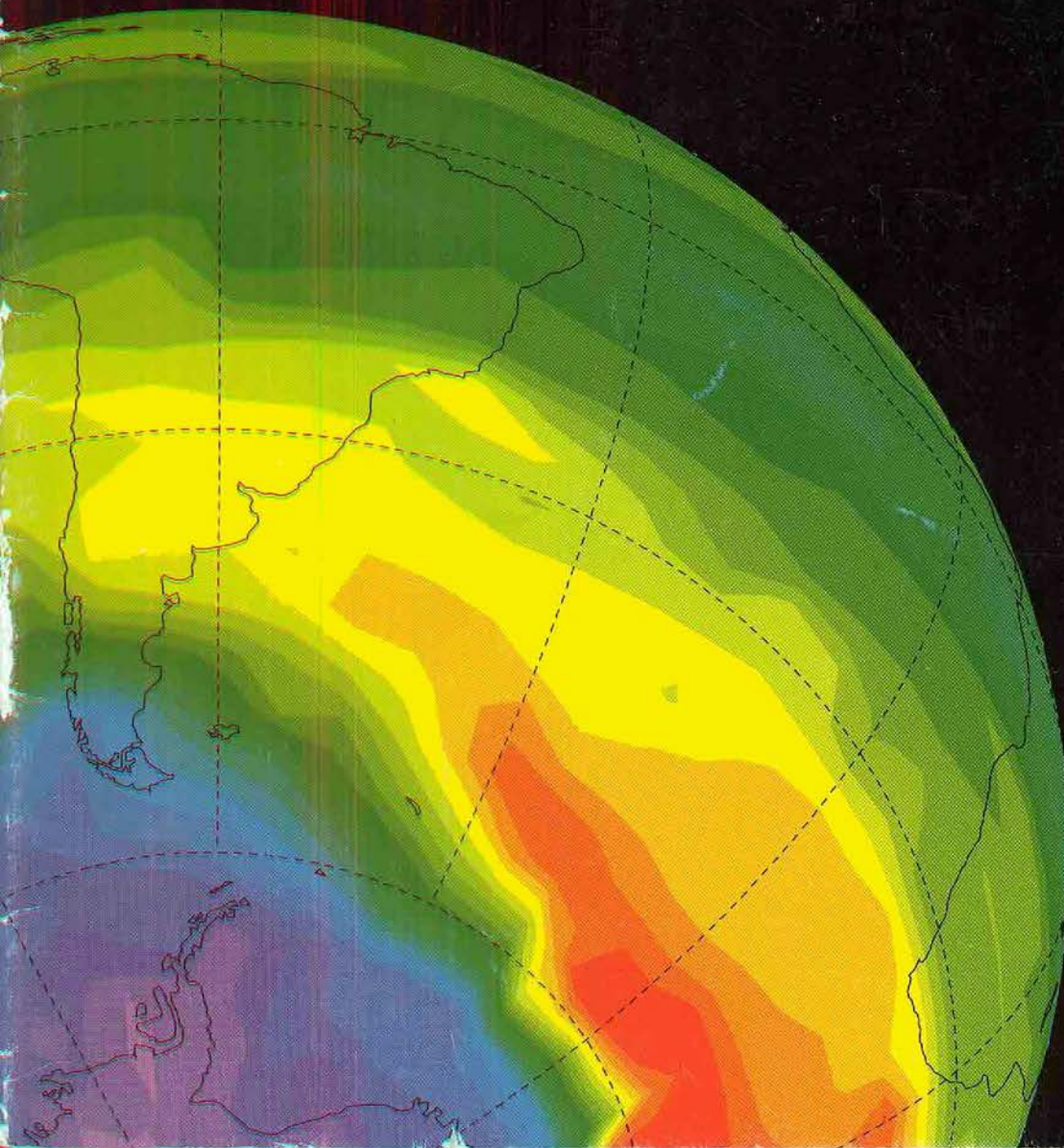# SCIENTIFIC ASSESSMENT OF OZONE DEPLETION: 1994

Executive Summary

National Oceanic and Atmospheric Administration
National Aeronautics and Space Administration
United Nations Environment Programme
World Meteorological Organization

Cover Figure: Observations of ozone abundances on 21 September 1991 in the Southern Hemisphere. The data are provided by the Microwave Limb Sounder on the NASA Upper Atmosphere Research Satellite. Light violet shows ozone depletion in the Antarctic ozone hole. Courtesy of Joe W. Waters, California Institute of Technology/Jet Propulsion Laboratory.

SCIENTIFIC ASSESSMENT OF OZONE DEPLETION: 1994

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SCIENTIFIC ASSESSMENT OF OZONE DEPLETION: 1994

PREFACE

The present document contains key summaries from the *Scientific Assessment of Ozone Depletion: 1994*. The full assessment report will be part of the information upon which the Parties to the United Nations Montreal Protocol will base their future decisions regarding protection of the stratospheric ozone layer.

Specifically, the Montreal Protocol on Substances That Deplete the Ozone Layer states (Article 6): ". . . the Parties shall assess the control measures . . . on the basis of available scientific, environmental, technical, and economic information." To provide the mechanisms whereby these assessments are conducted, the Protocol further states: ". . . the Parties shall convene appropriate panels of experts" and "the panels will report their conclusions . . . to the Parties."

Three assessment reports have been prepared during 1994 to be available to the Parties in advance of their meeting in 1995, at which they will consider the need to amend or adjust the Protocol. The two companion reports to the scientific assessment focus on the environmental and health effects of ozone layer depletion and on the technology and economic implications of mitigation approaches.

The scientific assessment summarized in the present document is the latest in a series of seven scientific reports prepared by the world's leading experts in the atmospheric sciences and under the international auspices of the World Meteorological Organization (WMO) and the United Nations Environment Programme (UNEP). The chronology of those scientific assessments and the relation to the international policy process are summarized as follows:

Year	Policy Process	Scientific Assessment
1981		*The Stratosphere 1981 Theory and Measurements.* WMO No. 11.
1985	Vienna Convention	*Atmospheric Ozone 1985.* 3 vol. WMO No. 16.
1987	Montreal Protocol	
1988		*International Ozone Trends Panel Report 1988.* 2 vol. WMO No. 18.
1989		*Scientific Assessment of Stratospheric Ozone: 1989.* 2 vol. WMO No. 20.
1990	London Amendment	
1991		*Scientific Assessment of Ozone Depletion: 1991.* WMO No. 25.
1992		*Methyl Bromide: Its Atmospheric Science, Technology, and Economics* (Assessment Supplement). UNEP (1992).
1992	Copenhagen Amendment	
1994		*Scientific Assessment of Ozone Depletion: 1994.* WMO No. 37.
(1995)	Vienna Amendment (?)	

The genesis of *Scientific Assessment of Ozone Depletion: 1994* occurred at the 4th meeting of the Conference of the Parties to the Montreal Protocol in Copenhagen, Denmark, in November 1992, at which the scope of the scientific needs of the Parties was defined. The formal planning of the present report was a workshop that was held on 11 June 1993 in

Virginia Beach, Virginia, at which an international steering group crafted the outline and suggested scientists from the world community to serve as authors. The first drafts of the chapters were examined at a meeting that occurred on 2 - 4 March 1994 in Washington, D.C., at which the authors and a small number of international experts improved the coordination of the text of the chapters.

The second draft was sent out to 123 scientists worldwide for a mail peer review. These anonymous comments were considered by the authors. At a Panel Review Meeting in Les Diablerets, Switzerland, held on 18 - 21 July 1994, the responses to these mail review comments were proposed by the authors and discussed by the 80 participants. Final changes to the chapters were decided upon, and the Executive Summary contained herein was prepared by the participants.

The group also focused on a set of questions commonly asked about the ozone layer. Based upon the scientific understanding represented by the assessments, answers to these common questions were prepared and are also included here.

As the accompanying list indicates, the *Scientific Assessment of Ozone Depletion: 1994* is the product of 295 scientists from the developed and developing world[1] who contributed to its preparation and review (230 scientists prepared the report and 147 scientists participated in the peer review process).

What follows is a summary of their current understanding of the stratospheric ozone layer and its relation to humankind.

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[1] Participating were Argentina, Australia, Austria, Belgium, Brazil, Canada, Chile, Cuba, Czech Republic, Denmark, Egypt, France, Germany, Greece, Hungary, India, Iran, Ireland, Israel, Italy, Japan, Kenya, Malaysia, New Zealand, Norway, Poland, Russia, South Africa, Sweden, Switzerland, Taiwan, The Netherlands, The People's Republic of China, United Kingdom, United States of America, and Venezuela.

EXECUTIVE SUMMARY

Recent Major Scientific Findings and Observations

The laboratory investigations, atmospheric observations, and theoretical and modeling studies of the past few years have provided a deeper understanding of the human-influenced and natural chemical changes in the atmosphere and their relation to the Earth's stratospheric ozone layer and radiative balance of the climate system. Since the last international scientific assessment of the state of understanding, there have been several key ozone-related findings, observations, and conclusions:

- **The atmospheric growth rates of several major ozone-depleting substances have slowed, demonstrating the expected impact of the Montreal Protocol and its Amendments and Adjustments.** The abundances of the chlorofluorocarbons (CFCs), carbon tetrachloride, methyl chloroform, and halons in the atmosphere have been monitored at global ground-based sites since about 1978. Over much of that period, the annual growth rates of these gases have been positive. However, the data of recent years clearly show that the growth rates of CFC-11, CFC-12, halon-1301, and halon-1211 are slowing down. In particular, total tropospheric organic chlorine increased by only about 60 ppt/year (1.6%) in 1992, compared to 110 ppt/year (2.9%) in 1989. Furthermore, tropospheric bromine in halons increased by only about 0.25 ppt/year in 1992, compared to about 0.85 ppt/year in 1989. The abundance of carbon tetrachloride is actually decreasing. The observed trends in total tropospheric organic chlorine are consistent with reported production data, suggesting less emission than the maximum allowed under the Montreal Protocol and its Amendments and Adjustments. Peak total chlorine/bromine loading in the troposphere is expected to occur in 1994, but the stratospheric peak will lag by about 3 - 5 years. Since the stratospheric abundances of chlorine and bromine are expected to continue to grow for a few more years, increasing global ozone losses are predicted (other things being equal) for the remainder of the decade, with gradual recovery in the 21st century.

