atmospheric concentrations of the CFC's at remote global surface sites show that the worldwide abundance of these species is increasing, reflecting the growing atmospheric burden resulting from several years of generally increasing release rates. Lastly, NOAA's balloon and aircraft measurements of ozone-related stratospheric constituents have added to the data set against which the model calculations are being compared and hence evaluated.

2. Current State of the Theory

These theoretical and experimental studies of NOAA, as well as those of many others, have substantially increased the confidence in the basic structure of the ozone-depletion hypothesis. These features can be summarized as follows.

2.1 Natural Background Vis-a-vis Man-made Perturbations

Atmospheric ozone is predicted to respond to the total chlorine loading of the stratosphere, irrespective of the origin of the chlorine atoms. Direct stratospheric and tropospheric observations, reported production rates of chlorinated compounds, and transport and photochemical calculations yield a current lower-stratospheric chlorine abundance of about 2.8 parts of chlorine per billion parts of air (ppb), of which about 0.6 ppb is measured to be the natural background. The latter arises primarily from methyl chloride (CH₃Cl) emissions from the oceans. The lower-atmospheric abundances of the man-made chlorine compounds are observed to be increasing substantially; those of the natural compounds are not. Less is currently known about the bromine compounds, but a similar picture is emerging.

 The man-made chlorine emissions to date have increased the stratospheric chlorine abundance by a factor of four over the natural background.

2.2 Man-made Chemical Compounds

The man-made chemical compounds of primary concern in ozone depletion are a class of hydrocarbons, the alkanes [i.e., C_nH_{2n+2} ; e.g., CH_{4} (n=1) and C_2H_{6} (n=2)], in which one or more of the hydrogen atoms have been replaced by a halogen [i.e., a chlorine (Cl), bromine (Br), or fluorine (F) atom]. An alkane with some, but not all, of the hydrogen atoms replaced by halogens is referred to as a partially halogened compound (example, CFC-22, which is CHF₂Cl). An alkane with all of its hydrogen atoms replaced by halogens is referred to as a fully halogenated compound (example: CFC-11, which is CFCl₃). As noted below, the degree of hydrogen-atom replacement and the particular halogens involved are the properties of the halogenated alkanes that determine their atmospheric and public-policy roles.

The overall factors governing the relative effectiveness of the emissions of a chemical compound to deplete ozone are:

- (1) the rate of release of the compound into the atmosphere,
- (2) the rate of removal of the compound in the lower atmosphere and its photodissociation rate in the stratosphere, and
- (3) the efficiency of the halogen fragments in destroying ozone in the stratosphere.

Factors (2) and (3) combine to yield a quantity defined as a compound's ozone depleting potential (ODP). Table I gives calculated ODP values for examples of halogenated chemicals, along with their approximate 1985 global production rates and calculated atmospheric lifetimes.

Table I. Ozone-related properties of man-made halogenated compounds.

Group	Compound Name	Formula	Ozone Depleting Potential	Approximate 1985 Global Production Rate (million kg/yr)	Lifetime (yr)
(a)	CFC-11 CFC-12 Carbon Tetrachlorid	CFCl ₃ CF ₂ Cl ₂ CCl ₄	1.00 1.1 1.0	340 440 1050**	65 130 50
	CFC-113	C ₂ F ₃ Cl ₃	0.8	160	90
	CFC-114	C ₂ F ₄ Cl ₂	1.0*	very low	180
	CFC-115	C ₂ F ₅ Cl	0.6*	very low	380
(b)	Halon-1301	CF ₃ Br	10*	10*	110*
	Halon-1211	CF ₂ BrCl	3*	10*	25*
(c)	CFC-22 Methyl Chloroform	CHF ₂ Cl C ₂ H ₃ Cl ₃	0.08 0.1	210*** 550	20 7
(d)	CFC-123	C ₂ HF ₃ Cl ₂	<0.05*	0	short*
	CFC-132b	C ₂ H ₂ F ₂ Cl ₂	<0.05*	0	5*
	CFC-134a	C ₂ H ₂ F ₄	0	0	10*

^{*} Preliminary estimate.

Group (a) contains fully halogenated chlorine compounds with ODP values near unity, where CFC-11 is the reference for the ODP scale. Group (b) consists of fully halogenated bromine compounds. Their ODP values are greater than unity, reflecting the fact that bromine atoms are considerably more effective in destroying ozone molecules than are chlorine atoms. The fully halogenated compounds of groups (a) and (b) have long atmospheric lifetimes because they are resistant to chemical reactions and hence removal in the lower atmosphere. Group (c) contains partially halogenated chlorine compounds. Their ODP values are substantially less than unity. These examples were in widespread commercial use in 1985. Group (d) contains partially halogenated compounds not produced in commercial quantities in 1985, but which may have potentially large applications in the future as possible substitutes for group (a) because they have ODP values significantly less than unity. The shorter lifetimes of groups (c) and (d) reflect the fact that their partial halogenation permits chemical removal from the lower atmosphere. Lastly, fluorine atoms play little or no direct role in ozone depletion [e.g., the ODP of C2H2F1 (CFC134a) is zero].

Table I shows that, at present, CFC-11 and CFC-12 combined are the largest contributors, approximately 70%, to the predicted depletion of ozone and that CFC-113 contributes an additional 10%, where a compound's approximate

^{**} Greater than 90% is used as an intermediate chemical and hence cannot be released to the atmosphere.

^{***} A portion is used as an intermediate chemical (see **).
(Sources: UNEP, 1987; DuPont, 1987 and private communication.)

contribution is the product of its ODP and production rate. It is clear that any proposed regulations that would allow substitution for CFC-11 and CFC-12 by other fully halogenated CFC's (e.g., CFC-114 or CFC-115) would not lower the risk to the ozone layer because of the large ODP values and the long lifetimes of these replacements. Although at current levels of production, a percentage reduction of CFC-11 and CFC-12 would lower the risk of ozone depletion more than an equivalent percentage reduction in the production of the other compounds listed in Table I, the high growth rates in the production of some of these other compounds (e.g., CFC-113 and the Halons) will be a source of concern if these rates continue over long periods of time.

- From a scientific perspective, any proposed regulatory policy should consider all of the fully halogenated compounds [groups (a) and (b), which are characterized by high ODP's and long lifetimes] as a group for the purposes of regulation.
- Chemical compounds that have low ODP values, such as CFC-22 and those in class (d), may have significant value as possible substitutes.

2.3 Emission/Production of the Halogenated Compounds

The input to stratospheric ozone models should be the total global emission rate into the atmosphere. Clearly, such data are not generally available because of the diverse emission modes (e.g., use of spray cans, leaking auto air conditioners, losses during servicing of refrigeration systems, diffusion out of insulating foams, and industrial releases). Approximate emission rates have been derived from reported annual production data and from estimates of the small unreported annual production, the allocation of production across the end uses, and the duration for which the compounds are stored in the various products. The left-hand portion of Figure 2 shows the estimated global annual emission rates through 1985 for the predominant fully halogenated chlorine compounds [group (a) in Table I].

Beyond 1985, three future growth-rate scenarios are depicted: a true global freeze at 1985 emission rates, a constant 1½% per year growth compounded annually, and a constant 3% per year growth compounded annually. The three are, of course, only hypothetical illustrative cases chosen for the purpose of examining a spectrum of predicted future stratospheric ozone responses. However, a glance at Figure 2 shows that they are not inappropriate choices, given the past emission rates. Moreover, a global freeze is a more realistic scenario than it first seems, since it is the net chlorine reaching the stratosphere (i.e., ODP) that is assumed constant. Clearly, this could be met by decreases in some sources and/or compounds to offset increases in others.

