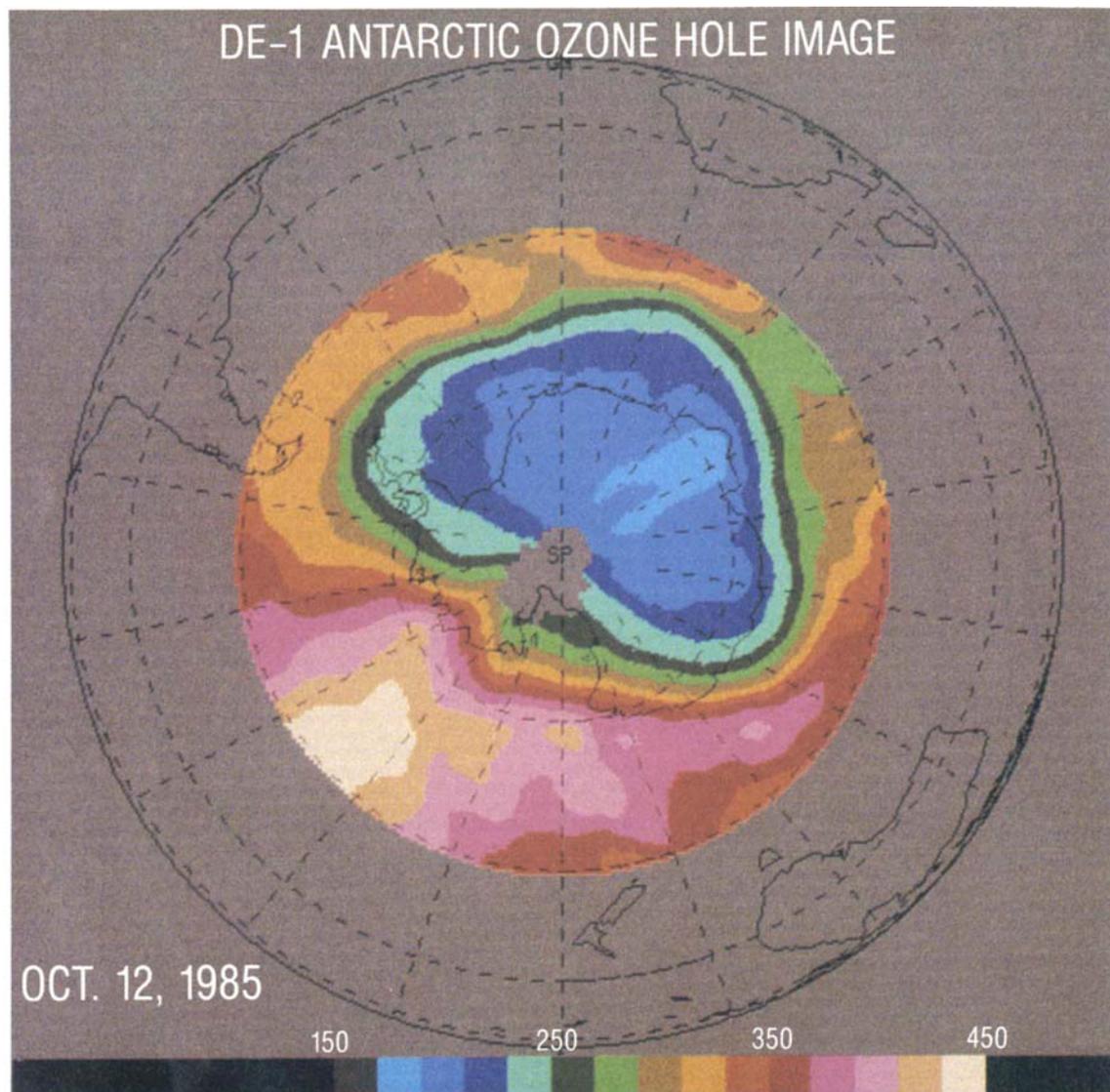


Total column ozone abundance (in Dobson units) from the South Pole measured on October 12, 1985, by the Spin-Scan Ozone Imager aboard the Dynamics Explorer I satellite. The large three-sided contours over Antarctica show the extraordinarily low levels of ozone that began occurring in the 1980s. This springtime phenomenon has been called the "ozone hole." (Image provided by G. M. Keating of the National Aeronautics and Space Administration Langley Research Center in Hampton, Virginia.)



The Endangered Ozone Layer

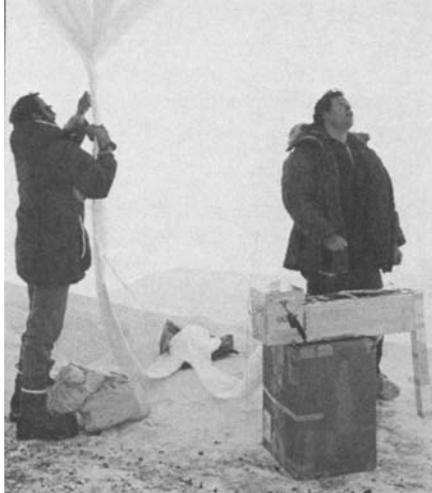
By Guy Brasseur

Depletion of the ozone layer has been much in the news over the last decade. Most recent discussion has centered on the shrinking amount of stratospheric ozone overlying Antarctica. Study of ozone, however, is not new (see box on page 8). But now, at the end of the 20th century, attention is focusing on the possible role of human activity in the perturbation of the ozone layer.

Although the concentration of ozone in the stratosphere is only on the order of a few parts per million (ppm), this molecule absorbs a significant amount of solar ultraviolet radiation and therefore protects the biosphere from several harmful effects. A partial destruction of the ozone layer by, for example, anthropogenic chemical compounds would undoubtedly have important human and environmental consequences—skin cancers, cataract problems, DNA damage, changes in the immune system, modifications in the life cycle of certain marine organisms, drop in the growth rate of several crop varieties, and other effects.

Absorption of solar energy by ozone also leads to significant heating of the atmosphere and is responsible for the temperature maximum (about 270 Kelvin) observed at an altitude of about 50 kilometers (km). The existence of the stratosphere—the atmospheric layer between approximately 12 and 50 km in altitude—is thus a direct consequence of the presence of ozone. Besides its interaction with solar ultraviolet radiation, ozone also emits and absorbs infrared terrestrial radiation and thus influences, to a certain extent, the radiative and meteorological processes that determine climate.

Most ozone is contained in the stratosphere. Smaller amounts also occur in the troposphere (the lowest 12 km of the



NSF/JIM HERPOLSHEIMER

The report in 1985 of a springtime "ozone hole" in the stratosphere above Antarctica stimulated new programs of research on atmospheric chemistry and dynamics. It also triggered widespread popular speculation as to the causes and effects of whatever processes accounted for the hole. This article reviews the present state of knowledge, the major uncertainties about what is happening, and the array of hypotheses put forward to explain these events and their implications for the future.

atmosphere) and are partially controlled by complex photochemical mechanisms related to atmospheric pollutants, topics not considered in this article.

Observations of Ozone

Because of its importance to the global environment and the threat of its gradual destruction, ozone is being continuously monitored in different regions of the world. Ozone density is measured from the ground by optical methods, in situ by chemical ozone-sondes, and from space by satellite-borne instruments. Measurements of the total column abundance (the number of ozone molecules per unit of area above the Earth's surface) indicate that, during almost the entire year, the amount of atmospheric ozone generally increases with latitude, reaching a maximum in early spring.¹ The morphology of the total ozone column is significantly different in the two hemispheres (see Figure 1 on page 9): the springtime ozone maximum is found near the pole in the Northern Hemisphere and near 50 or 60 degrees latitude in the Southern Hemisphere.

These findings might seem surprising since photochemical theory suggests that ozone, produced by solar radiation (see box on page 10), should be more abundant in low and midlatitudes and during the summer season. This paradox can easily be resolved by considering the photochemical lifetime of odd oxygen. This lifetime, which is on the order of a day in the upper stratosphere, increases downward to more than a year near the tropopause (the boundary between the troposphere and stratosphere). Thus, in the lower stratosphere, where the solar radiation that produces atomic oxygen and ozone is very weak, ozone behaves as a quasiconservative tracer

New Theories on Ozone Depletion

and is transported over very long distances before eventually being destroyed by chemical processes.