- **The atmospheric abundances of several of the CFC substitutes are increasing, as anticipated.** With phase-out dates for the CFCs and other ozone-depleting substances now fixed by international agreements, several hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs) are being manufactured and used as substitutes. The atmospheric growth of some of these compounds (*e.g.*, HCFC-22) has been observed for several years, and the growth rates of others (*e.g.*, HCFC-142b and HCFC-141b) are now being monitored. Tropospheric chlorine in HCFCs increased by 5 ppt/year in 1989 and about 10 ppt/year in 1992.

- **Record low global ozone levels were measured over the past two years.** Anomalous ozone decreases were observed in the midlatitudes of both hemispheres in 1992 and 1993. The Northern Hemispheric decreases were larger than those in the Southern Hemisphere. Globally, ozone values were 1 - 2% lower than would be expected from an extrapolation of the trend prior to 1991, allowing for solar-cycle and quasi-biennial-oscillation (QBO) effects. The 1994 global ozone levels are returning to values closer to those expected from the longer-term downward trend.

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- **The stratosphere was perturbed by a major volcanic eruption.** The eruption of Mt. Pinatubo in 1991 led to a large increase in sulfate aerosol in the lower stratosphere throughout the globe. Reactions on sulfate aerosols resulted in significant, but temporary, changes in the chemical partitioning that accelerated the photochemical ozone loss associated with reactive hydrogen (HO_x), chlorine, and bromine compounds in the lower stratosphere in midlatitudes and polar regions. Absorption of terrestrial and solar radiation by the Mt. Pinatubo aerosol resulted in a transitory rise of 1°C (globally averaged) in the lower-stratospheric temperature and also affected the distribution of ozone through circulation changes. The observed 1994 recovery of global ozone is qualitatively consistent with observed gradual reductions of the abundances of these volcanic particles in the stratosphere.

- **Downward trends in total-column ozone continue to be observed over much of the globe, but their magnitudes are underestimated by numerical models.** Decreases in ozone abundances of about 4 - 5% per decade at midlatitudes in the Northern and Southern Hemispheres continue to be observed by both ground-based and satellite-borne monitoring instruments. At midlatitudes, the losses continue to be much larger during winter/spring than during summer/fall in both hemispheres, and the depletion increases with latitude, particularly in the Southern Hemisphere. Little or no downward trends are observed in the tropics (20°N - 20°S). While the current two-dimensional stratospheric models simulate the observed trends quite well during some seasons and latitudes, they underestimate the trends by factors of up to three in winter/spring at mid- and high latitudes. Several known atmospheric processes that involve chlorine and bromine and that affect ozone in the lower stratosphere are difficult to model and have not been adequately incorporated into these models.

- **Observations have demonstrated that halogen chemistry plays a larger role in the chemical destruction of ozone in the midlatitude lower stratosphere than expected from gas phase chemistry.** Direct *in situ* measurements of radical species in the lower stratosphere, coupled with model calculations, have quantitatively shown that the *in situ* photochemical loss of ozone due to (largely natural) reactive nitrogen (NO_x) compounds is smaller than that predicted from gas phase chemistry, while that due to (largely natural) HO_x compounds and (largely anthropogenic) chlorine and bromine compounds is larger than that predicted from gas phase chemistry. This confirms the key role of chemical reactions on sulfate aerosols in controlling the chemical balance of the lower stratosphere. These and other recent scientific findings strengthen the conclusion of the previous assessment that the weight of scientific evidence suggests that the observed middle- and high-latitude ozone losses are largely due to anthropogenic chlorine and bromine compounds.

- **The conclusion that anthropogenic chlorine and bromine compounds, coupled with surface chemistry on natural polar stratospheric particles, are the cause of polar ozone depletion has been further strengthened.** Laboratory studies have provided a greatly improved understanding of how the chemistry on the surfaces of ice, nitrate, and sulfate particles can increase the abundance of ozone-depleting forms of chlorine in the polar stratospheres. Furthermore, satellite and *in situ* observations of the abundances of reactive nitrogen and chlorine compounds have improved the explanation of the different ozone-altering properties of the Antarctic and Arctic.

- **The Antarctic ozone "holes" of 1992 and 1993 were the most severe on record.** The Antarctic ozone "hole" has continued to occur seasonally every year since its advent in the late-1970s, with the occurrences over the last several years being particularly pronounced. Satellite, balloon-borne, and ground-based monitoring instruments revealed that the Antarctic ozone "holes" of 1992 and 1993 were the biggest (areal extent) and deepest (minimum amounts of ozone overhead), with ozone being locally depleted by more than 99% between about 14 - 19 km in October, 1992 and 1993. It is likely that these larger-than-usual ozone depletions could be attributed, at least in part, to sulfate aerosols from Mt. Pinatubo increasing the effectiveness of chlorine- and bromine-catalyzed ozone destruction. A substantial Antarctic ozone "hole" is expected to occur each austral spring for many more decades because stratospheric chlorine and bromine abundances will approach the pre-Antarctic-ozone-"hole" levels (late-1970s) very slowly during the next century.

- **Ozone losses have been detected in the Arctic winter stratosphere, and their links to halogen chemistry have been established.** Studies in the Arctic lower stratosphere have been expanded to include more widespread observations of ozone and key reactive species. In the late-winter/early-spring period, additional chemical losses of ozone up to 15 - 20% at some altitudes are deduced from these observations, particularly in the winters of 1991/2 and 1992/3. Model calculations constrained by the observations are also consistent with these losses, increasing the confidence in the role of chlorine and bromine in ozone destruction. The interannual variability in the photo-chemical and dynamical conditions of the Arctic polar vortex continues to limit the ability to predict ozone changes in future years.

- **The link between a decrease in stratospheric ozone and an increase in surface ultraviolet (UV) radiation has been further strengthened.** Measurements of UV radiation at the surface under clear-sky conditions show that low overhead ozone yields high UV radiation and in the amount predicted by radiative-transfer theory. Large increases of surface UV are observed in Antarctica and the southern part of South America during the period of the seasonal ozone "hole." Furthermore, elevated surface UV levels at mid-to-high latitudes were observed in the Northern Hemisphere in 1992 and 1993, corresponding to the low ozone levels of those years. However, the lack of a decadal (or longer) record of accurate monitoring of surface UV levels and the variation introduced by clouds and other factors have precluded the unequivocal identification of a long-term trend in surface UV radiation.

- **Methyl bromide continues to be viewed as a significant ozone-depleting compound.** Increased attention has been focused upon the ozone-depleting role of methyl bromide. Three potentially major anthropogenic sources of atmospheric methyl bromide have been identified (soil fumigation, biomass burning, and the exhaust of automobiles using leaded gasoline), in addition to the natural oceanic source. Recent laboratory studies have confirmed the fast rate for the $BrO + HO_2$ reaction and established a negligible reaction pathway producing HBr, both of which imply greater ozone losses due to emissions of compounds containing bromine. While the magnitude of the atmospheric photochemical removal is well understood, there are significant uncertainties in quantifying the oceanic sink for atmospheric methyl bromide. The best estimate for the overall lifetime of atmospheric methyl bromide is 1.3 years, with a range of 0.8 - 1.7 years. The Ozone Depletion Potential (ODP) for methyl bromide is calculated to be about 0.6 (relative to an ODP of 1 for CFC-11).

- **Stratospheric ozone losses cause a global-mean negative radiative forcing.** In the 1991 scientific assessment, it was pointed out that the global ozone losses that were occurring in the lower stratosphere caused this region to cool and result in less radiation reaching the surface-troposphere system. Recent model studies have strengthened this picture. A long-term global-mean cooling of the lower stratosphere of between 0.25 and 0.4°C/decade has been observed over the last three decades. Calculations indicate that, on a global mean, the ozone losses between 1980 and 1990 offset about 20% of the radiative forcing due to the well-mixed greenhouse-gas increases during that period (*i.e.*, carbon dioxide, methane, nitrous oxide, and halocarbons).

- **Tropospheric ozone, which is a greenhouse gas, appears to have increased in many regions of the Northern Hemisphere.** Observations show that tropospheric ozone, which is formed by chemical reactions involving pollutants, has increased above many locations in the Northern Hemisphere over the last 30 years. However, in the 1980s, the trends were variable, being small or nonexistent. In the Southern Hemisphere, there are insufficient data to draw strong inferences. At the South Pole, a decrease has been observed since the mid-1980s. Model simulations and limited observations suggest that tropospheric ozone has increased in the Northern Hemisphere since pre-industrial times. Such changes would augment the radiative forcing from all other greenhouse gases by about 20% over the same time period.

- **The atmospheric residence times of the important ozone-depleting gases, CFC-11 and methyl chloroform, and the greenhouse gas, methane, are now better known.** A reconciliation of observed concentrations with known emissions using an atmospheric model has led to a best-estimate lifetime of 50 years for CFC-11 and 5.4 years for methyl chloroform, with uncertainties of about 10%. These lifetimes provide an accurate standard for gases destroyed only in the stratosphere (such as CFCs and nitrous oxide) and for those also reacting with tropospheric hydroxyl radical, OH (such as HCFCs and HFCs), respectively. Recent model simulations of methane perturbations and a theoretical analysis of the tropospheric chemical system that couples methane, carbon monoxide, and OH have demonstrated that methane perturbations decay with a lengthened time scale in a range of about 12 - 17 years, as compared with the 10-year lifetime derived from the total abundance and losses. This longer response time and other indirect effects increase the estimate of the effectiveness of emissions of methane as a greenhouse gas by a factor of about two compared to the direct-effect-only values given in the 1991 assessment.

Supporting Scientific Evidence and Related Issues

OZONE CHANGES IN THE TROPICS AND MIDLATITUDES AND THEIR INTERPRETATION

- Analysis of global total-column ozone data through early 1994 shows substantial decreases of ozone in all seasons at midlatitudes (30° - 60°) of both hemispheres. For example, in the middle latitudes of the Northern Hemisphere, downward trends of about 6% per decade over 1979 - 1994 were observed in winter and spring and about 3% per decade were observed in summer and fall. In the Southern Hemisphere, the seasonal difference was somewhat less, but the midlatitude trends averaged a similar 4% to 5% per decade. There are no statistically significant trends in the tropics (20°S - 20°N). Trends through 1994 are about 1% per decade more negative in the Northern Hemisphere (2% per decade in the midlatitude winter/spring in the Northern Hemisphere) compared to those calculated without using data after May 1991. At Northern midlatitudes, the downward trend in ozone between 1981 - 1991 was about 2% per decade greater compared to that of the period 1970 - 1980.

