

STRATOSPHERIC OZONE 1999



United Kingdom Stratospheric Ozone Review Group

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A report from the Stratospheric Ozone Review Group including an update by the Ultraviolet Measurements and Impacts Review Group

Prepared at the request of the Department of the Environment, Transport and the Regions

Front cover

Base F (65°15′S, 64°16′W) of the Falkland Islands Dependencies Survey on the Argentine Islands (off the West coast of the Antarctic Peninsula) in January 1958. Ozone measurements were started here in 1957 and have been made continuously to the present day. The Dobson spectrophotometer was mounted in the small extension (coated with aluminium paint) in the centre of the roof. (In 1962 the name of the Survey was changed to the British Antarctic Survey. In 1977 the base was renamed Faraday Station. In February 1995 it was handed over to Ukraine and is now called Vernadsky.) (Courtesy of J.C. Farman)

Back cover

A Montgolfier Infra-Red balloon being launched at the Swedish Space Corporation base, Esrange (67°56'N, 21°04'E), in northern Sweden during February 1999. These long duration balloons have been recently developed by the Centre National d'Études Spatiales (CNES) for use in the Arctic stratosphere. They have flown for several weeks as part of the Third European Stratospheric Experiment on Ozone (THESEO) and valuable scientific measurements were made using instruments from the UK, France, Italy and Russia. (Courtesy of G. Hansford)

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Stratospheric Ozone 1999

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Executive Summary

In its six earlier reports the Stratospheric Ozone Review Group (SORG) has presented assessments of the scientific issues underlying the world-wide changes taking place in the ozone layer and the causes of these changes.

The reports showed that there had been a steady decrease in stratospheric ozone over all latitudes outside the tropics since the late 1970s, with the largest reductions in total ozone occurring over Antarctica each spring – the "ozone hole". Over the same period there had been a steady increase in the amounts of chlorine and bromine present in the stratosphere as a result of human activity. In SORG 1993 it was reported that the rate of increase of chlorine had slowed in response to regulation under the Montreal Protocol and, in SORG 1996, that the chlorine loading of the troposphere had started to decline.

It has been demonstrated unequivocally that chlorine and bromine compounds lead to chemical destruction of stratospheric ozone and that their presence could account quantitatively for the formation of the ozone hole. The 1996 report showed that chemical destruction of ozone had occurred in the Arctic stratosphere and that chlorine and bromine compounds were also implicated in the decline of ozone in middle latitudes.

Early reports noted that ultraviolet radiation at the earth's surface was expected to increase as a result of ozone loss and this had been observed under the ozone hole, where the effect is large. Ozone depletion had also been at least partly responsible for observed decreases in stratospheric temperatures as well as offsetting, in part, the surface warming effect arising from increases of greenhouse gases.

The implications of increased ultraviolet radiation (UV) at the surface were addressed in the first report of the Ultraviolet Measurements and Impacts Review Group (UMIRG) in 1996. This discussed the effects of UV on human and animal health, terrestrial and aquatic ecosystems, tropospheric chemistry and material degradation. Those aspects of the first UMIRG report not within the SORG remit have been updated in this report by UMIRG.

Update on ozone measurements

- The Antarctic ozone hole in each late winter and spring continues unabated. By early October, large areas over Antarctica in the altitude range from 14 to 21 km are practically devoid of ozone. The ozone hole has lasted longer during the 1990s.
- Observable chemical loss of ozone has occurred in all Arctic winters since 1991/92 despite the very different meteorological conditions. In stratospheric winters with long periods of low temperatures, conditions which are conducive for ozone depletion, the Arctic loss has been severe (estimated at around 50% near 20 km altitude in 1996/97). Even in the warmer stratospheric winters smaller but significant loss has

been reported (estimated at around 25% near 20 km in 1997/98). The ozone loss is broadly consistent with our understanding and strengthens the conclusion of earlier SORG reports that, despite reductions in halogen loading, large ozone loss in the Arctic can be expected into the 21st century, depending on the meteorological conditions in individual winters.

- In the late 1990s, annually averaged total ozone values from the TOMS satellite and ground-based measurements at northern mid-latitudes (25°–60°N) were 4–6% lower than those around 1980. The low values in 1992/93 following the eruption of Mt Pinatubo in 1991 have been a dominant feature of total ozone in the 1990s.
- Globally, annually averaged (60°S–60°N) total ozone values derived from ground-based measurements alone were 3–4 % lower in the late 1990s than in the 1970s. The ground-based measurements indicate that losses in the southern mid-latitudes are similar to those in the northern mid-latitudes. (The calibration of TOMS measurements since 1996 is preliminary. There is a 1–2% discrepancy between TOMS and ground-based measurements at low latitudes and in the southern hemisphere in this period. We have chosen to use the ground-based data here.)
- The observed mid-latitude ozone decline is broadly consistent with halogen-catalysed chemical destruction being the primary cause. Model studies can now explain the trend in ozone in middle latitudes since 1980 in terms of reactions involving chlorine and bromine compounds. However, other studies show that meteorological factors have also contributed to the trend. The precise quantification of the different processes is still required.
- The interpretation of the observed changes in total ozone since the eruption of Mt Pinatubo is difficult and in recent years depends quite strongly on the assumptions made about the influence of natural variability on ozone. The low ozone immediately following the eruption can be explained. The high ozone at northern mid-latitudes in 1998 is probably due to stronger atmospheric transport of ozone rich air. The updated observational record since WMO (1999) shows values closer to, but slightly above, the long term linear trend extrapolated from 1979–1991. However, we do not think that the observational record to date contains any evidence of an ozone recovery in response to reductions of ozone-depleting substances.
- Fifteen years of water vapour measurements above Colorado show an increase of 1%/year between 18 and 28 km. Satellite measurements over a shorter period suggest that an increase has occurred globally throughout the stratosphere. The lack of a complete explanation for the increase, which exceeds that expected from the increase in methane, is of concern because stratospheric water vapour is important for both climate and stratospheric chemistry.

Halocarbons in the Atmosphere

- The chlorine loading of the atmosphere should continue to fall and bromine loading is expected to peak early in the 21st century.
- There are substantial quantities of ozone depleting substances still in equipment which are likely to be released to the atmosphere in the future. Given the existing controls, the largest additional reduction that can be made to future chlorine and bromine loadings would be gained if these materials were removed and destroyed when the equipment containing them is decommissioned. Over the next 50 years, such reductions could amount to 5% in total cumulative chlorine loading and 25% in bromine.
- HCFC releases have a relatively small effect on stratospheric chlorine loading. A proposed regulation in the EU seeks to reduce production and use of HCFCs beyond the provisions of the Montreal Protocol with a phase-out early next century. If such a regulation were applied, globally it would enhance the reduction in total cumulative chlorine loading by slightly less than 1% over the next 50 years.
- Methyl bromide is the largest contributor of bromine to the stratosphere. Its budget is still uncertain despite a number of improvements in understanding since the 1994 WMO assessment.
- The methyl bromide ozone depletion potential (ODP) has been reevaluated downwards since the WMO 1994 report, based on a decreased estimate of the methyl bromide lifetime (which reduces the ODP) and an increased estimate of the ozone-depleting efficiency of bromine (which increases the ODP). The reduction in the lifetime also implies a larger flux of methyl bromide into the atmosphere than was assumed previously. Overall, there is no reduction in our assessment of the importance of methyl bromide in stratospheric ozone destruction.
- Analysis of air in Antarctic firn (unconsolidated snow) suggests concentrations of methyl bromide have increased by 20–25% during the 20th Century, with the most rapid growth occurring in the 1970s and 1980s. Despite the uncertainties in the atmospheric budget of methyl bromide, the firn air data suggest concentrations of methyl bromide to have been strongly influenced by human activities. It seems likely that less than half of the anthropogenic methyl bromide emissions are controlled under the Montreal Protocol.

Atmospheric Impacts

• For the period 1979–1997, the annual, global average temperature has decreased by 0.6 K/decade in the lower stratosphere and 3 K/decade in the upper stratosphere. Calculations indicate that much of the observed temperature

trend in the lower stratosphere can be explained by the observed ozone trend. In the upper stratosphere, the cooling is due largely to increases in CO_2 and decreases in ozone.

- Stratospheric ozone loss is estimated to have offset around 30% of the surface warming effect of increases in greenhouse gases since the late 1970s. One serious uncertainty in estimates of the global climate effect of ozone loss arises from inadequate knowledge of the vertical profile of ozone change in the tropics.
- Stratospheric halogen loading is not expected to return to a similar value to that which occurred before 1980 (when the ozone hole became clearly evident in the observational record) until about 2050. With the reduction in halogen loading, stratospheric ozone should slowly recover towards its pre-1980 concentrations. However, the future abundance of ozone will also be influenced by changes in other atmospheric gases and aerosols, and by interactions with the climate system, so that ozone recovery may not be a simple, slow return to earlier values.
- Spring Arctic and Antarctic ozone depletions are predicted to reach their peaks up to about 15 years later than the peak in halogen loadings because of the coupling between stratospheric climate change and ozone chemistry. The amount of maximum depletion and its timing are uncertain due to the complexity of this interaction.
- Sufficient observations exist to show a statistically significant anticorrelation between surface ultraviolet radiation (UV) and total ozone. Spectrally-resolved UV data from Antarctica and an increasing number of northern mid-latitude stations have shown increases in UVB radiation during the last decade, consistent with the locally observed ozone depletion.

Impacts of UV Change on the Biosphere

- The dose of UVB received by the human population is strongly dependent on behaviour. Social changes over several decades have served to increase exposure to UVB, which would have been exacerbated by ozone depletion.
- Climate change may modify ambient UV, for example by changes in cloud cover. Alterations in UK climate (such as warmer weather) are likely to further influence patterns of behaviour and thus doses of UV.
- Increased UVB would modify interactions between organisms in both terrestrial and aquatic systems, with unpredictable consequences for ecosystem function and diversity. Further uncertainty arises from the interaction between UVB and other environmental pressures.
- Localised extreme increases in UVB, for example over the Antarctic, may have wider consequences via aquatic food webs.



Background to the Seventh Report of the Stratospheric Ozone Review Group

This is the seventh in a series of reviews of our understanding of the stratospheric ozone layer and the risk to it from human activities. Prepared at the request of the Department of the Environment, Transport and the Regions, the reviews provide independent, up-to-date scientific assessments which inform the development of policy on the control of ozone depleting substances.

Since the last SORG report (1996), continued depletion of the ozone layer has been observed across the globe. New measurements and studies since 1996 have advanced our understanding of the factors which influence the state of the ozone layer and the impact of ozone depletion. The Department of the Environment, Transport and the Regions therefore asked for a new report reviewing the latest scientific information.

Concern that some halogenated hydrocarbons may destroy ozone in the upper atmosphere was first raised in the 1970s. Following the realisation that these man-made chemicals posed a threat to the ozone layer, the Vienna Convention for the Protection of the Ozone Layer was adopted by 28 countries in 1985. This committed signatories to a general obligation to take appropriate actions to protect the ozone layer and to co-operate on research. Shortly after the Vienna meeting, the ozone hole over the Antarctic was discovered, leading to renewed pressure to control potential ozone depleting substances. In 1987, the Montreal Protocol on Substances that Deplete the Ozone Layer was agreed and has since been ratified by over 160 countries. Initially, the Protocol imposed clear limits on the future production of CFCs and halons only and committed Parties to cutting production by 50% by the year 2000.

Over the next few years, the scientific links between stratospheric chlorine and bromine and ozone loss processes were firmly established, and the results from global monitoring networks showed continuing ozone depletion at all latitudes except the tropics. In response to this increased scientific understanding and a clear indication that existing controls were insufficient to prevent further build up of ozone depleting substances in the atmosphere, subsequent amendments to the Montreal Protocol (London, 1990; Copenhagen, 1992; Montreal, 1997) strengthened the control of ozone depleting substances. The phase-out of substances already regulated was accelerated, and other chemicals found to cause ozone depletion were included: carbon tetrachloride, methyl chloroform, HCFCs and methyl bromide (CH₃Br).

The Montreal Protocol has resulted in the successful phase-out of the production and consumption of several major classes of ozone depleting substances in developed countries. Production of halons ceased on 1 January 1994 and of CFCs, methyl chloroform and carbon tetrachloride on 1 January 1996. Within the European Union (EU), phase-out of CFCs and carbon tetrachloride was achieved one year earlier. In some cases provision was made to allow production for designated 'essential uses'. As a direct result of these controls, there has been a marked reduction in emissions of substances into the troposphere and, assuming that the Protocol is fully implemented, this should continue into the future thus reducing the atmospheric burden of chlorine and bromine.

For industrialised nations, current work under the Montreal Protocol is focused on the continuing phase-out of HCFCs and CH₃Br. Unlike most other ozone depleting substances there are both natural and man-made sources of CH₃Br emissions, while HCFCs are being used increasingly as transitional replacements for CFCs in some applications.

Controls on HCFCs and CH₃Br were strengthened at the meetings of the Parties held in Vienna in 1995 and Montreal in 1997. The Montreal Protocol provides a more relaxed control schedule on ozone depleting substances for developing countries (see Table 1). This only applies if the annual level of consumption of the substance is less than 0.3 kg per capita on the date of entry into force of the Protocol for that particular substance. EU controls on ozone depleting substances are already tighter than the Montreal Protocol, see Table 1. Negotiations are currently underway on a proposed new EC Regulation which would introduce even stricter controls. The main proposals include:

- Introduction of HCFC production controls with phase-out by 2025;
- General ban on the use and supply of all ozone depleting substances except HCFCs and CH₃Br;
- Ban on use of halon in fire protection systems deferred until 31 December 2002 with mandatory decommissioning of noncritical fire protection systems a year later;
- CH₃Br supply and production to be phased out before 2005, with interim cuts of 60% in 2001 and 75% in 2003;
- Tighter HCFC use controls.

Table 1

Substance	EC Regulation 3093/94	Montreal Protocol (Developed Countries)	Montreal Protocol (Developing Countries) Base level: 1995–97 Freeze from 1.7.99 50% cut: 2005 85% cut: 2007 Phase-out: 2010*		
CFCs production and consumption	Phase-out by 1.1.95*	Phase-out by 1.1.96*			
Carbon tetrachloride production and consumption	Phase-out by 1.1.95*	Phase-out by 1.1.96*	Base level: 1998–2000 85% cut: 2005 Phase-out: 2010*		
Halons production and consumption	Phase-out by 1.1.94*	Phase-out by 1.1.94*	Base level: 1995–97 Freeze from 1.1.2002 50% cut: 2005 Phase-out: 2010*		
1,1,1, Trichloroethane (Methyl chloroform) production and consumption	Phase-out by 1.1.96*	Phase-out by 1.1.96*	Base level: 1998–2000 Freeze from 1.1.2003 20% cut: 2005 70% cut: 2010 Phase-out: 2015*		
Methyl bromide production and consumption	Freeze at 1991 levels from 1.1.95 25% cut: 1.1.98	Freeze at 1991 levels from 1.1.95 25% cut: 1999 50% cut: 2001 70% cut: 2003 Phase-out: 2005*	Base level: 1995–98 Freeze from 1.1.2002 20% cut: 2005 Phase-out: 2015		
HCFCs consumption only	Freeze at 2.6% of CFC consumption in 1989 + total HCFC consumption in 1989: by 1.1.95 35% cut: 2004 60% cut: 2007 80% cut: 2010 95% cut: 2013 Phase-out: 2015	Freeze at 2.8% of CFC consumption in 1989 + total HCFC consumption in 1989: by 1.1.95 35% cut: 2004 65% cut: 2010 90% cut: 2015 99.5% cut: 2020 Phase-out: 2030	Base year: 2015 Freeze from 1.1.2016 Phase-out: 2040		

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Introduction

Although it was proposed in 1974 that stratospheric ozone could be depleted in chemical reactions involving the degradation products of chlorofluorocarbons (CFCs), it was not until 1985 that unequivocal evidence of ozone loss was reported in the scientific literature. In that year, scientists from the British Antarctic Survey described the polar ozone depletion, now known as the ozone hole, in which during six weeks in the spring the total ozone column decreases by more than half. It was subsequently shown that in the lower stratosphere almost all the ozone is removed from a layer at altitudes between about 13 and 20 km. These observations attracted great public interest, and aroused considerable scientific debate. At that time, photochemical theory had predicted ozone loss in the upper stratosphere by catalytic cycles involving the chlorine monoxide radical (ClO) and oxygen atoms. As the Antarctic ozone loss occurs in the lower stratosphere, it cannot be explained by that mechanism.

Previous SORG reports have described the advances in knowledge since 1985. It is now accepted that ozone in polar latitudes is mainly destroyed by additional catalytic cycles, which still involve ClO but do not involve atomic oxygen. These cycles are particularly efficient at the low temperatures always present in the Antarctic lower stratosphere in spring (and sometimes present in the Arctic). The new theory had also to explain the observations of large amounts of ClO. We now believe that this is due to heterogeneous reactions on polar stratospheric clouds (PSCs). In the lower stratosphere, most of the chlorine released from the CFCs is usually held in reservoirs (HCl and ClONO₂). In polar latitudes, reactions on the PSCs convert the reservoirs into more reactive forms capable of destroying ozone (see Box 1).

The global distribution of ozone is influenced strongly by atmospheric transport on a variety of timescales. The annual cycle (see SORG 1987) arises as a balance between photochemical production and destruction and transport, which moves ozone from its source region in the low latitude middle and upper stratosphere to its sink regions at higher latitudes. Long-term changes, or variability, in the large-scale stratospheric circulation have the potential to cause changes, or variability, in global column ozone amounts. Furthermore, large day-to-day variations in ozone measured at a particular site are known to be strongly related to meteorological conditions, with, for example, low ozone associated with tropospheric high pressure systems.

The meteorology in winter and spring in the Antarctic plays a major part in producing the ozone hole. There is a persistently strong circulation of westerly winds, or vortex, around the pole, associated with the very low polar temperatures during those seasons. The polar vortex inhibits substantial mixing of air between high and middle latitudes, thus preventing replenishment of ozone within the vortex by ozone-rich air from outside. In the Arctic, the mean winter and spring temperatures are higher than in the south, the abundance of PSCs is lower, and the vortex is more variable and breaks down earlier in the season than its southern hemisphere counterpart. Nevertheless, observations, discussed in SORG 1990, 1991, 1993 and 1996, have indicated that during the winter in northern high latitudes the chlorine can be in a form in which it can readily destroy ozone. SORG 1996 reported that in the cold stratospheric winters of 1994/95 and 1995/96 loss of ozone reached up to 50% locally in the Arctic lower stratosphere and that this loss could be attributed to chlorine- and bromine-catalysed ozone destruction.

Recent SORG reports have also considered the observed ozone decline in middle latitudes. SORG 1996 presented the latest evidence to show that chlorine-and bromine-containing compounds were implicated in the ozone loss but also discussed other factors which could have been important. The inability to explain the decline quantitatively was highlighted as a serious scientific concern.

SORG 1996 was able to report that the accumulation of halocarbons in the troposphere had peaked in 1993, with the peak in the stratosphere expected a few years later. Predicted chlorine and bromine loadings of the troposphere were updated to take account of further amendments to the Montreal Protocol and the most recent atmospheric observations. The peak in bromine loading was expected to occur between 2000 and 2010. The main form of bromine reaching the stratosphere is methyl bromide, which is partly controlled under the Montreal Protocol. SORG 1996 reported the unsatisfactory understanding of the budget of methyl bromide and its sources and sinks.

