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Radiative Forcing of Climate

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CONTENTS

Executive Summary	51	A2.5.4 Indirect Effects due to CH₄ Emissions	60
A2.1 Introduction	53	A2.5.4.1 Changes in Lifetimes due to Changes in OH	60
A2.2 Radiative Forcing	53	A2.5.4.2 Radiative Forcing Changes from CH ₄ -Induced Changes in Ozone	60
A2.3 The Global Warming Potential (GWP) Concept	53	A2.5.4.3 Effects of CH ₄ on Stratospheric Water Vapour	61
A2.3.1 Definition	53	A2.5.4.4 Oxidation to CO ₂	61
A2.3.2 Reference Molecule	54	A2.5.4.5 Indirect GWP for CH ₄	61
A2.3.3 Time Horizons for GWPs	54	A2.5.5 NO_x Emissions	61
A2.3.4 Limitations of Present GWPs	54	A2.5.6 CO and NMHC Emissions	61
A2.3.5 Direct GWPs of Well-Mixed Trace Gases	55	A2.5.7 Summary	61
A2.3.6 Indirect GWPs of Well-Mixed Trace Gases	55	A2.6 Radiative Forcing due to Aerosol Particles	62
A2.4 Radiative Forcing due to Stratospheric Ozone	55	A2.6.1 Tropospheric Particles	62
A2.4.1 Lower Stratospheric Losses	55	A2.6.1.1 Background	62
A2.4.2 The Greenhouse Effect of Ozone Losses	58	A2.6.1.2 New Findings	62
A2.4.3 Ozone Depletion and Stratospheric Temperature Changes	58	A2.6.1.3 Discussion	63
A2.5 Radiative Forcing due to Gases in the Troposphere	59	A2.6.2 Stratospheric Sulphate Particles	64
A2.5.1 Introduction	59	A2.7 Forcing Due to Solar Irradiance Changes	64
A2.5.2 Chemical Processes and Changes in O ₃ and OH	59	References	65
A2.5.3 Sensitivity of Radiative Forcing to Changes in Tropospheric Ozone	60		

EXECUTIVE SUMMARY

Since the 1990 IPCC Scientific Assessment (IPCC, 1990), there have been significant advances in our understanding of the impact of ozone depletion and sulphate aerosols on radiative forcing and of the limitations of the concept of the Global Warming Potential (GWP).

Radiative Forcing due to Changes in Stratospheric Ozone:

Observed global depletions of ozone (O_3) in the lower stratosphere have been used to calculate changes in the radiative balance of the atmosphere. Although the results are sensitive to atmospheric adjustments, and no General Circulation Model (GCM) studies of the implications of the O_3 changes on surface temperature have been performed, the radiative balance calculations indicate that the O_3 reductions observed during the 1980s have caused reductions in the radiative forcing of the surface-troposphere system at mid- and high latitudes. This reduction in radiative forcing resulting from O_3 depletion could, averaged on a global scale and over the last decade, be approximately equal in magnitude and opposite in sign to the enhanced radiative forcing due to increased chlorofluorocarbons (CFCs) during the same time period.

Radiative Forcing due to Changes in Tropospheric Ozone:

While there are consistent observations of an increase in tropospheric ozone (up to 10% per decade) at a limited number of locations in Europe, there is not an adequate global set of observations to quantify the magnitude of the increase in radiative forcing. However, it has been calculated that a 10% uniform global increase in tropospheric ozone would increase radiative forcing significantly compared to forcing by other greenhouse gases.

Radiative Effects of Sulphur Emissions:

Emissions of sulphur compounds from anthropogenic sources lead to the presence of sulphate aerosols which reflect solar radiation. This is likely to have a cooling influence on the Northern Hemisphere. For clear-sky conditions alone, the cooling caused by current rates of emissions has been estimated to be up to $1Wm^{-2}$ averaged over the Northern Hemisphere, a value which should be compared with the estimate of $2.5Wm^{-2}$ for the heating due to anthropogenic greenhouse gas emissions up to the present. The non-uniform distribution of anthropogenic sulphur sources coupled with the relatively short atmospheric residence time of sulphur compounds produce large regional variations in their

effects. In addition, sulphate aerosols may affect the radiation budget through changes in cloud optical properties.

Global Warming Potentials:

Gases can exert a radiative forcing both directly and indirectly: direct forcing occurs when the gas itself is a greenhouse gas; indirect forcing occurs when chemical transformation of the original gas produces a gas or gases which themselves are greenhouse gases. The concept of the Global Warming Potential (GWP) has been developed for policymakers as a measure of the possible warming effect on the surface-troposphere system arising from the emission of each gas relative to carbon dioxide (CO_2). The indices are calculated for the contemporary atmosphere and do not take into account possible changes in chemical composition of the atmosphere. Changes in radiative forcing due to CO_2 are highly non-linear with respect to changes in atmospheric CO_2 concentrations. Hence, as CO_2 levels increase from present values, the GWPs of the non- CO_2 gases would be higher than those evaluated here. For the concept to be most useful, both the direct and indirect components of the GWP need to be quantified.

Direct Global Warming Potentials:

The direct components of the GWPs have been recalculated, taking into account revised estimated lifetimes, for a set of time horizons ranging from 20 to 500 years, with CO_2 as the reference gas. The same ocean-atmosphere carbon cycle model as used in IPCC (1990) has been used to relate CO_2 emission to concentrations. While in most cases the values are similar to the previous IPCC (1990) values, the GWPs for some of the hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs) have increased by 20 to 50% because of revised estimates of their lifetimes. The direct GWP of methane (CH_4) has been adjusted upward, correcting an error in the previous IPCC report. The carbon cycle model used in these calculations probably underestimates both the direct and indirect GWP values for all non- CO_2 gases. The magnitude of the bias depends on the atmospheric lifetime of the gas, and the GWP time horizon.

Indirect Global Warming Potentials:

Because of our incomplete understanding of chemical processes, most of the indirect GWPs reported in IPCC (1990) are likely to be in substantial error, and none of them can be recommended. Although we are not yet in a position to recommend revised

numerical values, we do know that the indirect GWP for methane is positive and could be comparable in magnitude to its direct value. In contrast, the indirect GWPs for chlorinated and brominated halocarbons are likely to be negative. The concept of a GWP for short-lived, heterogeneously distributed constituents, such as carbon monoxide (CO), non-methane hydrocarbons

(NMHC), and nitrogen oxide (NO_x) may prove inapplicable, although these constituents will affect the radiative balance of the atmosphere through changes in tropospheric ozone and the hydroxyl radical (OH). Similarly, a GWP for sulphur dioxide (SO_2) is viewed to be inapplicable because of the non-uniform distribution of sulphate aerosols.

A2.1 Introduction

This section is an update of the discussions presented in Section 2 of the first Intergovernmental Panel on Climate Change Scientific Assessment (IPCC, 1990) concerning the radiatively and chemically important species in the atmosphere. The present update has five major objectives:

- (i) to extend the discussion on Global Warming Potentials (GWPs), and to re-evaluate their numerical values in view of revisions to the lifetimes of the radiatively active species;
- (ii) to characterize the radiative impacts of the observed stratospheric O₃ losses between 1979 and 1990;
- (iii) to discuss the changes in the concentrations of radiatively active gases occurring through chemical processes, and their implications for radiative forcing;
- (iv) to characterize the radiative forcing due to tropospheric and stratospheric sulphate aerosol concentrations;
- (v) to extend the discussion on the radiative forcing due to solar irradiance variations.

Some of the scientific details regarding the new developments have already appeared in the 1991 Scientific Assessment of Ozone Depletion (WMO, 1992).

A2.2 Radiative Forcing

The radiative forcing due to a perturbation in the concentration of a species is defined (see WMO, 1986; IPCC, 1990; WMO, 1992) by the net radiative flux change induced at the tropopause, keeping the concentrations of all other species constant. The change in the radiative flux is determined using a one-dimensional radiative transfer model in which the surface and tropospheric temperatures are held fixed at some reference values, while the stratospheric temperatures are allowed to relax to a new equilibrium in response to the perturbation. The definition assumes that the stratosphere undergoes a purely radiative adjustment, i.e., there is no change in the dynamical heating of the stratosphere due to the perturbation (WMO, 1986).