- Satellite and ozonesonde data show that much of the downward trend in ozone occurs below 25 km (*i.e.*, in the lower stratosphere). For the region 20 - 25 km, there is good agreement between the trends from the Stratospheric Aerosol and Gas Experiment (SAGE I/II) satellite instrument data and those from ozonesondes, with an observed annual-average decrease of $7 \pm 4\%$ per decade from 1979 to 1991 at 30° - 50°N latitude. Below 20 km, SAGE yields negative trends as large as $20 \pm 8\%$ per decade at 16 - 17 km, while the average of available midlatitude ozonesonde data shows smaller negative trends of $7 \pm 3\%$ per decade. Integration of the ozonesonde data yields total-ozone trends consistent with total-ozone measurements. In the 1980s, upper-stratospheric (35 - 45 km) ozone trends determined by the data from SAGE I/II, Solar Backscatter Ultraviolet satellite spectrometer (SBUV), and the Umkehr method agree well at midlatitudes, but less so in the tropics. Ozone declined 5 - 10% per decade at 35 - 45 km between 30°- 50°N and slightly more at southern midlatitudes. In the tropics at 45 km, SAGE I/II and SBUV yield downward trends of 10 and 5% per decade, respectively.

- Simultaneous *in situ* measurements of a suite of reactive chemical species have directly confirmed modeling studies implying that the chemical destruction of ozone in the midlatitude lower stratosphere is more strongly influenced by HO_x and halogen chemistry than NO_x chemistry. The seasonal cycle of ClO in the lower stratosphere at midlatitudes in both hemispheres supports a role for *in situ* heterogeneous perturbations (*i.e.*, on sulfate aerosols), but does not appear consistent with the timing of vortex processing or dilution. These studies provide key support for the view that sulfate aerosol chemistry plays an important role in determining midlatitude chemical ozone destruction rates.

- The model-calculated ozone depletions in the upper stratosphere for 1980 - 1990 are in broad agreement with the measurements. Although these model-calculated ozone depletions did not consider radiative feedbacks and temperature trends, including these effects is not likely to reduce the predicted ozone changes by more than 20%.

- Models including the chemistry involving sulfate aerosols and polar stratospheric clouds (PSCs) better simulate the observed total ozone depletions of the past decade than models that include only gas phase reactions. However, they still underestimate the ozone loss by factors ranging from 1.3 to 3.0.

- Some unresolved discrepancies between observations and models exist for the partitioning of inorganic chlorine species, which could impact model predictions of ozone trends. These occur for the ClO/HCl ratio in the upper stratosphere and the fraction of HCl to total inorganic chlorine in the lower stratosphere.

- The transport of ozone-depleted air from polar regions has the potential to influence ozone concentrations at middle latitudes. While there are uncertainties about the importance of this process relative to *in situ* chemistry for midlatitude ozone loss, both directly involve ozone destruction by chlorine- and bromine-catalyzed reactions.

- Radiosonde and satellite data continue to show a long-term cooling trend in globally annual-average lower-stratospheric temperatures of about 0.3 - 0.4°C per decade over the last three decades. Models suggest that ozone depletion is the major contributor to this trend.

- Anomalously large downward ozone trends have been observed in midlatitudes of both hemispheres in 1992 and 1993 (*i.e.*, the first two years after the eruption of Mt. Pinatubo), with Northern-Hemispheric decreases larger than those of the Southern Hemisphere. Global-average total-ozone levels in early 1993 were about 1% to 2% below that expected from the long-term trend and the particular phase of the solar and QBO cycles, while peak decreases of about 6 - 8% from expected ozone levels were seen over 45 - 60°N. In the first half of 1994, ozone levels returned to values closer to those expected from the long-term trend.

- The sulfur gases injected by Mt. Pinatubo led to large enhancements in stratospheric sulfate aerosol surface areas (by a maximum factor of about 30 - 40 at northern midlatitudes within a year after the eruption), which have subsequently declined.

- Anomalously low ozone was measured at altitudes below 25 km at a Northern-Hemispheric midlatitude station in 1992 and 1993 and was correlated with observed enhancements in sulfate-aerosol surface areas, pointing towards a causal link.

- Observations indicate that the eruption of Mt. Pinatubo did not significantly increase the HCl content of the stratosphere.

- The recent large ozone changes at midlatitudes are highly likely to have been due, at least in part, to the greatly increased sulfate aerosol in the lower stratosphere following Mt. Pinatubo. Observations and laboratory studies have demonstrated the importance of heterogeneous hydrolysis of N_2O_5 on sulfate aerosols in the atmosphere. Evidence suggests that $ClONO_2$ hydrolysis also occurs on sulfate aerosols under cold conditions. Both processes perturb the chemistry in such a way as to increase ozone loss through coupling with the anthropogenic chlorine and bromine loading of the stratosphere.

- Global mean lower stratospheric temperatures showed a marked transitory rise of about 1°C following the eruption of Mt. Pinatubo in 1991, consistent with model calculations. The warming is likely due to absorption of radiation by the aerosols.

POLAR OZONE DEPLETION

- In 1992 and 1993, the biggest-ever (areal extent) and deepest-ever (minimum ozone below 100 Dobson units) ozone "holes" were observed in the Antarctic. These extreme ozone depletions may have been due to the chemical perturbations caused by sulfate aerosols from Mt. Pinatubo, acting in addition to the well-recognized chlorine and bromine reactions on polar stratospheric clouds.

- Recent results of observational and modeling studies reaffirm the role of anthropogenic halocarbon species in Antarctic ozone depletion. Satellite observations show a strong spatial and temporal correlation of ClO abundances with ozone depletion in the Antarctic vortex. In the Arctic winter, a much smaller ozone loss has been observed. These losses are both consistent with photochemical model calculations constrained with observations from *in situ* and satellite instruments.

- Extensive new measurements of HCl, ClO, and $ClONO_2$ from satellites and *in situ* techniques have confirmed the picture of the chemical processes responsible for chlorine activation in polar regions and the recovery from those processes, strengthening current understanding of the seasonal cycle of ozone depletion in both polar regions.

- New laboratory and field studies strengthen the confidence that reactions on sulfate aerosols can activate chlorine under cold conditions, particularly those in the polar regions. Under volcanically perturbed conditions when aerosols are enhanced, these processes also likely contribute to ozone losses at the edges of PSC formation regions (both vertical and horizontal) just outside of the southern vortex and in the Arctic.

- Satellite measurements have confirmed that the Arctic vortex is much less denitrified than the Antarctic, which is likely to be an important factor in determining the interhemispheric differences in polar ozone loss.

- Interannual variability in the photochemical and dynamical conditions of the vortices limits reliable predictions of future ozone changes in the polar regions, particularly in the Arctic.

COUPLING BETWEEN POLAR REGIONS AND MIDLATITUDES

- Recent satellite observations of long-lived tracers and modeling studies confirm that, above 16 km, air near the center of the polar vortex is substantially isolated from lower latitudes, especially in the Antarctic.

- Erosion of the vortex by planetary-wave activity transports air from the vortex-edge region to lower latitudes. Nearly all observational and modeling studies are consistent with a time scale of 3 - 4 months to replace a substantial fraction of Antarctic vortex air. The importance of this transport to *in situ* chemical effects for midlatitude ozone loss remains poorly known.

- Air is readily transported between polar regions and midlatitudes below 16 km. The influence of this transport on midlatitude ozone loss has not been quantified.

TROPOSPHERIC OZONE

- There is observational evidence that tropospheric ozone (about 10% of the total-column ozone) has increased in the Northern Hemisphere (north of 20°N) over the past three decades. The upward trends are highly regional. They are smaller in the 1980s than in the 1970s and may be slightly negative at some locations. European measurements at surface sites also indicate a doubling in the lower-tropospheric ozone concentrations since earlier this century. At the South Pole, a decrease has been observed since the mid-1980s. Elsewhere in the Southern Hemisphere, there are insufficient data to draw strong inferences.

- There is strong evidence that ozone levels in the boundary layer over the populated regions of the Northern Hemisphere are enhanced by more than 50% due to photochemical production from anthropogenic precursors, and that export of ozone from North America is a significant source for the North Atlantic region during summer. It has also been shown that biomass burning is a significant source of ozone (and carbon monoxide) in the tropics during the dry season.

- An increase in UV-B radiation (e.g., from stratospheric ozone loss) is expected to decrease tropospheric ozone in the background atmosphere, but, in some cases, it will increase production of ozone in the more polluted regions.

- Model calculations predict that a 20% increase in methane concentrations would result in tropospheric ozone increases ranging from 0.5 to 2.5 ppb in the tropics and the northern midlatitude summer, and an increase in the methane residence time to about 14 years (a range of 12 - 17 years). Although there is a high degree of consistency in the global transport of short-lived tracers within three-dimensional chemical-transport models, and a general agreement in the computation of photochemical rates affecting tropospheric ozone, many processes controlling tropospheric ozone are not adequately represented or tested in the models, hence limiting the accuracy of these results.

TRENDS IN SOURCE GASES RELATING TO OZONE CHANGES

- CFCs, carbon tetrachloride, methyl chloroform, and the halons are major anthropogenic source gases for stratospheric chlorine and bromine, and hence stratospheric ozone destruction. Observations from several monitoring networks worldwide have demonstrated slowdowns in growth rates of these species that are consistent (except for carbon tetrachloride) with expectations based upon recent decreases in emissions. In addition, observations from several sites have revealed accelerating growth rates of the CFC substitutes, HCFC-22, HCFC-141b, and HCFC-142b, as expected from their increasing use.

- Methane levels in the atmosphere affect tropospheric and stratospheric ozone levels. Global methane increased by 7% over about the past decade. However, the 1980s were characterized by slower growth rates, dropping from approximately 20 ppb per year in 1980 to about 10 ppb per year by the end of the decade. Methane growth rates slowed dramatically in 1991 and 1992, but the very recent data suggest that they have started to increase in late 1993. The cause(s) of this behavior are not known, but it is probably due to changes in methane sources rather than sinks.