The transport of quasiconservative tracers (a quantity with weak sources or sinks) is complex, involving advection, diffusion, dissipation of large-scale planetary waves, breaking of gravity waves, and other processes. Both observational and theoretical studies have shown that in the lower stratosphere trace constituents or heat are transported by a large cell in both hemispheres: rising air motions over the tropics, poleward fluxes at midlatitudes, and downward motions over the pole. This circulation is stronger in winter, when planetary waves propagate and dissipate in the stratosphere, so that ozone is most efficiently transported during the winter season and the ozone column reaches its maximum in early spring.

The hemispheric difference in the ozone distribution is related to hemispheric differences in the dynamics of the stratosphere. Indeed, planetary waves, which transport tracers, are produced in the troposphere by the largest mountain ranges and continental or oceanic heating or cooling. As there are few mountains in the Southern Hemisphere, planetary wave activity is relatively weaker in the austral than in the boreal regions. Hence the poleward transport of heat and ozone differs between hemispheres: the temperature and the ozone density are significantly lower over the South Pole in September than over the North Pole in March.

Observations of ozone by satellite also show the vertical and meridional distribution of the density of this gas.² A comparison between these data and model calculations, based on our current knowledge of chemical and photochemical processes and on a detailed treatment of radiative and dynamical processes occurring in the stratosphere, shows good agreement except in the up-

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A CENTURY OF OZONE RESEARCH

The word "ozone" itself (from the Greek *ozein*, to smell) was introduced by Schoenbein in 1840 to characterize a chemical species with a pronounced odor.¹ Schoenbein, however, was not able to identify either the chemical structure of this gas nor its origin. Five years later de la Rive deduced from laboratory work that ozone was produced from oxygen,² and Soret in the 1860s found that the ozone molecule was formed by three oxygen atoms (O_3).³

Although Schoenbein had given the first indication of the presence of ozone in air, the existence of this gas in the atmosphere was established by Houzeau, who found that its concentration was linked to atmospheric conditions.⁴ Systematic observations of ozone at the Earth's surface were then made for about 30 years by Lévy at the Parc Montsouris in Paris.⁵ The data, however, were significantly altered by the presence of other oxidants in the atmosphere. Optical absorption prop-

erties of ozone were studied by Chapuis as early as 1880,⁶ by Hartley in 1881,⁷ and by Huggins and Huggins in 1890.⁸ After World War I scientists such as Fabry and Buisson in France,⁹ Cabannes and Dufay in California,¹⁰ Chalonge and Goetz in Switzerland,¹¹ and Dobson in England¹² used the data obtained from these spectroscopic studies to deduce ozone abundances by measuring the absorption of solar ultraviolet radiation. Dobson and his colleagues noted that the column abundance of ozone was directly linked to weather patterns: a maximum was clearly visible in low-pressure cells and a minimum was associated with high-pressure events. Furthermore, an unexpected seasonal variation was observed: the largest values were measured in spring and the lowest values in fall. Since these times, there has been considerable work done to understand the chemical and dynamical processes governing the distribution of ozone.

1. C. F. Schoenbein, "Recherches sur la nature de l'odeur qui se manifeste dans certaines actions chimiques," *Comptes Rendus de l'Académie des Sciences Paris* 10(1840):706.

2. M. de la Rive, "Sur l'ozone," *Comptes Rendus de l'Académie des Sciences Paris* 20(1845):20.

3. J. L. Soret, "Sur les relations volumétriques de l'ozone," *Comptes Rendus de l'Académie des Sciences Paris* 57(1863):604; J. L. Soret, "Recherches sur la densité de l'ozone," *Annales de Chimie et de Physique*, Paris, 4e série, 7(1866):113 and 13(1868):257.

4. A. Houzeau, "Méthode analytique pour reconnaître et doser l'oxygène naissant," *Comptes Rendus de l'Académie des Sciences Paris* 45(1857):873; A. Houzeau, "Sur le proportion d'ozone contenue dans l'air de la campagne et sur son origine," *Comptes Rendus de l'Académie des Sciences Paris* 74(1872):712.

5. A. Lévy, H. Henriet, and M. Bouyssi, "L'ozone atmosphérique," *Annales de l'Observatoire Municipal Montsouris* 6(1905):18.

6. J. Chappuis, "Sur le spectre d'absorption de l'ozone," *Comptes Rendus de l'Académie des Sciences Paris* 91(1880):985.

7. W. N. Hartley, "On the Absorption Spectrum of Ozone," *Chemical Society Journal* 39(1881):57.

8. W. Huggins and Mrs. Huggins, "On a New

Group of Lines in the Photographic Spectrum of Sirius," *Proceedings of the Royal Society of London* 48(1890):216.

9. C. Fabry and H. Buisson, "Sur l'absorption de l'ultraviolet par l'ozone et l'extrémité du spectre solaire," *Comptes Rendus de l'Académie des Sciences Paris* 156(1913):782; C. Fabry and H. Buisson, "A Study of the Ultraviolet End of the Solar Spectrum," *Astrophysical Journal* 54(1921):297.

10. J. Cabannes and J. Dufay, "Mesure de l'altitude de la couche d'ozone dans l'atmosphère," *Comptes Rendus de l'Académie des Sciences Paris* 181(1925):302; J. Cabannes and J. Dufay, "Les variations de la quantité d'ozone contenue dans l'atmosphère," *Journal de Physique et le Radium* 6e série, 8(1927):353.

11. D. Chalonge and F. W. P. Goetz, "Mesures diurnes et nocturnes de la quantité d'ozone contenue dans la haute atmosphère," *Comptes Rendus de l'Académie des Sciences Paris* 189(1929):704.

12. G. M. B. Dobson, "Atmospheric Ozone," *Gerlands Beitrage zur Geophysik* 24(1929):8; see also a series of papers published by Dobson and colleagues entitled "Measurements of the Amount of Ozone in the Earth's Atmosphere and Its Relation to Other Geophysical Conditions," *Proceedings of the Royal Society of London* (1926 to 1930).

per stratosphere and mesosphere, where models tend to underestimate the ozone density by 20 to 30 percent. This discrepancy between theory and observations suggests either errors in some of the currently accepted values of reaction rates or undiscovered chemical and photochemical processes that play an important role in the upper stratosphere and in the mesosphere. This uncertainty limits the accuracy of model predictions of future changes in the ozone amount.

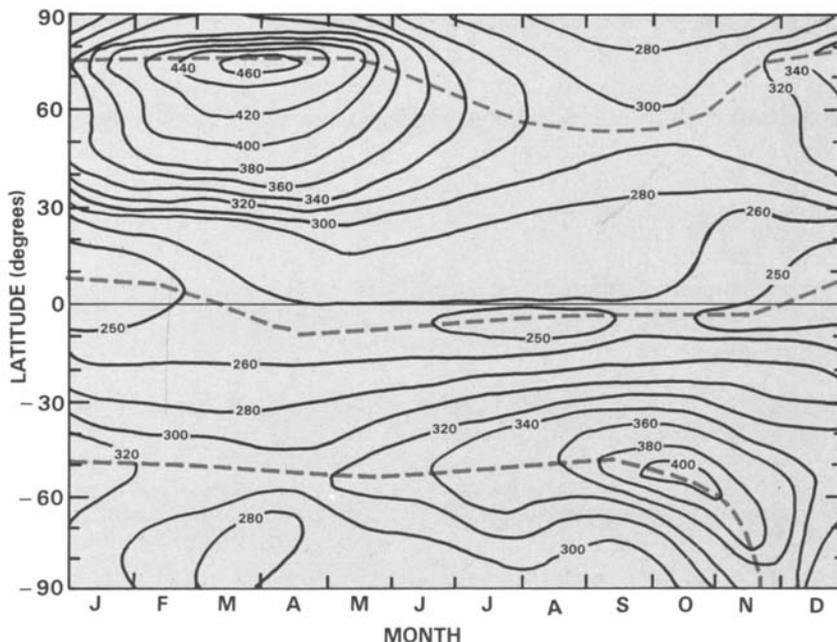
Perturbation of the Ozone Layer

Several chemical compounds resulting from industrial or agricultural processes, such as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), or the chlorofluorocarbons (CFCs), can affect the ozone layer with possible environmental and climatic consequences (see box on page 40). The atmosphere's response to increasing concentrations of these chemicals has been predicted by numerical models.