The radiative forcing is interpreted as a gain (positive) or loss (negative) for the surface-troposphere system as a whole. The motivation for this concept arises from radiative-convective modelling exercises, where the change in the global mean surface temperature can be related simply to the net radiative flux change at the tropopause (WMO, 1986). This has led to the adoption of the radiative forcing of the surface-troposphere system as a simple and convenient basis for estimating the potential climatic effects of various trace species.

A2.3 The Global Warming Potential (GWP) Concept

The aim of the GWP index is to offer a simple characterization of the relative radiative effects of the well-mixed species. It was created in order to enable policymakers to evaluate options that affect the emissions of various greenhouse gases, by avoiding the need to make repeated, complex calculations. IPCC (1990) discussed the GWP concept in considerable detail and only the salient features are addressed here. However, as noted below, there are serious limitations associated with the calculation of GWPs that constrain their practical utility.

A2.3.1 Definition

The Global Warming Potential is a measure of the relative, globally-averaged warming effect arising from the emissions of a particular greenhouse gas.

- It is a *relative* measure in that it expresses the warming effect compared to that of a reference gas (or 'molecule').
- It is a *global* measure in that it is derived from the globally- and annually-averaged net radiative fluxes at the tropopause, and thus describes the effects on the whole surface-troposphere system.
- It is a *time-integrated* measure of warming over a specified time horizon, taking account of the change with time of the species concentration.

The GWP of a well-mixed gas was defined in IPCC (1990) as the time-integrated change in radiative forcing due to the instantaneous release of 1kg of a trace gas expressed relative to that from the release of 1kg of CO₂. Calculation of the GWP for a particular species requires specification of the following:

- (i) the radiative forcing both of the reference gas and of the species, per unit mass or concentration change;
- (ii) the time horizon over which the forcings have to be integrated;
- (iii) the atmospheric lifetime both of the species and of the reference gas;
- (iv) the pathway of chemical breakdown of the species and the extent to which it gives rise to other greenhouse species, e.g., O₃ production from CH₄, NO_x, CO and NMHCs;
- (v) the present and future chemical state of the atmosphere, i.e., levels of the background concentrations of various species throughout the troposphere;
- (vi) the present and future physical state of the atmosphere, i.e., values of meteorological variables throughout the troposphere (e.g., temperature profile, cloud properties).

Factors (iii) and (iv) are intimately related to (v) and (vi) and are the sources of greatest uncertainty in the

calculation of GWPs - see Section A2.3.4 below.

It is possible to offer alternative definitions of GWP, for example based on sustained rather than pulse emissions (Wigley *et al.*, 1990). Such alternatives can lead to numerical values of GWP which are different from those under the present definition, but in general not so different as to alter the relative ranking of the important species.

A2.3.2 Reference Molecule

Given the conceptual framework of the GWP and its implications for policy-making, the choice of a reference molecule is dictated by the need to evaluate the results in terms of the dominant contributor in the greenhouse gas problem. IPCC (1990) therefore chose CO₂ as the reference gas for the determination of GWPs. Although another gas or surrogate would have a simpler atmospheric decay behaviour compared to CO₂ (e.g., CFCs; see Fisher *et al.*, 1990), the evaluation of GWPs presented here continues, after extensive review, to use CO₂ as the reference gas.

To avoid the need to use a single lifetime for CO₂, IPCC (1990) used a carbon cycle model to calculate the integrated radiative forcing for CO₂, specifically the ocean-atmosphere box diffusion model of Siegenthaler and Oeschger (1987; see also Siegenthaler, 1983) which assumed a net neutral biosphere. We adopt the same model for the direct GWP estimates in this assessment.

A2.3.3 Time Horizons for GWPs

Because greenhouse gases have a variety of removal mechanisms they have different residence times, or lifetimes, in the atmosphere. The calculated value of GWP thus depends on the integration period chosen. There is no single value of integration time for determining GWPs that is ideal over the range of uses of this concept, though the choice of a time-scale for integration in the GWP calculation need not be totally arbitrary (see IPCC (1990) and WMO (1992) for a discussion on the choice of time horizons). In this report, GWPs are calculated over time horizons of 20, 100 and 500 years (as employed in IPCC, 1990). It is believed that these three time horizons provide a practical range for policy applications.

A2.3.4 Limitations of Present GWPs

While the GWP, as defined in IPCC (1990), is a convenient and reasonably practical index for ranking the relative and cumulative impact of greenhouse gas emissions, it has the following limitations, some of which are very serious:

- (a) the modelling of radiative transfer within the atmosphere contains uncertainties, as was pointed out in IPCC (1990);
- (b) since the direct GWP is a measure of the global effect of a given greenhouse gas emission, it is most

appropriate for well-mixed gases in the troposphere (e.g., CO₂, CH₄, nitrous oxide (N₂O) and halo-carbons). The radiative forcing employed in the determination of GWPs does not purport to characterize the latitudinal and seasonal dependence of the change in the surface-troposphere radiative fluxes. Different well-mixed gases can yield different spatial patterns of radiative forcings (Wang *et al.*, 1991);

- (c) the GWP definition used here considers only the surface-troposphere radiative forcing rather than a particular response (e.g., surface temperature) of the climate system. While the surface-troposphere radiative flux perturbations can be related to temperature changes at the surface in the context of one-dimensional radiative-convective models (WMO, 1986), such a general interpretation for the temperature response either in three-dimensional General Circulation Models or in the actual surface-atmosphere system must be approached with caution. Further, although the GWP of a well-mixed gas can be regarded as a first-order indicator of the potential global mean temperature change due to that gas relative to CO₂, it is inappropriate for predicting or interpreting regional climate responses;
- (d) GWP values are sensitive to uncertainties regarding atmospheric residence times. Thus, revisions to GWP values should be expected as scientific understanding improves. Because CO₂ is used as the reference gas, any revision to the calculation of its integrated radiative forcing over time will change all GWP values. GWP results are also sensitive to the choice of carbon cycle model used to calculate the time-integrated radiative forcing for CO₂. In particular, because the Siegenthaler-Oeschger model has only an ocean CO₂ sink, it is likely to overestimate the concentration changes and to lead to an underestimate of both the direct and the indirect GWPs. The magnitude of this bias depends on the atmospheric lifetime of the gas, and the time horizon;
- (e) as defined here, GWPs assume constant concentration backgrounds at current levels. The calculated GWPs depend on the assumed background level(s). The indices are calculated for the contemporary atmosphere and do not take into account possible changes in the chemical composition of the atmosphere. Changes in radiative forcing due to CO₂, CH₄ and N₂O concentration changes are non-linear with respect to these changes. The net effect of these non-linearities is such that, as CO₂ levels increase from present values, the GWPs of all non-CO₂ gases would become higher than those evaluated here (see WMO, 1992);

(f) for the GWP concept to be most useful, both the direct and the indirect components need to be quantified. However, accurate estimates of the indirect effects are more difficult to obtain than those for the direct effects for the following reasons:

- (i) there are uncertainties in the details of the chemical processes as well as in the spatial and temporal variations of species involved in such transformations. As shown later, there is fair confidence in the sign of some of the indirect effects; however, precise estimates are lacking. Because of our incomplete understanding of chemical processes, it is now recognized that the uncertainties in the indirect components of GWPs reported in IPCC (1990) are so large that their use can no longer be recommended;
- (ii) for gases that are not well-mixed (e.g., tropospheric ozone precursors), the GWP concept may not be meaningful;
- (iii) further, while the GWP concept thus far has been applied to gases with perturbations only in the longwave spectra, it may not adequately account for the seasonally and latitudinally varying radiative effects due to inhomogeneously distributed species with a significant interaction in the solar spectrum (e.g., aerosols).

In conclusion, given the above limitations, great care must be exercised in applying GWPs in the policy arena.