SORG 1996 also presented new information on some of the consequences of stratospheric ozone loss. Increased ultraviolet radiation reaching the surface was shown to be correlated with low ozone amounts. It was suggested that stratospheric ozone loss could have offset about 30% of the surface warming effect due to increases in other greenhouse gases since 1979 and that the observed ozone depletion was believed to be the main cause of the observed cooling of the lower stratosphere.

In this SORG report, we give in Chapter 1 the latest assessments of ozone depletion and our current understanding of the causes of ozone loss. Chapter 2 reports the most recent measurements of the concentrations of ozone depleting substances in the atmosphere. In Chapter 3, we update information on surface ultraviolet radiation, and re-examine the links, in both directions, between ozone depletion and climate change. This chapter also presents projections of the future evolution of the ozone layer, including calculations with a new generation of threedimensional coupled chemistry/climate models and summarises the current views on the impact of passenger aircraft on ozone.

One of the principal reasons for the widespread concern about depletion of the stratospheric ozone layer is the anticipated increase in the levels of ultraviolet radiation (UV) received at the surface of the Earth, and the consequential effects on our environment and well-being. The 1996 report of the United Kingdom UVB Measurements and Impacts Review Group (UMIRG) discussed the effects of UV on human and animal health, terrestrial and aquatic ecosystems, tropospheric chemistry and material degradation, with particular reference to impacts in the UK. The main conclusions of that UMIRG report, excluding the atmospheric topics discussed by SORG, are summarised below.

Increased exposure to UV will increase the likelihood of deleterious effects on biological systems. In humans, these effects include sunburn and skin cancers, together with possible eye damage and changes in immune function. Risk is related to exposure and this is strongly affected by behaviour as well as UV flux in the environment. In land animals, similar deleterious effects can be expected. Management strategies could mitigate the risks to domestic animals, but wildlife may be more vulnerable. Aquatic creatures are also affected by UV, with the clarity of the water and the depth at which they reside influencing their exposure and hence potential response to changes in UV.

Increased UV can modify the development of flora and fauna, and interactions between species, with implications for biodiversity in

aquatic and terrestrial ecosystems. Possible reductions in agricultural yield could be accommodated by plant breeding for increased UV tolerance in annual crops, but this has limited practical value for perennial species and natural vegetation.

Natural and synthetic polymers widely used in construction, transport and agricultural industries are degraded by exposure to sunlight. At first the appearance may be affected but in the longer term the physical integrity may be compromised so that the material no longer performs as intended. Nevertheless, the UV stability of common polymers can be improved with currently available techniques.

Chapter 4 of this current SORG report is an update by UMIRG of our understanding of the potential impacts of UV change resulting from ozone depletion.

Recommendations from SORG are given in Chapter 5.



Box 1: Chemical processes leading to ozone depletion

The depletion of ozone in the polar lower stratosphere is attributed to a sequence of chemical reactions that starts with the conversion of halogen (Cl and Br) source gases with long lifetimes into less stable 'reservoir' forms. The Cl-containing reservoirs are then broken down by reactions involving polar stratospheric clouds (PSCs) to release high concentrations of reactive chlorine and chlorine monoxide radicals (Cl and ClO). This leads to catalytic cycles that can destroy ozone very rapidly in sunlight.

The Br-containing reservoirs are unstable and in daylight a large fraction of the bromine is in the form of Br and BrO. These radicals also participate in catalytic cycles which rapidly destroy ozone. At middle latitudes similar processes occur, although most of the stratospheric chlorine remains tied up in reservoirs and so the rate of chemical ozone depletion is much slower than in polar regions.

The reservoirs of chlorine are hydrochloric acid (HCl) and chlorine nitrate (ClONO₂). Dinitrogen pentoxide (N₂O₅) and nitric acid (HNO₃) are reservoirs of nitrogen oxides. All the gas phase reactions between these reservoirs are relatively slow, but fast reactions can occur on a suitable surface or in solution (heterogeneous reactions). Laboratory experiments suggest that the most important reactions are:

HCl + ClONO	$_2 \rightarrow$	$HNO_3 + Cl_2$	(1)
$CIONO_2 + H_2O$	$\tilde{O} \rightarrow$	HNO ₃ + HOCl	(2)
HCl + HOCl	\rightarrow	$H_2O + Cl_2$	(3)
N,O, + H,O	\rightarrow	2HNO ₃	(4)

The chlorine compounds formed in these reactions are readily photo-dissociated, even in low light conditions, releasing chlorine atoms (Cl). The following heterogeneous reactions involving bromine species are also important:

$BrONO_2 + H_2O \rightarrow$	HNO ₃ + HOBr	(5)
$HOBr + HCl \rightarrow$	$H_2O + BrCl$	(6)

The nitric acid formed in these reactions can remain in the particles, so that the gas phase concentrations of nitrogen oxides are reduced. This reduction, 'denoxification', slows down the rate of removal of ClO in the reaction:

 $ClO + NO_2 + M \rightarrow ClONO_2 + M$ (7)

where M is any air molecule, and so helps to maintain high levels of ClO in polar spring.

Particles which support heterogeneous reactions exist throughout the lower stratosphere, and are particularly enhanced in the wintertime polar lower stratosphere. Sulphate aerosol particles (droplets of aqueous sulphuric acid) are always present in the lower stratosphere. These droplets are formed following the oxidation of carbonyl sulphide, a naturally occurring sulphur compound that has a long enough lifetime to survive slow transport to the stratosphere. Another, highly sporadic, source of aerosol is sulphur dioxide, injected directly into the stratosphere during some volcanic eruptions. This can increase aerosol concentrations to many times the background.

At high latitudes during winter these liquid sulphate aerosols can grow, taking up water and nitric acid, to form polar stratospheric cloud (PSC) particles at temperatures near 195 K (–78°C). Solid PSC particles, containing water and nitric acid, can also form at these low temperatures. At lowers temperatures still (188 K) solid ice PSCs can exist.

Reactions (1)–(6) also occur on sulphate aerosol. The reactions involving chlorine species are temperature dependent and become much more efficient on liquid PSCs in the cold polar lower stratosphere. However, at mid-latitudes these aerosol reactions can perturb the chemistry, especially when large amounts of aerosol are present following volcanic eruptions. In the polar regions solid PSCs also support reactions (1)–(4) leading both to the release of active chlorine, and to denoxification. Thus there is the potential for widespread ozone destruction.

Following the heterogeneous production of active chlorine, sunlight can drive the following catalytic cycles in the polar region:

(I)	ClO + ClO + M	\rightarrow	$Cl_2O_2 + M$	and	(II)	ClO + BrO	\rightarrow	$Br + Cl + O_2$
	$Cl_2O_2 + hv$	\rightarrow	$Cl + ClO_2$			$Cl + O_3$	\rightarrow	$ClO + O_2$
	$ClO_2 + M$	\rightarrow	$Cl + O_2 + M$			$Br + O_3$	\rightarrow	$BrO + O_2$
	$2 \times (Cl + O_3)$	\rightarrow	$ClO + O_2)$			Net: $2O_3$	\rightarrow	3O ₂
	net: 2O ₃	\rightarrow	3O ₂			-		-

The dimer of the ClO radical (Cl_2O_2) involved in Cycle I is thermally unstable, and the cycle is most effective at low temperatures. It is thought to be responsible for most of the ozone loss in Antarctica, with a large contribution from Cycle II involving reactive bromine. In the warmer Arctic a larger proportion of the loss may be driven by Cycle II.

In the middle latitude lower stratosphere the coupled ClO + BrO (Cycle II) and BrO + HO_2 cycles are responsible for increased halogen-induced chemical depletion.

Observation and Understanding of Stratospheric Changes

- Over Antarctica, the ozone hole of late winter and spring continues unabated. By early October, the atmosphere between 14 and 21 km altitude in the core of the vortex is practically devoid of ozone.
- In each year of the 1990s, the final warming of the vortex, which effectively marks the end of the ozone hole, has taken place in December. By contrast, the final warmings of the 1960s to 1980s were earlier in the year, occasionally as early as mid-October.
- Chemical ozone depletion has been observed in all Arctic winters since 1991/92 despite the very different meteorological conditions from year to year. In two of the last four winters (1995/96 and 1996/97) temperatures in the low stratosphere were low and conditions were conducive for large ozone losses (around 50% loss near 20 km altitude). In the later two winters (1997/98 and 1998/99), although temperatures were higher, ozone loss, albeit smaller, still occurred. These losses are consistent with our current understanding.
- Models predict chemical ozone depletion in the Arctic vortex broadly in agreement with observations, although calculated loss rates are too small in some periods. Models also show that interannual variations in atmospheric dynamics exert a large influence on the average ozone column at northern high latitudes in spring.
- In the late 1990s, annually averaged total ozone values at northern mid-latitudes (25°-60°N) have been 4-6% lower than those around 1980. The low values in 1992/93 following the eruption of Mt Pinatubo in 1991 have been a dominant feature of total ozone in the 1990s.
- At other latitudes there has been a 1–2% discrepancy between the TOMS satellite record and the groundbased record since 1996. The ground-based measurements indicate that similar losses have been observed in the southern mid-latitudes as in the northern mid-latitudes. Globally (60°S–60°N) annually averaged total ozone values derived from ground-based measurements have been 3–4% lower than in the 1970s.
- There is no statistically significant trend in total ozone in the tropics. Using current measurement systems, it is not possible to derive reliable trends in the vertical distribution of ozone in the tropical lower stratosphere

and upper troposphere. A significant contribution to radiative forcing due to ozone change cannot be excluded.

- The observed mid-latitude ozone decline is broadly consistent with halogen-catalysed chemical destruction being the primary cause. Model studies can now explain the trend in ozone in middle latitudes since 1980 in terms of reactions involving chlorine and bromine compounds. However, other studies show that meteorological factors have also contributed to the trend. The precise quantification of the different processes is still required.
- The interpretation of the observed changes in total ozone since the eruption of Mt. Pinatubo is difficult and in recent years depends quite strongly on the assumptions made about the influence of natural variability on ozone. The low ozone immediately following the eruption can be explained. The high ozone at northern mid-latitudes in 1998 is probably due to stronger transport of ozone rich air from lower latitudes. The updated observational record since WMO (1999) shows values closer to, but slightly above, the long term linear trend extrapolated from 1979–1991. However, we do not think that the observational record to date contains any evidence of an ozone recovery in response to reductions of ozone-depleting substances.
- Fifteen years of water vapour measurements above Colorado show an increase of 1%/year between 18 and 28 km. Satellite measurements over a shorter period suggest that an increase is occurring globally throughout the stratosphere. The lack of a complete explanation for the increase, which exceeds that expected from increases in methane (the principal stratospheric source of water vapour), is of serious scientific concern because water vapour is an important greenhouse gas and is chemically active.

This chapter of SORG 1999 presents an update on stratospheric ozone observations and the interpretation of global ozone depletion. In section 1.1, the observational record of ozone is summarised and updated. In section 1.2, trends in stratospheric

water vapour and methane, two critical stratospheric constituents, are discussed. Finally, in section 1.3, the possible causes of past trends and changes in stratospheric ozone are considered qualitatively and quantitatively.

5

1.1 Observations of Ozone

1.1.1 Antarctic Ozone Loss

The sequence of deep and long-lasting ozone holes has continued. Figure 1.1 shows the now familiar state of the ozone layer over Antarctica in early October. In this sounding, the altitude range from 14 to 21 km is practically devoid of ozone. Prior to 1980, this layer would have contained 160 to 180 Dobson Units (DU) of ozone, or about 60% of the ozone column.

A similar state has been reached in each recent year despite interannual variations in the circulation of the stratosphere. Although these variations are less than in the Arctic, they are strong enough to affect the minimum ozone column or the area enclosed by a specific ozone column contour. With the present large halogen loading of the stratosphere, new records in such parameters are scarcely relevant – what is important now is that the cumulative effect of chemistry in the late winter and spring can reduce the partial pressure of ozone from typically 12-15 mPa at the peak of the winter ozone layer (at about 17 km) to less than 0.1 mPa by October.

The ozone holes of the 1990s have been long-lasting. Those in 1995, 1996 and 1998, remained evident until mid-December. In 1998 the minimum ozone column observed south of 40°S did not rise above 160 DU until early December. The ozone holes are long-lasting because the radiative heating of air in which ozone has been almost completely destroyed is very slow, and flow across isentropic surfaces is largely inhibited. The vortex in the lower stratosphere is thus now more stable than it was in the 1960s to 1980s, when the breakdown of the vortex took place in some years in early October.

This is illustrated in Figure 1.2, which further emphasises that the ozone hole has not got any better in recent years. Figure 1.3 shows that temperatures within the vortex now stay below the nominal threshold for PSC formation from May to October. In 1998, the temperatures in November and December were occasionally below the envelope of all previous years.



Figure 1.1

The vertical profiles of ozone at the South Pole on 29 July 1998 and on 3 October 1998. In October, the partial pressure at 17 km is nominally zero, less than the detection limit of the sensor. (Courtesy S. Oltmans, NOAA-CMDL.)



Figure 1.2

Minimum total ozone south of 40°S observed by TOMS in 1998 (points) and 1996 (line). The shaded area is the range of values observed between 1978 and 1994. The small value in August 1996 is an artifact of observations which can occur when the sun is close to the horizon. (Courtesy P. Newman, NASA-GSFC.)



Daily minimum temperatures for 1998 at 50 hPa (approximately 19 km) in the latitude band 65°–90°S (red). Also shown are the 1979–97 average minimum values (green), daily extreme minimum temperatures for any year (short dashed) and highest minimum for any year (thin line). The horizontal dashed line indicates the nominal threshold (–78°C) for PSC formation. (Courtesy M. Gelman, Climate Prediction Center, NOAA.)

1.1.2 Arctic

The mean ozone column in the Arctic region during winter exhibits large interannual variability. This variability is linked with interannual differences in meteorology, which influences ozone both through varying transport and through varying temperatures which modulate chemistry. SORG 1996 was published at a time when large chemical losses had been unequivocally observed in the Arctic lower stratosphere during the cold stratospheric winters 1994/95 and 1995/96. The recent winters have emphasised the role of dynamics in controlling the Arctic ozone abundance.

Our understanding of the chemical processes responsible for Arctic ozone loss has not changed significantly since SORG 1996. As in the Antarctic, the loss is caused by ClO and BrO species which become enhanced following reactions on polar stratospheric clouds (PSCs). However, we now know that these reactions can occur on both solid and liquid PSCs. Furthermore, in the Arctic small, mountain-wave PSCs may play an important role in relatively warm winters (Carslaw *et al.*, 1999).



Figure 1.4

Spring average total ozone values in the latitude range 63°–90° for each hemisphere. Also shown are similar northern hemisphere averages calculated with a three-dimensional chemical transport for 1992–1998 (dashed line) and the modelled wintertime chemical loss (solid line). (Modified and updated from Newman *et al.* (1997) and Chipperfield and Jones (1999).)

Figure 1.4 shows the mean column O_3 in the polar regions (63°–90°) in March and October. This figure clearly shows that, during the 1990s, the mean Arctic O_3 columns decreased through to 1997. Observations of the ozone profile inside the Arctic vortex (Figure 1.5) confirm that the winters 1994/1995, 1995/1996 and 1996/1997 did have less ozone in the lower stratosphere, a reduction of 50% compared to the 1992 winter at 18 km. However, following the very low mean column ozone of March 1997, the winters 1998 and 1999 showed much larger column ozone, and the observed profiles (Figure 1.5) do not exhibit clear indications of chemical ozone depletion.

Given the large variability of ozone in the Arctic, indirect techniques are necessary to infer chemical ozone loss from observations. Data from ozonesondes combined with trajectory calculations (Figure 1.6) shows ozone loss rates in early March 1997 of around 40 ppbv/day. These loss rates are as large as those observed in the previous cold winters of 1994/95 and 1995/96 and the earlier winter of 1991/92 (Rex *et al.*, 1999a). The ozone loss was particularly effective in March 1997 as the low temperatures



Ny-Ålesund/Spitsbergen (79° N,12° E)



Figure 1.5

Vertical distribution of ozone measured over Ny Ålesund in March for several years in the 1990s. These ozonesonde profiles were obtained on flights made inside the Arctic vortex. (Courtesy P. von der Gathen, AWI-Potsdam.)

persisted relatively late in the season (see below) and so occurred in conjunction with significant sunlight. In contrast, this technique gave much smaller O_3 loss rates (10 ppbv/day in late January and mid February) in the warmer, more dynamically disturbed winter of 1997/98. Müller *et al.* (1997) also derived Arctic O_3 loss by correlating ozone observations with those of longer lived tracers at the start of the winter and end of spring. This technique generally indicates larger O_3 depletion. In the cold winter 1996/97, the derived loss was 65% locally at 19 km, and around 75 DU in the column.

Chemical depletion of column ozone has also been inferred by using ground-based observations combined with transport calculations from a three-dimensional model (Goutail *et al.*, 1999). Results from this technique are shown in Figure 1.7 for the 6 winters from 1993/94 to 1998/99. This study indicates that chemical loss has occurred in all of these winters with the mean vortex column depletion by late March ranging from 5% to 32%. The study shows that the cold winters of 1994/95, 1995/96 and 1996/97 had the largest chemical loss, but the analysis also shows



Figure 1.6

Ozone loss rates (red) at altitudes of about 18-20 km deduced for five winters in the 1990s. The 70% confidence interval is marked. Also shown (blue shaded area) is the area of the northern hemisphere below the PSC existence temperature. (Rex *et al.*, 1999b.)



Chemical depletion of total ozone inside the Arctic vortex in the last six winters. The pink shaded area represents the chemical ozone loss (time evolution and cumulative loss in %) deduced from a semi-empirical approach which compares observed and modelled total ozone amounts (Goutail *et al.*, 1999). The lines show the losses of total ozone calculated by two chemical transport models: Reprobus (Lefèvre *et al.*, 1998), solid; and SLIMCAT (Chipperfield, 1999), dotted. (Courtesy F. Goutail, CNRS-SA.)

significant column loss for the warm winter of 1997/98 of 20%. The winter of 1998/99 was particularly warm in the stratosphere and the lowest chemical depletions are calculated.

In summary, chemical ozone depletion has been observed in all recent winters. The chemical ozone depletion appears to be larger in longer, colder winters consistent with the role of PSCs in initiating the loss. However, there remains disagreement in the precise extent of ozone depletion from different, indirect methods used to infer the loss from observations.

1.1.3 Mid- and Lower Latitudes

a) Introduction

Recent analyses of ozone data (WMO, 1999) confirm the now familiar features of the stratospheric ozone depletion which has occurred since the 1970s. At mid-latitudes in the northern hemisphere, there has been a statistically significant decline in total ozone in all seasons with larger reductions in winter and spring than in summer and autumn. The loss of ozone at southern mid-latitudes is also statistically significant throughout the year with a smaller seasonal variation. No statistically significant trends in total ozone have been observed in the tropics.

The last SORG report noted that the lowest ozone values occurred in 1992/93 in the aftermath of the eruption of Mt Pinatubo. In 1.1.3(b), the evolution of the ozone layer in recent years is described with special attention paid to the quality of the recent total ozone measurements and to the statistical techniques and assumptions used to derive long term trends. A number of concerns about the trends in the vertical distribution of ozone derived from different measurement systems were also noted in SORG 1996, in particular (a) the disagreement on the magnitude of the trends between 15 and 20 km over northern mid-latitudes and (b) the inconsistency between ozone trends in the tropics. Significant progress has been made since that time: the main conclusions of an international assessment of the trends in the vertical distribution of ozone are described in 1.1.3(c).

b) Total ozone in recent years

Total ozone over northern mid-latitudes (25°–60°N) is shown in Figure 1.8. Two sources of measurements are used: the network of ground-based instruments (January 1979 to December 1997) and the series of Total Ozone Mapping Spectrometers (TOMS) from January 1979 to April 1999. There is good agreement between these two records at northern mid-latitudes. The seasonal cycle has been removed from the measurements by using a statistical model. The record low values in 1992/93 still remain the lowest observed. With the exception of the 1997/98 northern winter, the total ozone values in the last four years at northern mid-latitudes are 4-6% lower than those around 1990. They are not obviously higher than those predicted by extrapolating the trend calculated for the period 1979–1991.