A report recently published by the World Meteorological Organization in Geneva, Switzerland, indicates that if the emission of CFCs into the atmosphere remains at its present level (about 650 kilotons per year), the average ozone column will be depleted and after more than a century will decrease to values 5 to 9 percent lower than at present.³ If the release of these chemicals continues to increase at a rate of about 3 percent per year, a dramatic reduction of the ozone column (of more than 10 percent) is expected as early as the year 2020. In these calculations, however, the atmospheric concentrations of other gases having potential effects on ozone and temperature were assumed to remain constant at their present values. In fact, concentrations of carbon dioxide, methane, and nitrous oxide are increasing at rates of approximately 0.5 percent per year, 1 percent per year, and 0.25 percent per year respectively.

Carbon dioxide has no direct chemical effect on ozone, but helps control the heat budget of the stratosphere and of the troposphere. As the ozone destruction rate depends on temperature in the middle and upper stratosphere,

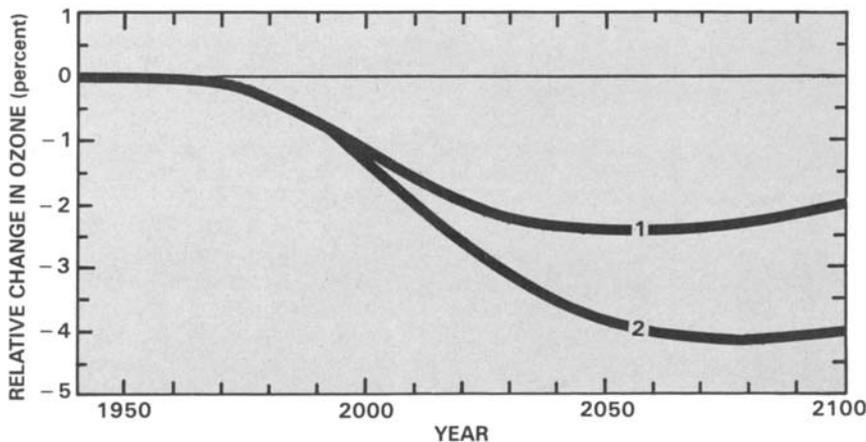
FIGURE 1. Ozone column abundance (Dobson units) as a function of latitude and month.



Note: The minimum and hemispheric maximums are shown in color.

SOURCE: J. London, "Radiative Energy Sources and Sinks in the Stratosphere and Mesosphere," in M. Nicolet and A. C. Aikin, eds., *Proceedings of the NATO Advanced Study Institute on Atmospheric Ozone: Its Variation and Human Influences* (Washington, D.C.: U.S. Department of Transportation, 1980), 703-21.

FIGURE 2. Change in the ozone column abundance relative to 1940 predicted by a chemical-radiative-transport one-dimensional model.



In these calculations, the density of CO_2 , CH_4 , and N_2O is assumed to increase by 0.5, 1.0, and 0.25 percent per year respectively. Trends in CFC-11, CFC-12, and CFC-113 are based on historical emissions until 1985 and on prescribed scenarios for the future. In case 1, a constant emission at the 1985 level is assumed for CFC-11 and CFC-12. In case 2, a 3 percent per year growth, with a capacity cap of 1.5 times the present level, is specified for CFC-11 and CFC-12. In both cases, the emission of CFC-113 is assumed to increase by 6 percent per year with an upper limit equal to the production of CFC-11.

an increase in carbon dioxide would slightly increase the ozone density in the 30- to 60-km region. More methane should result in larger ozone concentrations in the troposphere, especially in regions where the amount of nitrogen oxides is relatively high. Furthermore, methane in the stratosphere accelerates the conversion of active chlorine (Cl, ClO) into inactive chlorine (HCl), and therefore reduces the action of the

CFCs on ozone. Finally, more nitrous oxide will increase the amount of active nitrogen oxides in the stratosphere and consequently reduce the ozone density in this layer.

With all these effects simultaneously taken into account, the globally averaged ozone depletion is calculated to be less than 5 percent if the production of the CFCs remains at its present level. A model prediction of the change in the

ozone column abundance relative to the year 1940 has been developed for two prescribed scenarios of future changes in the emission of the perturbing gases (see Figure 2 on page 9). In case 1, the production of CFC-11 and CFC-12 is assumed to remain constant after the year 1985; in case 2, production is assumed to increase by 3 percent per year until a hypothetical capacity cap of 1.5 times the 1985 production level is

OZONE CHEMISTRY

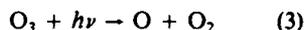
In 1930 Chapman suggested that the production of ozone in the stratosphere could result from the dissociation of molecular oxygen by solar ultraviolet radiation of wavelengths shorter than 242.4 nanometers:¹



He noted that once split the two oxygen atoms should immediately recombine with O₂ in the presence of a third body (M = N₂ or O₂), required to maintain the momentum of the colliding particles and to remove the energy released by the reaction. Thus ozone (O₃) is formed by:



The ozone is rapidly dissociated by solar radiation:



forming molecular (O₂) and atomic (O) oxygen. The O atom, however, recombines rapidly to reproduce an ozone molecule, so that process (3) is not a net loss of odd oxygen (O + O₃), but controls, together with reaction (2), the partitioning between atomic oxygen and ozone. In fact, the net destruction of odd oxygen occurs through a recombination process, also suggested by Chapman:



This reaction, however, is too slow to account for the ozone density observed in the stratosphere and in the mesosphere (50 to 85 kilometers in altitude). In the early 1950s Bates and Nicolet proposed an additional destruction process that partially resolves this discrepancy.² They suggested that odd oxygen is destroyed

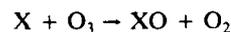
more efficiently in the mesosphere by radicals such as H, OH, and HO₂, which are produced by oxidation or photodestruction of water vapor and methane. In 1964 Hampson showed that several reactions involving these hydrogen radicals could also take place in the stratosphere.³ Even with these additional reactions, the chemical theory still predicted ozone densities larger than observed.

In the early 1970s Crutzen suggested that stratospheric ozone is destroyed rapidly by the nitrogen oxides (NO_x).⁴ Nitric oxide (NO) and subsequently nitrogen dioxide (NO₂), nitric acid (HNO₃), and other forms of odd nitrogen result from the oxidation in the stratosphere of nitrous oxide (N₂O) (see box on page 40). Nitrogen oxides are also directly produced by combustion processes, which essentially affect the lower troposphere. In addition, a small fraction of these compounds are released in the upper troposphere and lower stratosphere by commercial and military aircraft and are expected to affect the ozone layer.⁵

In 1974 Stolarski and Cicerone showed that chlorine could also deplete odd oxygen.⁶ Molina and Rowland found that chlorine was thus a real threat to ozone since the amount of this gas in the stratosphere was expected to increase from the continuous use of chlorofluorocarbons (CFCs).⁷ These chemical compounds, produced by industry at a rate of about 650 kilotons per year, are used for different applications. They can reach the middle and upper stratosphere where they are photolyzed and decomposed by the intense ultraviolet radiation at these altitudes. Active chlorine in the form of Cl or ClO is produced by this decomposition of the CFCs. Other

species belonging to the odd chlorine family such as HCl, HOCl, and ClONO₂ are also involved.

Reactive species of hydrogen, nitrogen, chlorine and bromine catalyze the recombination of ozone and atomic oxygen (reaction 4) through, for example, the following cycle:



where X can be H, OH, NO, Cl, or Br. Because radical X is constantly reproduced, ozone may be very efficiently destroyed by such catalytical mechanisms, even if the concentration of X is several orders of magnitude smaller than that of ozone.