- Despite the increased methane levels, the total amount of carbon monoxide in today's atmosphere is less than it was a decade ago. Recent analyses of global carbon monoxide data show that tropospheric levels grew from the early 1980s to about 1987 and have declined from the late 1980s to the present. The cause(s) of this behavior have not been identified.

CONSEQUENCES OF OZONE CHANGES

• The only general circulation model (GCM) simulation to investigate the climatic impacts of observed ozone depletions between 1970 and 1990 supports earlier suggestions that these depletions reduced the model-predicted warming due to well-mixed greenhouse gases by about 20%. This is consistent with radiative forcing calculations.

• Model simulations suggest that increases in tropospheric ozone since pre-industrial times may have made significant contributions to the greenhouse forcing of the Earth's climate system, enhancing the current total forcing by about 20% compared to that arising from the changes in the well-mixed greenhouses gases over that period.

• Large increases in ultraviolet (UV) radiation have been observed in association with the ozone hole at high southern latitudes. The measured UV enhancements agree well with model calculations.

• Clear-sky UV measurements at midlatitude locations in the Southern Hemisphere are significantly larger than at a corresponding site in the Northern Hemisphere, in agreement with expected differences due to ozone column and Sun-Earth separation.

• Local increases in UV-B were measured in 1992/93 at mid- and high latitudes in the Northern Hemisphere. The spectral signatures of the enhancements clearly implicate the anomalously low ozone observed in those years, rather than variability of cloud cover or tropospheric pollution. Such correlations add confidence to the ability to link ozone changes to UV-B changes over relatively long time scales.

• Increases in clear-sky UV over the period 1979 to 1993 due to observed ozone changes are calculated to be greatest at short wavelengths and at high latitudes. Poleward of 45°, the increases are greatest in the Southern Hemisphere.

• Uncertainties in calibration, influence of tropospheric pollution, and difficulties of interpreting data from broadband instruments continue to preclude the unequivocal identification of long-term UV trends. However, data from two relatively unpolluted sites do appear to show UV increases consistent with observed ozone trends. Given the uncertainties of these studies, it now appears that quantification of the natural (i.e., pre-ozone-reduction) UV baseline has been irrevocably lost at mid- and high latitudes.

• Scattering of UV radiation by stratospheric aerosols from the Mt. Pinatubo eruption did not alter total surface-UV levels appreciably.

RELATED PHENOMENA AND ISSUES

Methyl Bromide

• Three potentially major anthropogenic sources of methyl bromide have been identified: (i) soil fumigation: 20 to 60 ktons per year, where new measurements reaffirm that about 50% (ranging from 20 - 90%) of the methyl bromide used as a soil fumigant is released into the atmosphere; (ii) biomass burning: 10 to 50 ktons per year; and (iii) the exhaust of automobiles using leaded gasoline: 0.5 to 1.5 ktons per year or 9 to 22 ktons per year (the two studies report emission factors that differ by a factor of more than 10). In addition, the one known major natural source of methyl bromide is oceanic, with emissions of 60 to 160 ktons per year.

- Recent measurements have confirmed that there is more methyl bromide in the Northern Hemisphere than in the Southern Hemisphere, with an interhemispheric ratio of 1.3.

- There are two known sinks for atmospheric methyl bromide: (i) atmospheric, with a lifetime of 2.0 years (1.5 to 2.5 years); and (ii) oceanic, with an estimated lifetime of 3.7 years (1.5 to 10 years). The overall best estimate for the lifetime of atmospheric methyl bromide is 1.3 years, with a range of 0.8 to 1.7 years. An overall lifetime of less than 0.6 years is thought to be highly unlikely because of constraints imposed by the observed interhemispheric ratio and total known emissions.

- The chemistry of bromine-induced stratospheric ozone destruction is now better understood. Laboratory measurements have confirmed the fast rate for the $BrO + HO_2$ reaction and have established a negligible reaction pathway producing HBr, both of which imply greater ozone losses due to emissions of compounds containing bromine. Stratospheric measurements show that the abundance of HBr is less than 1 ppt.

- Bromine is estimated to be about 50 times more efficient than chlorine in destroying stratospheric ozone on a per-atom basis. The ODP for methyl bromide is calculated to be about 0.6, based on an overall lifetime of 1.3 years. An uncertainty analysis suggests that the ODP is unlikely to be less than 0.3.

Aircraft

- Subsonics: Estimates indicate that present subsonic aircraft operations may be significantly increasing trace species (primarily NO_x, sulfur dioxide, and soot) at upper-tropospheric altitudes in the North-Atlantic flight corridor. Models indicate that the NO_x emissions from the current subsonic fleet produce upper-tropospheric ozone increases as much as several percent, maximizing at northern midlatitudes. Since the results of these rather complex models depend critically on NO_x chemistry and since the tropospheric NO_x budget is uncertain, little confidence should be put in these quantitative model results at the present time.

- Supersonics: Atmospheric effects of supersonic aircraft depend on the number of aircraft, the altitude of operation, the exhaust emissions, and the background chlorine and aerosol loadings. Projected fleets of supersonic transports would lead to significant changes in trace-species concentrations, especially in the North-Atlantic flight corridor. Two-dimensional model calculations of the impact of a projected fleet (500 aircraft, each emitting 15 grams of NO_x per kilogram of fuel burned at Mach 2.4) in a stratosphere with a chlorine loading of 3.7 ppb, imply additional (*i.e.*, beyond those from halocarbon losses) annual-average ozone column decreases of 0.3 - 1.8% for the Northern Hemisphere. There are, however, important uncertainties in these model results, especially in the stratosphere below 25 km. The same models fail to reproduce the observed ozone trends in the stratosphere below 25 km between 1980 and 1990. Thus, these models may not be properly including mechanisms that are important in this crucial altitude range.

- Climate Effects: Reliable quantitative estimates of the effects of aviation emissions on climate are not yet available. Some initial estimates indicate that the climate effects of ozone changes resulting from subsonic aircraft emissions may be comparable to those resulting from their CO_2 emissions.

Ozone Depletion Potentials (ODPs)

- If a substance containing chlorine or bromine decomposes in the stratosphere, it will destroy some ozone. HCFCs have short tropospheric lifetimes, which tends to reduce their impact on stratospheric ozone as compared to CFCs and halons. However, there are substantial differences in ODPs among various substitutes. The steady-state ODPs of substitute compounds considered in the present assessment range from about 0.01 - 0.1.

- Tropospheric degradation products of CFC substitutes will not lead to significant ozone loss in the stratosphere. Those products will not accumulate in the atmosphere and will not significantly influence the ODPs and Global Warming Potentials (GWPs) of the substitutes.

- Trifluoroacetic acid, formed in the atmospheric degradation of HFC-134a, HCFC-123, and HCFC-124, will enter into the aqueous environment, where biological, rather than physico-chemical, removal processes may be effective.

- It is known that atomic fluorine (F) itself is not an efficient catalyst for ozone loss, and it is concluded that the F-containing fragments from the substitutes (such as CF_3O_x) also have negligible impact on ozone. Therefore, ODPs of HFCs containing the CF_3 group (such as HFC-134a, HFC-23, and HFC-125) are likely to be much less than 0.001.

- New laboratory measurements and associated modeling studies have confirmed that perfluorocarbons and sulfur hexafluoride are long-lived in the atmosphere and act as greenhouse gases.

- The ODPs for several new compounds, such as HCFC-225ca, HCFC-225cb, and CF_3I, have been evaluated using both semi-empirical and modeling approaches, and are found to be 0.03 or less.

Global Warming Potentials (GWPs)

- Both the direct and indirect components of the GWP of methane have been estimated using model calculations. Methane's influence on the hydroxyl radical and the resulting effect on the methane response time lead to substantially longer response times for decay of emissions than OH removal alone, thereby increasing the GWP. In addition, indirect effects including production of tropospheric ozone and stratospheric water vapor were considered and are estimated to range from about 15 to 45% of the total GWP (direct plus indirect) for methane.

- GWPs, including indirect effects of ozone depletion, have been estimated for a variety of halocarbons, clarifying the relative radiative roles of ozone-depleting compounds (*i.e.*, CFCs and halons). The net GWPs of halocarbons depend strongly upon the effectiveness of each compound for ozone destruction; the halons are highly likely to have negative net GWPs, while those of the CFCs are likely to be positive over both 20- and 100-year time horizons.

Implications for Policy Formulation

The research findings of the past few years that are summarized above have several major implications as scientific input to governmental, industrial, and other policy decisions regarding human-influenced substances that lead to depletion of the stratospheric ozone layer and to changes of the radiative forcing of the climate system:

• **The Montreal Protocol and its Amendments and Adjustments are reducing the impact of anthropogenic halocarbons on the ozone layer and should eventually eliminate this ozone depletion.** Based on assumed compliance with the amended Montreal Protocol *(Copenhagen, 1992)* by all nations, the stratospheric chlorine abundances will continue to grow from their current levels (3.6 ppb) to a peak of about 3.8 ppb around the turn of the century. The future total bromine loading will depend upon choices made regarding future human production and emissions of methyl bromide. After around the turn of the century, the levels of stratospheric chlorine and bromine will begin a decrease that will continue into the 21st and 22nd centuries. The rate of decline is dictated by the long residence times of the CFCs, carbon tetrachloride, and halons. Global ozone losses and the Antarctic ozone "hole" were first discernible in the late 1970s and are predicted to recover in about the year 2045, other things being equal. The recovery of the ozone layer would have been impossible without the Amendments and Adjustments to the original Protocol *(Montreal, 1987)*.

• **Peak global ozone losses are expected to occur during the next several years.** The ozone layer will be most affected by human-influenced perturbations and susceptible to natural variations in the period around the year 1998, since the peak stratospheric chlorine and bromine abundances are expected to occur then. Based on extrapolation of current trends, observations suggest that the maximum ozone loss, relative to the late 1960s, will likely be:

(i)   about 12 - 13% at Northern midlatitudes in winter/spring (*i.e.*, about 2.5% above current levels);
(ii)   about 6 - 7% at Northern midlatitudes in summer/fall (*i.e.*, about 1.5% above current levels); and
(iii)   about 11% (with less certainty) at Southern midlatitudes on a year-round basis (*i.e.*, about 2.5% above current levels).

Such changes would be accompanied by 15%, 8%, and 13% increases, respectively, in surface erythemal radiation, if other influences such as clouds remain constant. Moreover, if there were to be a major volcanic eruption like that of Mt. Pinatubo, or if an extremely cold and persistent Arctic winter were to occur, then the ozone losses and UV increases could be larger in individual years.