Total ozone values (12 month running means) over the United Kingdom are shown in Figure 1.9. The same overall pattern of a long term decline with appreciable interannual variability is apparent. As discussed in SORG 1996, the lowest values at Lerwick (60°N) occurred in early 1996, due to a combination of the tropospheric meteorology (a high pressure system elevating the tropopause) and a chemically depleted vortex being overhead. Higher values, more pronounced at the higher latitude site, were observed in 1998 but are within the previously observed variability.

The possible explanations for the long-term evolution of ozone are discussed in section 1.3.3. Here, a couple of additional points





Deviations in total ozone, area weighted over 25°-60°N. A seasonal trend model (not including the effects of the solar cycle or the quasi-biennial oscillation) was fitted to the ozone data over the period from January 1979 to May 1991. The upper panel shows the TOMS record (1/79-4/99) and the lower panel shows a ground-based series constructed from average ozone at stations in 5° latitude bands (1/79–12/97). Trend models are fit independently to the two data sets. The solid straight line represents the least squares fit to the deviations up to May 1991 and is extended as a dotted line through December 1997. (The TOMS record is updated from WMO (1999) courtesy of L. Bishop, Allied Signal.)

related to data quality and the statistical analysis are discussed. These are: (a) the quality of the currently available measurements, and (b) the calculation of the ozone trends.

The measurements from the ground-based network of instruments and from TOMS agree well $(\pm 1\%)$ in the northern mid-latitudes. However, at low latitudes and particularly in the southern midlatitudes, larger discrepancies (of perhaps 1-2% with TOMS measuring more ozone) have been reported for the period after 1996 when the Earth Probe (EP) TOMS went into orbit (see Figure 4.16 in WMO, 1999). There are two possible explanations:

The TOMS instrument response may be latitude dependant. TOMS measurements might be inadvertently calibrated to agree



1981

-1998

Figure 1.9

Total ozone over the United Kingdom as seen in the Lerwick (60°N) and the combined Bracknell and Camborne (51°N) records. A 12 month running average has been applied to the data. (Courtesy S.F.G. Farmer and M.J. Molyneux, UKMO.)

at northern mid-latitudes where the ground-based network is strongest and most of the comparisons of TOMS data have been made. Any problem might be enhanced for EP TOMS whose calibration is still considered preliminary (WMO, 1999). (Some support for this explanation comes from the trends derived from total ozone measurements from Lauder, New Zealand (45°S), which agree better with the trends from the earlier, version 6 TOMS data, than those from the later, version 7 TOMS data (G. Bodeker and R. McKenzie, private communication).);

The poorer coverage of ground-based stations in the tropics and southern hemisphere has, by chance, resulted in a bias relative to TOMS in recent years which was not present before.

10



Deviations in total ozone, area weighted over 25°-60°N. The seasonal trend model used to fit the ozone data included allowance for the effects of the solar cycle and the quasibiennial oscillation (c.f. Figure 1.8). The solid straight line represents the least squares fit to the deviations up to May 1991 and is extended as a dotted line through December 1997. The thinner smoother line is from a lowess local regression fit and does not include the TOMS data after January 1998 (triangles). (The TOMS record is updated from WMO (1999) courtesy of L. Bishop, Allied Signal.)

It is hard to know which explanation is correct. Here we choose to rely on the ground-based data for total ozone outside the northern mid-latitudes, as the calibration of EP TOMS is preliminary. The ground-based measurements indicate that similar depletion has occurred over southern mid-latitudes as over northern mid-latitudes, and that, globally (60°S-60°N), total ozone in the late 1990s has been 3-4% lower than in the 1970s (WMO, 1999; L. Bishop, private communication).

Whichever explanation is correct, care must be taken in discussions of the possible 'recovery' of the ozone layer in the tropics or southern mid-latitudes and in the global average until this data quality issue is resolved. There is a great danger of overinterpreting changes of 1% or so - after all this is the estimated stability of the observational network for total ozone.

A second caveat to be borne in mind when interpreting the recent data is related to how the ozone trends are calculated (i.e. whether the assumptions underlying the statistical analysis are valid). The statistical models used to calculate ozone trends attempt to account for a number of sources of variations in ozone in order to improve the estimates of the trends themselves. The standard approach is to allow for the effects of natural phenomena such as the quasi-biennial oscillation (QBO) and the 11 year solar cycle by assuming a linear relation between the strength of these phenomena and total ozone. The ozone trends are generally assumed to be linear. The details of the statistical analysis would be of solely academic interest if the relationship of total ozone with the QBO and particularly the solar cycle were well established and if the chemical destruction of ozone was really expected to be linear. In practice this is not the case.

The ozone values shown in Figure 1.8 are those found after removal of the seasonal cycle, but with no removal of any influence of the QBO or the solar cycle. For comparison, Figure 1.10 shows the total ozone after removal of all these effects and so in principle only the trend remains. In practice there is appreciable interannual variation in both figures, with a greater scatter (principally from the quasi-biennial oscillation) in Figure 1.8. Of most interest here, because of their influence on the trend, are the post-1996 values which are closer to the long term trend extrapolated from 1979-1991 when no allowance is made for the QBO or the solar cycle (i.e. in Figure 1.8). This comparison is important because recent work (Solomon et al., 1998; 1999) has suggested that the quasi-decadal cycle in total ozone previously described in trend analyses using the 11 year solar cycle might in fact be induced by the changes in stratospheric sulphate aerosol loading which have been very strongly influenced by the volcanic eruptions of El Chichon in 1982 and Mt Pinatubo in 1991 (Figure 1.11). In other words one of the assumptions underlying the statistical model which has been used in all recent major assessments has been questioned. A second assumption being questioned is that the long term trend is linear. These issues are considered further below in our discussion of the current understanding of the causes of the long term trend (section 1.3.3).

c) Trends in the vertical distribution of ozone

A thorough assessment of the various measurements of the vertical distribution of ozone (SPARC/IOC/GAW, 1998) has resulted in greater overall confidence in our knowledge of the trends. Figure 1.12 shows the best estimates (derived from four measurement systems) of the trends over northern mid-latitudes from 1980-1996. There are two clear maxima, one around 40 km (-6 to -8% per decade) and one around 15 km (-5 to -10% per decade) and statistically significant trends have occurred at all altitudes between 12 and 50 km. Furthermore, the seasonal variation of the ozone trends over northern mid-latitudes is greatest at altitudes between 10 and 20 km.

However, ozone trends below 20 km over other parts of the world must still be treated with caution because: (i) remaining





Stratospheric aerosol loading observed from 1976 to 1999. The aerosol loading is taken as the integrated column backscatter above the tropopause measured by the lidar at Garmisch-Partenkirchen, Germany (47°N, 11°E). Important volcanic eruptions are indicated by arrows. (Courtesy H. Jaeger and R. Muecke, IFU.)



Figure 1.12

The mean, annually averaged trend in the vertical distribution of ozone that has occurred over northern mid-latitudes from 1980-1996 (heavy solid line) is calculated using the trends derived from SAGE I/II, ozonesondes, Umkehr and SBUV measurements. The combined uncertainties are shown for 67% confidence limits (light solid lines) and 95% confidence limits (dashed lines). The results below 15 km are a mixture of stratospheric and tropospheric trends and the exact numbers should be viewed with caution. (SPARC/IOC/GAW, 1998.) uncertainties about the early SAGE measurements (1979–1981) limit confidence in the trends below 20 km from 1979; and (ii) there are insufficient correlative measurements outside the northern mid-latitudes to compare with SAGE trends. It remains a concern, worthy of mention because of the potential importance to climate change, that trends of ozone in the tropical lower stratosphere and upper troposphere still cannot be reliably derived using the currently available measurement systems.

1.2

Trends in Stratospheric Water Vapour and Methane

Water vapour is an important stratospheric constituent, exerting a major influence on both chemistry and radiation. A stratospheric cooling and a positive tropospheric radiative forcing are likely to be the most important consequences of an increase in water vapour (see Chapter 3).

Oltmans and Hofmann (1995) report an increase in water vapour in the lower stratosphere between 1981 and 1994 at one location. A recent extension to these balloon-borne measurements confirms that hydration of the stratosphere continues to increase at about 1%/year (Figure 1.13). This increase exceeds that expected from the methane oxidation source.

Satellite observations from HALOE show this trend to be present globally between 30 and 65 km since 1992 (Evans *et al.*, 1998; Nedoluha *et al.*, 1998). The combination of a long time series at one place and a short time series with trends at all altitudes and latitudes lends confidence to the detection of a global change.

Two separate analyses fit linear trends to the HALOE data over very similar periods, but there are subtle differences in their analysis procedures which result in significantly different trends. However, considering a realistic assessment of their probable systematic errors, they and the trends from balloon-borne measurements are consistent.

A global increase of water vapour of 1%/year could be caused by a warming of the tropical tropopause by about 0.1 K/year (as estimated from frost-point tables). However, some climate model calculations suggest that a cooling should have occurred there. Current observations of the temperature of the tropical tropopause are of insufficient quality to resolve the problem, although a recent reanalysis of model-assimilated data does show a cooling (Simmons *et al.*, 1999).

Methane (CH₄) is an important sink for atomic chlorine in a reaction producing HCl. Tropospheric CH₄ has increased from about 650 ppbv in the middle ages (Etheridge *et al.*, 1998) to its current value of 1730 ppbv. Tropospheric CH₄ is transported to the stratosphere, where it is oxidised to H₂O. Trends at the surface (Dlugokencky et al., 1998) are probably consistent with



Figure 1.13

Trends in stratospheric water vapour observed above Colorado by balloon-borne frost-point hygrometers from 1981 to 1997. The blue shaded area is the 95% confidence interval of the trend. (Courtesy S. Oltmans, H. Vömel and D. Hofmann, NOAA-CMDL.)

 CH_4 trends determined by HALOE at 30 km. Curiously, HALOE results show that CH_4 decreased in the upper stratosphere between 1992 and 1997. Explanations may be a slowing down of the meridional circulation in the stratosphere, or other circulation effects caused by Mt Pinatubo. Resolving these trends in CH_4 and global trends in H_2O in the lower stratosphere is essential for understanding the future evolution of the ozone layer.

1.3

Explanation of Past Trends and Changes

1.3.1 Antarctic

The simplest region of the stratosphere in which to explain the observed changes in ozone is the Antarctic springtime lower stratosphere. The meteorological conditions always lead to a strong, stable polar vortex within which temperatures are low enough (well



below the nominal 195 K threshold) to cause efficient chlorine activation on polar stratospheric clouds. The polar vortex, and low temperatures, persist into October which provides the opportunity for efficient ozone depletion in sunlit air masses contained within the polar vortex. This ozone loss is clearly dominated by catalytic cycles involving the CIO and BrO species, as discussed in earlier SORG reports (see Box 1 of the Introduction).

1.3.2 Arctic

The processes which lead to large ozone depletion in the Antarctic springtime lower stratosphere also occur in the Arctic. However, the meteorology of the northern hemisphere is much more variable, leading to large interannual differences in the strength of the polar vortex and the extent of low temperatures. Even within the extremes of this variability, no Arctic winter has as strong a vortex as the Antarctic, or such a long period of low temperatures. In the Arctic small-scale, mountain-wave PSCs play an important role in relatively warm winters (Carslaw *et al.*, 1999).

This large interannual variability strongly moderates the extent of chemical depletion which can occur through PSC processing. Activation of chlorine species on both solid and liquid PSCs is strongly dependent on temperature. Although liquid particles exist at higher temperatures, they only activate chlorine efficiently at temperatures near 195 K, at which solid particles become possible. Meteorological variability also strongly moderates the downward transport of ozone into the vortex during the winter. Indeed, the meteorological conditions which lead to large chemical depletion (a strong, stable vortex with low temperatures) are the conditions which also lead to weak transport of ozone, so that the low column ozone occurs for two complementary reasons. Conversely, a weak disturbed vortex is not only associated with higher temperatures, but will also induce downward transport of ozone rich air.

Recent Arctic winters illustrate this range of dynamical behaviour. In winter 1996/97 the polar vortex was very stable from January and persisted until early May. Temperatures low enough for PSCs occurred until March. Consequently large chemical ozone loss was inferred from observations (Figures 1.6 and 1.7). Figure 1.4 shows that the March 1997 mean column ozone was indeed the smallest recorded during the 1980s and 1990s. However, this small mean column was also influenced by the weak transport associated with this winter.

In contrast to the preceding three winters which were all relatively cold, the two most recent Arctic winters (1997/98 and 1998/99) have been characterised by very disturbed vortices and higher temperatures. Consequently the inferred chemical loss is smaller. The MATCH technique shows a large variation of the instantaneous O_3 loss rate near 20 km with different meteorological conditions (Figure 1.6). The accumulated column loss, as derived using 3-D models (Figure 1.7), shows less variation from year-to-year, except for the small depletion in 1998/99.

Three dimensional chemical transport models (3-D CTMs) have been developed during the 1990s and allow us to quantify the relative roles of transport and chemistry determining the ozone abundance in the Arctic. Results from two such models are shown in Figure 1.7. There is generally good qualitative agreement between the modelled loss and the losses derived from observations. However there are also important differences. For example, the 3-D CTMs generally underestimate the observationally derived loss; in 1993/94 there is a slight overestimate. It could be that the models are still missing some process, or the detail of some process, the importance of which varies with the meteorological conditions in a given winter.

Figure 1.4 also includes results from a 3-D model simulation from 1991 to 1998 (Chipperfield and Jones, 1999). The 3-D model does well in reproducing the magnitude and variability of the mean ozone column. The model results show that much of this variability is due to differences in transport. For example, the large increase from 1997 to 1998 of 80 DU is not simply due to less chemical loss (which decreases by about 20 DU) but is mainly caused by increased descent of ozone-rich air in the dynamically active winter of 1997/98.

The recent Arctic winters have continued to emphasise the variable nature of the Arctic polar vortex, which make determination of a chemical, or dynamical, trend difficult. These winters do show, however, that low mean ozone columns in the Arctic are produced by: 1) large chemical loss, which occurs when temperatures low enough for PSCs persist into March; and 2) stable meteorological conditions. The future evolution of Arctic ozone will therefore depend not only on the extent of chemical loss (linked to halogen loading), but also on possible climate change. Furthermore, the late breakdown of the polar vortex (as in 1997) will enable isolated air masses with low ozone to persist into periods of stronger insolation, for example, into May.

1.3.3 Mid- and Lower Latitudes

A major conclusion of SORG 1996 was that substances containing chlorine and bromine (halogens) were clearly implicated in the ozone decline at mid-latitudes, but that the magnitude of the observed loss was underestimated by the existing models. Since then a number of other mechanisms have been proposed as contributing to the ozone trends. In the first part of this section the possible mechanisms are described qualitatively, and in the second part these are discussed more quantitatively in the light of the observed ozone evolution and the derived trends.

a) Mechanisms affecting total ozone

Ozone amounts over mid-latitudes are affected by a number of chemical and dynamical processes.

(i) In situ chemical destruction of ozone over mid-latitudes

Solomon *et al.* (1998; 1999) have shown that the changing aerosol surface area over the last twenty years has significantly modulated



the effect that the gradually increasing halogen loading has had on stratospheric ozone. The substantially increased aerosol loading from the two major volcanic eruptions (El Chichon in 1982 and Mt Pinatubo in 1991) affected the chemical balance in the stratosphere and significantly enhanced any ozone loss above that which might have been expected from increasing halogen alone. Further, the timing of the eruptions producing a quasidecadal signal similar to the solar cycle variation. Stratospheric aerosol levels are now back to background levels (Figure 1.11) and so the enhancement of the halogen-catalysed ozone loss should currently be minimal.

Other advances in understanding chemical mechanisms have been made which can also account for ozone destruction in the lowermost stratosphere, where a large part of the ozone trend has occurred. These include activation on cirrus clouds, short term temperature fluctuations accelerating cold aerosol reactions, improved knowledge of the activation processes and of the bromine-chlorine coupling through heterogeneous reactions (Murphy and Ravishankara, 1994; Borrmann et al., 1997; Solomon et al., 1997; Solomon, 1999).

(ii) Transport of air from the Arctic vortex

Air transported from the polar vortex can affect the mid-latitudes in two ways. In SORG 1996 it was concluded that while activated air is exported with subsequent ozone loss from inside the Arctic vortex, it probably does not have a major impact on mid-latitude ozone. A more important mechanism was thought to be the export of ozone-depleted air from inside the vortex. This may initially occur during the winter as the vortex is gradually eroded but must happen at the end of the winter during the final warming and break-down of the vortex ('dilution'). These conclusions still hold and have since been better quantified (see below).

(iii) Circulation changes

The strong influence of tropopause height and stratospheric circulation on ozone amounts was discussed in SORG 1996. A number of studies had found some evidence for long term changes in the stratospheric circulation, including a strengthening of the Arctic vortex (Zurek et al., 1996) and changes in northern hemisphere geopotential heights (Labitzke and van Loon, 1995). Any such change would affect total ozone in these regions and so would influence the long term trends in ozone (Hood et al., 1997).

There is more evidence that regional changes in tropopause height have occurred. For example, a long term increase in tropopause height of 150±70m since the late 1960s has been observed over Hohenpeissenberg, Germany (Steinbrecht et al., 1998), consistent with the increase over much of Europe derived from meteorological analyses (Peters et al., 1996). This may be related to an increase in anticyclonic activity associated with the North Atlantic Oscillation (Hurrell, 1996): a tropospheric climate oscillation with intermittent changes between active and passive phases (Appenzeller et al., 1998). There has been a change to an active phase in the past two decades i.e. the period over which the trends in circulation, tropopause height and mid-latitude ozone have been observed. A strengthened polar vortex is also associated with the active phase. It is unclear what has triggered the transition to the active phase. In a climate system that exhibits distinct modes of natural variability, it is possible that anthropogenic activity may influence transitions between these modes (e.g. Graf et al., 1998), although there is no firm evidence of this.

Regardless of whether the circulation and tropopause height changes are of natural or anthropogenic origin, there remains the possibility that they may account for part of the observed ozone trend in mid-latitudes (Hood et al., 1997). Their relative importance compared with chemical depletion needs to be quantified.

b) Quantitative effect on total ozone

(i) Explanation of long-term trends

The observed behaviour of ozone over the last decades is a powerful constraint in assessing which mechanisms have contributed to the ozone decline and by how much. As discussed above the main two influences on stratospheric ozone are chemistry and dynamics.

The depletion of ozone by halogen chemistry is strongly modulated by the surface area of the stratospheric aerosol and so the time evolution of this chemical effect on ozone is a combination of the relatively monotonic increase in stratospheric chlorine and bromine amounts since the 1960s (see chapter 2) and the stratospheric aerosol loading which is much more variable, being dominated by volcanic eruptions (Figure 1.11). Model calculations show that this combined chemical effect could quantitatively explain nearly all the observed changes in ozone at northern mid-latitudes in the 1980s and 1990s, when short timescale variability is removed (Solomon et al., 1998; 1999).

Other studies have investigated dynamical mechanisms (described in 1.3.3(a)) which are not included in the above calculations. For instance, Hood et al. (1997) investigated the effect of changes in the winter-time stratospheric circulation on total ozone for the period from 1979–1991 (just before the Mt Pinatubo eruption). They concluded that a significant fraction (around 50%) of the total ozone trends at northern mid-latitudes could be accounted for by changes in transport and, further, that much of the longitude and latitude dependence of the trends could also be explained in this way.