A2.3.5 Direct GWPs of Well-Mixed Trace Gases

New direct GWPs (i.e., ignoring any radiative effects due to the products of chemical transformation) of several well-mixed species have been determined. For CFC-13, CFC-14, CFC-116, CHCl_3 and CH_2Cl_2 the lifetimes and radiative forcings are as given by Ramanathan *et al.* (1985); for the other compounds radiative forcings are as before (Tables 2.2 - 2.6 of IPCC, 1990) but lifetimes have been updated according to WMO (1992). Note that the lifetime of methane is here assumed to be 10.5 years, which accounts for a sink mechanism in the soil (see IPCC, 1990).

These new GWPs are listed in Table A2.1 for the three time horizons mentioned above. Changes in the lifetime and variations of radiative forcing with concentration change are neglected. Most of the new direct GWPs computed are generally within 20% of the values appearing in Table 2.8 of the IPCC (1990) study; the difference is entirely due to the differences in assumed lifetimes (WMO, 1992). HFC-125, HCFC-141b and HFC-143a, all have increases in GWPs exceeding 20% for the 100- and 500-year time horizons, while CF_3Br (Halon 1301) has a decrease of more than 20% for the 500-year time horizon; again, these differences are a manifestation

of the changes in the lifetimes. The direct GWPs for CH_4 here are substantially higher than those that can be inferred from IPCC (1990) owing to a typographical error appearing in that report. Note that CO, NMHC and NO_x have a negligible direct GWP component.

A2.3.6 Indirect GWPs of Well-Mixed Trace Gases

Because of our incomplete understanding of chemical processes, and their latitudinal and temporal dependence, it is not possible to quantify accurately the indirect GWPs. As noted above, the indirect GWPs reported in IPCC (1990) are likely to be in error and should not be used. In particular, the value for NO_x was probably overestimated by a substantial amount (Johnson *et al.*, 1992). Although we are not yet in a position to calculate new indirect GWPs, we can estimate the sign most likely for some compounds based on current understanding (see Table A2.1). For example, the indirect GWP for methane is positive and could be comparable in magnitude to the direct value. Because the weight of evidence suggests that halocarbons are largely responsible for the observed global stratospheric ozone loss over the past decade (WMO, 1992), chlorine- and bromine-containing compounds are likely to have negative indirect values. Although CO_2 is not itself involved in chemical reactions affecting the concentrations of radiatively active species, it could affect chemical processes through its influences on the atmospheric thermal structure. Other compounds such as CO, NMHC and NO_x indirectly affect the radiative balance of the atmosphere through changes in tropospheric ozone and OH (see Section A2.5). For such short-lived gases, however, the GWP concept may have little practical applicability.

A2.4 Radiative Forcing due to Stratospheric Ozone

A2.4.1 Lower Stratospheric Losses

There have been statistically significant losses of ozone in the middle and high latitudes between 1979 and 1990 which have important ramifications for radiative forcing (for details, the reader is referred to WMO, 1992). The TOMS satellite data (Stolarski *et al.*, 1991) point to a reduction in the column ozone, while the SAGE satellite data (McCormick *et al.*, 1992) and the ozonesonde data (Stachelin and Schmid, 1991) indicate that these losses have occurred mainly below 25km in the lower stratosphere. The weight of evidence suggests that these losses, both in the polar and the middle latitudes, are due in large part to the anthropogenic emissions of CFCs, as well as to other chlorine- and bromine-containing compounds (WMO, 1992).

These changes in ozone substantially perturb both solar and longwave radiation (WMO, 1986 and 1992). While the solar effects due to ozone losses are determined solely by

Table A2.1: Numerical estimates of the "Direct" GWP and the sign of the "Indirect" effect of several gases. The lifetimes of various non-CO₂ species follow WMO (1992) and Ramanathan et al. (1985). The carbon cycle model employed follows IPCC (1990). The 20, 100 and 500-year time horizons denote the time elapsed after a pulse release of the gas in consideration. The radiative forcing values follow IPCC (1990) and Ramanathan et al. (1985). The following important points should be noted regarding the entries in the Table:

1. The lifetimes of the various species are not as precisely known as the Table indicates; they are used for the GWP calculations primarily to ensure consistency with the lifetime values in the references cited.
2. This assessment does not calculate the "Total" (direct + indirect) GWP as did IPCC (1990). Note that CO, NMHC and NO_x are all short-lived gases having a negligible direct GWP component.
3. The indirect GWPs are uncertain but could conceivably be comparable in magnitude to the direct GWPs. Only the signs of the indirect effects are estimated here, based on current understanding. The estimates of the indirect effects for the chlorine- and bromine-containing compounds are based on the weight of evidence related to the observed lower stratospheric ozone depletion (WMO, 1992).
4. IPCC (1990) contained a typographical error for the indirect effect of methane which led to the inference of an incorrect value for its direct GWP. Also in that report, the indirect effect of NO_x was probably overestimated by a large factor (Johnson et al., 1992).

Gas	Lifetime (years)	Direct Effect for Time Horizons of			Sign of "Indirect" Effect
		20 years	100 years	500 years	
CO ₂	†	1	1	1	none ††
CH ₄	10.5	35	11	4	positive
N ₂ O	132	260	270	170	uncertain
CFC-11	55	4500	3400	1400	negative
CFC-12	116	7100	7100	4100	negative
CFC-13	400	11000	13000	15000	negative
CFC-14	>500	>3500	>4500	>5300	none †††
HCFC-22	15.8	4200	1600	540	negative
CFC-113	110	4600	4500	2500	negative
CFC-114	220	6100	7000	5800	negative
CFC-115	550	5500	7000	8500	negative
CFC-116	>500	>4800	>6200	>7200	none †††
HCFC-123	1.71	330	90	30	negative
HCFC-124	6.9	1500	440	150	negative
HFC-125	40.5	5200	3400	1200	none †††
HFC-134a	15.6	3100	1200	400	none †††
HCFC-141b	10.8	1800	580	200	negative
HCFC-142b	22.4	4000	1800	620	negative
HFC-143a	64.2	4700	3800	1600	none †††
HFC-152a	1.8	530	150	49	none †††
CCl ₄	47	1800	1300	480	negative
CH ₃ CCl ₃	6.1	360	100	34	negative
CF ₃ Br	77	5600	4900	2300	negative
CHCl ₃	0.7	92	25	9	negative
CH ₂ Cl ₂	0.6	54	15	5	negative
CO	months	-	-	-	positive
NMHC	days to months	-	-	-	positive
NO _x	days	-	-	-	uncertain

† The persistence of carbon dioxide has been estimated by explicitly integrating the box-diffusion model of Siegenthaler (1983); an approximate lifetime is 120 years.

†† CO₂ is not involved in chemical reactions affecting the concentrations of the radiatively active species. However, it could affect the relevant chemical reactions through its influences on the atmospheric thermal structure.

††† No currently known or negligible indirect effect.

the total column ozone amounts, the longwave effects are determined both by the amount and its vertical location (Manabe and Strickler, 1964; Ramanathan and Dickinson, 1979; WMO, 1986; Lacis *et al.*, 1990). The loss of ozone in the lower stratosphere induces three distinct radiative effects (Ramanathan *et al.*, 1985; Ramaswamy *et al.*, 1992):

- a reduction in the absorption by stratospheric ozone of incoming solar radiation, leading to an increase in the amount reaching the surface-troposphere system;
- in the absence of stratospheric temperature changes, a decrease in the emission of longwave radiation from the stratosphere into the surface-troposphere system which is opposite to the solar effect; and
- a reduction of the *in-situ* solar heating and a change in the convergence of the longwave radiation within the lower stratosphere. Because the thermal balance of the lower stratosphere is sensitive to radiative perturbations (Fels *et al.*, 1980; Shine, 1987; Kiehl *et al.*, 1988) this third effect results, in the absence of any compensatory dynamical heating, in a decrease of temperatures at these altitudes. This, in turn, leads

to a further reduction of the longwave emission into the troposphere, the magnitude of the reduction being sensitive to the decrease in stratospheric temperatures.