 While the restrictions in the 1970's on the use of CFC's in aerosol sprays halted the rapidly increasing emission rates of the preceding decade, global emissions have begun increasing again in recent years.

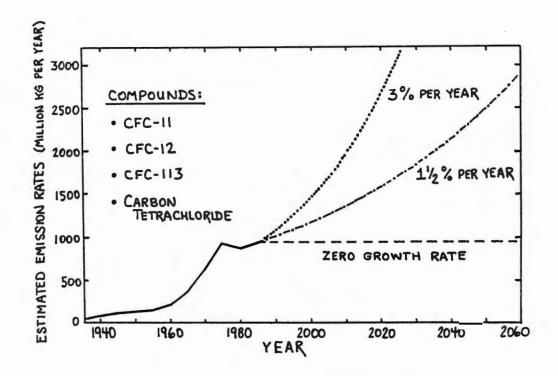


Figure 2. Estimated global annual emission rates of CFC-11, CFC-12, CFC-113, and carbon tetrachloride through 1985 and three hypothetical growth-rate scenarios. (Source: UNEP, 1987.)

2.4 Other Ozone-Related Trace Gases

The response of stratospheric ozone to chlorine emissions is sensitive to the abundances of other trace gases, notably carbon dioxide (CO2), methane (CHu), and nitrous oxide (N2O). The current theory predicts that increasing concentrations of carbon dioxide and methane increase the amount of ozone and those of nitrous oxide decrease it. Consequently, the predicted current and future ozone changes due to chlorine compounds depend on the current and future concentrations of these other trace gases. The source of CO2 is predominantly fossil fuel consumption, and hence fuel-usage projections are the basis for establishing possible future emission-rate scenarios. The one most commonly adopted in theoretical assessments is a future growth rate of approximately 0.5% per year. The sources of CH4 and N2O are poorly understood at present, but there are both man-made and natural contributions. The currently observed growth rates are about 1% per year for CH4 and 0.2% per year for N20. These are the future growth rates that are commonly adopted for CH4 and N2O in chlorine-perturbation calculations.

• The predicted ozone response to chlorinated and brominated compounds is sensitive to the scenarios assumed for the future emissions of $\rm CO_2$, $\rm CH_4$, and $\rm N_2O$. If the future growth rates of $\rm CO_2$ or $\rm CH_4$ are lower than assumed, then the predicted depletion of ozone would be greater. On the other hand, if the future growth rates of $\rm CO_2$ and $\rm CH_4$ are higher than assumed, then the predicted depletion would be lower.

2.5 Atmospheric Retention Times of the Fully Halogenated Compounds

As noted, the only significant loss of the fully halogenated compounds is photodestruction in the stratosphere. Today, this loss rate is much smaller than the emission rates of these compounds; thus, their stratospheric abundance would continue to grow for many decades, even if their emissions plateaued or declined moderately. For example, with a freeze at current emission rates, the stratospheric chlorine abundance would plateau after several decades to approximately 8 ppb (about three times the present level and ten times the natural abundance). On the other hand, since the growth in the stratospheric chlorine abundance is slow, differences of a few years in the timing of emission restrictions cannot produce dramatically different predictions. The details depend, of course, on the particular scenarios studied.

• From the standpoint of limiting the maximum concentrations of chlorine and bromine in the stratosphere, emission-rate restrictions done earlier need not be as severe as those done much later.

3. Theoretical Predictions and Implications

Based on the current understanding of atmospheric chemistry, radiation, and circulation, the response of stratospheric ozone to chlorine, bromine, and other trace-gas emissions is reflected in changes in the following properties of atmospheric ozone:

- total column amount of ozone overhead,
- variation of this column ozone with latitude, and
- vertical distribution of ozone.

The predicted response of each of these ozone properties is described below. The emphasis is on delineating the types of response and the atmospheric processes involved, rather than on detailed assessment calculations for specific policy options.

3.1 Predicted Atmospheric Response: Changes in the Total Vertical Column of Ozone

The atmospheric response that has been the subject of most theoretical scrutiny is the predicted CFC-induced change in the total column of ozone over-head. This is certainly an understandable initial focus. It is the column ozone that determines the amount of ultraviolet radiation reaching the ground. It has been the potential ultraviolet-induced biological effects that primarily have fueled the public-policy discussions regarding CFC regulations.

Figure 3 shows the 75-year changes in the global average column ozone that current one-dimensional (i.e., altitude) models predict for the three growth-rate scenarios of chlorinated and brominated compounds: zero, $1\frac{1}{2}$ per year compounded, and 3% per year compounded, beginning in 1985. Common to all three scenarios are the CO₂, CH₄, and N₂O increases cited above. The calculations include the species in groups (a), (b), and (c) of Table I (except for CFC-114 and CFC-115, whose current production rates are nearly zero).

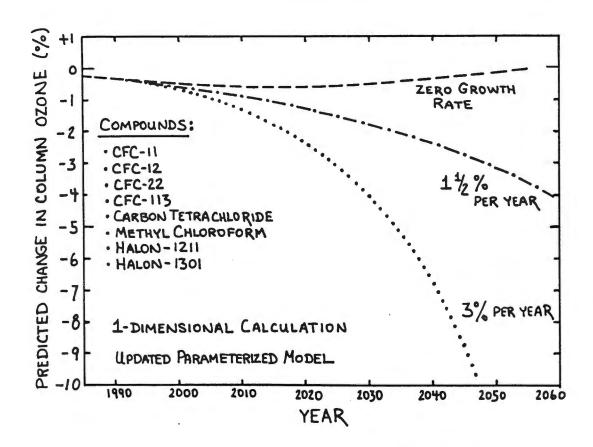


Figure 3. Predicted global average column ozone changes for three growth-rate scenarios of chlorinated and brominated compounds and continued increases of CO_2 , CH_4 , and N_2O emissions. (Source: UNEP, 1987.)

The global average column ozone depletion occurring in 1985 as a result of the emissions of chlorinated compounds during the preceding decades (see Figure 2) is predicted to be a fraction of a percent, as indicated in Figure 3. Furthermore, if the annual emission rates of the chlorinated and brominated compounds were to remain constant at the 1985 levels, the future column ozone loss is predicted to be approximately a percent or less over the next 75 years. In this case, the ozone losses due to the constant chlorine and bromine emissions are eventually offset by the ozone production due to the increasing $\rm CO_2$ and $\rm CH_{\mbox{\sc H}}$ emissions. If the adopted $\rm CH_{\mbox{\sc H}}$ growth rate had been 0.5% per year (rather than 1%), the offsetting effect would be less and the predicted column ozone depletion would grow to 2% by the year 2060. (This alternative scenario is not shown in Figure 3.)

For the two scenarios in which emissions grow at $1\frac{1}{2}$ and 3% per year, Figure 3 shows that the effect of the increasing chlorine/bromine concentrations dominates, and hence the column ozone loss grows steadily with time. Different one-dimensional models yield similar results. Furthermore, other emission-increase scenarios also show the same features, but, of course, differ in degree. The salient points of such emission-increase scenarios remain the same, which are the following.

The current understanding of the response of stratospheric ozone to increasing chlorine and bromine abundances predicts that a 3% annual growth rate risks a very large global-average column ozone depletion after 75 years. A 1½ annual growth rate is predicted to induce a 4% loss. Although public policy has not yet set an "acceptable" ozone loss, such growth rates risk a manmade perturbation that is comparable to or greater than what measurements indicate may be the decadal natural column ozone variation: 3 to 4% (see Figure 7 below and the associated text).

• A freeze of total chlorine and bromine emission rates is consistent with limiting predicted long-term perturbations in the global average column ozone to magnitudes less than the decadal natural variation, which is about 3 to 4%.