1. S. Chapman, "A Theory of Upper Atmospheric Ozone," *Memoirs of the Royal Meteorological Society* 3(1930):103.
2. D. R. Bates and M. Nicolet, "The Photochemistry of Water Vapor," *Journal of Geophysical Research* 55(1950):301.
3. J. Hampson, "Photochemical Behavior of the Ozone Layer," *Technical Note 1627* (Canadian Armament Research and Development Establishment, Quebec, 1964).
4. P. J. Crutzen, "The Influence of Nitrogen Oxide on the Atmospheric Ozone Content," *Quarterly Journal of the Royal Meteorological Society* 96(1970):320.
5. H. S. Johnston, "Reduction of Stratospheric Ozone by Nitrogen Oxide Catalysts from Supersonic Transport Exhaust," *Science* 173(1971):517; Earl W. Barrett, *Final Report to Climatic Impact Assessment Program, DOT-TST-76-23* (Washington, D.C.: Office of the Secretary of Transportation, 1975).
6. R. S. Stolarski and R. J. Cicerone, "Stratospheric Chlorine: A Possible Sink for Ozone," *Canadian Journal of Chemistry* 52(1974):1610.
7. M. J. Molina and F. S. Rowland, "Stratospheric Sink for Chlorofluoromethanes: Chlorine Atom Catalyzed Destruction of Ozone," *Nature* 249(1974):810.

reached. In both cases the model predicts a decrease in the global amount of ozone until the second half of the 21st century, essentially as a result of an increasing amount of chlorine in the stratosphere. The slow recovery predicted after the 2050-to-2080 time frame results from the assumed rapid growth in methane concentration.

Despite the limited changes calculated for the ozone column abundance, large variations are predicted for local ozone density and temperature. Ozone reductions of 60 to 75 percent, or temperature reductions of 25 to 30 K, should be expected by 2100 near 40 or 50 km in altitude (see Figures 3 and 4 on this page). An increase of tropospheric ozone, as well as a warming of the Earth's surface, is also predicted in the future. A comprehensive study of the greenhouse effects and the climatic impact of chemical compounds released into the atmosphere has been recently completed at the National Center for Atmospheric Research (NCAR) in Boulder, Colorado, by V. R. Ramanathan and several of his colleagues and by R. E. Dickinson and R. J. Cicerone.⁴ The impact of large changes in stratospheric ozone and temperature on the general circulation and the possible effects on the troposphere are not yet fully understood, as such a study requires complex multidimensional models with fully coupled dynamics, radiation, and chemistry. Such comprehensive models should become available with the next generation of high-speed computers.

Recent calculations have indicated that the response of ozone to anthropogenic perturbations will be dependent on latitude and season.⁵ The depletion of the ozone column, as calculated by a two-dimensional chemical-radiative-dynamical model, is indeed larger, by a factor of 2 to 3, in the polar regions than it is in the tropics, with a maximum effect in the spring (see Figure 5 on page 41).

Besides these predicted long-term changes, shorter-term variations also affect the ozone density. Most of these are related to rapid dynamical (meteor-

(continued on page 39)

FIGURE 3. Variation relative to 1940 of ozone density calculated as a function of altitude for years 2000, 2040, and 2100.

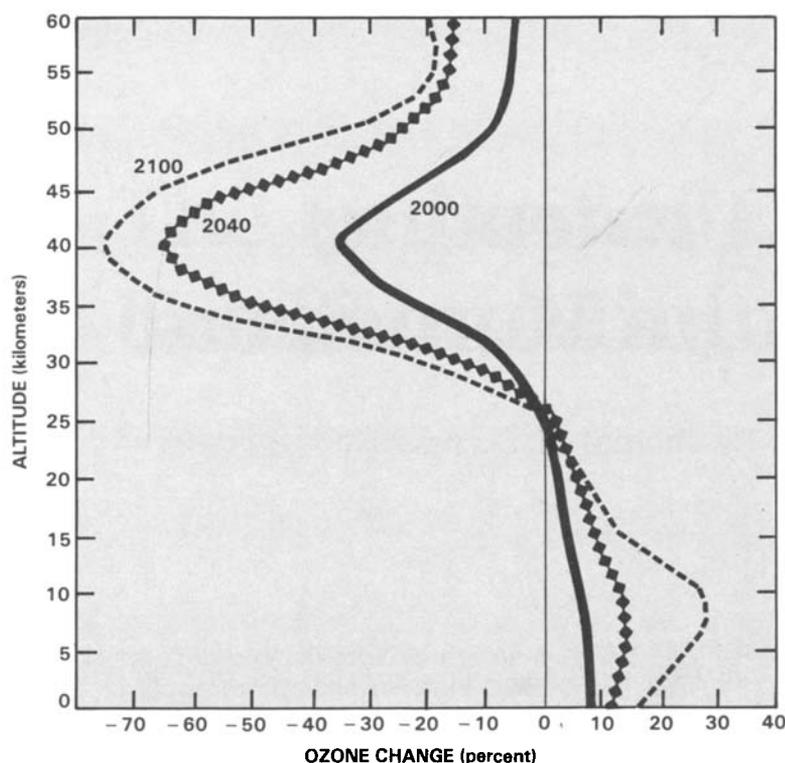
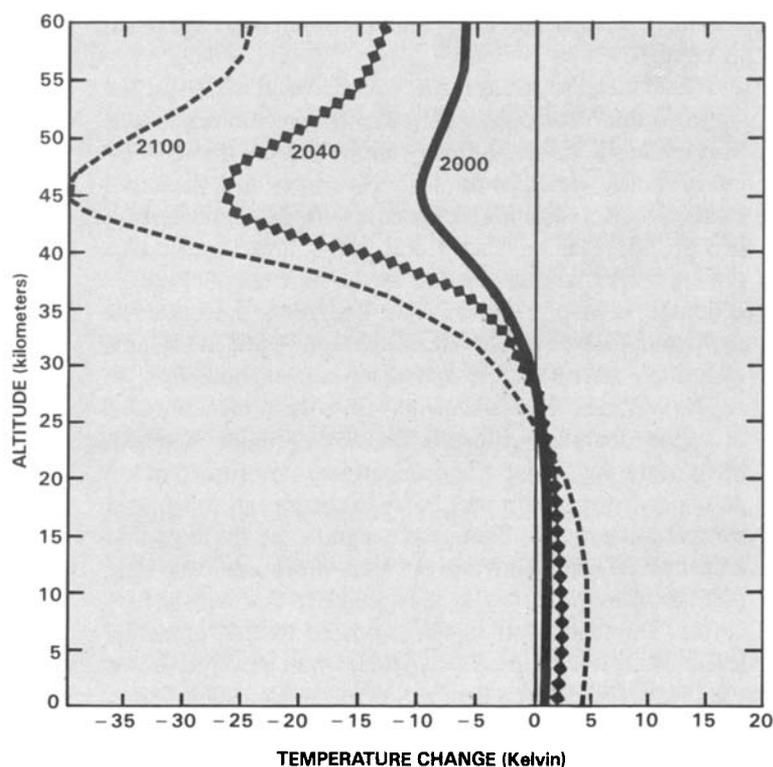


FIGURE 4. Variation relative to 1940 of temperature calculated as a function of altitude for years 2000, 2040, and 2100.



significantly elevated levels of radon and its progeny in excess of the EPA-recommended target value, remedial action should be taken as soon as is reasonable to reduce the increased risk associated with the exposure of young children.¹⁸