Two sets of radiative transfer modelling studies (WMO, 1992; Ramaswamy *et al.*, 1992), employing slightly different assumptions about the vertical profile of the lower stratospheric ozone loss over the past decade, have been performed and lead to similar conclusions. They suggest an enhancement of the ozone-induced longwave effects (effects b and c) over the solar (effect a) in the middle to high latitudes, corresponding to the larger ozone losses there. Because of the spatial variability in the ozone depletions and the presence of the solar component, the calculated ozone forcing depends on the season and the geographical region. Another factor that has a significant influence on the ozone radiative forcing is the meteorological state of the troposphere, particularly the distribution of clouds (WMO, 1992).

Figure A2.1 (from Ramaswamy *et al.*, 1992) illustrates, for the four seasons and for both hemispheres, the changes in radiative forcing due to ozone, due to CFCs alone, and

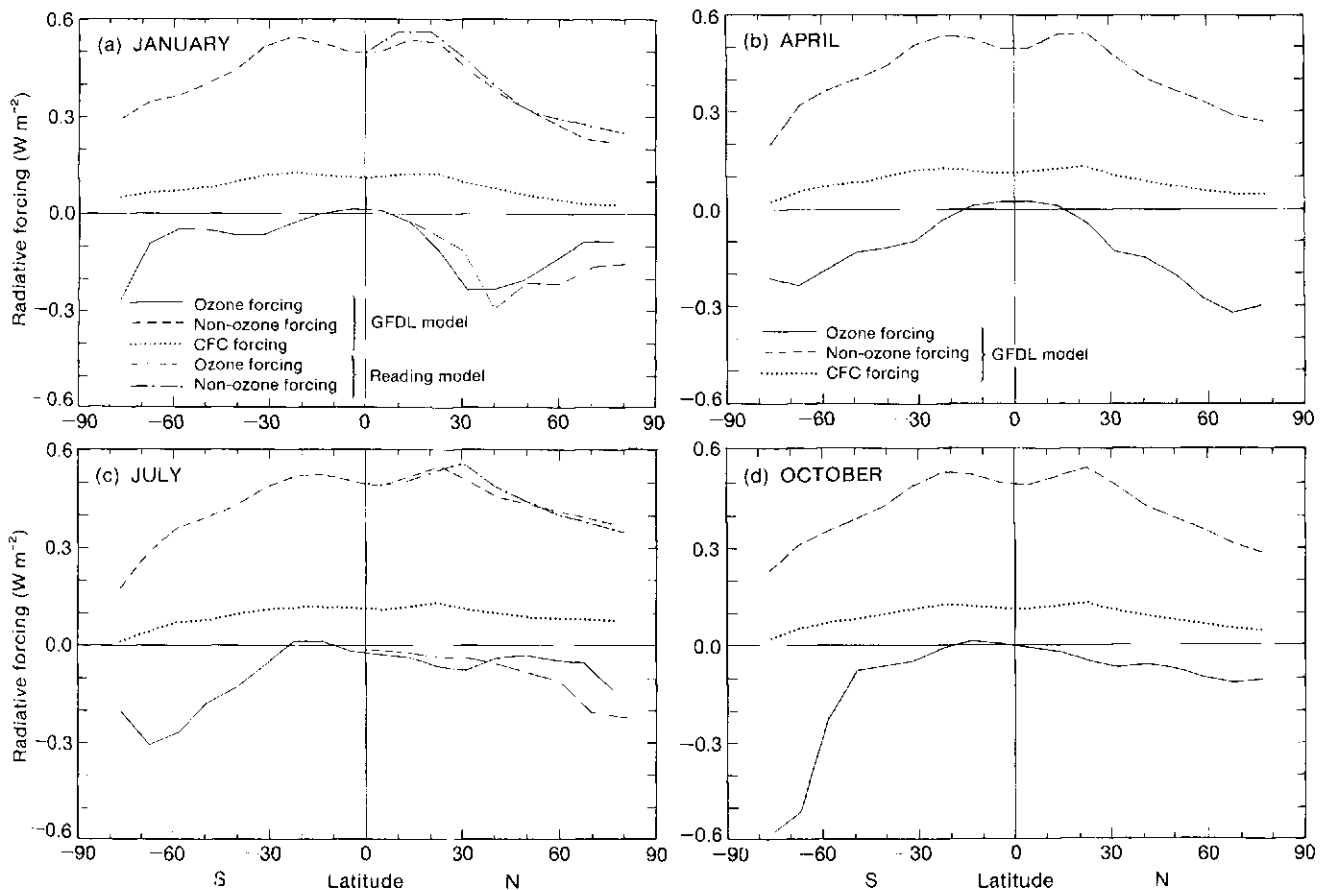


Figure A2.1: Latitudinal and seasonal dependence of the radiative forcing due to (i) the 1979-1990 increases in CFCs, (ii) the 1979-1990 increases in all the non-ozone gases (CO_2 , CH_4 , N_2O and the CFCs), and (iii) the 1979-1990 observed lower stratospheric losses of ozone (Stolarski *et al.*, 1991; McCormick *et al.*, 1992). Results from two different models are shown: (a) and (c) show University of Reading results (Northern Hemisphere January and July perturbations only), and (a) to (d) show GFDL results. All the results were obtained assuming stratospheric temperature equilibrium in the presence of a fixed dynamical heating (from Ramaswamy *et al.*, 1992).

due to the sum of the non-ozone gases (CO_2 , CH_4 , N_2O and the CFCs). The illustration indicates that, for the decade of the 1980s, the net ozone radiative forcing in middle to high latitudes is negative, being opposite in sign to the effects due to the non-ozone gases. Poleward of 30 degrees (N and S), the magnitude of the (negative) decadal ozone forcing becomes increasingly comparable to and can even exceed the (positive) CFC forcing over the same time period (WMO, 1992). In higher latitudes, the ozone forcing can counteract a significant fraction of the (positive) total non-ozone gas forcing over the same time period (WMO, 1992). When globally- and annually-averaged (Ramaswamy *et al.*, 1992) and assuming that there is no change in the dynamical heating of the stratosphere, the ozone forcing (-0.08 to -0.09Wm^{-2}), is comparable in magnitude ($\sim 80\%$) but opposite in sign to the decadal CFC greenhouse forcing (0.10 to 0.11Wm^{-2}). The globally-averaged ozone forcing is opposite in sign and is about 18% of the sum of the non-ozone decadal trace gas forcing (0.45 to 0.47Wm^{-2}).

It is emphasized that the ozone forcing is extremely sensitive to the altitude of the losses (Ramanathan *et al.*, 1985; Lacis *et al.*, 1990). There still is some uncertainty regarding the exact profile and the magnitude of the loss in the immediate vicinity of the tropopause. The SAGE profiles are available globally only from $\sim 17\text{km}$ and above, and suggest an increasing percentage of loss with decreasing altitude in the lower stratosphere. As an illustration of this sensitivity, let us suppose that the losses observed by TOMS are uniformly distributed through the entire stratospheric column (see WMO, 1986). For this to be the case, the principal ozone depletions would have to occur at altitudes higher than observed over the past decade, in which case a much smaller global ozone forcing (-0.01 to -0.04Wm^{-2}) would result. Thus, inferences about ozone forcing depend crucially on both the total column change as well as the change in the vertical profile.

The radiative forcing due to ozone is unique in two respects when compared to that due to the non-ozone gases. First, although there is an approximate global mean offset of the direct CFC forcing by the ozone losses occurring during the 1979-1990 period, this arises because of a significant negative forcing by ozone occurring only in the middle to high latitudes, in particular the radiative forcing due to increasing CFCs and decreasing ozone ranges from a net positive one at low latitudes to a net negative one at higher latitudes for the period considered (WMO, 1992). Because of the spatial dependence of the ozone losses and consequently the ozone forcing, the globally averaged results represent a considerable simplification and must be treated with caution.

Second, for ozone, unlike the other radiatively active species, both solar and the longwave interactions become significant. The negative surface-troposphere forcing at the

mid-to-high latitudes consists of a solar-induced warming tendency at the surface, combined with a longwave-induced cooling tendency of the troposphere (Ramanathan and Dickinson, 1979; WMO, 1992).