Dilution of vortex air into mid-latitudes has both chemical and dynamical aspects which are not necessarily well described in chemical transport models. The dilution of ozone-poor air from the Arctic vortex into northern mid-latitudes in the 1997 spring, following the large polar loss in that year, was estimated to have caused a reduction of 3% of the mid-latitude column by the time



the vortex had broken down in May 1997 (Knudsen et al., 1998). This corresponds to about one third of the observed long term change in TOMS total ozone. There is appreciable interannual variability in the impact of dilution with between 2% and 4% estimated for the 1993–96 springs, with the magnitude in a given year depending on the extent of springtime ozone loss in the vortex. Chipperfield (1999) calculated that, overall, PSC-related depletion processes in or at the edge of the vortex result in 2–3% less ozone at 50°N throughout the year. The effect was found to be larger in the southern hemisphere (5% less ozone at 50°S), reflecting the larger losses occurring inside the Antarctic vortex.

Until long-term modelling studies incorporating both chemical and dynamical effects have been performed, it will be hard to resolve the apparent discrepancy of the different quantitative explanations which between them can account for more than the observed long-term trend. Improved understanding of the relation of total ozone with the solar cycle will be needed to explain the quasi-decadal variation in total ozone.

(ii) Ozone in the 1990s

There have been significant features in the evolution of total ozone since the eruption of Mt Pinatubo in 1991. Figures 1.8 and 1.10 show that total ozone values at northern mid-latitudes were depressed following the eruption with minimum values in 1992/93. This behaviour was also seen in the tropics, but not in the southern hemisphere. Since about 1994 total ozone amounts have been roughly constant (Figure 1.8). As usual when considering factors affecting stratospheric ozone, both dynamic and chemical factors have to be taken into account.

Inter-annual variability in stratospheric transport is a more important factor on shorter time-scales than on longer ones. This issue has been examined in some recent modelling studies where 3-D models of the atmosphere were run using the observed winds in the period following the Mt Pinatubo eruption (Hadjinicalaou *et al.*, 1997; Chipperfield, 1999). These calculations indicate that most of the large negative anomaly in 1992/93 could be attributed to inter-annual variations in dynamics, with a possible radiative enhancement resulting from the stratospheric aerosols. The high values seen at high northern latitudes (Figure 1.9) in the 1997/98 and 1998/99 winters seem to be caused by a stronger Brewer-Dobson circulation than in the previous few winters.

During the mid to late 1990s, the rate of increase of stratospheric halogen loading decreased appreciably and stratospheric chlorine loading should peak in 1999 (see Chapter 2). The reduced growth in halogen loading should have caused a deceleration (but not an absolute decrease) in the resultant chemical ozone depletion. However the largest influence on the chemical depletion of ozone over the last few years was the reduction in the stratospheric aerosol loading. Model studies indicate that in 1992/93, the enhanced aerosol resulting from the eruption of Mt Pinatubo caused an enhanced chemical depletion of total ozone at northern mid-latitudes of 2–6% above that calculated for a constant background aerosol (Solomon *et al.*, 1999; Chipperfield, 1999).

The variation of the ozone column over northern mid-latitudes in the 1990s has been complex with low values after the Mt Pinatubo eruption in 1991 and higher values in the late 1990s. The interpretation of these latter values is difficult. Could they, for example, be signs of ozone recovery in response to the Montreal Protocol? We do not think so, for various reasons outlined above. First, in section 1.1.3, we have indicated that the statistical models used to determine trends have certain inadequacies. Short periods above or below the trend line must be interpreted with caution. In addition, we believe that interannual variability in the atmospheric transport of ozone is at least partly responsible for the high column amounts. Furthermore we have argued above that the reduction in the growth in stratospheric halogens, consequent on the Montreal Protocol, must be secondary to the chemical impact of the reduction in sulphate aerosol since the eruption of Mt Pinatubo. For all these reasons, we do not think that the observational record can be interpreted as showing any evidence of an ozone recovery in response to reductions of ozonedepleting substances.



Observation and Understanding of Stratospheric Changes

2 Halocarbons in the Atmosphere

- With several more years of observations of the effects of the Montreal Protocol on the consumption of ozone depleting substances, the scenarios for their future use are more constrained than in previous SORG reports.
- These better scenarios have not materially altered the pattern expected for future chlorine and bromine loadings of the atmosphere. The chlorine loading should continue to fall and bromine loading is expected to peak early in the 21st century.
- It is assumed that the Ozone Depleting Substances still in equipment will be released to the atmosphere in the future. The largest additional reduction that can be made to future chlorine and bromine loadings would be gained if these materials were removed and destroyed when the equipment containing them is decommissioned. Over the next 50 years, such reductions could amount to 5% in total cumulative chlorine loading and 25% in bromine.
- HCFC releases have a relatively small effect on stratospheric chlorine loading. A proposed regulation in the EU seeks to reduce production and use of HCFCs beyond the provisions of the Montreal Protocol. If such a regulation were applied globally, it would enhance the reduction in total cumulative chlorine loading by slightly less than 1% over the next 50 years.
- Methyl bromide is the largest contributor of bromine to the stratosphere. Its budget is still uncertain despite a number of improvements in understanding since the 1994 WMO assessment.
- Analysis of air in Antarctic firn (unconsolidated snow) suggests concentrations of methyl bromide have

increased by 20–25% during the 20th Century, with the most rapid growth occurring in the 1970s and 1980s. Despite the uncertainties in the atmospheric budget of methyl bromide, the firn air data suggest concentrations of methyl bromide to have been strongly influenced by human activities. It seems likely that less than half of the anthropogenic methyl bromide emissions are controlled under the Montreal Protocol.

- The methyl bromide ozone depletion potential (ODP) has been reevaluated downwards since the WMO 1994 report, based on a decreased estimate of the methyl bromide lifetime (which reduces the ODP) and an increased estimate of the ozone-depleting efficiency of bromine (which increases the ODP). The reduction in the lifetime also implies a larger flux of methyl bromide into the atmosphere than was assumed previously. Overall, there is no reduction in our assessment of the importance of methyl bromide in stratospheric ozone destruction.
- Atmospheric halon concentrations will continue to increase during the first decade of the 21st century. This is within the provisions of the Montreal Protocol. These emissions are dominated by release of material now in equipment, rather than arising from new production in the developing world.
- Illegal trade in ozone depleting substances, in the quantities that have been assumed, does not have a large effect on future chlorine and bromine loading.

2.1

Man-made Ozone Depleting Substances

Since the SORG 1996 report and the recently published 1998 WMO Science Assessment were written, much additional information on the use of halocarbons has been published. Coupled with more recent observations of atmospheric concentrations, these new data enable a better understanding of the factors controlling emissions and the construction of scenarios that are more constrained.

In particular, the historic production of CFCs, HCFCs and halons reported by nations to UNEP as required by the Montreal Protocol (UNEP, 1998a) now matches the values recorded independently by industry (AFEAS, 1999). Such good agreement suggests that the total production records are relatively certain. Emission estimates are much less certain but may be checked against, or even calculated from, atmospheric measurements. It is important to reduce the uncertainty in the size of the unemitted portion that is still being used in equipment (the so-called "bank") because, for CFCs, this now represents the principal source of future emissions.

2.1.1 Historical Production and Emissions

The Montreal Protocol imposes controls in different ways depending on a classification of a country's economy; this is more fully described in the Background to this report. In the developed world, consumption of CFCs was stopped in 1996 but limited production for use in less developed countries is allowed. Over the period 1986 to 1995 the annual productions of CFCs recorded in the UNEP and AFEAS databases match to within 1%, when based on the same set of nations and expressed in the same form (as ODPtonnes, the product of the Ozone Depletion Potential and mass). Additional production of CFCs, not recorded by industry, in less developed countries and those countries with economies in transition fell from a peak of 140,000 ODPtonnes in 1989 to 100,000 ODPtonnes in 1997 (UNEP, 1998a; personal communication, N. Sabogal, UNEP, 1998a).

Within the total for CFCs, Russian production declined to 17,000 ODPtonnes. On the other hand, production in China rose to 54,000 and in India to 22,000 ODPtonnes. Although these are clear increases that are nationally significant, there does not appear to be a rush to build CFC production capacity in the developing world to the pre-Protocol total that was over 1,000,000 ODPtonnes.

The UNEP data for HCFCs show that global production over the period 1992 to 1996 doubled from 15,000 to 30,000 ODPtonnes. Despite such a large increase, global HCFC production, expressed in this way, is still only of the order of 20% of current global CFC production. Furthermore, based on the observation that production of HCFCs by the developed world has remained constant at $93.2\pm0.3\%$ of the global total in each year, it seems that developing countries are not at present installing significant extra new capacity for the production of HCFCs.

Production of halons in developed economies ceased in 1993. Up till then, the industrial report for Halon-1211 and Halon-1301 (McCulloch, 1992) was systematically 5% lower than the total halon production reported to UNEP in the appropriate countries (UNEP, 1998a). All of the current 45,000 ODPtonnes of halon production is now in Russia and developing countries. Russian production has slumped from 27,800 ODPtonnes (thought to be mainly Halon-2402) in 1986 to less than 1000 in 1996. This value is lower than the quantity allowed for essential uses. Indian and Korean production has hardly changed over the same time period and most of the increase has occurred in China where production has increased almost fourfold. The quantities of Halon-1211 and Halon-1301 for which China has obtained compensation under the MultiLateral Fund of the Montreal Protocol and the agreed reduction schedule, viewed together, suggest that production of both halons there has now peaked.

A relative newcomer to the atmospheric observational record, Halon-1202 (CF_2Br_2) may have been manufactured for use in its own right or may be co-produced with Halon-1211 (CF_2ClBr) by overbromination and then subsequently released into the atmosphere. The observed increase in Halon-1202 in the atmosphere at Cape Grim, Tasmania (Fraser *et al.*, 1999) is consistent with a small (2 to 5%) by-product of CF_2Br_2 from Chinese manufacture of Halon-1211 being released into the atmosphere.

2.1.2 Calculation of Future Production and Emissions

The reports from the Technological and Economic Assessment Panel (for the Montreal Protocol) on HCFCs and halons (TEAP, 1999a; TOC, 1998), that analysed the uses of these compounds and their replacements, enable much better constraint of the scenarios for future production and emission. In the absence of this information, both SORG 1996 and Chapter 11 of the WMO 1998 Scientific Assessment (WMO, 1999) included future production at the maximum allowed under the Montreal Protocol. However, the analyses in the TEAP report suggest that this will overstate future production, particularly of CFCs and HCFCs. The countries of the developed world have never used up their quotas for sales to developing countries; and the developing countries, themselves, have not produced up to their allowances for "basic domestic needs". Furthermore, as described in 2.1.1 above, production by the former countries has fallen dramatically since 1994 with only a modest rise in the latter.

Accordingly, future production of CFCs has been revised heavily downwards in the scenario for SORG 1999, in comparison to the scenarios used in SORG 1996, Chapter 11 of WMO (1999) and Midgley and McCulloch (1999). The revised production totals some 140,000 tonnes/yr of CFCs early next century, much less than that allowed under the Protocol.

Illegal trade in CFCs is not addressed specifically in this report. In SORG 1996 it was considered to have little environmental impact at the levels of activity, estimated to be in the region of tens of thousands of tonnes. Although the practice still seems widespread (EIA, 1998), since the previous report there has been no new information on the level of activity and so there is no reason to amend this conclusion.

Under the Protocol, HCFCs are controlled in developed countries by a cap on consumption that will be progressively reduced to zero in the early decades of the 21st century. The details of the current regulations are described in the Background to this seventh report of SORG (see earlier). In the EU the cap is currently 7% smaller than that in the Protocol and the effective phase-out date is 2015 rather than 2020. Under a new proposed regulation, the cap in the EU should be reduced to 2% and the phase-out date brought forward to 2010 (Official Journal, 1999). Developing countries operating under Article 5 of the Protocol have until 2040 to phaseout consumption. The scenarios used here follow those in the TEAP report (TEAP, 1999b), with the minor adjustment for the proposed EU regulation, applied only in the EU.

As in previous scenario calculations, emissions of carbon tetrachloride (CCl₄) are linked to CFC-11 and -12 production (Simmonds *et al.*, 1998). Methyl chloroform production has now shrunk to 1500 tonnes from 718,000 tonnes in 1990; emissions have fallen accordingly. Consequently, atmospheric concentrations of methyl chloroform are falling sharply and it should not be present in the atmosphere after 2030.

2.1.3 Atmospheric Concentrations of Individual Compounds

Expressed as mixing ratios (parts per trillion or 1 in 10^{12}), the atmospheric concentrations of individual CFCs and related ozone depleting substances (ODS) measured over the past 10 years



are shown in Figure 2.1. This also shows predicted future concentrations of these chemicals, extrapolated from the measurements using the scenarios described above and current values for atmospheric lifetimes (Chapter 10 of WMO, 1999) in the single box model described in SORG 1990.

Concentrations of CCl_4 , CFC-113 and CFC-11 are currently falling and that trend should accelerate slightly in the early decades of the 21st century. CFC-12 concentrations should shortly stabilise and then start to fall.

The calculated future concentrations take into account the additional 44 pptv of CFC-11 and 25 pptv of CFC-12 that would result from total emission of the estimated 1 million tonnes of CFC-11 and 500,000 tonnes of CFC-12 still in systems throughout the world. If the developing countries continue to limit their use, these banks represent the largest source of future releases, and their capture and destruction is the only way to achieve significant additional reductions in atmospheric concentrations of CFCs. The additional contributions amount to a total of 5% of the total chlorine loading integrated until the middle of the 21st century.

In contrast to the CFCs, atmospheric concentrations of halons are expected to continue to rise during the next 10 years for Halon-1211 and 20 years in the case of Halon-1301. Figure 2.2 shows the historic measurements for all halons currently detected and their expected future concentrations. Compared with CFCs, less of the halons are emitted promptly. They are used almost exclusively as fire extinguishing agents, consequently the banks in current systems are large. Estimation of emissions from the bank is subject to large uncertainties. In the Technical Options Committee report (TOC, 1998), Halon-1211 emissions were calculated from estimates of the quantities in extinguishers using regional patterns of use. The results tended to understate observed atmospheric concentrations by about 25% (Fraser *et al.*, 1999). Previous industrial estimates (McCulloch, 1992) had tended to overstate emissions and concentrations. The values calculated for Figure 2.2 rely on the Technical Options Committee's estimates for historic and future emission functions, but are adjusted so that the calculated atmospheric concentration, allowing for interhemispherical gradient, fits the measured values in the period 1990 to 1997.

Historical Halon-1301 emissions seem overstated in the TOC report relative to atmospheric measurements, but the estimates in McCulloch (1992) are a good match (Chapter 1 of WMO, 1999). However, because the TOC report lists future emission functions year by year, these were used to calculate the Halon-1301 concentrations shown in Figure 2.2, again scaled to match atmospheric concentrations during the past few years.

If the halons in equipment were conserved and never emitted, the reductions in loadings would average 2 pptv each for Halon-1301 and Halon-1211 over the next 50 years, with a much smaller



Figure 2.1

Atmospheric concentrations of chlorofluorocarbons (CFCs) and chlorocarbons. The points show concentrations determined in background air at Mace Head, Ireland (Derwent *et al.*, 1998). The lines are projections of global average concentrations which, because these compounds are emitted predominantly in the northern hemisphere, will be slightly lower than the quantities determined at Mace Head.



Figure 2.2

Atmospheric concentrations of halons. The points show concentrations determined in background air at Cape Grim, Tasmania (Fraser *et al.*, 1999). For Halons 1211 and 1301, the lines are projections of global average concentrations. In this case, because the compounds are emitted predominantly in the northern hemisphere, global average concentrations will be higher than those determined at Cape Grim. The Halon-2402 line is an extrapolation of the Cape Grim data. contribution from Halon-2402. Together, these amount to 25% of the integrated total bromine loading of the atmosphere during that period.

In a departure from previous reports, the latest Technology and Economic Assessment Panel's report carries predictions for future HCFC production and use; both in developed and developing economies (TEAP, 1999b). These have been used to calculate the HCFC scenarios that follow. Like the CFC scenarios, the new HCFC scenarios are significantly smaller that those used in either SORG 1996 or Chapter 11 of WMO (1999); the expectations in the TEAP report are much lower than the total quantities of HCFCs that could legally be produced within the Montreal Protocol, particularly in developing countries, where there is no upper limit. However, the relatively constant proportion of production of HCFCs in developing countries during recent years, described in 2.1.1 above, tends to support the view that HCFC production there will follow the lower trend.

In the developed nations, consumption of HCFCs is beginning to hit the cap imposed by the Montreal Protocol, reflected in sales of HCFCs that remained constant between 1996 and 1997 (AFEAS, 1999). This is incorporated into the TEAP predictions. Also incorporated into the scenario used here is the recent revision of the European Regulation (Official Journal, 1999). This seeks to implement the last revision of the Montreal Protocol but goes further, reducing the level of the cap on consumption of HCFCs within the EU and setting a phase-out date of 2010. Applied only in the EU, the effect of the Regulation on atmospheric concentrations of HCFCs would be marginal (0.3% of total chlorine loading). Applied globally, the effect would be to reduce cumulative total chlorine loading between now and 2050 by slightly less than 1%.

Although the atmospheric concentration of HCFCs is set to double under the scenario used here, from 130 pptv in the mid 1990s to 260 pptv at the peak in 2008, their contribution then is 8% of the total of controlled ozone depleting substances. Figure 2.3 shows the current measurements and calculated future concentrations from the HCFCs.

2.2 Methyl Bromide

2.2.1 Atmospheric Concentrations and Budget

With a mean, global, background tropospheric concentration of 10 pptv (Chapter 2 of WMO, 1999; Lobert *et al.*, 1995; Schauffler *et al.*, 1998), methyl bromide is the largest carrier of bromine to the stratosphere. When considered as equivalent chlorine this amounts to a loading of 580 pptv: 13% of the total equivalent tropospheric chlorine loading (see section 2.3.1).

Analysis of air trapped in Antarctic firn (unconsolidated snow), suggests concentrations of methyl bromide have increased by 20-25% during the 20th Century, with the most rapid growth occurring in the 1970s and 1980s (Butler et al., 1999). Considerable uncertainties are involved in this type of analysis. Although profiles at two physically different Antarctic sites imply similar atmospheric histories for methyl bromide, a profile from Greenland is in direct contrast, indicating that atmospheric signals of methyl bromide are not always conserved in firn air. Assuming the Antarctic data provide a reliable atmospheric history for methyl bromide, these results suggest current atmospheric concentrations have been strongly perturbed, presumably by anthropogenic activities. This is a potentially important conclusion since, unlike other ozone depleting substances, the budget of atmospheric methyl bromide is not well understood, being complicated by a number of different, and poorly quantified, source and sink processes.

Shortly after the 1994 Scientific Assessment of Ozone Depletion (Chapter 10 of WMO, 1995) a lot of new work was published, such that SORG 1996 provided a considerably updated view of the budget. This included several major new findings such as the ocean being mostly undersaturated (Lobert *et al.*, 1995) and that the deposition to soil could be a significant, albeit quantitatively uncertain, sink of atmospheric methyl bromide (Shorter *et al.*,



Figure 2.3

Atmospheric concentrations of hydrochlorofluorocarbons (HCFCs). Concentrations of HCFC-22 determined in background air at La Jolla, California (crosses) and Cape Grim, Tasmania (downward pointing triangles) (up to 1996 from Miller *et al.*, 1998, thereafter personal communication, B. Miller, 1999). Concentrations of HCFC-141b (normal triangles) and HCFC-142b (plus signs) at Cape Grim (up to 1993 from Oram *et al.*, 1995, thereafter personal communication, D. Oram, 1999). In all cases, the lines are projected globally averaged concentrations.