NOTES

1. D. A. Holaday, D. E. Rushing, R. D. Coleman, P. F. Woolrich, H. L. Kusnetz, and W. F. Bale, "Control of Radon and Daughters in Uranium Mines and Calculations on Biologic Effects," Public Health Service Publication no. 494 (U.S. Department of Health, Education, and Welfare, Washington, D.C., 1957).
2. Colorado Bureau of Mines, "Colorado Mining Laws with Safety Rules and Regulations" (Colorado Department of Natural Resources, Division of Mines, Denver, 1962), 113; Staff Report of the Federal Radiation Council, *Guidance for the Control of Radiation Hazards in Uranium Mining*, Report no. 8 (Washington, D.C.: U.S. Government Printing Office, 1967); U.S. Mine Safety and Health Administration, 30 C.F.R. 57.5 (1986).
3. U.S. Congress, *Use of Uranium Mill Tailings for Construction Purposes*, Hearings conducted 28-29 October 1971, 91st Cong., 1st sess., 1971 (U.S. GPO, Stock no. 5270-1395).
4. Paul J. Peterson, Acting Surgeon General, "Surgeon General's Guidelines," Letter to Roy L. Cleere, Executive Director, Colorado Department of Health (Denver, 27 July 1970); 10 C.F.R. 712 (1970); the numeric value of 0.01 WL had as its origin the maximum permissible concentration of radon 222 as found in 10 C.F.R. Part 20 Table II, Column 1, Title 10, when adjusted for exposure of the general public.
5. U.S. General Accounting Office, *Controlling the Radiation Hazard from Uranium Mill Tailings*, RED-75-365 (Washington, D.C.: U.S. GAO, 21 May 1975).
6. Pub.L. 95-604.
7. U.S. Environmental Protection Agency, "Final Environmental Impact Statement for Remedial Action

Standards of Inactive Uranium Processing Sites, 40 C.F.R. (1986)," EPA 520/4-82-013-1 & 2 (Washington, D.C., 1982); 40 C.F.R. 192.

8. Allan C. B. Richardson, Chief, Guides and Criteria Branch, Office of Radiation Programs, EPA, "Radiation Protection Standards," Memorandum to Philip C. Nyberg, Health Physicist, Radiation Programs Branch, Region VIII, EPA (21 November 1985).
9. 10 C.F.R. 712 (1970); U.S. EPA, note 7 above; the procedures manual for GJRAP requires that the sampling procedure identify the annual average radon progeny concentration ± 50 percent at the 95 percent confidence level as required by 10 C.F.R. 712; see Colorado Department of Health, "Procedures Manual for the Grand Junction Remedial Action Program" (Colorado Department of Health, Radiation Control Division, Denver, 1986).
10. *State of Colorado v. Union Carbide Corporation and Umetco Minerals Corporation*, U.S. District Court, Colorado, Civil Action no. 83C2384; *State of Colorado v. Cotter Corporation*, U.S. District Court, Colorado, Civil Action no. 83C2389.
11. A Consent Decree was lodged with the U.S. District Court on 31 October 1986 in relation to the suit against Union Carbide and Umetco. The Consent Decree will not become final until completion of a public comment period and final approval by the court.
12. National Council on Radiation Protection and Measurements, "Exposures from the Uranium Series with Emphasis on Radon and Its Daughters," NCRP Report no. 77 (Bethesda, Md., 1984).
13. U.S. EPA, *A Citizen's Guide to Radon: What It Is and What To Do about It*, OPA-86-004 (Washington, D.C.: EPA, 1986).
14. Ibid; U.S. EPA, "Interim Indoor Radon and Radon Decay Product Measurement Protocols," EPA 520/1-86-04 (Washington, D.C., 1986).
15. Richard Guimond, EPA Office of Radiation Programs, Statement to the Office of Management and Budget (U.S. EPA, Washington, D.C., 1986).
16. "State of Colorado Indoor Radon Work Plan, November 1986," Draft (Colorado Department of Health, Denver, 1986).
17. Ibid.
18. U.S. EPA, note 13 above; National Council on Radiation Protection and Measurements, "Evaluation of Occupational and Environmental Exposures to Radon and Radon Daughters in the United States," NCRP Report no. 78 (Bethesda, Md., 1984).

Ozone

(continued from page 11)

ological) fluctuations and to annual, semiannual, and quasibiannual oscillations in the wind and temperature. Analyses of satellite data have also revealed that the ozone density in the upper stratosphere varies by about 1 percent over a solar rotation period of 27 days during which the solar irradiance at 205 nanometers (a spectral region contributing the most to photochemical ozone formation) varies typically by 2.5 percent.⁶ A solar-ozone relation has also been observed in the mesosphere. Extrapolation of such findings suggest that the ozone density near an altitude of 45 km should vary by 1 to 4 percent in response to the 11-year solar cycle. The amplitude of the corresponding modulation in the total ozone column should be roughly 1 percent.

Ozone Minimum Over Antarctica

The behavior of ozone over Antarctica in the austral spring (September to November) is very intriguing. Observations made from the ground and subsequently by satellite-borne instruments have revealed a dramatic decrease in total ozone in October, with the minimum column abundance getting smaller each year from 1979 to 1985. Before reviewing plausible explanations for this observed phenomenon, it might be useful to describe the appearance of the so-called ozone hole by differentiating between seasonal and year-to-year variations.

The seasonal change in the ozone column abundance has been dramatic since 1979. It is characterized by a decline during September and into October, with a recovery during November. The behavior of total ozone in a vertical column appears to be closely correlated with dynamical activity: low ozone amounts are found in regions where stratospheric geopotential (pressure) and temperature are low. In fact, during winter a very stable low-pressure system with very cold temperature builds up over the Antarctic conti-



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Bruce Barnham

TRACE GASES: POTENTIAL IMPACTS ON OZONE AND TEMPERATURE

A number of trace gases, partially or totally produced by human activity, are released into the atmosphere and contribute to the so-called greenhouse effect. They may also initiate several chemical reactions with potential effects on ozone abundance.

Carbon dioxide (CO₂), for example, is radiatively active, and thus plays a major role in the heat balance of the stratosphere. Continuous monitoring of the carbon dioxide concentration (presently on the order of 350 ppm) has revealed an increase on the order of 0.2 to 0.3 percent per year since 1958, probably resulting in large part from combustion processes (involving coal, oil, and natural gas) and land-use changes (from deforestation, for example).

Methane (CH₄) is the most abundant hydrocarbon in the atmosphere. A yearly increase in its concentration (presently about 1.7 ppm) is estimated to be on the order of 1 to 2 percent. Analyses of air bubbles trapped in polar ice cores suggest that the atmospheric concentration of methane has approximately doubled since the 17th century. This molecule initiates a complex set of chemical reactions that lead to the production of ozone in the troposphere (smog reactions) and that also interact with species belonging to other chemical families (such as chlorine).

Nitrous oxide (N₂O), which has a current surface concentration of 307 ppb, is the main source of nitrogen oxides in the stratosphere. It is produced by biological processes, mainly bacterial nitrification and denitrification, and by fossil-fuel and biomass combustion. Its growing concentration (about 0.2 to 0.3 percent per year) is believed to be largely associated with human activity. Analyses of air trapped

in ice cores from Antarctica suggest that the surface concentration of nitrous oxide might have been close to 285 ppb in the preindustrial era.

The chlorofluorocarbons (CFCs) are probably the species with the largest potential to destroy ozone in the near future. These organic molecules are used as aerosol propellants, refrigerants, foam blowing agents, and solvents. The species presently important for the ozone issue are essentially CFC-11 (CFC-11), CF₂Cl₂ (CFC-12), CCl₄ (carbon tetrachloride), CH₃CCl₃ (methyl chloroform), and to a lesser extent CF₂ClCFCl₂ (CFC-113), and CHClF₂ (CFC-22). There is also some concern that the halons 1211 (CF₂ClBr) and 1301 (CBrF₃), essentially used as fire-extinguishing material, could contribute to the destruction of ozone in the future. The global emissions of CFC-11 and CFC-12 increased by about 10 percent per year until 1974. Since then their release into the atmosphere has remained essentially at the same level (about 250 to 300 kilotons per year for CFC-11 and 350 to 400 kilotons per year for CFC-12). The emission of carbon tetrachloride is estimated to be fairly constant (on the order of 100 kilotons per year) while the release of methyl chloroform seems to be increasing by about 16 percent per year. A rapid growth is also evident for the emission of CFC-113.