3.2 Predicted Atmospheric Response: Latitudinal Dependence of Column Ozone Changes

The current theoretical understanding of the interaction of global circulation patterns and ozone chemistry predicts that more column ozone loss will occur at higher latitudes than at the equator. This latitudinal dependence has been examined with several different two-dimensional (i.e., altitude and latitude) models, with consistent results. Figure 4 shows this predicted variation with latitude for a freeze in CFC-11 and CFC-12 emissions, beginning in 1980, and continued growth in CH $_{\rm H}$ and N $_{\rm 20}$ emissions. The predicted column ozone depletions in the springtime (when the effect is a maximum) for the high latitudes are two to three times larger than the predictions of one-dimensional models. Therefore, even with a global freeze, column ozone losses of several percent are predicted to occur at the high-latitude locations (e.g., Alaska and Scandinavia) after 75 years.

 A significant decrease in total chlorine and bromine emission rates is consistent with limiting predicted long-term perturbations in the column ozone at high latitudes to magnitudes less than decadal natural variation.

3.3 Predicted Atmospheric Response: Changes in the Vertical Distribution of Ozone

The percentage changes in local ozone are negative in the upper strato-sphere and are positive in the lower stratosphere and below. For the scenario of 11/2 growth rate, the upper-stratospheric ozone loss is greater than the ozone production at the lower altitudes, and hence the predicted long-term change in the total column ozone is a substantial depletion, similar to that

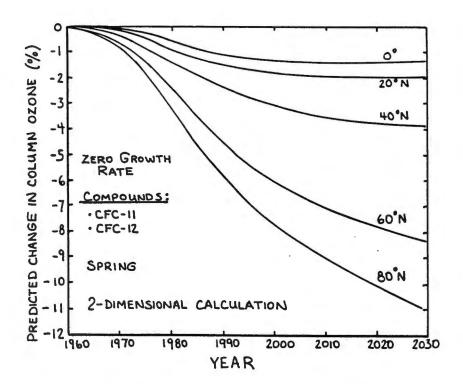


Figure 4. Predicted depletions in the column ozone for various latitudes for a freeze in CFC-11 and CFC-12 emissions at 1980 levels and continued increases of CH_{4} and N_{2}O emissions. Note: These are likely to be upper limits since the calculation did not include CO_{2} , whose offsetting effect would probably reduce the predicted depletions by about 30%. (Source: UNEP, 1986).

depicted by the dot-dashed curve in Figure 3. For the emissions-freeze case, there is near cancellation of the ozone losses in the upper stratosphere and the ozone production in the lower atmosphere. Hence, the predicted net change in the column ozone is nearly zero, similar to that depicted by the dashed curve in Figure 3.

Nevertheless, even though a global freeze is predicted to preserve the total ozone column within a few percent, the depletion in the upper stratosphere and the production at lower altitudes are themselves potentially of environmental significance. Each change arises from different processes and has different possible consequences. These are examined separately in Figure 6, which depicts the emissions-freeze case.

Upper-stratospheric ozone decrease

The upper parts of the solid curve and hatched area in Figure 6 shows that the long-term depletion of local ozone at 40 km is predicted to be about 25%. The dashed curve shows the depletions that are predicted for the CFC's alone, i.e., no effects of CO_2 , CH_4 , and N_2O_3 . As indicated, the predicted

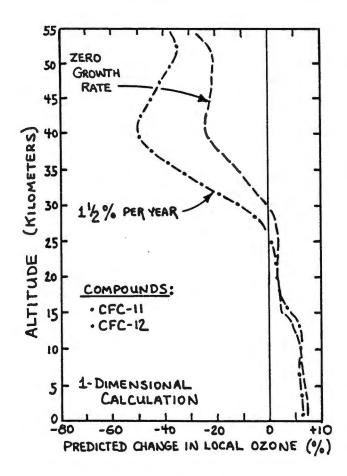


Figure 5. Predicted long-term changes in the vertical distribution of ozone for two CFC-11 and CFC-12 growth-rate scenarios and continued growth of CO_2 , CH_{4} , and N_2O emissions. The steady-state values that are reached are 8 and 15 ppb chlorine, respectively; doubled CO_2 and CH_{4} ; and a 20% increase in N_2O . (Source: WMO, 1985.)

depletion would be much larger were it not for the ameliorating effect of the other trace gases, primarily ${\rm CO_2}$, namely, a cooling that slows the temperature-dependent, natural ozone-destruction reactions.

The current understanding of the consequences of such a 25% ozone decrease is that it may produce a local temperature decrease of 5°C. The possible effects of such a cooling on stratospheric circulation patterns are current research topics.

The comparison of this predicted cooling to natural temperature variations is hampered by the lack of long-term temperature records at these altitudes. However, limited rocketsonde data for the western portion of the northern hemisphere suggest a decadal temperature decrease of 1-2°C. All of this decline may not be purely a natural variation, since some scientists claim that human-caused CO2 increases may be responsible for part of this cooling trend of the past decade. Nevertheless, a global freeze on chlorine and bromine emissions is predicted to allow upper-stratospheric temperature changes that are several times larger than what is currently the only estimate of decadal natural variation at those altitudes.

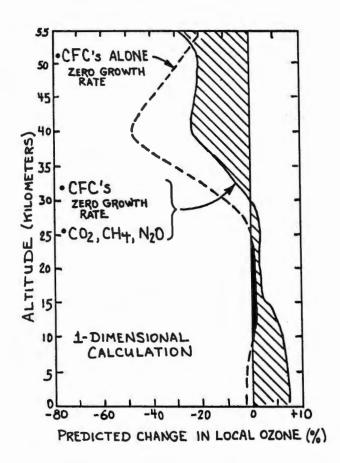


Figure 6. Predicted long-term changes in the vertical distribution of ozone for a constant emission rate of CFC-11 and CFC-12 since 1980 and continued growth in CO_2 , CH_4 , and N_2O emissions (solid curve). The dashed curve reflects the predicted changes due to the CFC's alone (constant 1980 rates). The steady-state abundances that are reached are 8 ppb chlorine, doubled CO_2 and CH_4 , and a 20% increase in N_2O . (Source: WMO, 1985).

- A significant decrease in total chlorine and bromine emission rates is consistent with limiting predicted long-term losses in the ozone at 40 km and associated decreases in local temperatures to magnitudes comparable to or less than the current (limited) observations of decadal natural variation, which is about 1 to 2°C.
- However, the couplings and feedbacks of chemistry, radiation, and circulation in the upper stratosphere have yet to be represented fully in three-dimensional models. Furthermore, the possible effects of the predicted upper-stratospheric cooling on life at the earth's surface are unknown at present.

Lower-atmospheric ozone production

The lower parts of the solid curve and hatched area in Figure 6 highlight the long-term ozone production that is predicted to occur below 25-30 km from

constant 1980-level emissions of the two CFC's and continued increases in the emissions of the other trace gases, primarily CH4. The lower dashed curve and the solid area identify the portion of the predicted ozone increase that is due to the presence of CFC's alone, namely, the ozone production resulting from enhanced ultraviolet radiation at those lower altitudes (a consequence of upper-stratospheric ozone losses). The area that is the difference between the hatched and solid areas represents the ozone that is predicted to be produced photochemically from CH4. Because of the complexities of the chemistry in the lower atmosphere (e.g., regional pollution effects), there is substantial uncertainty in this prediction.

The potential consequence of such ozone production, particularly those in the region of the temperature minimum at 10-15 km, is an enhanced "greenhouse effect". Specifically, at the higher pressures of the lower atmosphere, ozone absorbs infrared radiation from the earth and radiates a fraction of this outgoing energy back to the surface, thereby elevating the temperature.