1995). Furthermore, a re-evaluation of oceanic data led to an increase in the estimated ocean chemical sink (Yvon and Butler, 1996). These changes in our knowledge of individual sources and sinks identified an imbalance in the atmospheric budget of methyl bromide and, in SORG 1996, we highlighted this imbalance and suggested that there may be an additional unknown source of methyl bromide.

Despite new work since 1996, the known budget is still in a similar state. This is reflected in Chapter 2 of WMO (1999). Bacteria in tropical waters have been identified as a biological sink of methyl bromide (King and Saltzman, 1997), reducing the lifetime of atmospheric methyl bromide with respect to the ocean. The group who identified the soil sink have carried out a second study (Hines et al., 1998), this time at ambient concentrations of methyl bromide, the results of which confirm their original conclusions. An independent study (Serca et al., 1998) agrees with the deposition velocities previously determined, but has also shown that extrapolating globally using a different soil inventory leads to an even larger total sink. Recent studies (Baker et al., 1998; Chen et al., 1999) have shown the current global source of methyl bromide from automobiles to be relatively small (1-7 Gg/yr compared to a total of 120 Gg/yr estimated from other known sources or 200 Gg/yr calculated to be required to balance the sinks). If anything, the imbalance is even greater than before, but if certain sources and sinks are pushed to the extremes of their uncertainties, it is possible for the budget to balance. It is probable that there is an additional, as yet unknown source.

2.2.2 Atmospheric Lifetime

Based on new information on the strengths of individual sink processes, Chapter 2 of WMO (1999) gives a best estimate of the lifetime of atmospheric methyl bromide of 0.7 years. This reduction from 1.3 years in Chapter 10 of WMO (1995) is largely due to the additional consideration of the soil sink, which we considered in SORG 1996 and which led to our estimated lifetime of 0.8 years. The reduction to 0.7 years reflects a revision of the atmospheric oxidation rate and of the oceanic sink strength, with the inclusion of the biological loss process. Considering the uncertainties in the estimates of individual sink processes, the lifetime ranges from 0.3 to 1.3 years, illustrating the lack of our quantitative understanding. The range in the lifetime of 0.4 to 0.9 years as estimated by Chapter 2 of WMO (1999) assumes that the uncertainties in the individual sink processes will, to some extent, balance each other out.

2.2.3 Ozone Depletion Potential

Using a lifetime of 0.7 years, and reaction rates as estimated by DeMore *et al.* (1997), giving a Bromine Efficiency Factor (BEF) of 58, an ODP of 0.4 is calculated (Ko *et al.*, 1998; Chapter 2 of

WMO, 1999). This change in ODP from the value of 0.6 quoted in Chapter 10 of WMO (1995) results from the decrease in lifetime, discussed above, and an increase in the BEF from 48 based on the updated reaction rate recommendations. The estimate of the BEF is affected by uncertainties in the reaction rates and is also model dependent. Both the uncertainties in the lifetime and BEF will impact on the certainty of the ODP estimate. Furthermore, the ODP concept is only really applicable for compounds with a lifetime long enough for them to be well mixed throughout the global troposphere (~1 year).

2.2.4 Source Strengths

Based on 1992 production figures from the Methyl Bromide Global Coalition (MBGC, 1994), it was estimated in Chapter 2 of WMO (1999) that 41 (28–64) ktonnes of methyl bromide are released to the atmosphere through fumigation. According to the Methyl Bromide Technical Options Committee (MBTOC, 1998), in 1996, 71.425 ktonnes of methyl bromide were produced, 68.666 ktonnes were used in fumigation and 73 (43–87) % of this was released to the atmosphere (i.e. 50 (30–60) ktonnes).

Observations of methyl bromide over the western Pacific basin during the PEM-West A and B campaigns showed elevated concentrations to be associated with enhanced concentrations of CO, ethane, acetylene and tetrachloroethene (C_2Cl_4), suggesting a significant anthropogenic/urban source (Blake *et al.*, 1997). The only known source of this description is from automobiles, but its strength is estimated to be relatively small.

New work (Gan et al., 1998) published since the compilation of WMO (1999) provides evidence for a source of methyl bromide from plants, in particular Brassicaceae. The plants take up Br- from soil, produce methyl bromide and release it through the aboveground part of the plant. Relationships between the Brlevels in soil and the methyl bromide release rate have been used, along with the global biomass, to calculate global methyl bromide production rates from rapeseed plants (6.6±1.6 Gg/yr in 1996) and from cabbage (0.4±0.2 Gg/yr in 1996). This, along with observations from in vitro studies of discs from other plants (Saini et al., 1995), suggests that the contribution to atmospheric methyl bromide from the total plant biomass may be important, although it requires quantification. This may help to balance the current budget and, since this is a terrestrial source of methyl bromide, it is consistent with the observed north/south gradient. Moreover, the biomass of many cultivated crops has increased over the last few decades (FAO, 1966-1997) such that the source of methyl bromide from rapeseed, for example, is expected to have increased from about 1 Gg/yr in 1960 to nearly 8 Gg/yr in 1997. It should be noted that, although this source from cultivated crops is a result of human activity, it is not controlled by the Montreal Protocol.

In recent years the global source strength of methyl bromide from the oceans has been estimated assuming the open ocean to be a net sink and the coastal and upwelling areas to be a net source (Lobert *et al.*, 1995). However, a study in the open ocean of the North Atlantic (Baker *et al.*, 1999) has found the seawater to be supersaturated with methyl bromide with respect to the atmosphere (i.e. the net flux is from the ocean to the atmosphere). Furthermore, Baker *et al.* (1999) showed a strong seasonality in methyl bromide saturation in coastal waters of the North Sea, with supersaturation in the summer during a bloom of *phaeocystis* and undersaturation in the winter. Therefore it appears that the oceanic source is dependent on the presence of particular phytoplankton, which varies both temporally and spatially. Moreover, the chemical oceanic sink increases with temperature so will generally be at a minimum in polar waters and increase towards the tropics. The spatial variability of the biological sink is not known.

It is uncertain whether a new evaluation of the oceanic source strength of methyl bromide based on the temporal and spatial variability in these processes, which themselves are uncertain, would give a very different global value to that given in Chapter 2 WMO (1999). It is argued in WMO (1999) that it is unlikely any major "hot spot" would be found to greatly change the current estimate. However, it should be noted that the uncertainties quoted in the WMO (1999) oceanic source strength are based on the uncertainties in individual studies rather than those associated with the extrapolation of the results from these studies to the global estimate. With the observation of greater variability in the saturation of methyl bromide, the uncertainties in the global estimate may be larger.

2.2.5 Summary

Methyl bromide is the largest contributor of bromine to the stratosphere. Its budget is still uncertain despite a number of improvements in understanding since the 1994 WMO assessment.

The methyl bromide ozone depletion potential (ODP) has been reevaluated downwards since the WMO 1994 report, based on a decreased estimate of the methyl bromide lifetime (which reduces the ODP) and an increased estimate of the ozone-depleting efficiency of bromine (which increases the ODP). The reduction in the lifetime also implies a larger flux of methyl bromide into the atmosphere than was assumed previously. Overall, there is no reduction in our assessment of the importance of methyl bromide in stratospheric ozone destruction.

Analysis of Antarctic firn air suggests concentrations of methyl bromide have increased by 20–25% during the 20th Century, with the most rapid growth occurring in the 1970s and 1980s. The atmospheric budget of methyl bromide remains poorly understood, but the firn air data suggests concentrations of methyl bromide to have been strongly influenced by human activities. Assuming a lifetime of 0.7 (0.3–1.3) years and that 50 (30–60) ktonnes of methyl bromide used in agricultural

fumigation is released to the atmosphere, only about 20 (5–55) % of total emissions are controlled. The remaining 80 (45–95) % is of natural origin or is released to the atmosphere as a consequence of human activities not controlled under the Montreal Protocol. Note the large uncertainty in these numbers, illustrated by their possible ranges in brackets.

2.3

Halogen Loadings of the Atmosphere

2.3.1 Equivalent Tropospheric Chlorine Loading

In the Montreal Protocol, Ozone Depletion Potential (ODP) is used as a means of differentiating between ozone depleting substances based on their intrinsic potencies. However, because it does not take account of either the quantities emitted or when emissions occur, the ODP value for a compound is not indicative of environmental impact.

As in previous SORG reports (SORG, 1996), the anticipated environmental impact of ozone depleting substances is expressed as their contribution to future chlorine loading of the troposphere, on the premise that this shows the concentration of potential ozone depleters entering the ozone layer above the tropical tropopause. The values quoted are the atmospheric concentrations of the compounds expressed as their equivalent in chlorine so that the actual concentrations are simply multiplied by the number of chlorine atoms in each molecule. For bromine containing compounds, account has to be taken of the different effectiveness of chlorine and bromine. Chipperfield and Pyle (1998) describe how bromine and chlorine reactions are interdependent, so that the relative effectiveness factor for bromine over chlorine (α) actually increases with chlorine loading. In previous SORG Reports (SORG 1993; 1996) we used values of 40 and 46. In view of the higher values in the range 50 to 70 calculated by Chipperfield and Pyle and the calculations that show that α is not a simple function of chlorine loading, we have adopted a mean value of 58 for the whole of next century, as in Chapter 11 of WMO (1999).

Figure 2.4 shows the future contributions to Equivalent Tropospheric Chlorine Loading from methyl chloride, methyl bromide, CFCs, carbon tetrachloride, methyl chloroform, HCFCs and halons. The graph can be compared directly with Figures 2.1, 2.4, 2.7 & 2.8 in SORG 1996 that show the contributions individually.

The best estimates of the development in time of chlorine and bromine loadings are shown in Figure 2.5. The analysis covers the period prior to significant perturbation by man-made compounds, through the present day, to the future as described





Figure 2.4

Projected equivalent tropospheric chlorine loadings from ozone depleting substances. The differing potency for ozone depletion by chlorine and bromine is adjusted as described in the text. Because there is not enough information to justify change, methyl chloride and that fraction (about 80%) of methyl bromide that is assumed not to be controlled are treated as being constant.

above. For chlorine, the historical loadings were calculated from observed atmospheric concentrations and the known emissions of the compounds (AFEAS, 1999). For bromine, historical emissions of halons were used (McCulloch, 1992), coupled with determinations of the atmospheric concentrations of methyl bromide inferred from analyses of firn air samples described by Butler *et al.* (1999).

2.3.2 Equivalent Effective Stratospheric Chlorine Loading

Figure 2.6 compares directly with Figure 11.4 of WMO (1999), in which the contributions are expressed as Equivalent Effective Stratospheric Chlorine loadings. This form of presentation includes an overall adjustment of the tropospheric loadings by a factor to take account of the transport time between the troposphere and the stratospheric ozone layer. The contributions from individual halocarbons are individually adjusted to allow for their different effects on the ozone layer. The relative effectiveness values for ozone depletion are calculated in a 2-D model by Daniel *et al.* (1995). The delay due to transport is set at three years. These effectiveness factors parameterising the differences between chlorine compounds are much smaller than the factor for the difference between chlorine and bromine (α), which equals 58 as described above. They range from 1.11 for



Figure 2.5

Historical and projected total chlorine and bromine loadings of the troposphere. Values for the individual halogens approximate to the inputs to the stratosphere of reactive chlorine (scaled on the left axis) and reactive bromine (scaled on the right as bromine and on the left as equivalent chlorine). The combined loading is expressed as equivalent chlorine. The line at 2.5 ppbv of equivalent chlorine is indicative of the loading during the latter half of the 1970s.



Figure 2.6

Projected equivalent effective stratospheric chlorine (EESC) from ozone depleting substances. Calculated concentrations are adjusted for the potency for ozone depletion of individual substances and for the average transport time to the stratospheric ozone layer. HCFC-123 (CF₃CHCl₂) to 0.35 for HCFC-22 (CF₂HCl). Because the principal HCFCs have fractional effectiveness factors, the plot shows that the effective contribution from HCFCs is lower than the comparable tropospheric chlorine loading plot would indicate. Differences in scenarios are the sole reason for differences between this figure and that in WMO (1999). Nevertheless, we calculate that the equivalent effective stratospheric chlorine will have fallen by 20% in 2020 and 45% in the middle of next century.

2.4 Conclusion

Despite the greater certainty of the numbers used in the scenarios, the halogen loadings reproduced here are similar to those that were shown in the previous SORG 1996 and Chapter 11 of WMO (1999). This is because the changes affect mainly the HCFCs and halons so that, although the expected reductions from previous scenarios are significant in each of these categories, the effect on the total halogen loading (however this is calculated) is comparatively small.

It is now clear from atmospheric observations that equivalent chlorine loading is at a maximum and, all other things being equal, return to loadings that predate the Antarctic ozone hole are still expected to occur towards the middle of next century. None of the changes that have occurred since the last SORG report was published have materially altered this expectation.

Stratospheric Ozone 1999



- Measurements from satellite and radiosondes indicate that for the period 1979–1997, the annual, globally averaged temperature in the stratosphere decreased at a rate varying between 0.6 K/decade in the lower stratosphere and 3 K/decade in the upper stratosphere.
- The dominant cause of the decrease in temperature in the lower stratosphere is believed to be ozone depletion. In the upper stratosphere, the decrease in temperature is due largely to increases in CO₂ and decreases in ozone.
- When stratospheric ozone changes are incorporated into General Circulation Models, the height of the transition from warming (due to increased concentrations of greenhouse gases) to cooling (due to ozone depletion) occurs at about 12 km, in better agreement with the observed temperature trends than when ozone depletion is not included.
- Stratospheric ozone loss is estimated to have offset about 30% of the radiative forcing due to increases in greenhouse gas concentrations since the late 1970s. One serious source of uncertainty in the estimates of the ozone forcing results from inadequate knowledge of the vertical profile of ozone change in the tropical lower stratosphere.
- The increase in UV radiation reaching the troposphere, as a result of ozone depletion, is estimated to have contributed 20–40% of the reduction in methane growth rates in recent years, and 25–40% of the decrease in surface concentrations of carbon monoxide.
- With the anticipated reduction in halogen amounts, ozone is expected to return to similar values to those observed before 1980, by about the year 2050.

- Spring Arctic and Antarctic ozone depletions are predicted to reach their peaks up to about 15 years later than the peak in halogen loadings because of the coupling between stratospheric climate change and ozone chemistry. The amount of maximum depletion and its timing are uncertain due to the complexity of this interaction.
- Comparisons between atmospheric simulations and observations of ozone and temperature trends are complicated by the presence of significant signals from stratospheric aerosol of volcanic origin.
- Sufficient observations now exist to show the statistically significant anticorrelation between surface UV and ozone amounts. Spectrally-resolved UV data from Antarctica and an increasing number of northern mid-latitude stations show continuing increases in UVB radiation during the last decade, consistent with the locally observed ozone depletion.
- Both spectral and broadband UV data series for the UK show small but statistically insignificant increases in UV during the past decade, consistent with the magnitude of the decrease in ozone.
- New estimates of UV from satellite data for the years 1979–1992 show that the zonally averaged trends in annual erythemal UV were greatest in high southern latitudes. In the northern hemisphere the estimated percentage increase was greater at 60°N than at 40°N, and there were no significant changes at latitudes lower than 30°N.

3.1 Introduction

Many of the issues concerning the impacts of ozone depletion which were raised in the last report (SORG, 1996) have been investigated in more detail. A major international report (WMO, 1999, Chapters 5, 7, 9, 10 and 12) has also just been published discussing these issues at length. In this chapter, recent progress is outlined with an emphasis on new results, which were too late to be included in the WMO report.

Although there is still large uncertainty in some temperature measurements, satellite data now indicate clear evidence of stratospheric cooling due to both ozone decrease and increases in the concentrations of other greenhouse gases (section 3.2). Further emphasis has also been placed on the impact of chemistry on climate and the converse, and this has led to the development of coupled chemistry-climate models for the prediction of future ozone amounts (section 3.3). These models are still in their infancy, but the development work has now advanced to the point where multi-decadal coupled chemistry-climate simulations are possible. This has required not just improvements in computer technology, but also improvements in the performance of climate model simulations in the stratosphere. There are still some difficult problems to solve, such as the simulation of the quasibiennial oscillation (section 3.3.4) and the observed mid-latitude ozone loss still lacks a complete explanation, but progress is continuing rapidly.

Longer data records for UV have now become available and the expected increase in UV at the ground with decreasing ozone is no longer limited to isolated reports. It is now seen as a persistent upward change, both at individual locations and regionally (section 3.4). Changes in UV over a few years have now been detected by an increasing number of ground-based instruments.

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Techniques combining modelling with observations have now become available to estimate surface UV from satellite data globally for the period 1979–1992 (section 3.4.6).

SORG 1996 considered briefly the effects of aircraft emissions on ozone. This issue has been reviewed in depth in a special report of the Intergovernmental Panel on Climate Change. A brief summary of that report is presented in section 3.5.

3.2

Stratospheric Temperature Trends and Greenhouse Gases

SORG 1996 reported that the observed long-term cooling of the lower stratosphere was broadly consistent with that calculated by models, when observed ozone depletions were imposed. Since SORG 1996, there has been considerable activity in both quantifying and attempting to explain the temperature trends. Under the auspices of the WMO SPARC project, a Stratospheric Temperature Trends Assessment (STTA) panel has been established (Ramaswamy *et al.*, 1999) and, related to this, the area has been reviewed in WMO (1999), Chapter 5.

3.2.1 Observational Techniques

Several issues limit our confidence in both deducing temperature trends and attributing a cause to them (WMO, 1999, Chapter 5). One major problem is the quality of the observational datasets. Most data on stratospheric temperature have been collected for operational meteorological purposes, rather than for detecting decadal scale trends. Radiosondes have incomplete global coverage and there have been frequent changes in instrumentation over the past few decades. Most satellite trends have been derived from data from a series of different instruments, and there are difficult issues in cross-calibrating them. In addition, satellite instruments measure temperatures over a broad depth of atmosphere, so that they are not directly comparable to radiosonde data, which have much better vertical resolution. A second major problem in attributing observed temperature changes to specific causes is that there is incomplete information on the geographical and vertical distribution of changes in ozone (and other constituents), which are a vital input to modelling studies.

A relatively new source of data is the analyses of meteorological fields constructed by weather forecasting centres. These analyses combine data from radiosondes, satellites and numerical models to produce a dynamically consistent view of the atmosphere which is then used as the initial condition from which to make forecasts.

The analysed datasets have also been immensely valuable beyond weather forecasting. For example, they are used to provide winds in trajectory analyses and chemical transport models (such as those used in Chapter 1) and are used to advance understanding of the seasonal evolution of the atmosphere. However, when they are used for trend studies, they are reliant on the quality, consistency and mix of the radiosonde and satellite data, as well as the underlying performance of the numerical model. Indeed, changes in the configuration of the model can result in false trends; recently the analyses have been repeated (for example at the European Centre for Medium-range Weather Forecasts) to produce so-called "reanalyses" using a stable model configuration. Initially data has been reanalysed for the period since 1979, when satellite data became more readily available; the reanalyses are now being extended back to the 1960s.