These chemical compounds—together with methyl chloride (CH₃Cl), which is produced by the oceans, biomass burning, and fungi—are the major sources of active chlorine in the stratosphere. (For more on trace gases, see Jill Jäger, "Climatic Change: Floating New Evidence in the CO₂ Debate," *Environment*, September 1986.)

came from scientists of the British Antarctic Survey.⁷ Their data, taken over the Halley Bay Station near the Weddell Sea, clearly showed an ozone reduction of about 40 percent between October 1979 and October 1985 (see Figure 6 on page 42). Observations made since 1979 by the Total Ozone Mapping Spectrometer (TOMS) and by the Solar Backscatter Ultraviolet (SBUV) experiment, both on board the Nimbus 7 satellite, confirmed the findings of the British scientists.⁸ As noted by scientists from the National Aeronautics and Space Administration (NASA) in a recently published issue of *Geophysical Research Letters*, the region of low total ozone is roughly centered over the South Pole and is surrounded by a ring of maximum total ozone.⁹ The low-ozone region meanders around the pole in response to meteorological activity in the lower stratosphere. The monthly mean total ozone in October, which was on the order of 260 Dobson units (D.U.) in 1979, declined to about 150 D.U. in 1985 (the lowest amount found anywhere). A reduction of 70 D.U. (about 17 percent) was also found in the maximum ozone region near 55 degrees south latitude between 1979 and 1985. (One hundred D.U. correspond to a layer of pure ozone 1 millimeter thick at standard temperature and pressure.)

Observations of the vertical profile of ozone, made from balloon-borne instruments over the Japanese station of Syowa, Antarctica,¹⁰ and more recently over the U.S. station of McMurdo,¹¹ clearly indicate that most of the column ozone depletion is confined to a layer between 12 and 25 km in altitude. Analyses of satellite data tend to suggest that a modest decrease (up to about 15 percent) also takes place at higher altitudes.¹²

Chemical Theories

Several attempts have been made to explain the rapid decrease in the spring-time ozone column abundance since its discovery. The first hypothesis, already considered by the British scientists who discovered the trend, was that ozone could have been destroyed at a fast rate

ment and is surrounded by a belt of strong westerly winds, forming the so-called polar vortex. Because of weak planetary wave activity in the Southern Hemisphere, this vortex tends to isolate the polar cap region from lower latitude regions and to prevent ozone and heat from converging into polar regions. The ozone decrease (whose cause remains to be explained) comes to an end in November, when the low-pressure and

low-temperature cell is replaced by a high-pressure system with higher temperature. The time at which this "final warming event" takes place (during which strong transient dynamical processes produce significant meridional transport of ozone) appears to be important for understanding changes in the seasonal variation of ozone.

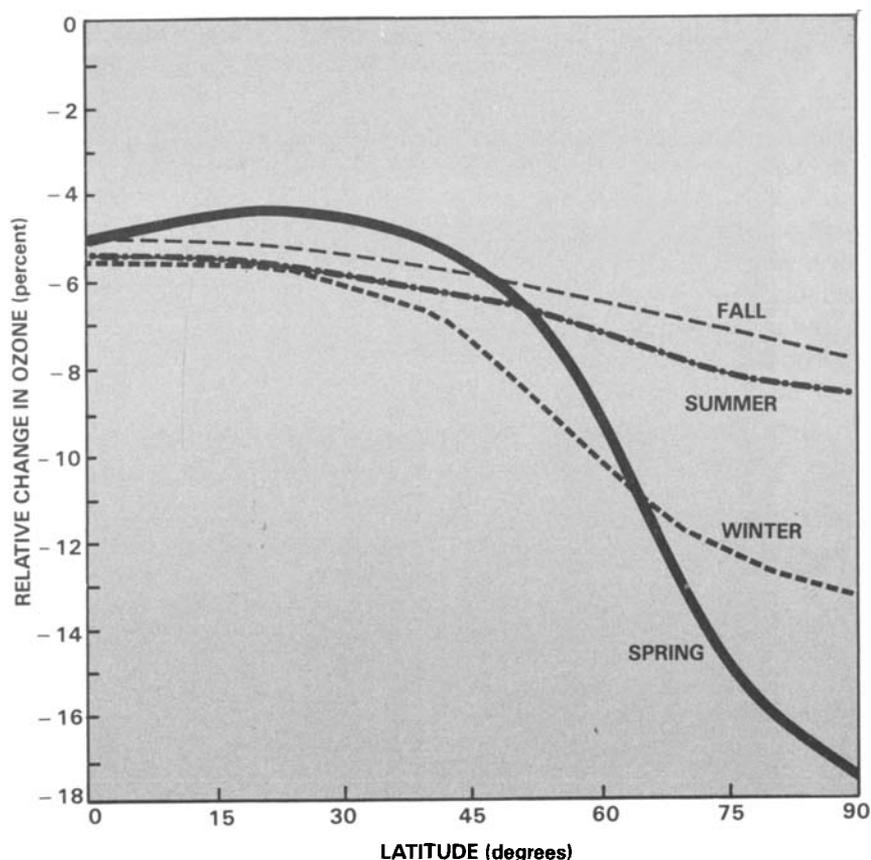
The first report of a secular (year-to-year) trend in ozone during October

as a result of increasing concentration of CFCs. Model calculations, however, using a chemical scheme with classical homogeneous gas phase chemistry, do not predict the large ozone depletions that are observed. Indeed, in the lower stratosphere, most chlorine atoms are tied up into inactive "reservoir" species, such as HCl or ClONO₂. If chlorine is responsible for the destruction of ozone in polar regions, these chemical reservoirs have to be destroyed in spring in order to assure that the concentration of active ClO becomes as large as about 1 part per billion (ppb) in the lower stratosphere. Furthermore, in order to avoid reformation of ClONO₂, the atmospheric content of nitrogen dioxide (NO₂) should be low.

S. Solomon from the National Oceanic and Atmospheric Administration (NOAA) in Boulder, R. R. Garcia from NCAR, I. S. Rowland from the University of California at Irvine, and D. J. Wuebbles from the Lawrence Livermore National Laboratory in Livermore, California,¹³ have suggested that the chlorine reservoirs (essentially ClONO₂ and HCl) as well as the nitrogen oxides (essentially in the form of N₂O₅ during the polar winter) are destroyed inside the polar stratospheric clouds that are observed over Antarctica during wintertime.¹⁴ P. J. Crutzen and F. Arnold in West Germany have noted that the disappearance of nitric acid (HNO₃) from the gas phase, after its condensation onto the ice particles in these polar stratospheric clouds, would lead to large increases in the density of OH radicals, which have the potential of destroying HCl to produce active chlorine (Cl, ClO).¹⁵ Moreover, the condensation of HNO₃ would lead to a drastic reduction of odd nitrogen in the gas phase.

M. B. McElroy and several of his colleagues at Harvard University have suggested that if active chlorine is indeed released in sufficiently large amounts and if nitrogen oxides are removed from the lower stratosphere, the reaction between chlorine and bromine oxides could explain the observed ozone trend (see box on page 43).¹⁶ According to this theory, the presence of ClO and

FIGURE 5. Reduction of the ozone column abundance as a function of latitude and for different seasons.



Note: The reduction is based on simulated simultaneous increases in the amount of Cl_x (from 0.82 to 9.51 ppb), CO₂ (from 316 to 413 ppm), CH₄ (from 1.32 to 2.46 ppm), and N₂O (from 292 to 331 ppb).