Like CO_2 , the CFC's and CH $_{\parallel}$ are also "greenhouse molecules". Therefore, the CFC's and CH $_{\parallel}$ contribute both directly and indirectly to "greenhouse" warmings, the latter being the associated increases in lower-atmospheric ozone. Current radiative models predict that the emissions of CFC's through 1980 have committed the planet to an eventual warming that is about 10% of that produced by the past CO_2 emissions, (i.e., about 10% of 0.6°C). Essentially all of this CFC contribution is believed to be due to their direct "greenhouse" role. The time delay in the arrival of the warmer surface temperatures is due primarily to the thermal inertia of the oceans.

There is a distinct need for new global-warming calculations to be made for trace-gas scenarios that are under public discussion presently, e.g., a global freeze in the late 1980's on the fully halogenated compounds. Based on the limited number of future emission scenarios that have been examined in radiation models, it is estimated that a freeze of CFC-11 and CFC-12 emissions at the present rates would have committed, by the year 2030, the planet to a direct and indirect greenhouse warming that could be (very approximately) a fourth of that due to $\rm CO_2$ (i.e., about 1/4 of 1.3°C). The uncertainties in the long-term predictions of current climate models is probably a factor of three, due primarily to the incompleteness with which the cloud feedback processes can be incorporated at present.

Surface temperature changes have been measured directly for about a century and have been inferred from tree-ring variations for an additional century and a half. This 250-year record reveals natural variations of ±0.5°C, with changes of 0.5°C occurring on both decadal and centennial time scales. Thus, the approximate 0.3°C long-term warming that is predicted to be associated with a 1980's freeze of CFC emissions is comparable to the observed long-term natural variations.

• A decrease in the total emission rates of chlorinated and brominated compounds is consistent with limiting their predicted commitment to a long-term "greenhouse" warming such that it is less than the decadal natural variation, which is about 0.5°C.

3.4 Uncertainties in Predictions

All of the responses examined above are the results of theoretical predictions. If they are to be taken as forecasts of how the atmosphere will actually behave over the next 75 years, then the uncertainties in these forecasts arise from two primary sources:

- (1) the inability to forecast what the actual trace gas abundances will be and
- (2) deficiencies in the framework of the theoretical models of the atmosphere.

For the first, the issue is not so much the future production rates of the chlorinated and brominated compounds, since the current public-policy discussions are examining the various scenarios of these emissions as regulatory options. Rather, this source of uncertainty arises from the inability to forecast the future $\rm CO_2$, $\rm CH_4$, and $\rm N_2O$ abundances, which are outside of the present policy discussions. The effect of this uncertainty is highlighted by recognizing that the small global average column ozone change in the scenario for zero growth of the fully halogenated compounds in Figure 3 is a cancellation of multiple trace-gas effects.

The second source of uncertainty, model deficiencies, is more subtle and difficult to assess. While the current satisfactory consistency between the predictions of different models is very gratifying, it cannot be taken as proof that all important atmospheric processes are known and have been correctly included. The basic test of a model is that it closely simulates the observed ozone-related features of the present atmosphere. Many of these features have been characterized by observations. Disagreements between a few of the observations and model predictions do exist, which places some limits on the confidence in the predictive abilities of the models. A particularly unsettling discrepancy is the apparent underprediction by about 30% of ozone itself in the upper stratosphere. Furthermore, the treatment of dynamical processes in the two-dimensional models is still a subject of current research. However, in general, there is good agreement between observations and predictions for a large number of atmospheric features.

- Since stratospheric ozone is sensitive to changes in other trace gases besides chlorinated and brominated compounds, an additional uncertainty in the assessments of possible man-made perturbations is the difficulty in predicting the future abundances of the other trace gases, particularly $\rm CO_2$ and $\rm CH_4$.
- While the models based on the present theories simulate much of the present stratosphere quite well, they are not perfect, which places a factor of two to three uncertainty on their predictive abilities.
- Nevertheless, based on present understanding, most atmospheric scientists believe that if the present growth rates of chlorinated and brominated compounds continue unabated indefinitely, then it is highly likely that substantial ozone depletions will occur in the next century, particularly at the higher altitudes and latitudes.

4. NOAA Future Research

NOAA research will continue to focus on reducing the uncertainties in the predictions of possible stratospheric ozone perturbations. The major emphases will be the following near-term research efforts:

- In collaboration with the National Aeronautics and Space Administration (NASA), the reactive nitrogen content of the lower stratosphere will be measured in both hemispheres with an instrumented research aircraft. The reactive nitrogen sources, abundance, and latitudinal distribution are important parameters in controlling the rate of chlorine-induced ozone loss.
- The processes by which ozone-related compounds are exhanged between the stratosphere and the lower atmosphere will be characterized better by direct airborne measurements (in collaboration with NASA) and accompanying theoretical investigations.
- CFC-113, CFC-22, Halon 1301, and Halon 1211 are being added to the ground-based monitoring program. The atmospheric abundances of these species are the fastest growing of the chlorinated and brominated compounds.
- Theoretical simulation of ozone-influencing atmospheric circulation will be improved with higher resolution in three-dimensional models, thereby better defining the scales of ozone variation.
- An improved treatment of radiation will be incorporated into one- and two-dimensional models and tested by making vertical-distribution measurements of solar radiation.
- Interactive three-dimensional models of ozone with climatic feedbacks will be explored, with an emphasis on improving the understanding of the interactions between CO₂, ozone, and temperature changes.

C. GLOBAL OZONE DEPLETION OBSERVATIONS

Similar to the advance of the theory of the stratospheric ozone response to chlorine and bromine emissions, there has been substantial effort to monitor the variation of ozone in order to detect human-caused depletions.

1. NOAA Current Research

NOAA has contributed to this accumulating data base that shows how the stratospheric ozone abundance varies in space and time. Specifically, NOAA scientists are operating several ground-based "Dobson" instruments that monitor the total amount of ozone overhead and that estimate how it is distributed vertically (via the "Umkehr" method). Balloon-borne ozonesondes are also providing regular measurements of the vertical distribution at selected sites. In addition, NOAA has a Solar Backscatter Ultraviolet instrument (SBUV-2) on its TIROS satellite series that, once the testing is complete, will provide similar information on a global scale. This instrument represents a monitoring continuation of the NASA research version (SBUV-1) launched several years

earlier on the NIMBUS 7 satellite. NOAA scientists are among the analysts who scrutinize this accumulating data base for trends arising from natural variation and for the first signs of possible human-caused depletions.

2. Current State of the Observations

As explained above, current models predict that the present global column ozone depletions due to the past emissions of chlorinated compounds should be small, less than a percent (see Figure 3). At high latitudes, the predicted current column ozone depletions are probably less than a few percent (see Figure 4). Based on one-dimensional, time-dependent calculations similar to those leading to Figure 5, the predicted current ozone depletions at 40 km, which is where the percentage changes due to chlorine chemistry are expected to be the largest, are approximately five percent. How consistent are these predictions with the present stratospheric ozone data record?

2.1 Global-Average Column Ozone Changes

Column ozone observations are the longest-running ozone record. The ozone column has been observed both from above and below.

Ground-based observations

The ground-based Dobson network, which currently consists of approximately 85 stations, has provided global column ozone data from about 1960 to the present. The 25-year record up through 1985 is given in Figure 7.

The center portion of Figure 7 shows the temporal behavior of the average column ozone for the globe. The biennial variation is evident in this data set, as are longer-term trends over decades. Global column ozone increased by about 3% through the 1960's, remained more invariant during the 1970's, and has decreased about 4-5% in the 1980's.

There are individual Dobson stations that provide a longer data record. The most notable of these is Arosa, Switzerland, whose measurements began in 1932. These data suggest that column ozone was, at that site, about 3% lower during the decade of the 1930's compared to that of the 1970's.