• **Approaches to lowering stratospheric chlorine and bromine abundances are limited.** Further controls on ozone-depleting substances would not be expected to significantly change the timing or the magnitude of the peak stratospheric halocarbon abundances and hence peak ozone loss. However, there are four approaches that would steepen the initial fall from the peak halocarbon levels in the early decades of the next century:

(i)   If emissions of methyl bromide from agricultural, structural, and industrial activities were to be eliminated in the year 2001, then the integrated effective future chlorine loading above the 1980 level (which is related to the cumulative future loss of ozone) is predicted to be 13% less over the next 50 years relative to full compliance to the Amendments and Adjustments to the Protocol.
(ii)   If emissions of HCFCs were to be totally eliminated by the year 2004, then the integrated effective future chlorine loading above the 1980 level is predicted to be 5% less over the next 50 years relative to full compliance with the Amendments and Adjustments to the Protocol.
(iii)   If halons presently contained in existing equipment were never released to the atmosphere, then the integrated effective future chlorine loading above the 1980 level is predicted to be 10% less over the next 50 years relative to full compliance with the Amendments and Adjustments to the Protocol.
(iv)   If CFCs presently contained in existing equipment were never released to the atmosphere, then the integrated effective future chlorine loading above the 1980 level is predicted to be 3% less over the next 50 years relative to full compliance with the Amendments and Adjustments to the Protocol.

- **Failure to adhere to the international agreements will delay recovery of the ozone layer.** If there were to be additional production of CFCs at 20% of 1992 levels for each year through 2002 and ramped to zero by 2005 (beyond that allowed for countries operating under Article 5 of the Montreal Protocol), then the integrated effective future chlorine loading above the 1980 level is predicted to be 9% more over the next 50 years relative to full compliance to the Amendments and Adjustments to the Protocol.

- **Many of the substitutes for the CFCs and halons are also notable greenhouse gases.** Several CFC and halon substitutes are not addressed under the Montreal Protocol (because they do not deplete ozone), but, because they are greenhouse gases, fall under the purview of the Framework Convention on Climate Change. There is a wide range of values for the Global Warming Potentials (GWPs) of the HFCs (150 - 10000), with about half of them having values comparable to the ozone-depleting compounds they replace. The perfluorinated compounds, some of which are being considered as substitutes, have very large GWPs (*e.g.*, 5000 - 10000). These are examples of compounds whose current atmospheric abundances are relatively small, but are increasing or could increase in the future.

- **Consideration of the ozone change will be one necessary ingredient in understanding climate change.** The extent of our ability to attribute any climate change to specific causes will likely prove to be important scientific input to decisions regarding predicted human-induced influences on the climate system. Changes in ozone since pre-industrial times as a result of human activity are believed to have been a significant influence on radiative forcing; this human influence is expected to continue into the foreseeable future.

COMMON QUESTIONS ABOUT OZONE

Ozone is exceedingly rare in our atmosphere, averaging about 3 molecules of ozone for every ten million air molecules. Nonetheless, atmospheric ozone plays vital roles that belie its small numbers. This Appendix to the *World Meteorological Organization/United Nations Environment Programme (WMO/UNEP) Scientific Assessment of Ozone Depletion: 1994* answers some of the questions that are most commonly asked about ozone and the changes that have been occurring in recent years. These common questions and their answers were discussed by the 80 scientists from 26 countries who participated in the Panel Review Meeting of the *Scientific Assessment of Ozone Depletion: 1994*. Therefore, this information is presented by a large group of experts from the international scientific community.

Ozone is mainly found in two regions of the Earth's atmosphere. Most ozone (about 90%) resides in a layer between approximately 10 and 50 kilometers (about 6 to 30 miles) above the Earth's surface, in the region of the atmosphere called the stratosphere. This stratospheric ozone is commonly known as the "ozone layer." The remaining ozone is in the lower region of the atmosphere, the troposphere, which extends from the Earth's surface up to about 10 kilometers. The figure below shows this distribution of ozone in the atmosphere.

While the ozone in these two regions is chemically identical (both consist of three oxygen atoms and have the chemical formula "O_3"), the ozone molecules have very different effects on humans and other living things depending upon their location.

Stratospheric ozone plays a beneficial role by absorbing most of the biologically damaging ultraviolet sunlight called UV-B, allowing only a small amount to reach the Earth's surface. The absorption of UV radiation by ozone creates a source of heat, which actually forms the stratosphere itself (a region in which the temperature rises as one goes to higher altitudes). Ozone thus plays a key role in the temperature structure of the Earth's atmosphere. Furthermore, without the filtering action of the ozone layer, more of the Sun's UV-B radiation would penetrate the atmosphere and would reach the Earth's surface in greater amounts. Many experimental studies of plants and animals, and clinical studies of humans, have shown the harmful effects of excessive exposure to UV-B radiation (these are discussed in the WMO/UNEP reports on impacts of ozone depletion, which are com-

panion documents to the WMO/UNEP scientific assessments of ozone depletion).

At the planet's surface, ozone comes into direct contact with life-forms and displays its destructive side. Because ozone reacts strongly with other molecules, high levels are toxic to living systems and can severely damage the tissues of plants and animals. Many studies have documented the harmful effects of ozone on crop production, forest growth, and human health. The substantial negative effects of surface-level tropospheric ozone from this direct toxicity contrast with the benefits of the additional filtering of UV-B radiation that it provides.

With these dual aspects of ozone come two separate environmental issues, controlled by different forces in the atmosphere. In the troposphere, there is concern about *increases* in ozone. Low-lying ozone is a key component of smog, a familiar problem in the atmosphere of many cities around the world. Higher than usual amounts of surface-level ozone are now increasingly being observed in rural areas as well. However, the ground-level ozone concentrations in the smoggiest cities are very much smaller than the concentrations routinely found in the stratosphere.

There is widespread scientific and public interest and concern about *losses* of stratospheric ozone. Ground-based and satellite instruments have measured decreases in the amount of stratospheric ozone in our atmosphere. Over some parts of Antarctica, up to 60% of the total overhead amount of ozone (known as the "column ozone") is depleted during September and October. This phenomenon has come to be known as the Antarctic "ozone hole." Smaller, but still significant, stratospheric decreases have been seen at other, more-populated regions of the Earth. Increases in surface UV-B radiation have been observed in association with decreases in stratospheric ozone.

The scientific evidence, accumulated over more than two decades of study by the international research community, has shown that human-made chemicals are responsible for the observed depletions of the ozone layer over Antarctica and likely play a major role in global ozone losses. The ozone-depleting compounds contain various combinations of the chemical elements chlorine, fluorine, bromine, carbon, and hydrogen, and are often described by the general term *halocarbons*. The com-
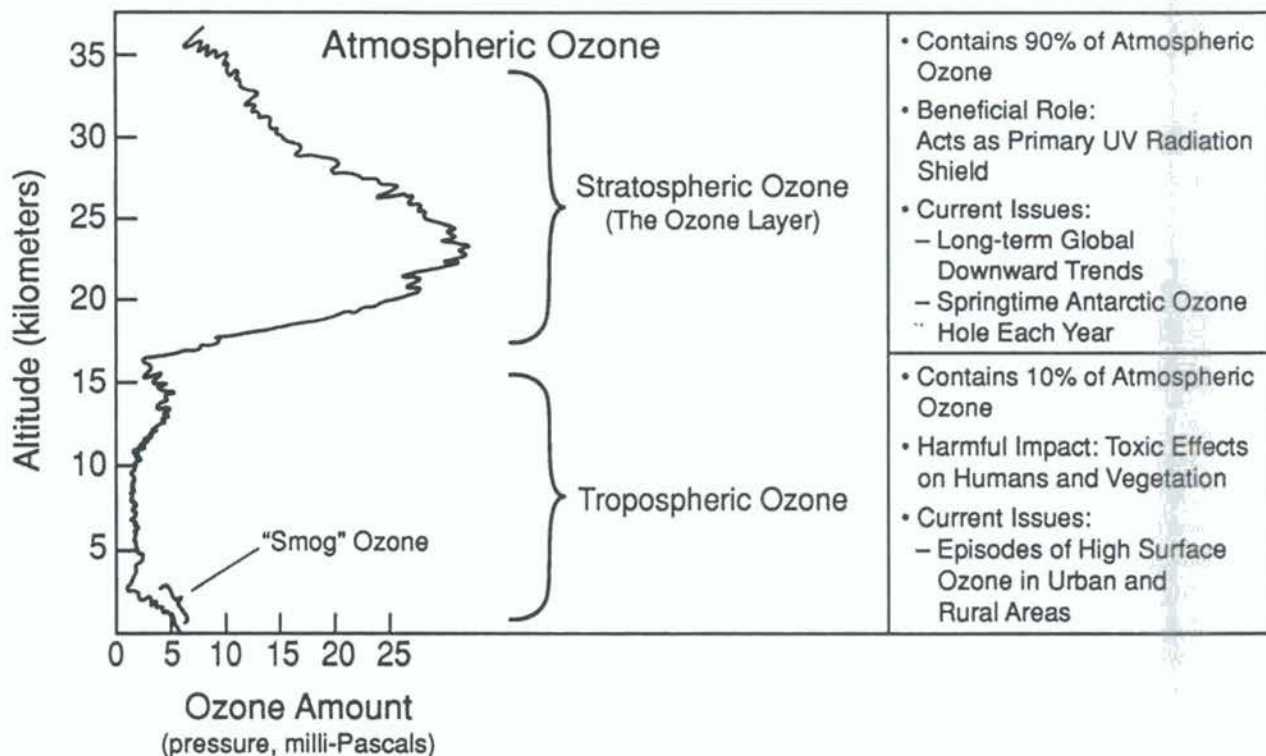
pounds that contain only carbon, chlorine, and fluorine are called *chlorofluorocarbons*, usually abbreviated as *CFCs*. CFCs, carbon tetrachloride, and methyl chloroform are important human-made ozone-depleting gases that have been used in many applications including refrigeration, air conditioning, foam blowing, cleaning of electronics components, and as solvents. Another important group of human-made halocarbons is the *halons*, which contain carbon, bromine, fluorine, and (in some cases) chlorine, and have been mainly used as fire extinguishants. Governments have decided to discontinue production of CFCs, halons, carbon tetrachloride, and methyl chloroform, and industry has developed more "ozone-friendly" substitutes.