3.2.2 Observed Trends

An overall picture of temperature changes in the lower stratosphere over the past two decades can be seen in Figure 3.1. This shows the global mean temperature change derived from Channel 4 of the Microwave Sounding Units on board a series of US operational satellites (see e.g. Christy, 1995). Channel 4 senses emission by the atmosphere from altitudes between 13 and 21 km and so gives a broad picture of the overall behaviour of the lower stratosphere. Two main features are apparent in Figure 3.1. First, the strong warmings around 1983 and 1992 are due to the stratospheric aerosols resulting from the eruptions of the El Chichon and Mount Pinatubo volcanic eruptions. The aerosols lead to an increased absorption of both solar radiation and infrared radiation emitted from below, and so cause a local



Figure 3.1

Changes in global-mean lower stratospheric temperature (13-21 km) derived from the Microwave Sounding Units on-board US operational satellites. The temperature is plotted as an anomaly from the 1984–1990 mean. (Updated from Christy 1995, using data from web site http://www.atmos.uah.edu/ essl/msu.)

warming (although the aerosols act to *cool* the surface). The second feature is a cooling of about 1 K over the 20 year period. Although it is possible to interpret this cooling as a long-term trend since the late 1970s, Pawson *et al.* (1998) have also pointed out that it could be interpreted as two step-changes after the two volcanic eruptions, followed by a relatively constant temperature.

For the first time, STTA attempted a direct comparison of most of the available trend analyses, by comparing trends at similar altitudes. As an example, Figure 3.2 shows the temperature change at 50 mbar as a function of latitude using the radiosonde analyses. It shows broad agreement on the magnitude of the cooling in the northern mid-latitude lower stratosphere. This intercomparison has increased our confidence in the robustness of the observed temperature trends.

Over the period between 1979 and 1994, both satellite and radiosonde data indicate a global and annual mean cooling of the lower stratosphere of 0.6 K/decade. In the northern mid-latitude (30–60°N) lower stratosphere, the cooling is 0.75 K/decade between 1979–1994, a clear acceleration over the longer term (1966–1994) cooling of 0.3 K/decade. In winter/spring lower stratospheres in both polar regions, a cooling of 3 to 4 K/decade is indicated, but the high variability makes the statistical significance of these trends harder to establish. In the upper stratosphere, satellites are the only sources of information. Data from the Stratospheric Sounding Unit indicates a cooling of 2 to 3 K/decade near 50 km over the period between 1979–1997 (Figure 3.3).

3.2.3 Stratospheric Climate Change

In the upper stratosphere, models (see WMO 1999, Chapter 5) indicate that the cooling is due both to decreases in ozone and increases in carbon dioxide. In the lower stratosphere, ozone depletion is the dominant contributor to the cooling (WMO,



Figure 3.2

Zonal mean decadal temperature trends at 50 hPa as a function of latitude over the 1966–1994 period from different datasets. (Taken from Ramaswamy *et al.* (1999).)

Temperature change 1979-1997 K/decade -2.00 -1.00 Pressure hPa .0.50 10 0.00 So -0.50 1 00 -1.00 1.00 0.50 100 -50 0 50 Latitude

Figure 3.3

Annual mean temperature trend for the period 1979 to 1997 from SSU/MSU radiance data, after removal of the solar cycle. The light shading indicates statistically significant cooling at the 95% confidence level and successively darker shading indicates cooling at significantly greater than 1 K/decade and 2 K/decade. (Taken from Scaife *et al.* (1999).)

1999, Chapter 5), reaffirming the conclusions of earlier assessments. Figure 3.4 shows results from a General Circulation Model (GCM) on progressively adding different climate change mechanisms (adapted from Hansen *et al.*, 1997). Increases in well-mixed greenhouse gases (mainly carbon dioxide) result in a cooling in the mid stratosphere (above about 20 km (50 mbar)). When ozone changes are included, the cooling extends into the lower stratosphere.

However, there remain outstanding questions. Firstly, the model studies have had to make assumptions about the imposed vertical distribution of ozone change. Most GCM studies (e.g. Ramaswamy *et al.*, 1996; Tett *et al.*, 1996; Hansen *et al.*, 1997; Folland *et al.*, 1998) have assumed that the ozone change is restricted to heights within a few kilometres of the tropopause. There is a strong dependence of the modelled cooling on assumptions made about the vertical profile. This has been demonstrated by Forster and Shine (1997) (albeit in a so-called fixed dynamical heating model that neglects the potential impact of changes in circulation on temperatures).

Second, it is possible that trends in stratospheric water vapour have contributed to a cooling of the lower stratosphere which can, at some altitudes, cause a cooling which is a significant fraction of that due to ozone loss (Forster and Shine, 1997). Although observations of stratospheric water vapour changes are limited (see Chapter 1) this indicates that we cannot yet be certain that ozone is the dominant cause of the observed temperature changes. In the case of stratospheric water vapour changes, as their cause(s) are not yet certain, this limits the confidence with which the observed coolings can yet be fully attributed to human activity.

A third outstanding question concerns the inherent variability of the atmosphere and the extent to which the observed trends in temperature are larger than this variability. One particular issue, discussed in Chapter 1, concerns recent changes in the tropospheric circulation, which can be characterised by the so-called North Atlantic Oscillation, and the related so-called Arctic Oscillation (Thompson and Wallace, 1998). The Arctic Oscillation can be characterised by the difference in surface pressure between the Arctic and mid-latitudes. Over the past 30 years there has been a tendency to a decrease in Arctic surface pressure, a strengthening of the mid-latitude westerly flow in the troposphere and an associated cooling of the polar stratosphere. Hence, in searching for causes of the observed cooling it is necessary to include such variability in addition to the "local" causes, such as changes in the concentration of water vapour and



Figure 3.4

Change in global-mean annual mean temperature profile between 1979 and 1995 based on linear trends calculated using the NASA GISS GCM, as different climate forcing mechanisms are added, one-by-one, to the model. The calculations use observed sea surface temperature variations. (From WMO 1999, adapted from Hansen *et al.*, 1997.)

ozone in the lower stratosphere. It remains an open question as to why there has been a shift in the circulation. It could be unforced natural variation, be linked to human-induced climate change in the troposphere or be due to the changes in stratospheric ozone or other constituents (see Thompson and Wallace, 1998 and Ramaswamy *et al.*, 1999 for discussion).

3.2.4 Tropospheric Climate Change

SORG 1996 noted that changes in stratospheric ozone also affect tropospheric and surface climate. Ozone loss in the lower stratosphere acts to cool the surface and troposphere and so opposes, to a certain extent, the warming effect of increased concentrations of carbon dioxide and other greenhouse gases. SORG 1996 concluded that the ozone loss might have offset about 30% of the greenhouse gas effect since 1979. At that time, the impact of ozone changes on climate had been largely assessed using radiative forcing; this quantifies the impact of the ozone changes on the Earth's energy budget but does not compute the impact on, for example, surface temperature. Since then, GCM studies (e.g. Tett *et al.*, 1996; Hansen *et al.*, 1997) have started to report the impact of ozone changes on the simulations of recent climate change.

WMO (1999, Chapter 10) estimated a global and annual mean radiative forcing due to stratospheric ozone loss since the late 1970s of -0.2±0.15 Wm⁻², similar to that reported in SORG 1996. (The quoted uncertainty in this estimate is an indication of the spread of values from the available literature, rather than a formal indication of uncertainty.) This estimate did not take into account the revised ozone trends presented in that report. Forster (1999) has now estimated the forcing using the new trends together with updated estimates of the vertical profile of the ozone change, from SPARC/IOC/GAW (1998). There are severe problems in estimating changes in ozone in the tropical lower stratosphere. Stratospheric Aerosol and Gas Experiment satellite data, used in different ways, can indicate either an increase or a decrease; this severely reduces our confidence in radiative forcing estimates. When only statistically significant ozone changes are used, Forster's "best guess" for the forcing between 1979-1997 is -0.10±0.02 Wm-2. This is significantly less than the central value given by WMO (1999), but still important at the 15% level, compared to well-mixed greenhouse gases.

The GCM studies which have incorporated stratospheric ozone changes indicate that in the past 15–20 years the ozone loss will have resulted in a surface cooling of about 0.1 K (Tett *et al.*, 1996; Hansen *et al.*, 1997). Again, this is roughly 30% of the warming that these models simulate for changes in greenhouse gas concentrations. However, such a 0.1 K change is no larger than the unforced internal variability, and it does not lead to a significant improvement in the agreement between models and observations. In contrast, in the upper troposphere and lower stratosphere, the ozone changes do improve agreement between modelled and observed temperature changes.

One significant aspect of observed climate change is the height of the transition from "tropospheric" warming to "stratospheric" cooling. On including the ozone changes, this transition height is reduced, compared with calculations accounting for changes in well-mixed greenhouse gases alone, bringing models and observations into better agreement (e.g. Hansen *et al.*, 1998; Folland *et al.*, 1998). Figure 3.4 shows that the transition from warming to cooling changes from about 20 km (50 mbar) for well-mixed greenhouse gas changes only, to about 12 km (200 mbar) when ozone changes are included.

3.2.5 Impact of Stratospheric Ozone Loss on Tropospheric Composition

As reported in SORG 1996, an additional impact of stratospheric ozone changes is that the consequential increase in UV radiation reaching the troposphere affects photochemical processes there. There has been little change in our understanding of this area since then. It has been estimated that the increased UV since 1979 has contributed 20–40% to the reduction in methane growth rates in recent years, and 25–40% of the decrease in surface concentrations of carbon monoxide (WMO, 1999, Chapter 10).

3.3 Future Trends in Ozone

Predictions of the future behaviour of ozone have been obtained using two-dimensional (2-D, latitude-height) and 3-D (latitudelongitude-height) models. Since the last report (SORG, 1996) new predictions have appeared in WMO (1999, Chapter 12) and this section provides further explanation of some of the results presented there together with some new results.

3.3.1 Future Recovery of Ozone

It is important to consider when ozone is likely to recover following the ozone losses of the 1980s and 1990s. Recovery in the sense of negligible impact of chlorine compounds is unlikely to occur before about 2050, when chlorine levels are expected to have reduced considerably and the Antarctic ozone hole will become small. By this time, the recovery will be to a different atmosphere, in the sense that increases in greenhouse gases (GHGs) will have cooled the stratosphere resulting in a reduction in homogeneous ozone loss, while increases in N₂O will have increased NO_x to counteract this effect. Changes in H₂O will likely have complicated effects due to both radiative and chemical impacts. Thus, the ozone vertical distribution is likely to be different in the year 2050 from that during the period before substantial ozone depletion was observed, due to changes in temperature, transport, water vapour and nitrogen species. The date of ozone recovery could alternatively be defined as the year when ozone has increased significantly from its minimum. However, as interannual variability is large in the north, several years after the actual minimum may need to elapse before we can be certain that recovery has occurred.

3.3.2 Future Ozone Predictions – Two-Dimensional Models

Two-dimensional model results imply an increase in ozone soon after upper atmospheric chlorine levels peak, which is expected in about the year 2000. A range of model results is shown in WMO (1999), Chapter 12, indicating that the overall structure of recent observed local and column ozone trends has been reproduced. However, many of these models have difficulty in providing even a reasonable simulation of established features such as the Antarctic ozone hole. Therefore, their main advantage is in investigating the impacts of a range of constituent changes on general quantities such as globally averaged ozone.

The main conclusions of WMO (1999), Chapter 12, were that increases in N2O and aerosol amounts will delay recovery of ozone while increases in CH₄ will have the opposite effect. Increases in N₂O increase NO_x and lead to mid-stratospheric ozone losses by gas phase destruction. Increases in aerosols provide sites for heterogeneous chemistry leading to ozone loss in the presence of halogen molecules. In contrast, increases in CH_4 lead to increased production of HCl and a reduction in the amount of active chlorine. Figure 3.5 shows the projected changes in total ozone for a range of 2-D models. All the models are consistent in showing a minimum in ozone after the Mt Pinatubo eruption. There is then a gradual increase in ozone amounts, taking several decades to return to 1980 values, but the details differ between one model and the next. These results also suggest that ozone could be significantly perturbed if another eruption were to occur within the next few years, before halogen amounts have reduced substantially.

3.3.3 Ozone Predictions of Three-Dimensional Models in Polar Regions

To simulate the important coupling processes between ozone and climate, and to simulate realistic interannual variations, coupled chemistry-climate models are required. These need to be threedimensional and hence are very demanding on computing resources. Nonetheless, results from four 3-D models are presented in WMO (1999), Chapter 12 and they show the same or greater ozone depletion in the Arctic in 2015 as 1995, despite the assumed reduction in chlorine of between 3% and 16%. The two models that have been integrated beyond 2010 suggest that the minimum Arctic ozone will be attained during the decade 2010 to 2020, although these results are subject to large uncertainty.



Figure 3.5

Percentage change in 65°S–65°N annual average column ozone relative to the year 1979, for a range of 2-D models for the period 1980 to 2050. The TOMS observations for the period 1979 to 1997 are indicated by the solid circles. (From WMO, 1999.)



Figure 3.6

Minimum total column ozone south of 65°S for a range of 3-D models. The GISS model results are given by open circles. The bars at 1990 and 2015 show the one standard deviation variability on the ECHAM3/CHEM over the last ten years of their simulations. The CNRS results are shown by the large open circle, with the bars indicating the range of values over 5-year simulation. The UKMO results are indicated by triangles. Observations from TOMS for the period 1979–1997 are given by closed circles. (Based on WMO, 1999.)

In one particular model simulation (Shindell et al., 1998) the effect of increasing GHGs is to produce a more symmetric Arctic vortex with reduced amplitude planetary waves and a resultant reduction in the frequency of stratospheric warmings. This significantly affects ozone amounts since heterogeneous chemistry is favoured during cold winters without stratospheric warmings; while during winters with stratospheric warmings substantial transport of ozone into polar regions occurs and temperatures increase sufficiently to prevent further heterogeneous chemistry (see e.g. Austin and Butchart, 1994). Although a reduction in major warmings in the early part of the 1990s has been observed (Pawson and Naujokat, 1997), it is not clear that this can necessarily be attributed directly or indirectly to increasing GHGs. For example, in other climate model simulations (e.g. Mahfouf et al., 1994; Butchart et al., 1999), a different sensitivity to increasing GHGs was noted. Despite the uncertainties regarding the simulation of stratospheric warmings, some delay in Arctic ozone recovery is likely because of the impact of climate change on polar stratospheric clouds (Waibel et al., 1999).

Results from a range of models for the Antarctic ozone hole are shown in Figure 3.6 for the period 1975 to 2070 together with measurements up to 1997 from TOMS. Despite the fact that the processes are quantitatively better understood than for the Arctic, there is still considerable variability between the individual models. Of the models which provide results in the second half of the next century, one model (Shindell *et al.*, 1998) shows a near steady recovery in spring Antarctic ozone whereas in the UKMO results (Austin *et al.*, 1999, plus unpublished results) the impact of increasing greenhouse gases is predicted to lead to increased production of ice polar stratospheric clouds in the lower stratosphere, and to enhanced heterogeneous reactions. Consequently, even though the halogen amounts are projected to decrease gradually, it does not follow that steady recovery of Antarctic ozone will occur.

3.3.4 Remaining Uncertainties

Improved models with higher resolution and more complete chemistry are needed to permit continuous multi-decadal simulations. Such simulations would also allow studies to investigate interannual variability and to predict more reliably the impact of GHG increases on ozone amounts. Without a full indication of model interannual variability it is difficult to separate the effects of model internal variability from systematic effects due to GHG and halogen concentration changes. To date, chemistry-climate models of sufficient complexity have not been simulated for a sufficient period to address this problem fully.

The relatively poor simulation in the frequency of stratospheric warmings mentioned above is also a limitation, particularly in view of the tendency of some models to cluster warmings together. Hamilton (1998) has shown that this clustering can be removed by imposing a quasi-biennial oscillation (QBO) in the

tropical stratospheric winds. Hence, accurate simulations of ozone may not be available until the QBO can be adequately simulated in models.

An additional uncertainty is the impact of tropospheric chemistry. For example, Dameris *et al.* (1998) suggested that tropospheric ozone could increase and counteract to some extent any future stratospheric ozone decrease. Also, although the effects of the aerosols produced from major volcanic eruptions are now better known, their timing is uncertain and large temporary perturbations to ozone might occur while halogen loadings remain high. Clearly, to resolve all these problems requires the development of a chemical model capable of treating both the tropospheric and stratospheric regions effectively as well as accurate scenarios of the ozone source and sink molecules.

3.4 UV Radiation

Stratospheric ozone is a major absorber of solar UV, but is not the sole influence on UV radiative transfer. On a global basis, the sun's position (determined by latitude, time of day and time of year) is the most important factor determining daily UV doses, and significant surface altitude can also be a more important determinant of UV than regional differences in ozone column (Sabziparvar et al., 1998). Other atmospheric variables (aerosols, pollutants, and most noticeably cloud) can have a dramatic effect on local UV at the surface and are highly variable on several timescales: day to day, season to season, and year to year. As stratospheric ozone also undergoes significant short-term fluctuations about its longer term seasonal cycles, the relationship between trends in stratospheric ozone and UV at the ground becomes confused. This implies that a long time series of UV measurements is required to enable a change in UV resulting from changing ozone to be distinguished from natural variability.

At the time of the last SORG report (SORG 1996) existing UV data records were generally too short to identify trends in UV. The extraordinary depletion of ozone over Antarctica during the spring results in a large and easily identified increase in UV, but elsewhere ozone decline had been slow and gradual with less opportunity to identify a cause-effect relationship. Nonetheless, clear anti-correlations between UV and ozone had been observed at several mid-latitude sites, both in the seasons following the eruption of Mt Pinatubo, when ozone was exceptionally low, and on individual days when particularly low ozone for a location coincided with clear skies. Short periods of anomalously high UV had thus been identified, but were insufficient to determine continuing change against the very variable UV signal. The measured data were supported by modelling exercises that used a satellite-derived ozone climatology to calculate UV in the absence of cloud for the years 1979-1995. A gradual increase in

UV at mid-latitudes was indicated. Thus we concluded that current levels of UV were higher than they would otherwise have been without ozone depletion, but natural variability is so great that a trend could not be clearly identified from the short data records available.

3.4.1 Update on Observations

Since the last report, data records have become several years longer. Also, there have been continuing efforts to improve the stability of UV instruments at individual sites and to improve the agreement between sites (Webb, 1997; Thompson et al., 1997; Webb et al., 1998; SUSPEN, 1998). With the support of meteorological data and the availability of spectrally-resolved data, it is now possible to identify a general upward trend in UV. Reanalysis of erythemally effective UV (to account for a potential shift in calibration and other instrument problems) from a network of broadband meters in the USA dating back to the 1970s has also shown, contrary to previous reports, that UV did not decrease during the 1970s and 1980s but showed some small (statistically insignificant) increases as ozone depletion first began to take effect (DeLuisi and Barnett, 1992; Weatherhead et al., 1997). Unfortunately, this network has not been maintained throughout the past decade and there is no contiguous set of UV data for the ozone depletion era.

The last few years have seen an extension of UV data records to the point where a medium term (of order 10 years) rate of change of UV might be calculated. However, with a relatively short time series with respect to the variability of the signal, the addition of an extra year's data can have a large effect on the overall rate of change for the series. In addition many of the current UV data series began around the time of the Mt Pinatubo eruption and resulting low ozone anomaly (Chapter 1), thus giving potentially high UV at the start of the data record. It is also important to note that ozone trends are generally presented as zonal averages from a large number of measurement stations, or a global satellite view, while the UV data below represent local measurements from a single site. Thus while the UV data should be consistent with local ozone trends, it may not appear so well matched to the reported zonal or global ozone trends.