The experimental uncertainty in the Dobson technique has been examined extensively. The relative accuracy (i.e., for trend determinations) is considered to be 1-2%. Consequently, it is very likely that much of the variation shown in Figure 7 truly reflects changes in global ozone.

Satellite observations

The SBUV-1 instrument on the NIMBUS 7 satellite has made measurements of column ozone since 1979 and provides far more extensive global coverage than the Dobson data set. Unpublished accounts of the satellite data cite a 7.4% decrease in global column ozone over its eight-year record. The overall calibration of the SBUV-1 sensor is known to be drifting since launch, making this estimate very uncertain. One way to account for this drift is to correct for it by matching the data to the ground-based Dobson measurements for the limited number of overpasses of the network sites. This recalibration results in a

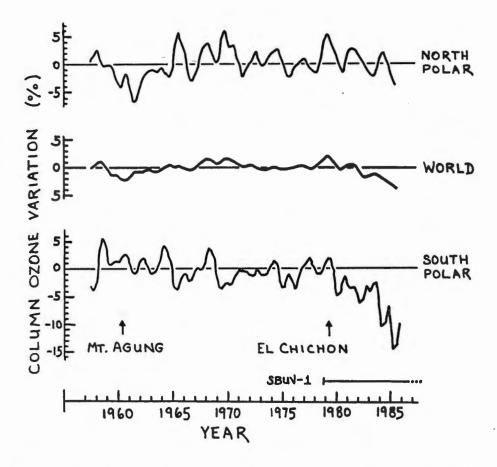


Figure 7. Seasonally averaged column ozone observed by the ground-based Dobson network. The occurrence of several potentially ozone-altering events are noted. The measurement period of the SBUV-1 satellite instrument is also indicated. (Source: Angell, 1987.)

4.4% decreasing trend. Therefore, the spatially limited number of Dobson data and the globally extended satellite observations show the same downward trend from 1979 to 1985, if this recalibration is adopted.

Interpretation

Thus, both the ground-based Dobson data and the satellite data demonstrate that global column ozone changes on several time scales. A variety of possible radiative, chemical, and dynamical causes have been considered, both natural and man-made: variation in solar output radiation, episodic aerosol injection from major volcanic eruptions like El Chichon in 1982, circulation anomalies like the major El Niño - Southern Oscillation event in 1982-83, the chemical perturbations introduced by the atmospheric nuclear bomb tests in 1961-62, and the growing stratospheric abundance of man-made chlorine compounds during the last decade. For obvious reasons, considerable attention has been focused on the downward trend that began about 1979. The question is not whether an ozone decrease is indeed occurring. Experimentally, there is little doubt about this. Rather, the question relates to why. Specifically, how much of this global decrease in column ozone is due to a depletion caused by man-made chlorine compounds?

If the 3% increase of the sixties is due to unexplained natural variation (and not a possible recovery from the 1961-62 bomb tests), then it cannot be ruled out that the 4-5% decrease of the last decade also may be due largely to such variation. Indeed, analyses of these data, which include an accounting for the external ozone forcing by solar activity, do not reveal any statistically significant ozone depletions in recent years. Specifically, the solar sunspot cycle was in a strongly decreasing phase over this period, which may account for about 2% of the decline. Furthermore, about 1% is due to the large loss in a specific region, Antarctica (which is examined in Section III below). Thus, there is an "unexplained" decrease of less than 2% in global ozone over the seven years as evidenced by the Dobson and satellite data. When this 1-2% unexplained decrease is considered against the observed variability seen in the 25-year Dobson record, there appears to be no inconsistency in accepting that this is within the range of natural variation, and hence it alone cannot be considered as conclusive evidence of an anthropogenic influence.

This conclusion is consistent with the current theory of stratospheric ozone chemistry, which predicts that the present global column ozone depletions due to past releases of chlorinated compounds should be less than 1% (see Figure 3). A depletion of this magnitude would be very difficult to identify against the background of poorly understood natural variation. It is particularly ironic that, during the period 1979-1985 over which the Dobson and satellite data are being intently scrutinized for a possible chlorine-induced ozone loss, two major geophysical events of the century occurred: El Chichon's 1982 eruption and the 1982-1983 El Niño.

Current theory predicts that the chlorine-induced column ozone depletion should be larger at high latitudes, perhaps a few percent by 1985 (see Figure 4). The upper part of Figure 7 shows the Dobson data record for the north polar region. No downward trend for the last decade is obvious, although the larger biennial oscillation at these latitudes and the uncertainty introduced by the limited number of stations (three) make it difficult to judge. There are unpublished comments regarding column ozone depletions at the northern latitudes based on the SBUV-1 data, but without the opportunity to examine the details, it is difficult to assess their meaning. Thus, there is no unequivocal observational evidence of a decrease in column ozone at high latitudes. This does not "disprove" the chlorine-induced ozone-depletion theory, since the predicted current depletions, even at high latitudes, are still relatively small compared to the short-term variations in the published ozone data.

Discussion of the lower part of Figure 7 is deferred until Section III below.

2.2 Ozone Vertical-Distribution Changes

Although the existing data record for the vertical distribution of ozone is not as long as that for the column ozone, it does begin to offer the opportunity to search for a possible chlorine-induced "signature" in the trends. The vertical distribution of ozone has been measured by ground-based, balloon-borne, and satellite instruments.

Ground-based observations

The Umkehr variation of the Dobson method provides estimates of the altitude distribution of ozone. Hence, a time series of Umkehr data can reveal the temporal change in local ozone at different stratospheric altitudes. The method has been applied consistently only since about 1967, so the 18-year data record is about three-quarters that of column ozone from the Dobson network. Currently, there are 13 Umkehr stations. Figure 8 shows the temporal variation of ozone in two altitude layers above the north temperate latitudes (where most of the Umkehr sites happened to be located).

Current theory predicts that the largest altitude-dependent ozone depletions due to chlorine emissions should be in the 48-32 km altitude region (see the discussion associated with Figure 5). Indeed, if these data are taken at face value, they exhibit a distinct overall downward trend of about 5% over the past decade. Moreover, the data for the lower altitudes, 32-24 km, show no significant trend.

Balloon-borne observations

Until recently, almost all of the sites at which ozonesondes are launched regularly have been at the north temperate latitudes. There are currently about 12 such sites, and the data extend back to 1967. Therefore, the temporal and spatial coverage of the ozonesonde network is limited much as the Umkehr network is. In addition, the accuracy with which the expendable sondes can measure ozone deteriorates in the upper stratosphere. Hence, there is a dearth of reliable trend data from ozonesondes in the critical 48-32 km alti-

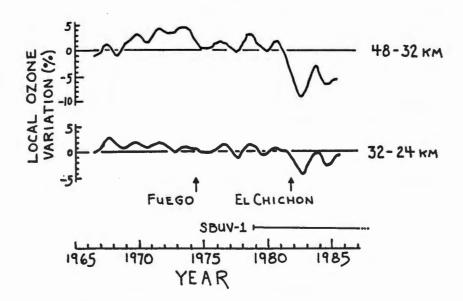


Figure 8. Ozone in the 48-32 and 32-24 km altitude ranges observed by the Umkehr stations in north temperate latitudes. The arrows denote the dates of the indicated volcanic eruptions, and the measurement period of the SBUV-1 satellite instrument is also noted. (Source: Angell, 1987.)

tude range. However, the sonde data for the lower layer, 32-24 km, do provide a valuable confirmation of the Umkehr finding of the absence of any prominant trend.

Satellite observations

The SBUV-1 instrument, in addition to column ozone, can yield the vertical ozone distribution, in a fashion similar to that of the Umkehr method. Hence, its 8-year data record potentially can yield the temporal trends of ozone over a range of altitudes and latitudes. Unpublished statements refer to a 15% decrease of global-average ozone at 45 km over the past eight years and a 7% decrease at 33 km. The average of these decreases is about twice as large as the decline based on the Umkehr data for the 48-32 km layer. The ozone decreases reported tended to be larger at higher latitudes.