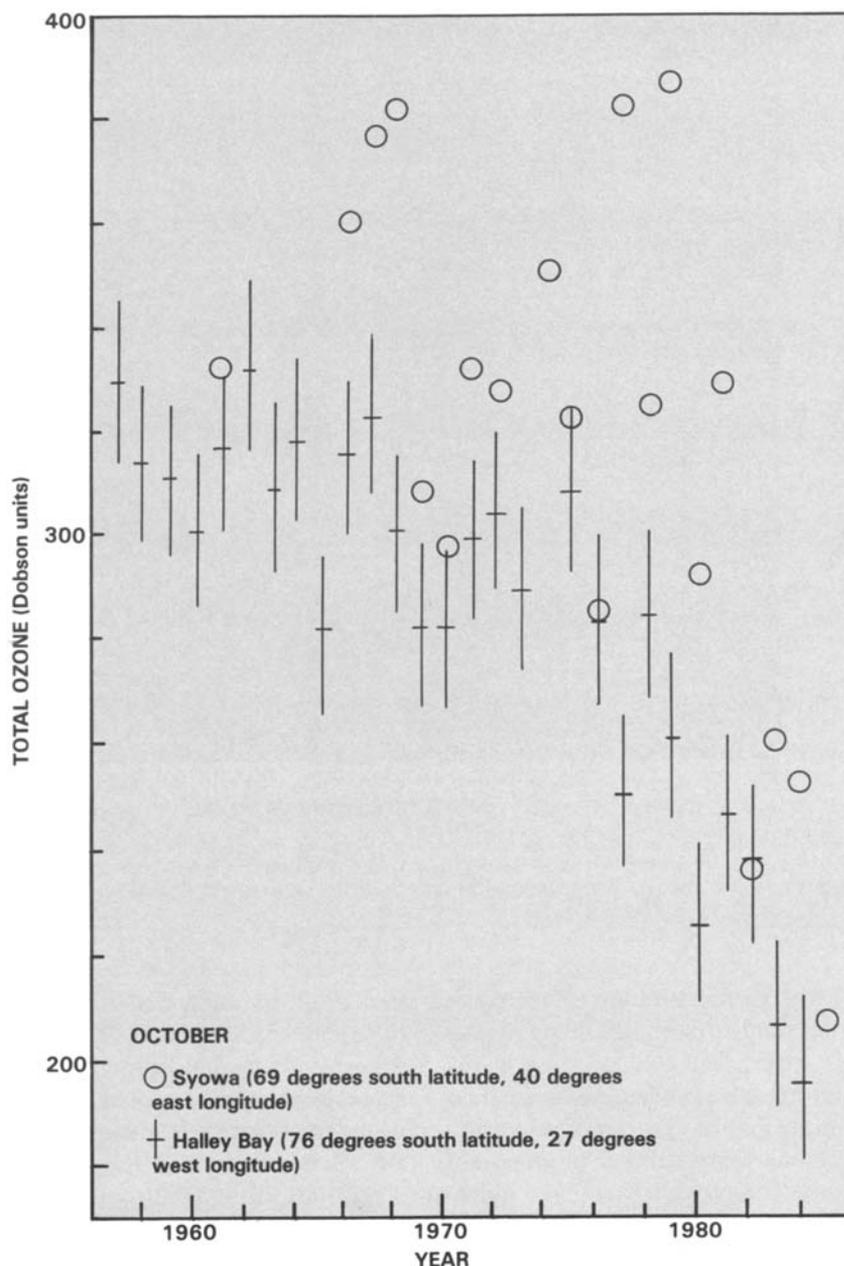
BrO with a concentration on the order of 1 ppb and 20 parts per trillion respectively would lead to a decrease in the ozone column abundance of more than 2 percent per day, a rate significantly larger than observed. Both bromine and chlorine compounds have large industrial sources. B. Krüger and P. Fabian from West Germany have suggested that, in the presence of clouds (particles), acceleration of the photolysis of chlorinated and brominated hydrocarbons could take place and contribute to the observed ozone decline.¹⁷

Besides these theories invoking anthropogenic effects, other hypotheses based on natural mechanisms have been advanced. L. B. Callis from the NASA Langley Research Center and M. Natarajan from SASC Technologies believe that ozone could have been

destroyed by increased amounts of nitrogen oxides produced in the thermosphere (extending beyond 85 km into space) during the exceptional maximum of the last solar cycle and injected into the lower stratosphere through strong subsidence in the polar regions during winter.¹⁸ These two scientists base their assumption on the fact that satellite observations suggest a significant positive trend in the stratospheric nitrogen oxide content between 1979 and 1982.

In order to establish the type of mechanism involved in the springtime ozone decrease, the U.S. National Science Foundation, NOAA, and NASA organized a scientific expedition to the U.S. Antarctic station of McMurdo in October 1986. The purpose was to measure the key species involved in each of the theories suggested. As none of

FIGURE 6. Monthly averaged column abundance of ozone in October measured since the 1960s over the Antarctic stations of Halley Bay and Syowa.



SOURCE: S. Chubachi and R. Kajiwara, "Total Ozone Variation at Syowa, Antarctica," *Geophysical Research Letters* 13(1986):1197.

the numerous data collected have yet been reviewed and published, only some of the findings will be reported here.

For example, it appears that the concentration of ClO at 50 millibars (mb) is lower than 1 ppb in volume, which is

lower than that assumed in certain model predictions but not necessarily lower than the concentration required to involve the CFCs. The column abundance of nitrogen dioxide observed over McMurdo appears to be considerably smaller than that required in the theory

of Callis and Natarajan. The role of the polar stratospheric clouds remains uncertain. But, to the team present in Antarctica, the fact that the ozone profiles observed over McMurdo exhibit layering—indicating that the ozone depletion occurs within height differences of only a few hundred meters—suggests that the ozone depletion might be associated with springtime evaporation of the polar stratospheric clouds.

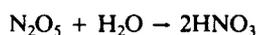
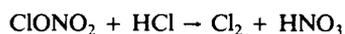
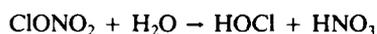
Dynamical Theories

The high correlation between total ozone and temperature in the lower stratosphere suggests to some scientists, such as R. S. Stolarski and his colleagues at the NASA Goddard Space Flight Center, that the observed ozone trend could be a direct consequence of a secular decline in stratospheric temperature over the South Pole.¹⁹ Such a climatic shift might have been triggered by volcanic eruptions or by changes in sea-surface temperature differences. In fact, at the Japanese station of Syowa (but not necessarily at other stations), the monthly mean temperature at 100 mb in November decreased by about 15 K between 1979 and 1985.²⁰ Temperature decreases of this magnitude are considerably larger than the expected change that would result from the ozone depletion itself. A number of dynamicists therefore claim that the change in the seasonal variation of ozone is most probably the result of a substantial reduction in wintertime planetary-scale wave activity.

J. D. Mahlman and S. B. Fels at the NOAA Geophysical Fluid Dynamics Laboratory have shown, for example, that most features observed over Antarctica since 1979 are consistent with this hypothesis.²¹ K. K. Tung from Clarkson University claims that as the sun returns to Antarctica in early spring, a "diabatic" circulation with upward motion at high latitudes and downward motion at midlatitudes will take place.²² This circulation will tend to redistribute ozone in spring with lower values over the pole and higher values at midlatitudes. Observational support for this theory is given by Stolarski and M.

OZONE DEPLETION OVER ANTARCTICA: POSSIBLE CHEMICAL MECHANISMS

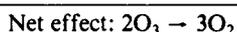
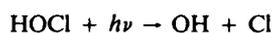
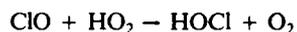
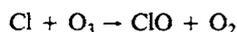
If the ozone depletion occurring in spring over Antarctica is due to the action of chlorine, "inactive chlorine reservoirs" (essentially ClONO₂ and HCl) have to be destroyed and the concentration of "active chlorine" (essentially ClO in the lower stratosphere) should become dramatically larger than under standard conditions. Low densities of ClONO₂ also require low concentrations of NO₂ and therefore a strong destruction mechanism for nitrogen oxides. These conditions could be achieved if the following heterogeneous reactions (in the presence of particles) occurred rapidly inside the polar stratospheric clouds:



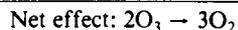
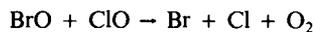
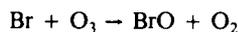
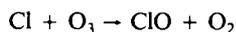
The nitric acid molecules produced by these reactions are believed to be removed from the gas phase by condensation onto solid particles. Cl₂ and HOCl are rapidly photolyzed when the sun returns over Antarctica in spring to produce active chlorine (Cl and ClO).

Several catalytical cycles could play an important role in the destruction of ozone.