A2.4.2 *The Greenhouse Effect of Ozone Losses*

The weight of evidence suggests that the observed stratospheric ozone losses are due to heterogeneous chemical reactions involving chlorine- and bromine-containing chemicals (halocarbons), particularly the anthropogenic emissions of CFCs (WMO, 1992). The computed ozone radiative forcings indicate that the indirect chemical effects due to the halocarbons have substantially reduced the radiative contributions of the CFCs to the greenhouse forcing over the past decade (WMO, 1992). Thus, the net greenhouse impact attributed to the CFCs taken together, including the indirect as well as direct contributions, may be significantly reduced because of the halocarbon-induced destruction of ozone.

Three-dimensional General Circulation Model (GCM) simulations of the impacts on the Earth's climate due to the ozone losses have not been performed as yet, so estimates of the effects on the surface temperature are not available. In an investigation of the climatic effects due to the Northern Hemisphere mid-latitude ozone changes around the tropopause during the decade of the 1970s, Lacis *et al.* (1990) estimated a cooling of the surface at those latitudes. One-dimensional globally- and annually-averaged radiative-convective models indicate that, while a loss of ozone in the lower stratosphere leads to a surface cooling, ozone losses in the middle and upper stratosphere, as predicted from homogeneous gas-phase chemistry models, yield a warming of the surface (Ramanathan *et al.*, 1985).

A2.4.3 *Ozone Depletion and Stratospheric Temperature Changes*

The indirect effect of CFCs on the climate system due to depletion of ozone in the lower stratosphere is critically sensitive to the actual temperature change and its distribution in the lower stratosphere. Because atmospheric circulation can change in response to radiative perturbations, the dynamical contribution to the heating could also change, thereby contributing to the actual temperature change in the stratosphere (Dickinson, 1974).

The observed global ozone depletion has not yet been simulated in a GCM, but simulations for the following scenarios of ozone changes have been performed: (a) a uniform decrease of O_3 throughout the stratospheric column (Fels *et al.*, 1980; Kiehl and Boville, 1988), (b) a homogeneous gas-phase chemical model prediction of ozone depletion (Kiehl and Boville, 1988), which is different from the observed losses, and (c) observed springtime depletion in the Antarctic region (Kiehl *et al.*,

1988). The resulting stratospheric temperature changes in these studies indicate that, unless the column depletions are large (>50%), the GCM response is similar to the response indicated by the fixed dynamical heating model but there are some latitude-dependent differences. The GCM studies of Rind *et al.* (1990, 1991) suggest that dynamically forced temperature changes can result from subtle interactions between changes in the atmospheric structure, upper tropospheric latent heat release, and the forcing and transmission of planetary waves and gravity waves.

Turning to the observed temperature trend (see Section C3.3), the long-term temperature trends in the lower stratosphere suggest a cooling. While this would be in accord with the ozone-induced radiative cooling tendency of the lower stratosphere (Lacis *et al.*, 1990; WMO, 1992; Miller *et al.*, 1992) considerable difficulties remain in attributing a part or all of the observed stratospheric temperature trends to the ozone losses. Other physical factors, such as possible changes in the physical state of the troposphere, volcanic aerosols, change in stratospheric circulation, etc., which are not accounted for in the determination of the radiative forcing, could also be contributing to the trends. Thus, significant questions remain with regard to the influence of ozone losses on stratospheric temperatures.

A2.5 Radiative Forcing due to Gases in the Troposphere

A2.5.1 Introduction

Indirect greenhouse gas effects, induced by changes in the chemistry of the troposphere, are likely to be significant. Gases which are key compounds for the indirect effects (and for the oxidation processes in the troposphere) are O₃ and OH.

Ozone is a greenhouse gas by itself. It is formed *in situ* in the atmosphere by photochemical processes. Tropospheric O₃ concentrations are influenced by the distribution of CH₄, CO, NO_x and NMHCs, leading to indirect greenhouse contributions for these gases. OH is of importance for the greenhouse effect because it controls the loss of a large number of greenhouse gases in the troposphere (CH₄, HFCs, HCFCs, etc.), thereby determining their chemical lifetimes. Furthermore, O₃ and OH will be affected by the release of these gases, leading to important feedback effects on their lifetimes. Additional indirect greenhouse effects arise from CH₄, CO and NMHCs since CO₂ is the end product of their chemical oxidation in the troposphere.

Since the tropospheric chemical processes determining the indirect greenhouse effects are highly complex and not fully understood, the uncertainties connected with estimates of the indirect effects are larger than the

uncertainties of those connected to estimates of the direct effects. Added to these uncertainties are the limitations of the current models in formulating the distribution and spatial variability of NO_x in the troposphere, or the impact of clouds on gas phase chemistry (see Chapter 5, WMO, 1992).

This sub-section examines the key processes for O₃ and OH formation, the role of gaseous emissions of CH₄, NO_x, CO and NMHC in changing the O₃ and OH distributions (and thereby leading to indirect effects), and uncertainties in estimating these impacts. The impact of increased UV fluxes in the troposphere due to reduced ozone columns, and enhanced water vapour content resulting from enhanced temperatures are not included. Both effects are likely to enhance the OH levels in the troposphere.

A2.5.2 Chemical processes and changes in O₃ and OH

Although tropospheric O₃ only makes up about 10% of all ozone in the atmosphere, its presence is central to the problem of the oxidizing efficiency of the troposphere. O₃ photolysis is the primary source of OH radicals as well as being an oxidizing species itself. Through the formation of OH it determines the cleansing efficiency of the troposphere. Ultimately, therefore, ozone is one of the most important constituents in determining the chemical composition of the troposphere.

The production of ozone depends on the concentrations of NO_x (WMO, 1992, Chapter 5). As the latter compounds are short-lived, and their concentrations vary strongly in the troposphere, ozone production is believed to vary significantly throughout the troposphere. There is also an *in situ* chemical loss of ozone in the troposphere in areas where NO_x concentrations are particularly low (less than approximately 20 pptv). This occurs in the middle troposphere and in remote oceanic areas where there are no NO_x sources (Liu *et al.*, 1987). Ozone sonde measurements reported in WMO (1992) indicate that ozone has increased by 1-1.5% per year in the free troposphere over Europe during the last 20 to 25 years (Staelin and Schmid, 1991). A similar trend in tropospheric ozone has previously been reported for stations influenced by regional air pollution (Logan, 1985; Bojkov, 1987; Penkett, 1988).

Methane is oxidized primarily in the troposphere (>90%) through reaction with the hydroxyl radical. Since this reaction also provides a substantial fraction of the OH loss in the troposphere, there is a strong interaction between OH and CH₄. This causes OH to decrease when CH₄ increases, leading to a further increase in CH₄ - a positive feedback (Chameides *et al.*, 1977; Sze, 1977; Isaksen, 1988).

Because of the central role O₃ and OH play in tropospheric chemistry, the chemistry of CO, CH₄, NMHC and NO_x is strongly intertwined, making the interpretation

of emission changes rather complex. In general, increases in the emissions of hydrocarbons and of CO may lead to increases in the global average O₃, but to reductions in OH levels (Isaksen and Hov, 1987). The result will be a slower atmospheric loss of methane and other species controlled by OH (e.g., HFCs, HCFCs). The impact of NO_x changes is different. Increased emissions of NO_x lead to increases in both globally averaged O₃ and OH (Isaksen and Hov, 1987). The increased OH levels reduce the lifetime of methane. The important consequence of this is that NO_x emissions have opposing effects on the two greenhouse gases O₃ and CH₄.

Significant changes in the global distribution of OH may have occurred over the last two centuries as the trace gas composition of the troposphere has changed dramatically. Key compounds like CII₄ and CO have increased in concentrations. This is expected to have led to reduced OH levels. On the other hand, increases in the concentrations of NO_x that are believed to have occurred (although it has not been possible to measure any changes) will have tended to reduce OH levels. The net effect is difficult to estimate, and there are no direct measurements of OH which can give reliable information on the global distribution or changes over time. Several indirect methods have, however, been used to derive a global distribution of OH (WMO, 1992). A recent estimate of the OH trend is the analysis of Prinn *et al.* (1992). They deduce a trend in global OH from the ALE/GAGE CH₃CCl₃ record of +1.0±0.8% per year over the past decade. Their result was based on a simple tropospheric box model.