Interpretation

The Umkehr data record shows ozone decreases that are consistent with the theoretical predictions based on past chlorine emissions, namely, about 5% decadal loss of ozone in the upper stratosphere. This record is the strongest of the existing observational evidence that supports the reality of current ozone loss due to man-made chlorine compounds. However, it is not without its uncertainties. As noted by the arrows in Figure 8, two major volcanic eruptions occurred during this time period. The Umkehr technique is quite sensitive to interference by stratospheric dust. Corrections must be made using ground-based lidar data on the vertical dust distribution. Only a few Umkehr sites have accompanying lidars; hence, the correction must often be made using assumed dust distributions. As a result, the uncertainties of this data set are difficult to quantify.

The most thought-provoking data set of ozone vertical distributions is the unpublished record from the SBUV-1 satellite instrument. Since the larger ozone decreases are associated reportedly with the higher altitudes and latitudes, key features of the chlorine-induced ozone losses would appear to be present. If these measurements are indeed correct, the magnitude of this reported decrease is substantially larger than what current theory is predicting, which would imply model deficiencies. However, these data are not yet accepted by the scientific community. The calibration difficulties have been discussed above. Since the global three-dimensional coverage afforded by a satellite provides a very sensitive way to search for the predicted chlorine induced "signature", the correctness of these data is a pressing issue that needs to be settled.

3. Implications of the Trends Observations

The stratospheric scientific community currently is $\underline{\text{divided}}$ as to whether the existing data on ozone trends provide sufficient evidence to believe that a chlorine-induced ozone destruction is occurring now. The main points $\underline{\text{for}}$ and against are as follows:



The current global stratospheric ozone data record DOES show compelling evidence of a chlorine-induced ozone depletion:

- Global column ozone is lower now than it has been at any time since such observations began.
- Indeed, the steepest downward trend of global ozone of extended duration that has been seen over the past 25 years of the Dobson network is the decrease that has been occurring over the last few years, which is the period of increasing stratospheric chlorine abundance.
- The data from the SBUV-1 satellite instrument indicate that the recent global column ozone decrease may be even larger than originally believed.
- The ozone vertical-distribution data record from the Umkehr network show ozone decreases in the upper stratosphere over the past decade that agree both in magnitude and altitude with the predictions of the chlorine-induced ozone-depletion theory.
- The SBUV-1 vertical-distribution data show ozone decreases over the past eight years that are larger at high altitudes and latitudes, a pattern that agrees with the chlorine-induced theory.
- The magnitudes of these high-altitude and high-latitude ozone decreases are even larger than theory predicts!



The current global stratospheric ozone data record DOES NOT show compelling evidence of a chlorine-induced ozone depletion:

- When the whole Dobson data record is considered, there is no compelling statistical evidence of a chlorine-induced ozone depletion of global column ozone, even at high latitudes, during the last several years.
- In particular, the often-cited time period of 1979-1985 includes four unique <u>natural</u> anomalies and potential ozone perturbations: (a) it began with the <u>highest</u> global column ozone on record, (b) it contained the powerful El Chichon volcanic eruption, (c) it included the

strong 1982-83 El Niño, and (d) it spanned a period of strongly decreasing solar output. The one possible man-made contribution to the 4-5% ozone decrease of this period cannot, at present, be distinguished from the potential contributions of these natural causes.

- The SBUV-1 data for 1979-1985 do not show unequivocally larger column ozone decreases than the Dobson data. Specifically, the SBUV-1 instrument has no onboard calibration standard, and hence the known drift in the sensor's sensitivity is difficult to account for unambiguously. When the SBUV-1 data are calibrated with those from the overpassed Dobson sites, the two data sets, which are vastly different in spatial coverage, yield consistent global column ozone trends.
- The upper-stratospheric Umkehr data of the past decade neither confirm nor disprove the chlorine-induced ozone-depletion theory. This method of obtaining ozone vertical distributions is very sensitive to volcanic dust, such as that from El Chichon. The uncertainties introduced from incomplete corrections for this effect are not well-quantified.
- The large downward ozone trends at high altitudes and high latitudes that are reportedly in the SBUV-1 data cannot be accepted as proof of a chlorine-induced ozone depletion. These results have not been published yet. As a result, they have not yet had the scrutiny of the scientific community that they clearly deserve, particularly in light of the calibration difficulties.
- Lastly, the unpublished statements regarding this data set appear to have an internal contradiction: the chlorine-induced ozone-depletion theory is evoked to explain the observed pattern of the high-altitude, high-latitude ozone decreases, but it is rejected as insufficient in explaining the magnitude of those decreases. One is not allowed to play it both ways!

It is clear that "the jury is still out" on the implications of observed stratospheric ozone trends.

4. NOAA Future Research

NOAA's ozone-trends research will continue to focus on improving the monitoring record. Specifically, the near-term research will consist of the following tasks.

- Participation in the Dobson and Umkehr networks obviously will continue. It is particularly important to establish the future near-term behavior of the present sharp downward trend in global column ozone and upper-stratospheric ozone.
- NOAA Dobson and Umkehr sites will be improved. In particular, expanded lidar measurements of the stratospheric dust distribution will increase the accuracy of ozone data.

- NOAA will continue to implement the current SBUV-2 instrument on the TIROS satellite, to plan to extend these types of measurements through the mid-1990's, and to design an improved instrumental system for monitoring ozone and other related species on future satellite series after TIROS.
- NOAA scientists have joined colleagues from NASA, the Chemical Manufacturers Association (CMA), and several universities in an independent evaluation of the existing Dobson, Umkehr, and satellite data sets. The goal is to address the inconsistencies, reanalyze data, intercompare in detail, re-estimate uncertainties, and put together, insofar as possible, a consistent picture of global ozone trends and their implications by the end of 1987.
- NOAA has joined NASA and CMA in planning a ground-based network of several sites that will use modern remote-sensing techniques to detect stratospheric change and will employ expanded data-handling and assimilative capacities to aid and speed interpretation. The results will have several critical and immediate payoffs. First, the covariation of several stratospheric chemical species and properties is key input to improve theory, as well as providing an assessment of its adequacy. Moreover, the data would serve as the absolute calibration for satellite sensors, provide an independent monitoring system for quality-control intercomparisons with the satellite systems, act as backup in case of satellite or launch-vehicle problems, and include chemical species not obtainable presently by satellites.

III. ANTARCTIC STRATOSPHERIC OZONE DEPLETION

A. THE PROBLEM

In contrast to the search for the small beginnings of the predicted global chlorine-induced ozone loss and the debate over its reality, an unprecedented and unanticipated phenomenon was discovered recently that has riveted the attention of ozone scientists, policy makers, industrialists, and environmentalists alike, namely the Antarctic "ozone hole". Here, the amount lost is considerable -- half of the ozone over Antarctica disappears or fails to appear in a very puzzling fashion -- and the agreement between the ground-based, satellite, and balloon-borne ozone data for that region leave no doubt about the reality of this phenomenon.

The data that revealed the existence of the phenomenon are shown in Figure 9. In the Antarctic spring (and only there and only at that season), a substantial fraction of the column ozone disappears. The fractional decrease has increased, on the average, over the past several years. The magnitude of the decrease and the nature of its occurrence could not be explained by previously existing ozone-loss theory (i.e., that described in Sections IIA and IIB). The fact that nothing of this magnitude had occurred earlier in the south polar latitudes is shown by the 25-year column ozone record given in the lower part of Figure 7. Indeed, this recent decrease in Antarctic ozone discovered by the British Antarctic Survey is far larger than the variability observed at any time anywhere on the globe. What is causing it?