3.4.2 UV Measurements in the UK

Spectrally-resolved data have been routinely collected at Reading (UK, 51.5°N) for the years 1993–98, and were therefore gathered under varying amounts of cloud. Although the impacts of cloud are slightly wavelength dependent (Kylling *et al.*, 1997; Celarier *et al.*, 1999), they can be largely eliminated by taking a ratio between two UV bands, simplifying the time series of absolute measurements. By contrast the effects of changes in solar zenith angle (SZA) are strongly wavelength dependent and so the analysis is performed in narrow SZA bands. Hence in the results described here the monthly mean values of the ratio of

erythemally-effective UV (UV_{ery}) to UVA are analysed for SZAs between 75° and 80°. The advantage of this choice of SZA is that it occurs in every month of the year whereas a band of smaller SZA does not occur in winter.

Figure 3.7 shows a clear anticorrelation between the seasonal cycles of ozone and the UV ratio (Bartlett and Webb, 1999). Supporting data from a visible photodiode indicates that there was interannual variability in cloud (with summer 1995 being least affected by cloud), but no trend in cloud cover. As UVA is only very weakly affected by ozone, any increase in the ratio indicates an increase in UV_{ery} associated with reduced ozone. Over the 6-year period 1993–1998 the change in monthly mean ozone values at Camborne (closest ground-based station to Reading) was -2% with a correspondingly small (statistically insignificant) increase in UV_{ery}: UVA of +3.5%.

The National Radiological Protection Board (NRPB) has six sites monitoring erythemally effective UV and UVA throughout the length of the UK. The longest running site is at Chilton (51.6°N), with records dating back to 1988. Analysis of these data shows a small but statistically insignificant increase of 5% in 9 years in annual totals of erythemally effective UV (Driscoll *et al.*, 1998). However, there has been no attempt to remove the effect of cloud during the analysis and data from the early years show large departures from the annual average, perhaps due to particularly sunny or cloudy summers. In the later years the data are qualitatively more consistent with the changes seen at Reading; annual integrated UV increases from 1992 to 1997 with a particularly high value in 1995 when the summer was notably less cloudy than usual. Rates of change have not been determined



Figure 3.7

Monthly means of the ratio of erythemally-effective UV to UVA at solar zenith angles between 75° and 80° at Reading for the period 1993 to 1998 (lower panel) in comparison with monthly mean ozone measurements taken from a Dobson spectrophotometer at Camborne (upper panel).

from other NRPB sites that have data records of 4–6 years in length. Several of the sites identified short periods of high UV during low ozone events and clear skies, especially in March 1996 and April/May 1997 (Driscoll *et al.*, 1998).

3.4.3 UV Measurements in Europe

Elsewhere in Europe, Zerefos *et al.* (1997) report on the changes in UV at four stations between 40.5° and 64.1°N, with data records of between 3 and 6 years in length. They analysed monthly means of absolute UV irradiances at 305nm (which is well absorbed by ozone) at a solar zenith angle of 63 degrees. For three of these stations the ozone trend varied between -1.5 and +0.5% per year with corresponding UV changes of +9% and -3%per year. Results from the fourth station at Garmisch Partenkirchen highlighted the effect that cloud can have on UV amount: this site recorded very high monthly mean UVB for October 1995, which has been attributed to the exceptionally high number of clear days in that month, and shows that cloud conditions cannot be neglected in such analyses, especially in the short-term.

3.4.4 UV Measurements Elsewhere

One of the longest spectral data records is that from Toronto, Canada, where from 1989–1997 changes in daily average UV fluxes for the months May to August (when most UV exposure is expected) equate to 15% per decade at 300nm, for an equivalent decrease in ozone of 4.3% per decade (WMO 1999, Chapter 9). Radiation at 300nm is more strongly absorbed by ozone than longer wavelengths, or the weighted erythemal UV waveband, and therefore more responsive to changes in ozone (Figure 3.8). The change in the flux at 324nm (a wavelength of weak ozone absorption) for the same period was zero.

In Antarctica, where ozone depletion is larger than elsewhere, spectral UV monitoring has been in operation at three stations: since 1988 (Palmer), 1989 (McMurdo) and 1991 (South Pole). All three stations show significantly elevated UV irradiances during periods of corresponding low ozone. Absolute average doses for a given period depend on latitude (controlling solar elevation and day length) and cloud amount as well as ozone. Nonetheless, the maximum daily average doses from Palmer station (furthest from the Pole at 64.8°S) during the Austral spring are higher than those of San Diego (32.8°N) in mid-summer. The data from all sites has been tested for trends in the daily average DNAweighted irradiances. The most compelling trend was found at Palmer station for the month of November, where the trend of +15% per year since 1988 had a correlation coefficient of 0.86 (n=9). The corresponding change in the waveband 400-600nm, unaffected by ozone, was +2.7% per year with a correlation coefficient of 0.52 (Booth et al., 1998).



3.4.5 Determining UV Trends

While these observations confirm that UV has increased with decreased ozone, they should not yet be used to infer long-term trends since the data records are still too short to detect statistically significant trends (except in Antarctica), given the large variability in ozone and cloud. A medium-term trend can also be altered significantly by the addition of an extra year or two of data (Zerefos *et al.*, 1997).

The UV observations discussed above originate from a small number of sites, using different instruments with varying data formats and analysis techniques. They all show that UV has changed (generally increased) in the opposite sense to ozone at the same location. However, each local trend can be influenced by local climate conditions and pollution, and quantitative trends should not be applied to wider regions. In the following section, a new technique to determine surface UV from satellite measurements is described. The data from this technique could complement the ground-based data and provide a coarse resolution, but global, view of UV fluxes.

3.4.6 Determination of UV Fluxes from Satellite Data

The advantages of satellite data are their global coverage and uniformity. A single well calibrated instrument (TOMS) on board the Nimbus 7 satellite operated for the period 1979–1992 and has been used for the following analysis. Unfortunately, the advantage of uniformity of data is limited to this period, after which this TOMS instrument ceased to operate. Measurements of



Figure 3.8

UV irradiance trends for May-August at Toronto, Canada in % per decade as a function of wavelength. The ozone absorption spectrum is also shown for comparison, suggesting that much of the UV change is due to ozone change. (Taken from Wardle *et al.* (1997).)

backscattered radiances have been used to derive simultaneous measurements of ozone, cloud, aerosol, ground reflectivity and extraterrestrial solar radiation, although the influences of aerosols, tropospheric ozone and clouds at low altitude cannot be completely determined. A UV radiative transfer model was then used to estimate UV at the Earth's surface (Herman et al., 1996). This has provided UV irradiances since 1979, well before many of the current ground-based stations became operational. However the fluxes are not a direct measurement but come from a calculation based on knowledge of many parameters and their effects on UV, giving uncertainties in the resulting data that depend on the accuracy of the input data and mechanisms of the model. These new estimates of UV from satellite data for the years 1979-1992 (Herman et al., 1996), over the latitude range 65°N to 65°S, show that the zonally averaged trends in annual erythemal UV were largest in high southern latitudes (9±6% per decade at 60°S). In the northern hemisphere the corresponding estimate was 3.7±3% at 60°N and 3±2% at 40°N. There were no significant changes at latitudes lower than 30°N and no change in the associated UVA.

Comparison of TOMS-derived UV and ground-based measurements of UV from Toronto showed good agreement, but with some seasonal differences (Eck *et al.*, 1995; Labow *et al.*, 1999). The satellite method, with all its potential advantages, now needs further validation against ground-based stations.

3.5 Impact of Aircraft

The exhaust emissions from the growing fleet of passenger aircraft provide a potentially significant perturbation to atmospheric composition and consequently may influence climate and ozone. This concern has been addressed recently in assessments of the possible influence of both subsonic aircraft flying in the tropopause region and supersonic aircraft (High Speed Civil Transport, HSCT) flying in the stratosphere (15–21 km altitude) (Brasseur et al, 1998; IPCC, 1999). The emissions of concern for ozone are water, NO_x, sulphur gases, and aerosols and soot. These pollutants can affect the gas phase and heterogeneous chemistry which influences ozone concentrations, and hence temperature and transport, in the lower stratosphere and upper troposphere. Most emissions occur in the main flight corridors of the northern hemisphere mid-latitudes. However the impact in the stratosphere can be more widespread, due to large scale atmospheric transport of the pollutants.

The assessment of the impact of subsonic aircraft on ozone has been made with tropospheric chemical/transport models, using global emissions of NO_x from the current fleet and from postulated future fleets. Models used to assess the impact of subsonic aircraft do not yet have an adequate representation of

chemistry and dynamics in the critical tropopause region. Consequently the results summarised below have considerable uncertainty.

Subsonic emissions result in significant increase in the average concentrations of NO_x (factor of about 2 increase at 12 km) and a decrease in the lifetime of CH_4 as a result of increases in tropospheric OH concentration. In all models ozone is predicted to increase but the impact is restricted to northern mid- and high-latitudes. For the latitude band 30–60°N, the estimated maximum yearly average tropospheric ozone increase at cruise altitudes (above background with no aircraft) is 6% in 1992 and 13% in 2050. The response is nearly linear with changes in NO_x emission. The effects on the column of ozone are +0.4 % (1992) and +1.2% (2050).

The assessment of the impact of HSCT aircraft has been made for IPCC using nine stratospheric models. A projected fleet of 500 HSCT was assumed, with a common emission scenario for NO_x (Emission index, EI = 5) and H₂O (EI = 1230). The HSCT fleet was assumed to replace a part of the subsonic fleet so that the total impact would be diagnosed. The following effects were concluded for a 2015 atmosphere:

Column change in O₃ slightly negative (-0.4% with a range of 0-1.1%), reflecting a negative change of 40 to 60 ppbv at 30 km and a positive change of 15 ppbv at 20 km.

- Significant perturbations to stratospheric NO_y and water vapour; at 20 km altitude in the northern hemisphere, Δ NO_y ~ 5%; ΔH₂O ~ 10%.
- The water emissions are predicted to add ~0.1 Wm⁻² radiative forcing; this estimate has a large uncertainty (~ a factor of 3).
- The mainly northern hemisphere emissions of water would affect aerosol reactivity (PSC and sulphate) in both hemispheres. Ozone losses are due mainly to changes in H₂O with a smaller contribution from NO_x.
- The response to sulphur emission depends on how much of the fuel sulphur is converted to sulphate particles in the aircraft exhaust plume, with larger O₃ loss from larger conversion to aerosols.

The effects are highly dependent on the altitude of injection due to the changing chemical and dynamical regimes in the lower stratosphere. The overall impact on ozone of this probably unrealistically large projected fleet of HSCTs is small compared with perturbations due to halogens.



4 The Impacts of UV Changes

- UVB can damage the skin and eyes, and cause changes in the immune system. The dose of UVB received by the human population is strongly dependent on behaviour. Social changes over several decades have served to increase exposure to UVB, and this would have been exacerbated by ozone depletion.
- Incident UV at the ground is modified not only by stratospheric ozone depletion but also by climate change, for example by changes in cloud cover. Alterations in UK climate (such as warmer weather) are likely to further influence patterns of behaviour and thus doses of UV. Increases in temperature could also exacerbate the effects of UVB on materials.
- 4.1 Introduction

In 1996 the Ultraviolet Measurements and Impacts Review Group (UMIRG) published a report 'The Potential Effects of Ozone Depletion in the United Kingdom' to complement the SORG 1996 report.

In that report we detailed the potential effects of ozone depletion, mediated through increased ultraviolet radiation, on human and animal health, terrestrial and aquatic ecosystems, materials and atmospheric chemistry, and the resulting socioeconomic consequences. The biological and chemical effects of UV described in the report are general, while more specific impacts were discussed as they relate to the UK, its population, and expected changes in ozone and UV. In this chapter of SORG 1999 we update the information from UMIRG 1996 in light of recent research on UV effects, and the contents of the preceding chapters of this report.

The effects of solar UV radiation depend on the doses received by the population concerned, and these depend on:

- 1. The UV flux in the environment;
- 2. Patterns of behaviour which bring about exposure to UV in the environment;
- 3. Any actions taken by the population to mitigate the effects of such exposure.

Some populations, those with mobility, have the ability to influence their exposure by moving in or out of the sun. The clearest example is the human population, which has a great deal of control over location and also the use (or not) of protective devices such as clothing and sunscreens. Dose control in plants is limited to orientation of leaves and the development of protective pigments (the botanical equivalent of a tan), while

- Increased UVB alters interactions between organisms in both terrestrial and aquatic systems, with unpredictable consequences for ecosystem function and diversity.
 Further uncertainty arises from the interaction between UVB and other environmental pressures.
- Localised extreme increases in UVB, for example over the Antarctic, may have wider consequences via aquatic food webs.

orientation in many planktonic organisms is severely restricted. Inanimate materials (paints and polymers) receive doses that depend simply on the incident UV of the environment into which they are placed.

UV irradiances and other changes in the atmosphere associated with ozone depletion have been discussed in chapter 3. The latest understanding of the consequences of those changes are discussed below.

4.2 Effects on Human Health

4.2.1 Physiological Effects

Evidence continues to accumulate that DNA photodamage plays a major role in the adverse effects of solar UV. Human studies in vivo show that 300 nm is the most effective wavelength for such damage and provide evidence that DNA is a chromophore for sunburn (Young et al., 1998): susceptibility to sunburn is a risk factor for skin cancer. DNA is also a candidate chromophore for UV-induced cytokines (Nishigori et al., 1996; Kibitel et al., 1998; Petit-Frere et al., 1998) that are thought to play a major role in the inflammatory and immunosuppressive effects of UV. Furthermore, there is increasing evidence that UVB-induced mutation of a tumour suppressor gene (p53) is important in human non-melanoma skin cancer (Ziegler et al., 1996). Recent data also suggest a role for UVB-induced p53 mutation in malignant melanoma (Zerp et al., 1998). Photoageing is another consequence of exposure to sunlight. Recent studies have shown that single and repeated sub-erythemal exposures of UVB are able to induce synthesis of collagen-degrading enzymes in human skin in vivo (Fisher et al., 1996; 1997). It is hypothesised that UVB plays an important role in photoageing.

Recent estimates (UNEP, 1998b) suggest that the increased risk of cataract and skin cancer due to ozone depletion would not have been adequately controlled by implementation of the Montreal Protocol alone, but can be achieved through implementation of its later amendments (Copenhagen 1992 and Montreal 1997). These estimates indicate that under the Montreal Amendments, incidences of cataract and skin cancer (all types) will peak around the mid-part of the next century at additional incidences of just under 3 per 100,000 and about 7 per 100,000, respectively. For the UK population of approximately 60 million this would imply 1800 and 4200 additional cases of these two diseases per annum. Thereafter the increase in disease rates attributable to ozone depletion is expected to return almost to zero by the end of the next century (as skin cancer typically results from several decades of UV exposure the response of the disease follows later than changes in exposure). Some estimates for increases in nonmelanoma skin cancer as a result of ozone layer depletion have been based on adjustments of a mouse action spectrum for differences in ex vivo epidermal UV transmission between the two species (de Gruijl and van der Leun, 1994). However, recent human data suggest that UVB damage to the crucial epidermal basal layer is greater than would be predicted from UV transmission measurements (Young et al., 1998), which would increase such risk estimates for non-melanoma skin cancer.

Although there is substantial evidence that exposure to ultraviolet radiation can also modulate immune reactions in humans, reliable quantitative risk estimates for the effects of UVB on the immune system and the consequence of this on infectious diseases are not yet possible (Garssen *et al.*, 1998). However, recent human studies *in vivo* show that cell-mediated immunity is readily suppressed, locally and systemically, by a moderate solar simulated radiation sunburn over a small area of skin (Kelly *et al.*, 1998). There is also now some evidence that solar exposure may have a role in non-Hodgkin's lymphoma (Bentham, 1996; Hartge *et al.*, 1996), although the evidence for this remains controversial.

The quantitative risk estimates for skin cancer are only valid if all other factors which determine risk, notably human behaviour, remain unchanged. Public health campaigns aimed at encouraging people to reduce their sun exposure by sun avoidance and the use of photoprotective measures such as sunscreens, clothing and (for the eyes) UVB filtering sunglasses, may achieve a reduction in average population UVB exposures, and presumably cataract and skin cancer rates, which could more than offset the adverse effects of ozone depletion (Hill and Boulter, 1996). Some preventive measures, however, may not be as effective as commonly believed. The role of sunscreens in the prevention of skin cancer is controversial with some studies suggesting that sunscreen use is associated with increased risk of malignant melanoma (most recently by Autier et al., 1998). The reasons for this observation are not known. One of several possibilities is that sunscreen use encourages people to stay in the sun longer. It is also widely believed that a tan is photoprotective but recent human studies have shown that tanning offers very

modest protection against sunburn (Sheehan *et al.*, 1998). It is evident, therefore, that human behaviour may well have a greater impact on individual exposure to sunlight, and consequent risks of eye and skin damage, than atmospheric changes, but the contribution is complex and not easy to generalise.

On the other hand, the calculated risks imply full compliance with restrictions on the production and consumption of ozone-depleting chemicals throughout the world (targets to date have been met, see chapter 2). If, in the future, compliance does not continue, damage to the ozone layer could be greater than hitherto expected and biological impacts could be more severe (UNEP, 1998b). Also, as noted in section 3.3, changes in greenhouse gases will likely enhance ozone depletion and lead to a larger risk than the previous calculations imply.

4.2.2 Behavioural Influences

In our earlier UMIRG report we stressed that the large increase in skin cancer in the UK (and elsewhere) in the past half century (Figure 4.1) is likely to have been the result of changes in lifestyles and behaviour rather than in UV flux. One important factor has probably been the growth of holiday travel to foreign destinations and recent data (ONS International Passenger Survey, 1998) confirm that this trend has continued unabated in the 1990s. Furthermore, in recent years the most rapid increases in foreign holiday travel have been to long-haul destinations at low-latitude, where UV levels are typically high. For example, holiday visits to the USA (where Florida is the most popular destination) increased 15-fold in the 20 years up to 1997. It therefore seems



Figure 4.1 Annual melanoma deaths in the UK population from 1949–1997.



likely that changing patterns of holidaymaking continue to be an important factor tending to increase the overall UV doses received by the UK population and any associated health risks. Closer to home, recent results from the General Household Survey (ONS, 1998) confirm that the long-term growth in outdoor leisure activities has continued in the 1990s with consequential increases in sunlight exposure. The daily ambient erythemal UV in the UK shows a clear sky summer to winter ratio of about 40:1, with day-to-day variations as a result of cloud cover. However population UV exposure will be subject to even greater variation due to differences in individual behaviour. This is illustrated in the results of a recent study (Diffey and Gies, 1998) which reported the daily summer sun exposure received by children in England (50-55°N) and Queensland, Australia (21-27°S). Whilst the median daily personal exposure in Queensland was twice that received in England, there was a wide overlap between the two distributions. On any one day the daily solar UV exposure of 17% of English children exceeded the Queensland median, and the exposure of 26% of Queensland children was less than the English median. Future social trends remain uncertain but a continuation of such increases would add to the UV doses experienced by the population and the risks associated with them. Superimposed on this would be any increase in UV in the environment as a result of ozone depletion.

4.2.3 Variability of Population Exposure

Another area of future uncertainty relates to the possible effects of greenhouse gas-related climate change. Most attention has been given to the effects of changes in UV flux as a result of stratospheric ozone depletion. What has frequently been overlooked is that greenhouse gas induced climate change may also independently influence population exposure to sunlight. One reason is that clouds have a large effect on the amount of UVB reaching the surface and therefore changes in cloud cover would affect exposure. Another factor is that a change to warmer conditions could encourage behaviour associated with greater exposure to the sun (more time outdoors, greater participation in outdoor leisure activities, more sunbathing, lighter clothing that exposes more of the body). The result might be a marked change in patterns of behaviour that would increase population exposure to sunlight and the health risks associated with it.