A2.5.3 Sensitivity of Radiative Forcing to Changes in Tropospheric Ozone

The radiative forcing due to increases in tropospheric ozone has been investigated in earlier reports (WMO, 1986). Even though tropospheric ozone amounts are less than in the stratosphere, their effective longwave optical pathlength is comparable to that in the stratosphere (Ramanathan and Dickinson, 1979), thus rendering them radiatively important. In particular, the upper tropospheric concentrations are most significant (Lacis *et al.*, 1990). A 10% uniform increase with height in the concentrations of tropospheric ozone from current levels at 40°N (January conditions) yields a positive radiative forcing of about 0.1Wm⁻² (WMO, 1992). Although the radiative effect of increases in tropospheric ozone could be extremely important in the greenhouse forcing of climate, there is, at present, insufficient evidence that such increases are actually taking place globally, especially in the radiatively significant upper tropospheric regions. In the absence of data from which meaningful global trends can be derived, it is not possible at this stage to quantify the current contribution of tropospheric ozone to the global greenhouse radiative forcing.

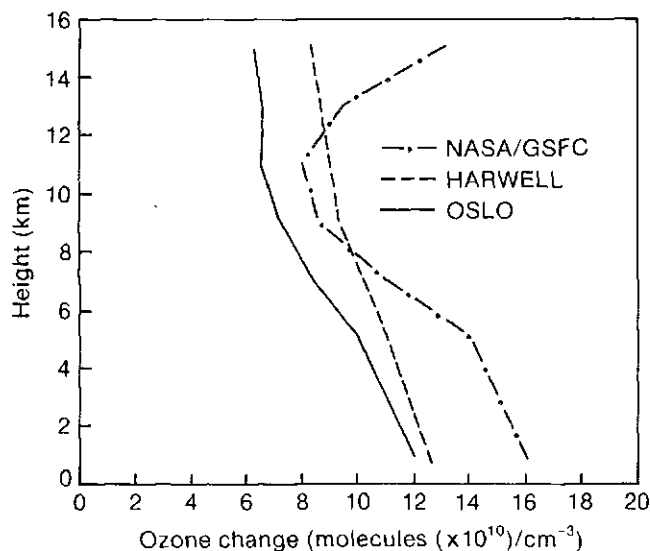


Figure A2.2: Calculated global average ozone increases due to a doubling (compared to current levels) of surface emissions of methane (2-D model calculations with the Harwell and the Oslo models referred to in WMO, 1992) and a doubling of the surface concentrations of methane (1-D model calculations with the NASA/GSFC model). In the latter case, ozone production is most likely underestimated compared to the other two cases because it implies smaller emission increases (see discussion in the text).

A2.5.4 Indirect Effects due to CH₄ Emissions

A2.5.4.1 Changes in Lifetimes due to Changes in OH

Enhanced surface emissions of CH₄ cause increased ozone levels which show moderate variations with latitude and season (WMO, 1992). The increase is most pronounced in the lower troposphere and decreases with height. Figure A2.2 shows calculated global and seasonal average ozone increases (in molecules per cm³) with height for three different tropospheric models resulting from a doubling of surface CH₄ (fluxes and concentrations).

The positive feedback on methane through the impact on the OH distribution is found to be substantial. Furthermore, the feedback is non-linear: it increases with increasing CH₄ emission in the sense that the relative increase in steady-state concentration for a given increase in emissions increases faster than the relative increase in emissions. For example, a 10% increase in emission leads to a 13 to 14% increase in steady-state concentration, while a doubling of emissions leads to a 150% increase in concentration. In a similar way the feedback will affect the concentrations when emissions are reduced, but it will become less significant at lower concentrations.

A2.5.4.2 Radiative Forcing Changes from CH₄-Induced Changes in Ozone

Methane has an indirect effect on the radiation balance through its influence on ozone. Ozone changes in the troposphere due to increased CH₄ surface fluxes (Figure

A2.2) have been used to calculate the change in radiative forcing at different latitudes and seasons using a radiative-convective model (W.-C. Wang, personal communication). The relative effect of ozone on the long wavelength radiation (compared to the effect of CH₄ alone) varies between 10 and 22%. The globally-averaged effect is around 14%.

The CH₄ effect in these calculations includes the direct effect and the indirect effect resulting from reduced OH levels (the positive feedback effect). Approximately 0.7 of the methane effect is a direct effect, and 0.3 is due to the OH feedback. The calculated indirect global effect through ozone increases is therefore approximately 20% of the direct methane effect.

A2.5.4.3 Effects of CH₄ on Stratospheric Water Vapour

Methane has an indirect effect through its oxidation in the stratosphere to water vapour. In IPCC (1990) it was assumed that this would enhance the methane forcing by 30% over its direct value in the absence of N₂O absorption-band overlap (see Table 2.2 in IPCC, 1990). More recent modelling studies have shown that this enhancement is highly uncertain. In WMO (1992) the range is 22 to 38%, while Lelieveld and Crutzen (1992) give a value of 5%. These differences may partly reflect the different types of numerical experiment performed to calculate the effect. They may also reflect the fact that changes depend critically on the vertical profile of the water vapour change (A.A. Lacis, personal communication).

A2.5.4.4 Oxidation to CO₂

Oxidation of CH₄ leads to formation of CO₂ and thus contributes indirectly to greenhouse warming. The contribution will depend on the time horizon used. It should be noted that only oxidation of fossil fuel-related CH₄ leads to a CO₂ increase; most CH₄ emitted into the atmosphere is short-term recycled biogenic CO₂.

A2.5.4.5 Indirect GWP for CH₄

While the results given here demonstrate the importance of a number of processes in amplifying the direct radiative forcing effect of increasing methane concentrations, they cannot be applied to directly scale up the direct GWP for methane. The experiments performed give only the steady-state changes due to a sustained emission change, whereas the GWP definition used here considers the integrated time-dependent response to a pulse emission.

A2.5.5 NO_x Emissions

The calculated global ozone changes from increases in NO_x surface emissions show large seasonal, latitudinal and height variations (WMO, 1992). The impact on ozone drops off rapidly with height in the troposphere. This is

significant as the impact on surface temperatures from ozone changes is likely to be height dependent in the troposphere with the largest effect resulting from changes in the upper troposphere (Wang and Sze, 1980; Lacis *et al.*, 1990). Furthermore, the calculations show that the results are highly model sensitive, leading to large differences (more than a factor of 2) in ozone impact between the 2-D models used.

Calculation of the impact on ozone from NO_x emissions from aircraft indicate that this source of NO_x may be more than an order of magnitude more efficient in enhancing O₃ levels than surface emissions of NO_x. The enhancement occurs also at higher altitudes (middle and upper troposphere) where O₃ changes have larger effects on surface temperatures (Wang and Sze, 1980; Lacis *et al.*, 1990).

Increased NO_x levels are expected to increase global amounts of OH, and thus to lead to a reduction in CH₄. NO_x increases therefore have an opposite effect on the abundance of the two greenhouse gases, O₃ and CH₄. Calculations also indicate that the radiative forcings caused by NO_x-induced changes in O₃ and CH₄ could be of the same magnitude. Taking all this into account, estimates of the radiative forcing of NO_x changes are extremely uncertain and cannot be reliably made at present.

There is a consistent picture emerging from the calculations of OH sensitivity to enhanced fluxes of source gases showing that increased NO_x emissions lead to increased OH and thereby to increased oxidation in the troposphere, while increases in the emissions of the other source gases lead to reduced OH values.

A2.5.6 CO and NMHC Emissions

Ground-based emissions of CO and NMHC lead to O₃ production, but these are found to be less efficient ozone producers on a global scale than CH₄. OH is also increased. The increases show pronounced global and seasonal variations making estimates of indirect GWP highly uncertain.