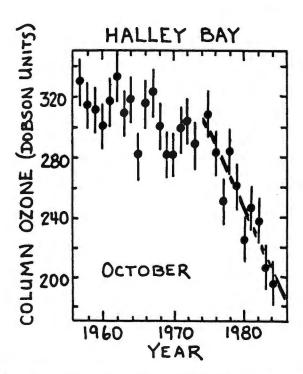


Figure 9. Column ozone observations by the British Antarctic Survey at Halley Bay, Antarctica, in the austral spring. (Source: Farman, Gardener, and Shanklin, 1985.)

 The existence of growing and substantial seasonal losses of ozone in the Antarctic stratosphere is established beyond doubt and is unique among ozone observations.

B. CURRENT STATE OF THE SCIENCE

1. NOAA Current Research

NOAA researchers have responded to this puzzle with new theoretical and experimental studies. Shortly after the discovery of the "ozone hole" was announced by the British Antarctic Survey in 1985, NOAA scientists sought to explain this dramatic phenomenon in terms of two separate theories: natural climate change and man-made chemical perturbation, both theories recognizing certain unique features of the Antarctic region, such as atmospheric circulation patterns, extremely low temperatures, and polar stratospheric clouds.

The paucity of chemical data at the time for the Antarctic meant that it was difficult to decide which of these two theories, or a separate solar-cycle theory, was closer to the truth. Therefore, in the summer of 1986, NOAA joined with NSF, NASA, and CMA to rectify this data-short situation by helping organize, support, and participate in the National Ozone Expedition (NOZE). A NOAA scientist served as the team leader, and she and her colleagues helped make up a 20-investigator team that greatly added to the chemical information regarding the occurrence of the "ozone hole".

In addition, NOAA analysts have scrutinized the existing meteorological data for Antarctica to obtain a more detailed look at temperature trends and other physical parameters that might provide correlative clues regarding the mechanisms that could be responsible for the ozone decrease.

2. "Ozone Hole" Theories

Two general groups of theories, chemical and dynamical, have been proposed to explain the seasonal loss of ozone over Antarctica and its past increase in magnitude.

2.1 Chemical theories

The current chemical theories of the "ozone hole" fall into two classes, depending upon the identity of the chemical species hypothesized to be reacting with the ozone: man-made chlorine and bromine compounds and naturally occurring nitrogen compounds.

Man-made chlorine and bromine compounds

The fact that the seasonal Antarctic ozone loss began about the same time that stratospheric chlorine was increasing naturally led to man-made chlorine compounds being an early suspect. Besides, the ozone scientific community had been sensitized to halogens. However, while the chlorine-induced ozone-depletion theory does indeed predict that larger losses should occur at the high latitudes, the "ozone hole" is of a far greater magnitude than the few percent predicted for 1986 by the current global theory (c.f., Figures 4 and 9). In addition, no such ozone loss was occurring over the northern polar latitudes (Figure 7). The key parameter in a chlorine-induced ozone depletion is the

fraction of stratospheric chlorine that is in a reactive form; the conventional global theory simply predicts too small a fraction to yield an ozone loss the size of the observed "ozone hole".

However, it was recognized that some of the unique physical conditions of the Antarctic stratosphere may cause a substantially higher fraction of reactive chlorine to exist there. Namely, the Antarctic stratosphere is the coldest of any region on the planet. Indeed, polar stratospheric clouds are present frequently in the Antarctic winter, much more so than in the Arctic. Laboratory studies have shown that the reactive form of chlorine can be liberated by chemical reactions that occur on surfaces, such as cloud particles.

Therefore, while there is a variety of differing details, the halogen theories of the "ozone hole" propose that the chemical reactions on the surfaces of polar stratospheric clouds "accelerate" the halogen-ozone chemistry compared to that elsewhere. The photochemical ozone loss is predicted to begin with the arrival of sunlight to the Antarctic region. With further warming, transport from ozone-richer northern latitudes later fills in the "hole". Since these theories tag man-made halogens as the cause of the ozone decrease, they predict that the "ozone hole" will deepen, on the average, with increasing stratospheric chlorine abundances.

Natural nitrogen compounds

It is well known that nitrogen compounds also catalytically deplete ozone. Indeed, such chemistry is part of the natural ozone balance in the stratosphere. Furthermore, it was the possible depletion of the ozone layer by enhanced reactive nitrogen compounds from the envisaged supersonic aircraft fleets that first brought the stratosphere to the attention of chemists in the early 1970's. Natural small-scale perturbations of the stratospheric nitrogen-ozone chemistry have been studied, confirming the chemical relations between reactive nitrogen compounds and ozone.

The ozone hole has been proposed to be the result of an unusually large and growing perturbation of the nitrogen chemistry over Antarctica. Specifically, it is suggested that large amounts of reactive nitrogen compounds were created in the highest parts of the atmosphere by the relatively intense solar activity occurring around 1980 and that these compounds were transported downwards to the upper stratosphere during the polar night. It was pointed out that such long-range and downward transport may be particularly effective for the cold, isolated Antarctic region. Hence, the arrival of sunlight in Antarctic spring would find a large abundance of reactive nitrogen compounds. A substantial photochemical loss of ozone in the upper stratosphere would commence, which would later be replenished, as noted above, by warming-induced mixing of ozone-rich air from more-northerly latitudes.

Therefore, while this theory employs chemistry, the chemicals are natural, and human influences are not implicated. Furthermore, since the driving force is periodic solar activity, this theory predicts that the currently occurring "ozone hole" is apt to diminish.

2.2 Dynamical Theories

As noted above, it is generally agreed that transport processes terminate the seasonal "ozone hole" phenomenon. However, a number of theories propose that dynamical processes may also explain its formation. Several such processes have been identified.

Because the Antarctic region gets exceedingly cold during the winter, the return of sunlight in the austral spring and the resultant warming may induce a net upward air motion, which would carry the ozone-poor air of the lower atmosphere (see Figure 1) into the lower stratosphere, which would yield a reduced column of ozone. The recent trend toward lower column ozone values is explained by possible increased heating associated with higher aerosol abundances arising from the recent volcanic eruptions (see Figure 8).

Larger-scale transport processes also have been proposed as causes of the lower ozone over Antarctica. In particular, the reduction of wintertime planetary wave activity in the southern hemisphere has been noted, which would lead to reduced downward transport of ozone into Antarctica during winter and spring. It was also noted that this would imply a colder region, which would enhance springtime upward air motions, and delay the final warmings. Similar transport processes have been proposed as being responsible for the larger-than-usual ozone levels just northward of the Antarctic region reported by satellites, i.e., a result of redistribution.

Therefore, the dynamical theories of the "ozone hole" recognize that large- and small-scale transport processes undergo natural variation on the scale of years to decades and propose that such "climatogical" changes can result in a gradual redistribution of ozone in the Southern Hemisphere, resulting in lower values in Antarctica. Such a cause is a natural ephemeral process that could well reverse in the future, leading to a return of the Antarctic ozone patterns of past decades.

 Halogen-chemical, sunspot-induced-chemical, and dynamical/climatechange theories have been proposed as causes of the "ozone hole". The first evokes man-made perturbations; the latter two evoke natural processes.

2.3 Chemical, Radiative, and Dynamical Couplings

As noted in Section II.B.3.3, there are well-known couplings between ozone, temperature, and circulation, e.g., absorption of radiation by ozone induces heating, temperature gradients cause air motion, and transport processes redistribute ozone. Thus, cause and effect are often difficult to separate. Nevertheless, correlations can be instructive in establishing the scope of the phenomenon.