It seems clear that a number of environmental, socio-economic and behavioural factors may be increasing population exposure to solar UV radiation. What is less clear is whether the effects of such trends may have been mitigated by a deliberate move to more cautious patterns of behaviour in the sun (ONS Health in England 1996, 1997). Hints that such changes might be underway come from the divergence in trends in melanoma incidence and mortality by sex where the upward trend for females is slower than that for males (Mackie, 1998). Interestingly, survey evidence (ONS Health in England 1996, 1997) shows that women's knowledge of the importance of sun-avoidance measures is

greater than for men and they are less likely to have experienced recent sunburn. However, it should be recognised that, because skin cancers may take at least 15 years to develop, it will be some time before any current changes in behaviour show up in incidence or mortality figures.

4.3 **Effects on Animal Health**

Since the last report there have been few advances in knowledge of animal effects, distinct from those covered under physiological effects above. These contribute to better understanding of molecular and cellular effects common to animal and human tissues with animals often being used as models to describe possible effects for man. The situation affecting animals of economic and social value is summarised below.

The effects of UVB on animals are similar to those on humans, and are also directly related to exposure, a product of incidence and duration. Duration varies with longevity of the target animals and their behaviour. It will also vary with their susceptibility based on their genetic makeup and environment, including nutrition. The longevity of animals varies with their ecology, size and 'usefulness'. It is, on average, shortest for wildlife and longest for companion animals. Therefore it may be that any impact on short-lived wildlife is transitory and currently observable. There are examples of behavioural changes to limit exposure, for example Mims (1996) studied mosquito larvae which aggregated under absorbing film rather than UVB transmitting film when the population was exposed to radiation containing UVB. On farm animals the effects are most likely to be observed in some sheep and cattle. Companion animals are likely to be most affected, with the effects related to those suffered by their keepers. Any major impacts would not be detected for some time.

4.4 **Effects on Ecosystems**

The direct effects of UV on ecosystems have become clearer since the last UMIRG report, and it is now evident that in some ecosystems the interactions and indirect effects may be particularly important factors. Primary production in aquatic ecosystems is still considered to be at risk from increased UVB radiation resulting from ozone depletion (Hader et al., 1998). However, most recent evidence from terrestrial ecosystems, including agriculture, indicates that direct effects of increased UVB on the growth or survival of plants are unlikely to be significant in terms of productivity (Caldwell et al., 1999). Nevertheless, there is evidence that effects of increased UVB may modify the interactions between species, with implications for



community structure in both aquatic and terrestrial systems (Franklin and Forster, 1997; Santas *et al.*, 1997; Paul *et al.*, 1997). Interactions between UVB and a range of natural and anthropogenic factors (e.g. temperature, aquatic contaminants and increases in atmospheric CO_2 concentration) are increasingly recognised, but their long-term consequences for plants and animals are not known.

4.4.1 Aquatic Ecosystems

The primary producers in aquatic ecosystems are phytoplankton and, as noted in UMIRG 1996, any change in phytoplankton populations is likely to have further implications for the rest of the aquatic foodweb, and ultimately fishing resources. As aquatic creatures often migrate large distances in the oceans, changes to phytoplankton in one region may be manifested as a change in some other species in a different part of the world. UVB has been shown to have a detrimental effect on phytoplankton, but in their natural setting in the oceans the effect of UVB on phytoplankton necessarily depends on the characteristics of different water masses. Coastal waters, with their higher concentrations of phytoplankton, suspended particulates and dissolved organic matter have much greater absorption of UVB than the clear ocean waters of, for example, the seas around Antarctica. Hojerslev (1988) reported the depth of 90% attenuation of UVB at only 0.5 m in the German Bight and 3.5-6 m in the Fladen Ground of the northern North Sea. Vertical mixing brings phytoplankton to the surface so that mixing processes in the water column are crucial to understanding possible effects of elevated UVB. This movement is not constant and depends on wind stress, tides and the balance between heat gain from insolation and diurnal heat loss. The effect of UVB is therefore likely to vary in different oceanic regions. Shallow seas with high tidal energy have high suspended sediment concentrations and, with a shallow depth of UVB penetration, any increase in UVB is likely to have less impact than in regions where light penetrates deeper in the water column (e.g. the Southern Oceans). The effect of UVB on marine productivity will depend on the response of phytoplankton to the constantly varying light which cells experience as they are moved vertically within the light gradient.

A further complication comes from field studies that have shown that UVB may modify chemical contaminants, with the resultant product being more toxic than the parent compound (Palenik *et al.*, 1991). A number of authors have demonstrated that phototoxicity and resultant mortality are a function of both polyaromatic hydrocarbon dose and UV (Arfsten *et al.*, 1996; Pelletier *et al.*, 1997; Swartz *et al.*, 1997; Boese *et al.*, 1997). These studies have indicated that UV can activate and increase the toxic effect of a single pollutant and produce a detrimental effect much greater than that caused by either the pollutant or UV alone. As such, caution must be exercised when comparing impacts on similar species from polluted and non-polluted environments.

4.4.2 Terrestrial Ecosystems

In terrestrial plants, studies using environmentally realistic UVB doses given outdoors or with appropriate doses of visible light (photosynthetically active radiation: 400-700 nm) have revealed little significant effect on photosynthesis (Allen et al., 1998). Furthermore, recent papers, mostly about crops, tend to confirm that increases in plant-weighted UVB radiation within the range expected from moderate ozone depletion have rather little effect on the growth, biomass and yields under field conditions (Caldwell et al., 1999). These data support the suggestion that ozone depletion is unlikely to have significant, direct effects on primary productivity in terrestrial systems. However, environmentally realistic UVB doses can significantly alter plant morphology, chemical composition, plant genetic processes and developmental patterns in some plant species. These effects may cause significant changes to the interactions between plant species (Caldwell, 1997) and between plants and other organisms. UVB damage may accumulate in perennial species (e.g. trees: Sullivan and Teramura, 1990), and there are indications that it can cause heritable damage (Musil, 1999) or trigger detrimental genetic changes (Walbot, 1999). Such effects are poorly understood and there are many potential influences of such changes so that the consequences for ecosystems are not easily predicted.

There is increasing evidence that enhanced UVB radiation can affect interactions between plants and other organisms including herbivores, and both pathogenic and non-pathogenic fungi (Paul *et al.*, 1997; Newsham *et al.*, 1998). While modified interactions may be due to the effects of UVB on the chemical composition of plant tissues, the precise nature of these changes remain poorly defined. Overall the outcome of UVB effects on these interactions remain unpredictable with contrasting observations in different systems.

The effects of UVB on the interaction between plant material and the microbes that cause decomposition are also variable. In some systems increased UVB decreases the rate of decomposition, measured as mass loss, (Gerhke *et al.*, 1995), but increases it in others (e.g. Newsham *et al.*, 1999). An additional factor underlying this variability is direct photodegradation of dead material (i.e. physical breakdown due to absorption of UVB), although the magnitude of photodegradation due to environmentally relevant increases in UVB is not clear. Consequently, an increasing body of literature suggests that UVB-induced changes in decomposition could have consequences for nutrient cycling in terrestrial ecosystems (Caldwell *et al.*, 1999). As yet, the magnitude of changes in nutrient cycling remain poorly defined.

Given these biological uncertainties and the unresolved duration of ozone depletion, the possibility of significant impacts on certain components of aquatic and terrestrial ecosystems cannot be excluded. In particular, the loss of vulnerable species of phytoplankton could have global consequences for aquatic food webs.

4.5 Effects on Materials

There have been few advances in understanding of material effects since the last report. The current situation is summarised below. Organic polymers, both natural (e.g. wood, horn, natural rubber) and synthetic (e.g. polyvinyl chloride, polymethylmethacrylate, polycarbonate, polyolefins), are most at risk from increased UV (Davis and Sims, 1983; Allen, 1992). When exposed to sunlight (especially UVB), degradation can occur, and is exacerbated if combined with moisture and high temperatures. At first only the appearance may be affected, but in the longer term loss of physical integrity and function can occur (Halliwell and Gardiner, 1994). The very wide range of organic polymers and their applications make it difficult to generalise the potential effects of increased UVB. In the most extreme cases with a polymer exclusively sensitive to UVB the rate of degradation is expected to increase in proportion to the increase in UVB. Other systems may be less affected. However, the majority of polymer systems in the UK are heavily stabilised against UV degradation and it is thought that significant ozone depletion would be necessary before serious damage was observed.

The technology to improve the stability of polymers to UV degradation already exists, so the durability of susceptible materials can be improved if necessary, albeit at some cost. However materials currently in use may have a shorter lifetime than originally expected if UV increases throughout their intended lifetime.

5 Recommendations for Future Work

5.1 Introduction

In earlier SORG reports we made a number of wide-ranging recommendations for future scientific research, including the need for international cooperation and coordination of research, especially within the EU. The overall aim of the recommendations was to provide a firm basis for an independent assessment of the state of the ozone layer, so that policy could be informed by the best available scientific advice. UK scientists have been able to make major contributions to the improved understanding of ozone depletion and have played a prominent role in European research programmes. However, despite the scientific and political progress made since 1988, the problem of ozone depletion is not solved; many important uncertainties remain and have been highlighted in this report. Stratospheric halogen loading should now be around its peak. Recovery of the ozone layer will depend not just on the halogen loading but also on complex interactions within the coupled chemistry climate system. Detection of recovery and its attribution to policy requires a more complete scientific understanding. Hence, we feel that it is timely to reiterate the need for continued basic research into the ozone layer.

The general scientific issues were highlighted in SORG 1996 and restated here. Unless our understanding is improved we cannot predict with confidence the future evolution of the global ozone layer; we will not be able to predict with confidence the possibly crucial role played by interactions between chemical processes and climate, and we will be unable to assess with confidence the impact of the projected expansion of sub- and supersonic aircraft on ozone. There is also a need to continue to investigate the properties and behaviour of newly developed chemical substances, for example, those used as substitutes for CFCs, so that their environmental impact can be assessed. These are all issues of global importance. It is vital that research programmes continue to address vigorously the fundamental scientific problems so that the new and ongoing issues can be resolved.

5.2 Aims of Future Research

As a result of this assessment, the following specific aims have emerged as being a necessary part of a balanced programme.

- To monitor the evolution of ozone and associated parameters, including other trace species and ultraviolet at the surface.
- To improve the analysis, quantification and attribution of past changes in ozone and associated parameters.

- To advance the capability for prediction of ozone, including knowledge of processes that could either shorten or lengthen the period of recovery; we have particularly identified the interactions between climate change and ozone change. The Montreal and Kyoto protocols are interconnected in this respect, so that decisions taken under one protocol have the potential to influence the aims of the other protocol.
- To strengthen the underpinning science to make those predictions, including instrument development and laboratory measurements.
- In view of the interconnections between tropospheric and stratospheric processes, it is important to encourage a more integrated approach to the scientific investigation of the relevant processes.

5.3 A Balanced Programme

The quantitative understanding of the ozone layer is usually expressed within a numerical model of the atmosphere. Numerical models must include the best possible descriptions of the important physical and chemical processes. These descriptions are based on fundamental laboratory studies and knowledge of atmospheric structure. The models must be validated against a range of atmospheric data that will test the model performance over a wide range of space and timescales.

We welcome developments within the NERC atmospheric science programme which has become better resourced in recent years. A new 5-year £9 million programme, UTLS OZONE, is studying the upper troposphere and lower stratosphere while earlier laboratory and field instrument programmes are to be continued, at a somewhat reduced level of funding, in a new programme, COSMAS. Strategic modelling developments are supported by the NERC UGAMP programme. Research on lower stratospheric ozone will also be a theme within the EU Framework Programme 5. Planned new satellite projects (e.g. HIRDLS and ENVISAT) will provide global measurements of unprecedented detail for ozone research. These initiatives provide a framework for future basic research on the ozone layer in the UK. Nevertheless, targetted research on specific policyrelated issues may not be fully covered. For example, we perceive an ad hoc approach to long term monitoring of ozone and associated parameters, to quantifying the source of emissions and to evaluation of laboratory data which are vital as input to models and analyses of their output. We recommend closer liaison between NERC and government agencies, including the UK Meteorological Office and DETR to ensure that these elements of the programme are properly implemented.

5.4 Effects Research

The long term monitoring of UV is particularly important in considering the future focus of effects research and provides the link between SORG and UMIRG activities: the basic research in the different effects fields can then be set against the background of the existing UV climate and actual changes in UV.

While there is still a need for basic, focussed effects research (e.g. to understand the biological significance of UV-induced

immunosuppression), it is becoming clear that the response of individual systems to UV cannot be viewed in isolation. Changes in behaviour (e.g. sheltering, sunscreen use), interactions between organisms and synergy between UVB and other environmental pressures can all confound the isolated system response. Quantification of UVB effects on different populations and ecosystems requires the integration of several, frequently interacting, responses to UV. We recommend that these broader issues involving multidisciplinary research are addressed alongside continuing targetted effects research.

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2-D Two-dimensional.

3-D Three-dimensional.

AFEAS Alternative Fluorocarbons Environmental Acceptability Study.

Bank The quantity, particularly of Halons, remaining in equipment and not yet emitted into the atmosphere.

BEF Bromine efficiency factor.

Brassicaceae The plant family of brassicas which includes oil seed rape as well as cabbage, brussels sprouts and similar vegetables.

Bromine loading The total concentration, in the troposphere, of compounds which could transport bromine into the stratosphere.

CFC Chlorofluorcarbon: a compound consisting of carbon, chlorine and fluorine only; historically used as refrigerants, foam blowing agents and aerosol propellants; each distinct chemical is designated by a unique number, e.g. CFC-11.

Chlorine loading The total concentration, in the troposphere, of compounds which could transport chlorine into the stratosphere.

Chromophore Molecule, or part of molecule, that absorbs UV energy. This may result in structural changes that initiate biological responses.

CNRS Centre National de la Recherche Scientifique, France.

COSMAS Core-Strategic Measurements for Atmospheric Science.

Cytokine Class of natural chemical that regulates immune function. Cytokines are released from a wide range of cells.

DETR Department of the Environment, Transport and the Regions (UK).

DNA Deoxyribonucleic acid

DU Dobson Units.

EIA Environmental Investigation Agency, a non-governmental organisation.

EASOE European Arctic Stratospheric Ozone Experiment.

EESC Equivalent effective stratospheric chlorine.

EP Earth Probe

Erythemal UV The incident UV spectrum weighted with the erythemal action spectrum and integrated across the full UV spectrum (for sunlight this means both UVB and UVA wavelengths). This quantity then indicates the effectiveness of the radiation for producing erythema (sunburn).

EU European Union.

FAO Food and Agriculture Organisation of the UN.

Firn Unconsolidated snow.

GCM General circulation model.

GHG Greenhouse gas.

GISS Goddard Institute for Space Studies.

HALOE HALogen Occultation Experiment.

Halon A compound consisting of carbon, bromine, fluorine and, in some cases, chlorine; halons have been identified as ozone depleting substances.

HBFC Hydrobromofluorocarbon: a compound consisting of carbon, bromine, fluorine and hydrogen.

HCFC Hydrochlorofluorocarbon: a compound consisting of carbon, chlorine, fluorine and hydrogen; used as a replacement for CFCs; individual compounds are designated by unique numbers, e.g. HCFC-22.

HFC Hydrofluorocarbon: a compound consisting of carbon, fluorine and hydrogen; used as a replacement for CFCs and HCFCs; individual compounds are designated by unique number, e.g. HFC-134a.

hPa hectopascal (unit of pressure – same as mbar).

IFMA Instruments for Field Measurement in the Atmosphere.

K Temperature in degrees Kelvin.

kg Kilogrammes.

km Kilometres.

LSAC Laboratory Studies for Atmospheric Chemistry.

mbar Millibar (unit of pressure – same as hPa).

mPa MilliPascal (unit of pressure).

MLF MultiLateral Fund – A fund controlled under the Montreal Protocol and supported by a levy on developed countries that subsidises projects to enable developing countries to stop using ozone depleting substances.

Montreal Protocol The international treaty governing identification of substances as ozone depleters and measures to control their production and consumption.

NASA National Aeronautics and Space Administration (USA).

NERC Natural Environment Research Council (UK).

NH Northern Hemisphere.

nm Nanometres

NRPB The National Radiological Protection Board, UK.

ODP Ozone Depletion Potential. This provides a relative measure of the expected impact on ozone per unit mass emission of a gas compared to that expected from the same mass emission of CFC-11 integrated over the atmospheric lifetimes of both compounds.

ODPtonnes The mathematical product of the ozone depletion potential of a substance and mass, giving a crude measure of environmental impact.

ODS Ozone Depleting Substance. A chemical compound that has been identified within the Montreal Protocol to be capable of depleting stratospheric ozone.



ONS Office of National Statistics, UK.

PEM-West A & B Pacific Exploratory Mission – West (Experiments A & B).

ppbv Parts per billion by volume.

pptv Parts per trillion by volume.

PSC Polar stratospheric cloud.

QBO Quasi-Biennial Oscillation.

SAGE Stratospheric Aerosol and Gas Experiment.

SBUV Solar Backscatter Ultraviolet (radiometer).

SESAME Second European Stratospheric Arctic and Midlatitude Experiment.

SH Southern Hemisphere.

SOAZ Système d'Analyses par Observations Zénithales.

SORG Stratospheric Ozone Review Group.

SPARC Stratospheric Processes and their Role in Climate.

SSU/MSU Stratospheric Sounding Unit/Mesopheric Sounding Unit.

Stratosphere Altitude between about 12 km and 50 km.

STTA Stratospheric Temperature Trends Assessment.

SZA Solar zenith angle

TEAP Technology and Economic Assessment Panel of the Montreal Protocol.

TOC Technical Options Committee of TEAP.

TOMS Total Ozone Mapping Spectrometer.

Troposphere Altitude between the ground and about 12km.

UGAMP Universities' Global Atmospheric Modelling Programme

UKMO United Kingdom Meteorological Office.

UMIRG Ultraviolet Measurements and Impacts Review Group (UK).

UNEP United Nations Environment Programme.

UTLS OZONE Upper Troposphere Lower Stratosphere Ozone Programme of NERC.

UV Ultraviolet.

UVA Ultraviolet radiation with wavelengths ranging from 320–400nm. Most solar UV is UVA (\geq 95%) but UVA is generally very much less biologically effective than UVB.

UVB Ultraviolet radiation of shorter wavelenghts ranging from 280–320nm. Solar UVB is from ~ 290–320nm, the short wavelengths being filtered by the ozone layer. UVB, readily absorbed by skin chromophores, is the cause of most of the detrimental effects of solar UV.

UV fluxes Amount of radiation incident on a given area in a given time.

WMO World Meteorological Organisation.

Chemical Formulae

BrO	bromine monoxide	NO _x	nitrogen oxides (NO, NO_2 , NO_3)
BrONO ₂	bromine nitrate	HNO_3	nitric acid
CCl_4	carbon tetrachloride	SO ₂	sulphur dioxide
CH ₃ CCl ₃	methyl chloroform 1,1,1-trichloroethane	H_2SO_4	sulphuric acid
CF_2Cl_2	CFC-12 dichlorodifluoromethane	HCl	hydrogen chloride
CFCl ₃	CFC-11 fluorotrichloromethane	HBr	hydrogen bromide
CH_4	methane	OClO	chlorine dioxide
CH_3Br	methyl bromide	OH	hydroxyl radical
CHF ₂ Cl	HCFC-22 chlorodifluoromethane	ClO _x	total active chlorine
ClO	chlorine monoxide	IO	iodine monoxide
ClONO ₂	chlorine nitrate	Cl_y	total inorganic chlorine
N_2O_5	dinitrogen pentoxide	Br _y	total inorganic bromine
HOCl	hydrochlorous acid		

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