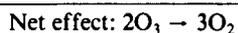
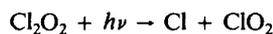
• The "classical" chlorine cycle in the lower stratosphere:



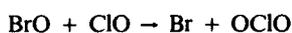
• A cycle involving bromine compounds:



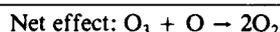
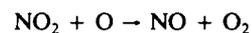
• A cycle involving the Cl₂O₂ dimer:



If ClONO₂ is indeed destroyed in the polar regions in spring, the diurnal variation of ClO should be determined by the formation and destruction rates of Cl₂O₂ and the behavior of OClO produced by:



If, on the other hand, intrusions of nitrogen oxides from the upper atmosphere are responsible for the ozone destruction in the lower stratosphere, the cycle involved is:



R. Schoeberl.²³ From an analysis of the TOMS data, these two scientists have found that the total amount of ozone from 44 degrees south latitude to the pole for a given year is approximately conserved from August to November and, more specifically, that as the ozone minimum over Antarctica declines the ozone maximum outside the polar vortex increases.

An interesting analysis has been made in West Germany by K. Labitzke on the behavior in winter and spring of the lower stratosphere over the two polar regions.²⁴ The analysis shows that vortices over both the Arctic and Antarctica have become stronger since 1979 and that the timing of the final warming over Antarctica has been simultaneously delayed—probably, according to

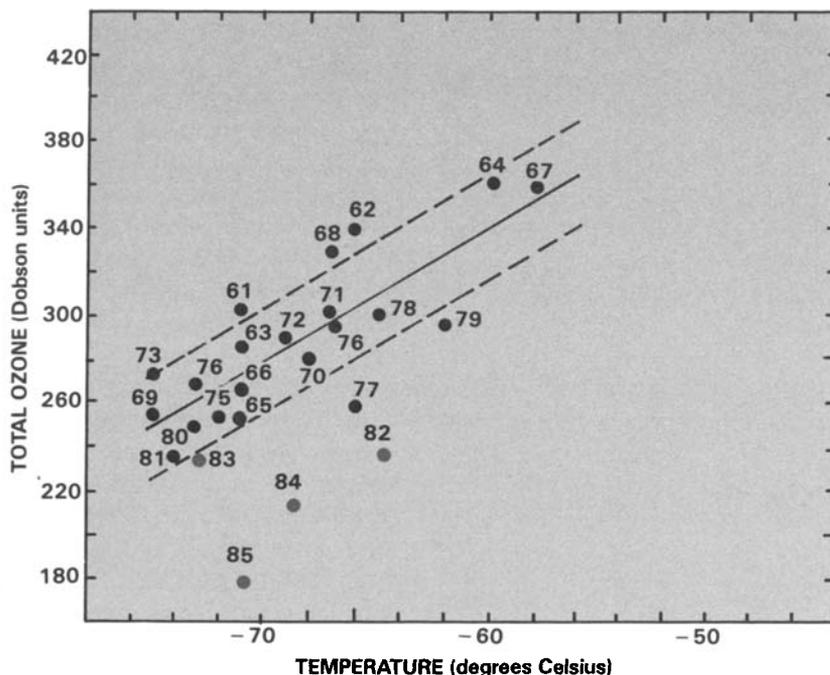
this analysis, as a result of increased stratospheric aerosol content following recent volcanic eruptions. Labitzke noted that the relation between total ozone and the 50-mb temperature found for three stations (Halley Bay, Syowa, and the South Pole), on the basis of a regression made for monthly mean October data between 1961 and 1981, did not hold after 1982, when the ozone column became much too low for the observed temperature (see Figure 7 on page 44). She suggested that this abnormal behavior, probably linked to the delay in the final warming, would allow more time for chemistry to destroy ozone within the polar vortex.

An important question is how the changes in the dynamics of Arctic regions, noted by Labitzke, have affected ozone near the North Pole. Not much attention has yet been given to this problem. Analysis of the TOMS data, however, suggests an ozone decrease in these regions of 50 D.U. (16 percent) between 1979 and 1985.²⁵ The respective roles of chemistry and dynamics in this behavior are not yet known.

Which Theory?

Because of the lack of observational as well as laboratory data, it remains difficult at this juncture to fully adopt or reject any of the theories that have been recently proposed. It is not yet known if the amount of active chlorine in the lower stratosphere is sufficiently large to destroy in 20 to 30 days as much as 40 percent of the ozone confined in the polar vortex. It is not known with absolute certainty if chlorine compounds such as OClO or Cl₂O₂ (see box on this page) are effectively present in the lower stratosphere; neither is the concentration of bromine compounds over Antarctica known. Furthermore, it is not known if nitric acid is effectively removed from the gas phase or how effective heterogeneous processes are in polar stratospheric clouds. An important indication, however, is that the abundance of nitrogen oxides detected in spring over Antarctica is the lowest ever observed in the world and that a

FIGURE 7. Relation between the monthly average ozone column and the 50-millibar temperature over the South Pole in October between 1961 and 1985.



Note: The color points corresponding to years 1982 through 1985 clearly show an anomalous situation.

SOURCE: K. Labitzke, "The Lower Stratosphere over the Polar Regions in Winter and Spring: Relation between Meteorological Parameters and Total Ozone," *Annales Geophysicae* (submitted).

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large fraction of the ozone destruction seems to take place in relatively thin vertical layers. Scientists favoring a dynamical explanation stress the fact that chemical theories are "designed" to explain the springtime ozone decrease in the cold vortex but fail to account for the ozone increase observed at the edge of the vortex. The members of the Antarctica expedition, however, claim that the very low value of nitrous oxide observed in the vortex as well as the study of air mass motions made during the campaign do not give evidence for the upward motions that are needed to support dynamical theories. The explanation of the dramatic change in the behavior of ozone over Antarctica might thus be more complex than first expected and probably involves a combination of chemical and dynamical processes. Although this unpredicted and as yet unexplained phenomenon appears to be confined to lower stratospheric layers over the Antarctic continent, its explanation might involve presently unknown mechanisms that could play a role in the behavior of ozone and additional chemical species in other parts of the terrestrial atmosphere. Thus it appears important to further monitor ozone in the stratosphere with enough precision to detect any global or local significant change in its concentration. Moreover, the comprehensive campaign scheduled to take place in Antarctica in spring 1987 should provide additional information about the mechanisms involved in the rapid ozone change over the South Pole during spring.

ACKNOWLEDGMENTS

Helpful comments and useful suggestions from R. E. Dickinson, J. C. Gille, M. Hitchman, J. London, C. Smythe, and S. Solomon are gratefully acknowledged.

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Overview

(continued from page 5)

ronmental groups, they might see things in a different light."¹⁶

In whatever way environmental groups might decide to get involved in nuclear weapons issues—whether they join arms control groups in the strategic debate in Washington, D.C., or whether they try to add an environmental dimension to the debate—it seems that more environmental groups, continuing the clear trend of the past five years, will begin to address what most environmentalists already see as the "ultimate environmental issue."

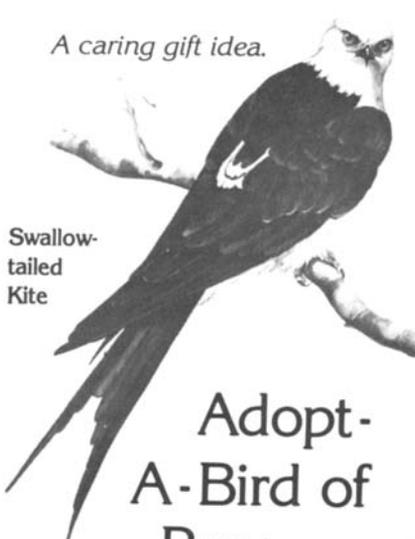
It is likely that disagreements between proenvironment hawks and doves over whether to address the issue will subside as the federal budget gets tighter. It may become impossible for politicians and supporters of the environmental movement to give priority to both military spending and environmental spending. In the future, military concerns may compete for funds so fiercely with environmental concerns that both politicians and advocacy groups will have to choose which concern is more important to them. This may shake some of the more ideological hawks out of the environmental movement and open the way, within certain financial limits, for more environmental groups to play a role in the debate over nuclear weapons.

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