The dynamical theories for the "ozone hole" generally imply that a cooling of 15 to 20°C should be associated with the long-term Antarctic ozone decrease if transport processes were responsible. However, if chemical processes were solely responsible for the phenomenon, the decrease in ozone, which is the principal heat source in the stratosphere, would also cause a cooling, albeit smaller. In addition, lower temperatures would imply more polar strato-

spheric clouds, which introduce another temperature-chemistry coupling. Hence, temperature trends have been sought in several Antarctic-ozone studies.

Unfortunately, the current results are not fully consistent. One investigation reported about 20°C changes, others claim that only 5-10°C changes have occurred. The data sets include those from both satellite measurements, as well as from balloon-borne radiosondes. The differences in these early analyses no doubt reflect, in part, the dearth of sounding sites in Antarctica, the understandable difficulty of making balloon soundings under those conditions, and the relatively short period of satellite observations.

The paucity of chemical and physical data at the time the theories
of the "ozone hole" were proposed prevented the assessment of which
one, ones, or if any were correct.

3. 1986 Ground-based Chemical Observations

The National Ozone Expedition (NOZE) was at McMurdo Station, Antarctica, for the occurrence of the 1986 "ozone hole". Four teams focused a variety of ground-based apparatus and balloon-borne instruments on the chemical and aerosol characteristics of the phenomenon. Their observations that are reported thus far are the following:

3.1 Ozone

The ozone hole is a strongly seasonal phenomenon. The NOZE expedition did a great deal to clarify the understanding of its seasonal nature. The expedition arrived in Antarctica in late August to find total ozone levels and vertical profiles that were rather normal for that latitude and time of year. In the next several weeks, they witnessed about a 35% decrease of the total ozone overhead. Therefore, in addition to the strong evidence provided by the historical data extending back to the 1960's and 1950's showing a recent downward trend in Antarctic ozone, the behavior observed in a single spring season showed the growth of the hole very clearly.

Observations were made of the ozone abundance with two independent ground-based techniques, as well as from balloons. The excellent agreement of these data and the different principles involved in each measurement confirm that the change cannot be due to instrumental artifacts (such as volcanic or other natural contaminants).

In addition to confirming the decrease in column ozone, the balloon-borne measurements also revealed the changes that occurred in the vertical distribution of ozone. These data demonstrated that the ozone decrease occurred primarily in the \underline{lower} stratosphere, 12 to 22 km, often in sharply defined layers.

3.2 Nitrogen Compounds

Nitrogen dioxide was measured with two independent techniques. The observed abundances of nitrogen dioxide were only about one third as large as Arctic observations or that predicted by "standard" global atmospheric theoretical models discussed in Section II.B above. The nitrogen dioxide abundances also did not change with time of day as would be expected and as has been ob-

served at other latitudes. These observations demonstrate that the nitrogen photochemistry of the Antarctic stratosphere is anomalous.

3.3 Chlorine Compounds

Chlorine monoxide is closely tied to the possible depletion of Antarctic ozone due to halocarbons. The abundance of chlorine monoxide was established near 20 km. These data suggested that this compound was present in the Antarctic stratosphere in larger quantities than observed elsewhere. A related molecule is chlorine dioxide. This molecule was measured and found to be present at extremely high levels. The observed amount of chlorine dioxide was about 20-50 times greater than that occurring elsewhere or predicted by the "standard" global models, clearly demonstrating that the chlorine chemistry in the Antarctic stratosphere is also perturbed.

3.4 Implications

The observed chlorine and nitrogen perturbations are roughly consistent with the theory that the "ozone hole" is due to the chlorinated and brominated compounds. However, the uncertainties in the abundance of other chlorine and bromine species and in the chemical reactions that link the observed chlorine species to ozone depletions preclude, at present, treating the observations as proof of the halogenated chemical theory. The low nitrogen abundance and the occurrence of the ozone loss in the lower, rather than upper, stratosphere does make the solar-cycle theory more difficult to accept. The long-term record of meteorological variables continues to suggest a role of dynamical processes in the "ozone hole". Thus, while the role of the chlorinated and brominated compounds seems now somewhat more likely and that of the solar cycle seems less likely, the detailed cause of the "ozone hole" has not yet been established with certainty. Until the cause is better established, the question as to the significance to global ozone cannot be answered.

- Measurements made on the 1986 National Ozone Expedition to Antarctica revealed that the abundance of the reactive chlorine compounds is highly elevated compared to other regions of the globe. Remaining uncertainties, however, prevent unequivocal identification of CFC's as the cause of the "ozone hole". Until the cause is established, the implications of the Antarctic ozone losses for global ozone are not known.
- Because the current theories of the cause of the "ozone hole" require the unique physical features of the region, the failure of the "standdard" global ozone-depletion theories to predict its occurrence does not necessarily imply that these theories have major shortcomings regarding their predictions in other regions.

4. NOAA Future Research

The fact that the chlorine chemistry in the Antarctic stratosphere is now $\frac{\text{known}}{\text{processes}}$ to be highly perturbed and the continuing need to define the dynamical processes make it mandatory to understand their roles in the "ozone hole" in much more detail. Hence, NOAA plans a greatly expanded effort during 1987. First, the agency's scientists will take an improved instrument back to NSF's

McMurdo Base. As in 1986, one of these NOAA scientists will be the leader of the expedition. Others will provide year-round ozonesonde balloon launches from the South Pole during 1987. In addition, NOAA will join NASA, NSF, CMA, and university investigators in an aircraft expedition to probe the chemistry and dynamics at the stratospheric altitudes where the ozone loss is occurring. NOAA scientists will have five chemical instruments on board NASA's ER-2 and DC-8 aircraft, and one of these investigators will serve as mission scientist. NOAA is also helping to provide satellite ozone data that will locate the "ozone hole" for flight planning and define its shape.

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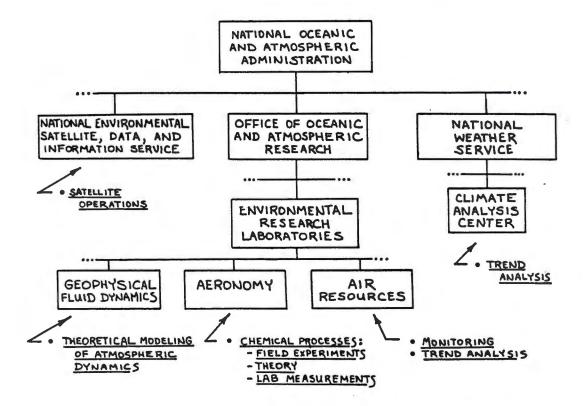
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V. NOAA ORGANIZATIONAL UNITS ENGAGED IN STRATOSPHERIC OZONE RESEARCH



VI. AUTHORS

This summary was prepared by the following authors with the substantial help of numerous of their NOAA colleagues:

Daniel L. Albritton	Aeronomy Laboratory, Environmental Research Laboratories
James K. Angell	Air Resources Laboratory, Environmental Research Laboratories
Jerry D. Mahlman	Geophysical Fluids Dynamics Laboratory, Environmental Research Laboratories
Alvin J. Miller	Climate Analysis Center, National Weather Service
James T. Peterson	Air Resources Laboratories (Geophysical Monitoring for Climate Change), Environmental Research Laboratories
Walter G. Planet	Office of Research and Applications, National Environmental Satellite, Data, and Information Service

VII. ACKNOWLEDGEMENTS

The initial version of this report was prepared in conjunction with the hearings of the United States Senate Committee on Environment and Public Works 12-13 May, 1986. Constructive comments on that version were given by Mack McFarland, Rafe Pomerance, and Steve Shimberg. In preparing related material and presentations over the last several months, valuable input and advice were provided by John Hoffman, Richard Johnson, Jim Margitan, J. R. Spradley, and Bob Watson.