Two responses are natural when a new problem has been identified: cure and prevention. When the problem is the destruction of the stratospheric ozone layer, the corresponding questions are: Can we repair the damage already done? How can we prevent further destruction? Remedies have been investigated that could (i) remove CFCs selectively from our atmosphere, (ii) intercept ozone-depleting chlorine before much depletion has taken place, or (iii) replace the ozone lost in the stratosphere (perhaps by shipping the ozone from cities that have too

much smog or by making new ozone). Because ozone reacts strongly with other molecules, as noted above, it is too unstable to be made elsewhere (e.g., in the smog of cities) and transported to the stratosphere. When the huge volume of the Earth's atmosphere and the magnitude of global stratospheric ozone depletion are carefully considered, approaches to cures quickly become much too expensive, impractical, and potentially damaging to the global environment. Prevention involves the internationally agreed-upon Montreal Protocol and its Amendments and Adjustments, which call for elimination of the production and use of the CFCs and other ozone-damaging compounds within the next few years. As a result, the ozone layer is expected to recover over the next fifty years or so as the atmospheric concentrations of CFCs and other ozone-depleting compounds slowly decay.

The current understanding of ozone depletion and its relation to humankind is discussed in detail by the leading scientists in the world's ozone research community in the *Scientific Assessment of Ozone Depletion: 1994.* The answers to the common questions posed below are based upon that understanding and on the information given in earlier WMO/UNEP reports.

Atmospheric Ozone

Altitude (kilometers) — Ozone Amount (pressure, milli-Pascals)

Stratospheric Ozone (The Ozone Layer)

• Contains 90% of Atmospheric Ozone
• Beneficial Role:
  Acts as Primary UV Radiation Shield
• Current Issues:
  – Long-term Global Downward Trends
  – Springtime Antarctic Ozone Hole Each Year

Tropospheric Ozone

"Smog" Ozone

• Contains 10% of Atmospheric Ozone
• Harmful Impact: Toxic Effects on Humans and Vegetation
• Current Issues:
  – Episodes of High Surface Ozone in Urban and Rural Areas

How Can Chlorofluorocarbons (CFCs) Get to the Stratosphere If They're Heavier than Air?

Although the CFC molecules are indeed several times heavier than air, thousands of measurements have been made from balloons, aircraft, and satellites demonstrating that the CFCs are actually present in the stratosphere. The atmosphere is not stagnant. Winds mix the atmosphere to altitudes far above the top of the stratosphere much faster than molecules can settle according to their weight. Gases such as CFCs that are insoluble in water and relatively unreactive in the lower atmosphere (below about 10 km) are quickly mixed and therefore reach the stratosphere regardless of their weight.

Much can be learned about the atmospheric fate of compounds from the measured changes in concentration versus altitude. For example, the two gases carbon tetrafluoride (CF_4, produced mainly as a by-product of the manufacture of aluminum) and CFC-11 (CCl_3F, used in a variety of human activities) are both much heavier than air. Carbon tetrafluoride is completely unreactive in the lower 99.9% of the atmosphere, and measurements show it to be nearly uniformly distributed throughout the atmosphere as shown in the figure. There have also been measurements over the past two decades of several other completely unreactive gases, one lighter than air (neon) and some heavier than air (argon, krypton), which show that they also mix upward uniformly through the stratosphere regardless of their weight, just as observed with carbon tetrafluoride. CFC-11 is unreactive in the lower atmosphere (below about 15 km) and is similarly uniformly mixed there, as shown. The abundance of CFC-11 decreases as the gas reaches higher altitudes, where it is broken down by high energy solar ultraviolet radiation. Chlorine released from this breakdown of CFC-11 and other CFCs remains in the stratosphere for several years, where it destroys many thousands of molecules of ozone.

Measurements of CFC-11 and CF₄
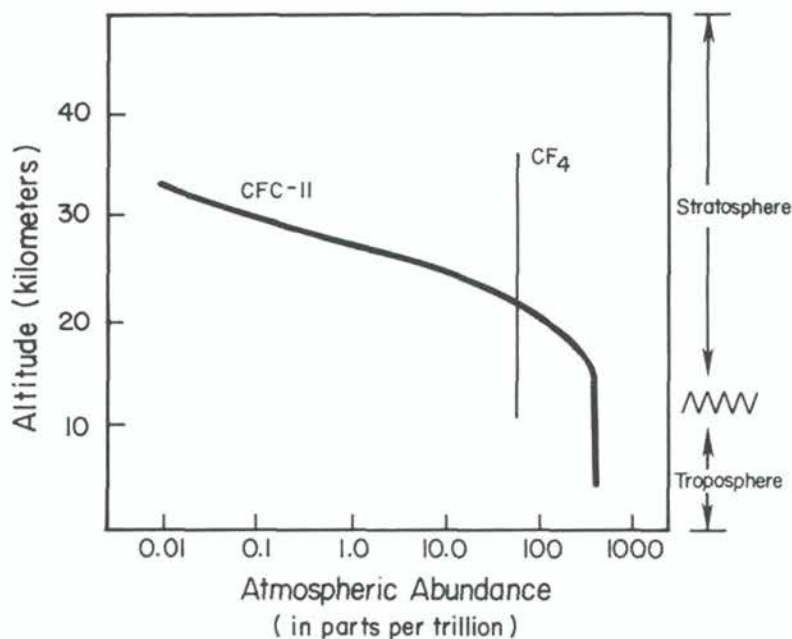
What is the Evidence that Stratospheric Ozone is Destroyed by Chlorine and Bromine?

Laboratory studies show that chlorine (Cl) reacts very rapidly with ozone. They also show that the reactive chemical chlorine oxide (ClO) formed in that reaction can undergo further processes which regenerate the original chlorine, allowing the sequence to be repeated very many times (a "chain reaction"). Similar reactions also take place between bromine and ozone.

But do these ozone-destroying reactions occur in the real world? All of our accumulated scientific experience demonstrates that if the conditions of temperature and pressure are like those in the laboratory studies, the same chemical reactions will take place in nature. However, many other reactions including those of other chemical species are often also taking place simultaneously in the stratosphere, making the connections among the changes difficult to untangle. Nevertheless, whenever chlorine (or bromine) and ozone are found together in the stratosphere, the ozone-destroying reactions must be taking place.

Sometimes a small number of chemical reactions is so important in the natural circumstance that the connections are almost as clear as in laboratory experiments. Such a situation occurs in the Antarctic stratosphere during the springtime formation of the ozone hole. During August and September 1987 — the end of winter and beginning of spring in the Southern Hemisphere — aircraft equipped with many different instruments for measuring a large number of chemical species were flown repeated-ly over Antarctica. Among the chemicals measured were ozone and chlorine oxide, the reactive chemical identified in the laboratory as one of the participants in the ozone-destroying chain reactions. On the first flights southward from the southern tip of South America, relatively high concentrations of ozone were measured everywhere over Antarctica. By mid-September, however, the instruments recorded low concentrations of ozone in regions where there were high concentrations of chlorine oxide and vice versa, as shown in the figure. Flights later in September showed even less ozone over Antarctica, as the chlorine continued to react with the stratospheric ozone.

Independent measurements made by these and other instruments on this and other airplanes, from the ground, from balloons, and from satellites have provided a detailed understanding of the chemical reactions going on in the Antarctic stratosphere. Regions with high concentrations of reactive chlorine reach temperatures so cold (less than approximately -80°C, or -112°F) that stratospheric clouds form, a rare occurrence except during the polar winters. These clouds facilitate other chemical reactions that allow the release of chlorine in sunlight. The chemical reactions related to the clouds are now well understood through study under laboratory conditions mimicking those found naturally. Scientists are working to understand the role of such reactions of chlorine and bromine at other latitudes, and the involvement of particles of sulfuric acid from volcanoes or other sources.

Measurements of Ozone and Reactive Chlorine from a Flight into the Antarctic Ozone Hole
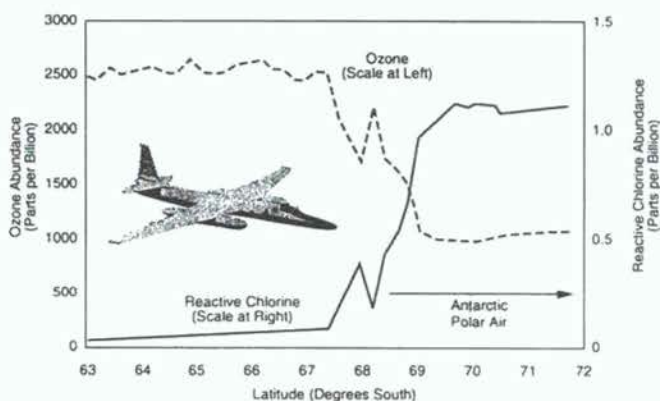
Does Most of the Chlorine in the Stratosphere Come from Human or Natural Sources?

Most of the chlorine in the stratosphere is there as a result of human activities.

Many compounds containing chlorine are released at the ground, but those that dissolve in water cannot reach stratospheric altitudes. Large quantities of chlorine are released from evaporated ocean spray as sea salt (sodium chloride) aerosol. However, because sea salt dissolves in water, this chlorine quickly is taken up in clouds or in ice, snow, or rain droplets and does not reach the stratosphere. Another ground-level source of chlorine is its use in swimming pools and as household bleach. When released, this chlorine is rapidly converted to forms that dissolve in water and therefore are removed from the lower atmosphere, never reaching the stratosphere in significant amounts. Volcanoes can emit large quantities of hydrogen chloride, but this gas is rapidly converted to hydrochloric acid in rain water, ice, and snow and does not reach the stratosphere. Even in explosive volcanic plumes that rise high in the atmosphere, nearly all of the hydrogen chloride is scrubbed out in precipitation before reaching stratospheric altitudes.

In contrast, human-made halocarbons – such as CFCs, carbon tetrachloride (CCl_4) and methyl chloroform (CH_3CCl_3) – are not soluble in water, do not react with snow or other natural surfaces, and are not broken down chemically in the lower atmosphere. While the exhaust from the Space Shuttle and from some rockets does inject some chlorine directly into the stratosphere, this input is very small (less than one percent of the annual input from halocarbons in the present stratosphere, assuming nine Space Shuttle and six Titan IV rocket launches per year).