A2.5.7 Summary

A summary of the indirect contributions to radiative forcing from CH₄, NO_x, CO and NMHC is given in Table A2.2. The Table gives the sign of the contribution from the individual processes and the net effect, but no absolute values for the indirect effects are presented.

For CH₄, all the indirect contributions are believed to be positive and therefore they add to the direct GWP. The sum of their contributions is likely to be significant, possibly similar in magnitude to the direct effect. Indirect effects for methane will thus add substantially to the direct values given in Table A2.1. CO and NMHC will also make positive indirect contributions, although they are believed to be less significant than the contribution from CH₄ and

Table A2.2: Summary of the impact of emissions of CH₄, NO_x, CO and NMHC on OH, on O₃ and those other greenhouse gases affected by changes in OH

Increased emission of:	Effect on concentrations of		
	OH	O ₃	CH ₄ , HCFCs, HCFs
CH ₄	-	+	+
NO _x	+	+	-
CO	-	+	+
NMHC	-	+	+

more difficult to assess due to temporal and spatial variations in concentrations. For NO_x, uncertainties are large and it is not possible to give even the sign of the indirect effect. It should be noted, however, that NO_x emitted from aircraft in the upper troposphere could have a large impact on the chemistry and, thereby, on the radiative forcing on the surface-troposphere-system.

Although indirect GWP values were given in IPCC (1990), we are now aware of additional complications affecting such calculations and are less sure of the results. As a consequence, no indirect GWP values are given in this report. For methane, however, the indirect contributions to the GWP are likely to be significant, possibly as large as the direct effect.

A2.6 Radiative Forcing due to Aerosol Particles

A2.6.1 Tropospheric Particles

A2.6.1.1 Background

Aerosol particles influence the Earth's radiative balance directly by scattering and absorption of shortwave (solar) radiation. An increase in concentrations of aerosol particles will enhance this effect. Aerosol particles also absorb and emit longwave (infrared) radiation, but this effect is usually small because (a) the opacity of aerosols decreases at longer wavelengths and (b) the aerosols are most concentrated in the lower troposphere where the atmospheric temperature, which governs emission, is practically the same as the surface temperature (Coakley *et al.*, 1983; Grassl, 1988).

Aerosol particles also serve as sites on which cloud droplets form (cloud condensation nuclei, CCN). Increased concentrations of aerosol particles have the potential, therefore, to alter the microphysical, optical and radiative properties of clouds, changing their reflective properties and possibly their persistence.

By providing additional surfaces for condensation and heterogeneous chemistry, aerosol particles may also influence the chemical balance of gaseous species in the atmosphere. This may be especially important for the balance of O₃ in the lower stratosphere.

In the unperturbed atmosphere, the principal aerosol constituents contributing to light scattering are sulphate from biogenic gaseous sulphur compounds and organic carbon from partial atmospheric oxidation of gaseous biogenic organic compounds. Seasalt and windblown soil dust contribute substantially at some locations but their effect on the global climate is generally unimportant because the particles are large and usually short-lived and thus transported only short distances. Other aerosol substances may also be locally and regionally important, especially those that are sporadic such as from volcanoes, wildfires, and windblown dust from deserts.

Submicrometre (diameter <10⁻⁶m) anthropogenic aerosols are produced in the atmosphere by chemical reactions of primarily sulphur-, but also nitrogen- and carbon-containing gases, predominantly sulphur dioxide. These particles are efficient light scatterers, so changes in them may alter the energy balance of the atmosphere. The overall effect also depends on changes in the amount of energy absorbed. Light absorption is dominated by particles containing elemental carbon produced by incomplete combustion of carbonaceous fuel. The light scattering effect is dominant at most latitudes, but absorption might dominate at high latitude, especially over highly reflective snow- or ice-covered surfaces (Blanchet, 1989). Over surfaces with low albedo (<0.1) characteristic of most of the surface of the Earth, anthropogenic aerosols cool rather than warm the Earth (Coakley *et al.*, 1983); thus in global terms the scattering by sulphate should dominate over absorption by elemental carbon.

A2.6.1.2 New findings

The most important new information that has become available since IPCC (1990) refers to the backscattering by sulphate aerosols. Based on simulations of the global distribution of sulphate aerosols (Langner and Rodhe, 1991), Charlson *et al.* (1990, 1991 and 1992) used previously acquired information on scattering and backscattering coefficients per unit mass of sulphate to estimate the impact of anthropogenic sulphur emissions on the shortwave radiation balance in cloud-free regions. The

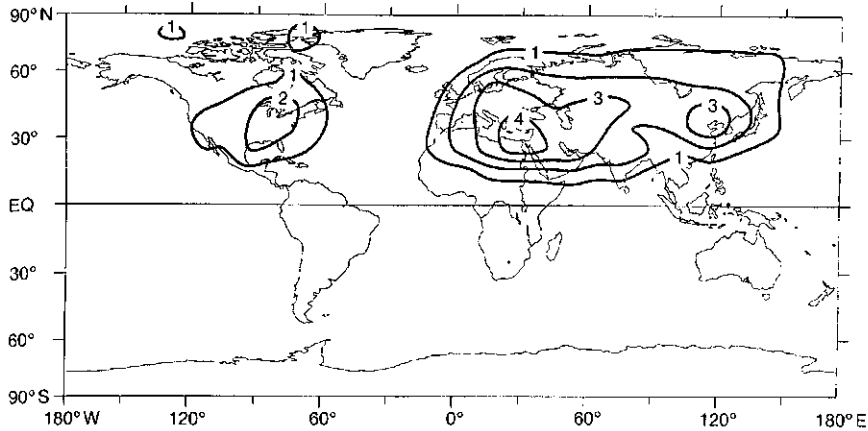


Figure A2.3: Calculated increase of reflected flux to space due to tropospheric sulphate aerosols derived from anthropogenic sources. Unit: Wm^{-2} (from Charlson *et al.*, 1991).

geographical distribution of the calculated increase of reflected solar flux to space is shown in Figure A2.3. The authors concluded that the effect for current emission levels, averaged over the Northern Hemisphere, corresponds to a negative forcing at the Earth's surface of about 1Wm^{-2} , with about a factor of two uncertainty. This is comparable (but of opposite sign) to the forcing due to anthropogenic CO_2 ($+1.5\text{Wm}^{-2}$) or to the direct forcing of the other greenhouse gases ($+1\text{Wm}^{-2}$). In addition to the direct effect on climate of sulphate aerosols, there is an indirect effect - via changes in CCN and cloud albedo - which tends to act in the same direction (i.e., towards a cooling) with a magnitude that has not yet been reliably quantified (Charlson *et al.*, 1990 and 1992; Kaufman *et al.*, 1991).

A very important implication of this estimate is that the net anthropogenic radiative forcing over parts of the Northern Hemisphere during the past century is likely to have been substantially smaller than was previously believed. A quantitative comparison between the positive forcing of the greenhouse gases and the negative forcing due to sulphate is complicated by the fact that the latter is much less uniformly distributed geographically than the former, cf. Figure A2.3.