Several pieces of evidence combine to establish human-made halocarbons as the primary source of stratospheric chlorine. First, measurements (see the figure below) have shown that the chlorinated species that rise to the stratosphere are primarily manufactured compounds (mainly CFCs, carbon tetrachloride, methyl chloroform, and the HCFC substitutes for CFCs), together with small amounts of hydrochloric acid (HCl) and methyl chloride (CH_3Cl) which are partly natural in origin. The natural contribution now is much smaller than that from human activities, as shown in the figure below. Second, in 1985 and 1992 researchers measured nearly all known gases containing chlorine in the stratosphere. They found that human emissions of halocarbons plus the much smaller contribution from natural sources could account for all of the stratospheric chlorine compounds. Third, the *increase* in total stratospheric chlorine measured between 1985 and 1992 corresponds with the known increases in concentrations of human-made halocarbons during that time.

Primary Sources of Chlorine Entering the Stratosphere
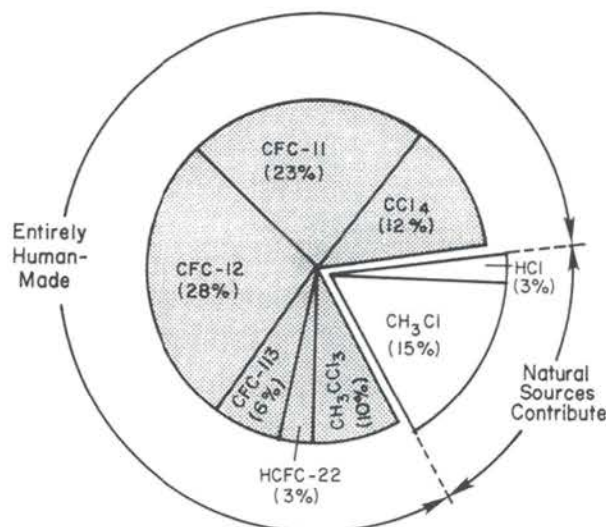
Can Changes in the Sun's Output Be Responsible for the Observed Changes in Ozone?

Stratospheric ozone is primarily created by ultraviolet (UV) light coming from the Sun, so the Sun's output affects the rate at which ozone is produced. The Sun's energy release (both as UV light and as charged particles such as electrons and protons) does vary, especially over the well-known 11-year sunspot cycle. Observations over several solar cycles (since the 1960s) show that total global ozone levels decrease by 1-2% from the maximum to the minimum of a typical cycle. Changes in the Sun's output cannot be responsible for the observed long-term changes in ozone, because these downward trends are much larger than 1-2%. Further, during the period since 1979, the Sun's energy output has gone from a maximum to a minimum in 1985 and back through another maximum in 1991, but the trend in ozone was downward throughout that time. The ozone trends presented in this and previous international scientific assessments have been obtained by evaluating the long-term changes in ozone concentrations after accounting for the solar influence (as has been done in the figure below).

Global Ozone Trend (60°S–60°N)
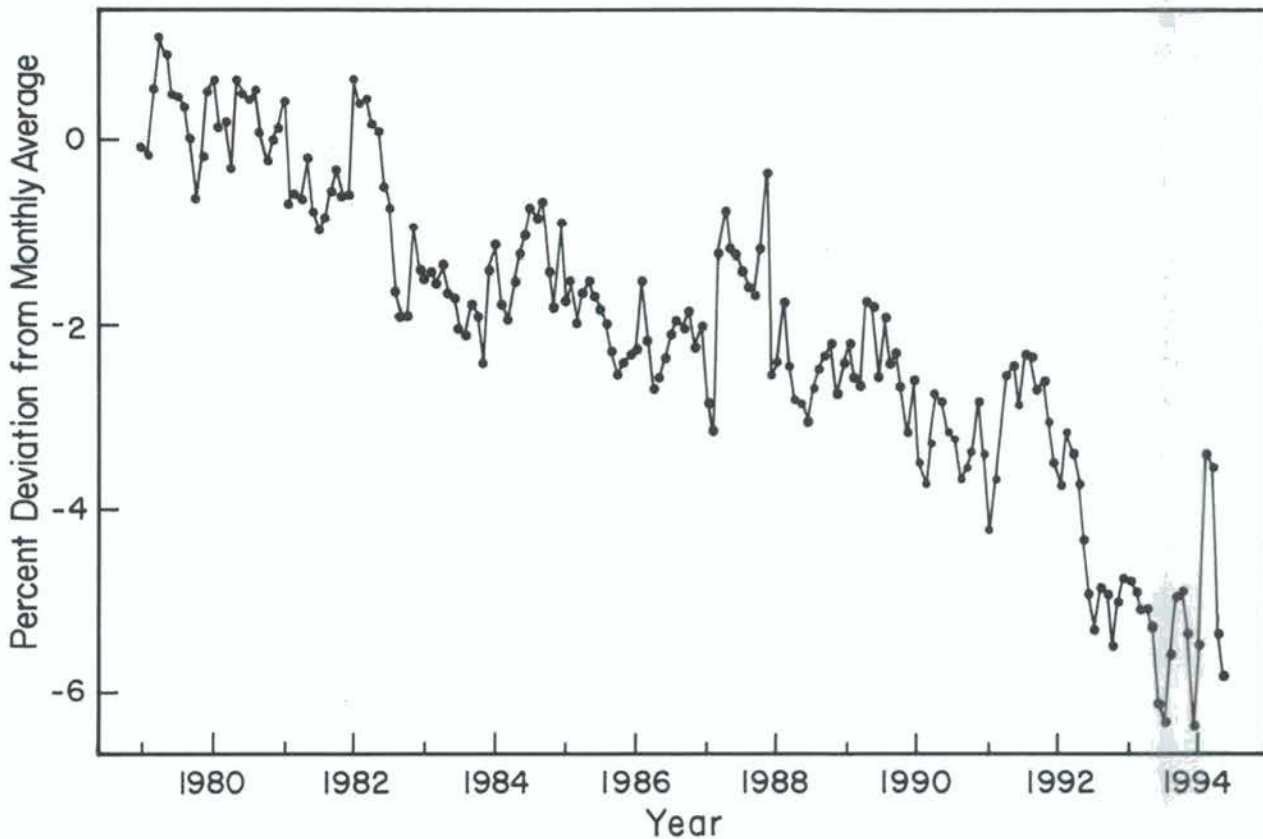
When Did the Antarctic Ozone Hole First Appear?

The Antarctic ozone hole is a new phenomenon. The figure shows that observed ozone over the British Antarctic Survey station at Halley Bay, Antarctica first revealed obvious decreases in the early 1980s compared to data obtained since 1957. The ozone hole is formed each year when there is a sharp decline (currently up to 60%) in the total ozone over most of Antarctica for a period of about two months during Southern Hemisphere spring (September and October). Observations from three other stations in Antarctica, also covering several decades, reveal similar progressive, recent decreases in springtime ozone. The ozone hole has been shown to result from destruction of stratospheric ozone by gases containing chlorine and bromine, whose sources are mainly human-made halocarbon gases.

Before the stratosphere was affected by human-made chlorine and bromine, the naturally occurring springtime ozone levels over Antarctica were about 30-40% lower than springtime ozone levels over the Arctic. This natural difference between Antarctic and Arctic conditions was first observed in the late 1950s by Dobson. It stems from the exceptionally cold temperatures and different winter wind patterns within the Antarctic stratosphere as compared to the Arctic. This is not at all the same phenomenon as the marked downward trend in total ozone in recent years referred to as the ozone hole and shown in the figure below.

Changes in stratospheric meteorology cannot explain the ozone hole. Measurements show that wintertime Antarctic stratospheric temperatures of past decades have not changed prior to the development of the hole each September. Ground, aircraft, and satellite measurements have provided, in contrast, clear evidence of the importance of the chemistry of chlorine and bromine originating from human-made compounds in depleting Antarctic ozone in recent years.

A single report of extremely low Antarctic winter ozone in one location in 1958 by an unproven technique has been shown to be completely inconsistent with the measurements depicted here and with all credible measurements of total ozone.

Historical Springtime Total Ozone Record for Halley Bay, Antarctica (76°S)

October Monthly Averages
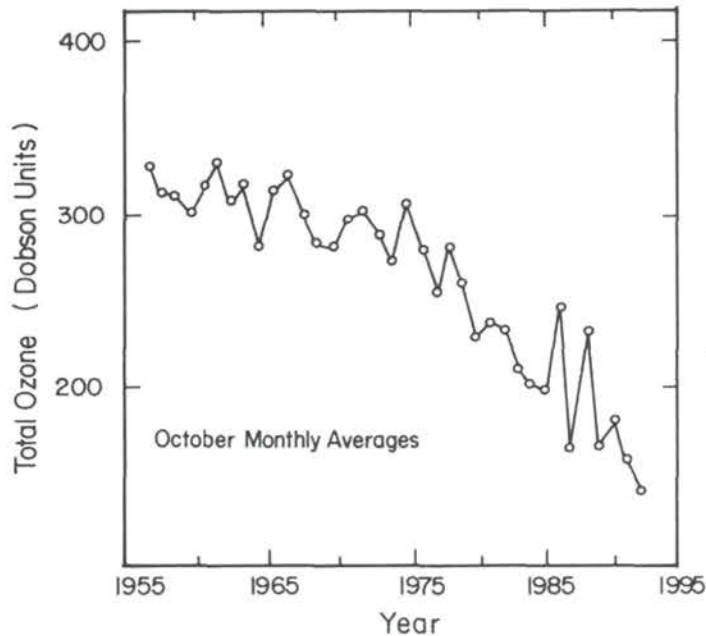
Why is the Ozone Hole Observed over Antarctica When CFCs Are Released Mainly in the Northern Hemisphere?

Human emissions of CFCs do occur mainly in the Northern Hemisphere, with about 90% released in the latitudes corresponding to Europe, Russia, Japan, and North America. Gases such as CFCs that are insoluble in water and relatively unreactive are mixed within a year or two throughout the lower atmosphere (below about 10 km). The CFCs in this well-mixed air rise from the lower atmosphere into the stratosphere mainly in tropical latitudes. Winds then move this air poleward — both north and south — from the tropics, so that air throughout the stratosphere contains nearly the same amount of chlorine. However, the meteorologies of the two polar regions are very different from each other because of major differences at the Earth's surface. The South Pole is part of a very large land mass (Antarctica) that is com-

pletely surrounded by ocean. These conditions produce very low stratospheric temperatures which in turn lead to formation of clouds (polar stratospheric clouds). The clouds that form at low temperatures lead to chemical changes that promote rapid ozone loss during September and October of each year, resulting in the ozone hole.

In contrast, the Earth's surface in the northern polar region lacks the land/ocean symmetry characteristic of the southern polar area. As a consequence, Arctic stratospheric air is generally much warmer than in the Antarctic, and fewer clouds form there. Therefore, the ozone depletion in the Arctic is much less than in the Antarctic.

Schematic of Antarctic Ozone Hole

1979   1986   1991
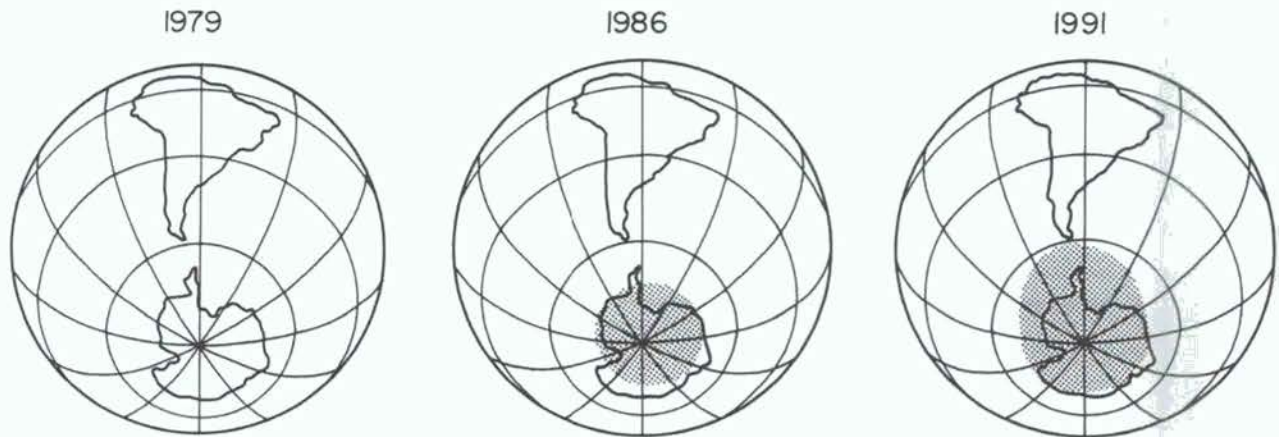
Is the Depletion of the Ozone Layer Leading to an Increase in Ground-Level Ultraviolet Radiation?

The Sun emits light over a wide range of energies, with about two percent given off in the form of high-energy, ultraviolet (UV) radiation. Some of this UV radiation (UV-B) is especially effective in causing damage to living things, including sunburn, skin cancer, and eye damage for humans. The amount of solar UV radiation received at any particular location on the Earth's surface depends upon the position of the Sun above the horizon, on the amount of ozone in the atmosphere, and upon local cloudiness and pollution. Scientists agree that in the absence of changes in clouds or pollution, decreases in atmospheric ozone will increase ground-level UV radiation.

The largest decreases in ozone during the last decade have been observed over Antarctica, especially during each September and October when the "ozone hole" forms. During the last several years, simultaneous measurements of UV radiation and total ozone have been made at several Antarctic stations. As shown in the figure below, when the ozone amounts decrease, UV-B increases. Because of the ozone hole, the UV-B intensity at Palmer Station, Antarctica, in late October, 1993, was more intense than found at San Diego, California, at any time during all of 1993.

In areas where small ozone depletion has been observed, UV-B increases are more difficult to detect. Detection of UV trends associated with ozone decreases can also be complicated by changes in cloudiness or by local pollution, as well as by difficulties in keeping the detection instrument in precisely the same condition over many years. Prior to the late 1980s, instruments with the necessary accuracy and stability for measurement of small long-term trends in ground-level UV-B were not employed. Recently, however, such instruments have been used in the Antarctic because of the very large changes in ozone being observed there. When high-quality measurements have been made in other areas far from major cities and their associated air pollution, decreases in ozone have regularly been accompanied by increases in UV-B. The data from urban locations with older, less specialized instruments provide much less reliable information, especially because good simultaneous measurements are not available for any changes in cloudiness or local pollution.

Increases in Erythemal (Sunburning) UV Radiation Due to Ozone Reductions

Change In Ozone
(Spring vs. Autumn, for the Same Solar Angle)
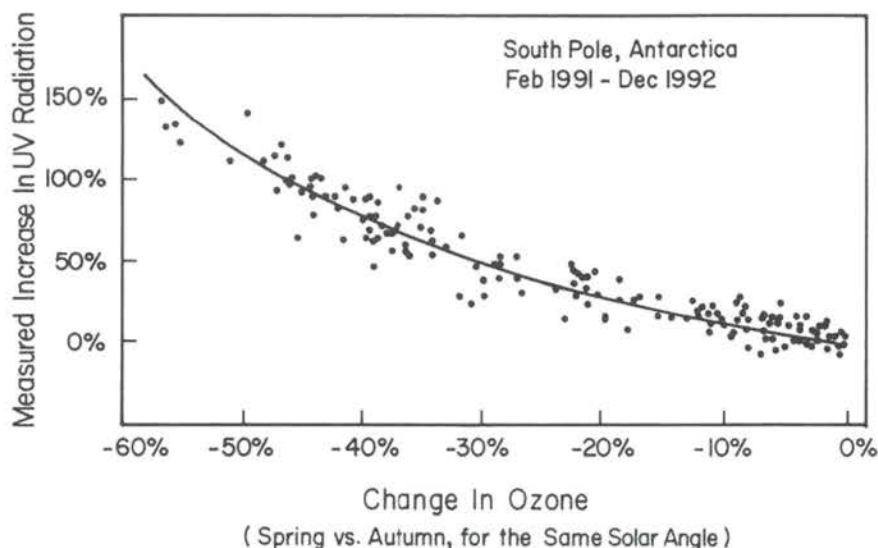
How Severe Is the Ozone Depletion Now, and Is It Expected to Get Worse?

Scientific evidence shows that ozone depletion caused by human-made chemicals is continuing and is expected to persist until chlorine and bromine levels are reduced. Worldwide monitoring has shown that stratospheric ozone has been decreasing for the past two decades or more. Globally averaged losses have totaled about 5% since the mid-1960s, with cumulative losses of about 10% in the winter and spring and 5% in the summer and autumn over locations such as Europe, North America, and Australia. Since the late-1970s, an ozone "hole" has formed in Antarctica each Southern Hemisphere spring (September / October), in which up to 60% of the total ozone is depleted. The large increase in atmospheric concentrations of human-made chlorine and bromine compounds is responsible for the formation of the Antarctic ozone hole, and the weight of evidence indicates that it also plays a major role in midlatitude ozone depletion.

During 1992 and 1993 ozone in many locations dropped to record low values: springtime depletions exceeded 20% in some populated northern midlatitude regions, and the levels in the Antarctic ozone hole fell to the lowest values ever recorded. The unusually large ozone decreases of 1992 and 1993 are believed to be related, in part, to the volcanic eruption of Mount Pinatubo in the Philippines during 1991. This eruption produced large amounts of stratospheric sulfate aerosols that temporarily increased the ozone depletion caused by human-made chlorine and bromine compounds. Recent observations have shown that as those aerosols have been swept out of the stratosphere, ozone concentrations have returned to the depleted levels consistent with the downward trend observed before the Mount Pinatubo eruption.

In 1987 the recognition of the potential for chlorine and bromine to destroy stratospheric ozone led to an international agreement (The United Nations Montreal Protocol on Substances that Deplete the Ozone Layer) to reduce the global production of ozone-depleting substances. Since then, new global observations of significant ozone depletion have prompted amendments to strengthen the treaty. The 1992 Copenhagen Amendments call for a ban on production of the most damaging compounds by 1996. The figure shows past and projected future stratospheric abundances of chlorine and bromine: (a) without the Protocol; (b) under the Protocol's original provisions; and (c) under the Copenhagen Amendments now in force. Without the Montreal Protocol and its Amendments, continuing human use of CFCs and other compounds would have tripled the stratospheric abundances of chlorine and bromine by about the year 2050. Current scientific understanding indicates that such increases would have led to global ozone depletion very much larger than observed today. In contrast, under current international agreements, which are now reducing and will eventually eliminate human emissions of ozone-depleting gases, the stratospheric abundances of chlorine and bromine are expected to reach their maximum within a few years and then slowly decline. All other things being equal, the ozone layer is expected to return to normal by the middle of the next century.

In summary, record low ozone levels have been observed in recent years, and substantially larger future global depletions in ozone would have been highly likely without reductions in human emissions of ozone-depleting gases. However, worldwide compliance with current international agreements is rapidly reducing the yearly emissions of these compounds. As these emissions cease, the ozone layer will gradually improve over the next several decades. The recovery of the ozone layer will be gradual because of the long times required for CFCs to be removed from the atmosphere.

Ozone-Damaging Stratospheric Chlorine/Bromine
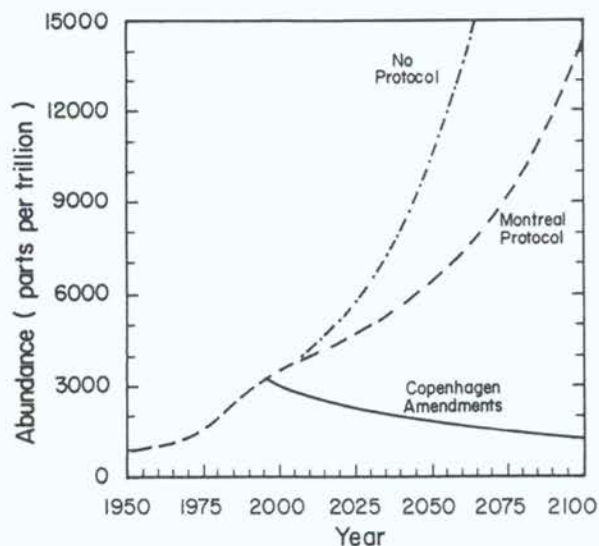
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Future Chlorine/Bromine Loading *(Susan Solomon and Donald J. Wuebbles)*

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