A2.6.1.3 Discussion

Future changes in the forcing due to sulphate aerosols and greenhouse gases will depend on how the corresponding emissions vary. Because of the short atmospheric lifetimes of sulphate and its precursors, atmospheric concentrations will adjust within weeks to changes in emissions. This is a very different situation from that for most greenhouse gases which have effective lifetimes of decades to centuries. For example, the concentration of CO_2 will continue to rise for more than a century even if emissions are kept constant at today's level. This difference is illustrated in Figure A2.4 (from Charlson *et al.*, 1991), which shows schematically

how the climate forcings due to CO_2 and aerosol sulphate could change if the global fossil fuel consumption levelled off and eventually were reduced. More detailed calculations have been given by Wigley (1991). Because of the rapid growth in emissions during the past decades both the enhanced greenhouse forcing due to CO_2 and the opposite forcing due to aerosol sulphate have grown

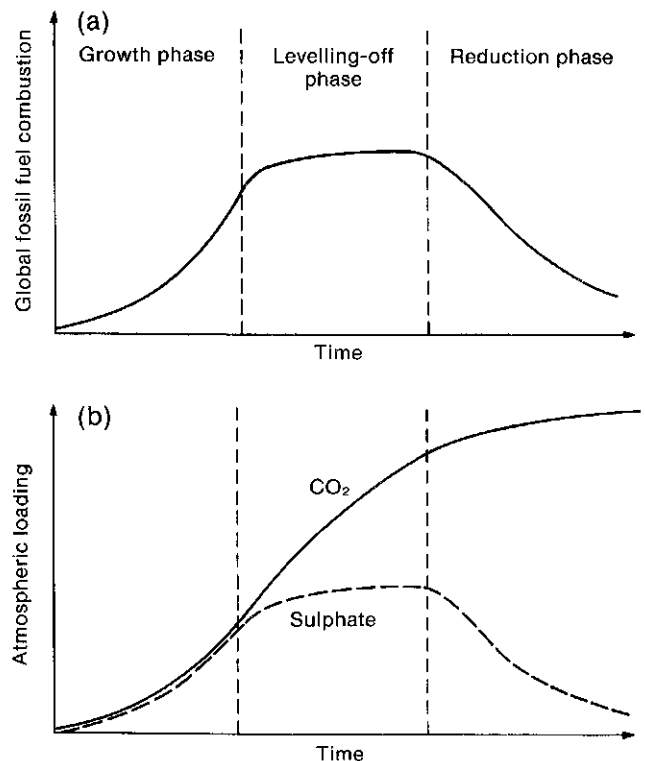


Figure A2.4: Schematic illustration of the effects of a scenario of future global fossil fuel combustion (a) on the atmospheric loadings of CO_2 and sulphate (b). The differences in response arise from their different atmospheric residence times. CO_2 produces a positive radiative forcing (heating) and sulphate a negative forcing (cooling) (from Charlson *et al.*, 1991).

accordingly. During a levelling off phase, the greenhouse forcing due to CO_2 will continue to grow whereas the aerosol forcing will remain constant. During a decay phase, the greenhouse forcing will start to level off and the aerosol forcing decline. This simple example illustrates that the relative importance of these two major anthropogenic forcing agents in the future will depend critically on the character of changes in fossil fuel use (large-scale desulphurization measures would also have to be considered).

Because of the very different character of the forcing due to aerosol sulphate as compared to that of the greenhouse gases, no attempt is made here to define a negative Global Warming Potential (GWP) for anthropogenic sulphur emissions. In addition to the well-known negative deleterious environmental effects of sulphates - including acidification - there are several reasons why increases in sulphur emissions cannot simply be considered as trade-offs against reductions in greenhouse gas emissions. Among these are: the very different horizontal and vertical distributions of radiative forcing due to sulphates compared with greenhouse gases, the great uncertainty about their effects on clouds and the subsequent effects on the climate system, and the fact that the distribution of sulphates globally is largely inferred from models rather than being directly measured. Although in a global sense the negative forcing due to aerosols may offset a substantial part of the positive greenhouse forcing, the differences in spatial distribution of greenhouse radiative forcing and aerosol effects mean that increases in sulphates can never be expected to compensate for the climatic effects of greenhouse gas increases.

It is clear that a better quantitative description of the climate influence of anthropogenic aerosols is necessary in models of past, present and future climate. Modelling studies need to take into account sulphate aerosol concentrations and their radiative influence as a function of location and time as governed by emissions of sulphur gases. To do this, models should accurately represent the direct light scattering effect and also the influences of these aerosols on cloud optical, radiative, and persistence properties. At present, the influences on clouds in particular cannot be estimated with confidence. Further, there is only a meagre data base of observations with which to validate the models of atmospheric chemistry, transport and removal processes that are required to relate aerosol concentrations to precursor emissions. There is, similarly, little observational information on the relationships between aerosol microphysical properties and cloud microphysical properties, between aerosol and cloud microphysical properties and their radiative properties, and between cloud microphysical properties and cloud persistence.

A2.6.2 Stratospheric Sulphate Particles

Observations over the past decade (lidar, satellite, balloon, sunphotometer) indicate that the stratospheric aerosol concentration throughout most of the 1980s remained higher than that measured in 1979 (a relatively quiescent period). This is probably largely attributable to the major El Chichon volcanic eruption in 1982 together with the effects of a few other minor eruptions (McCormick and Trepte, 1987). Anthropogenic sources may have provided an additional contribution (Hofmann, 1990). With the recent major eruption of the Mt. Pinatubo volcano, there is now a fresh accumulation of particulates in the stratosphere (optical depth estimated to be between 0.1 and 0.3 one month after the eruption; M. P. McCormick, personal communication). For more details, the reader is referred to the special issue of *Geophysical Research Letters* (Vol. 19, 149-218, 1992).

The radiative effects due to these particles may be already manifest in an observed warming of the lower stratosphere (see Section C4.2.4.2; also Labitzke and McCormick, 1992). Radiative forcing calculations indicate that these aerosols can also be expected to exert a significant negative but transient radiative forcing ($\sim 0.5 \text{ W m}^{-2}$ or more in magnitude) on the surface-troposphere system over the next few years (Hansen *et al.*, 1992). This is opposite in sign to the greenhouse gas-induced forcing. General Circulation Model simulations (Hansen *et al.*, 1992) suggest that such an aerosol-induced forcing could yield a temporary cooling tendency at the surface and dominate the global surface temperature record in the next year or more (see Section C4.2.4.2).

A2.7 Forcing Due to Solar Irradiance Changes

For a recent review of changes in solar irradiance, see Lean (1991). A 1% change in total irradiance is equivalent to a radiative forcing of 2.4 W m^{-2} at the top of the troposphere, comparable to the total enhanced greenhouse forcing to date. Considerably smaller changes than this could, if sustained for a number of years, noticeably affect global climate and either enhance or offset the effects of increasing greenhouse gas concentrations. It is necessary, therefore, to monitor future and, if possible, reconstruct past irradiance changes with an accuracy of substantially better than $\pm 1\%$.

To obtain better accuracy, it is necessary to place instruments high in or above the atmosphere. Continuous observations require satellite instrumentation and the available record spans only the period from 1978 to the present. These data show a strong link between solar magnetic activity (sunspots, faculae and the background "active" network radiation) and total irradiance on time-scales of days to years. During the sunspot minimum of 1986, total irradiance was about 0.1% less than during the

previous (1980) maximum, with the reduction in output from bright features outweighing the decreased blocking effect of sunspots (Foukal and Lean, 1988, 1990; Lean, 1989). Changes since 1986 have continued to parallel the 11-year sunspot cycle (Willson and Hudson, 1991).

The small amplitude of the observed solar-cycle-related irradiance changes does not preclude the existence of additional lower-frequency effects operating on the 10 to 100 year time-scale (Foukal and Lean, 1990; Lean, 1991). To investigate this possibility further, there have been attempts to extend the observational record back before 1980 by using rocket- and balloon-based measurements (Fröhlich, 1987). These show an apparent change in irradiance between the late 1960s and the late 1970s of around 0.4%. There is considerable doubt, however, about the representativeness of these values (measured over time-scales of a day) and about the absolute accuracy of the instruments used to obtain them (Lean, 1991). Although Reid (1991) argues against these problems, it is clearly difficult to identify a long-term trend using extremely noisy daily data from instruments of uncertain accuracy.

Apart from these data, there are no useful direct irradiance measurements prior to 1978, so various authors have tried to deduce irradiance forcing indirectly. For example, Reid (1991) has suggested that low-frequency irradiance changes run parallel to the envelope of sunspot activity, which shows quasi-cyclic behaviour with a roughly 80-year period, and Friis-Christensen and Lassen (1991) have hypothesized that low-frequency irradiance changes are related to changes in the length of the solar cycle. In both cases, there is a strong visual correspondence between the solar irradiance proxy and global-mean temperature changes over the past 100 years - see Section C4.2.1. These results are intriguing, but they have yet to be fully evaluated in terms of the implied changes in solar forcing compared to greenhouse forcing (Kelly and Wigley, 1990).

An entirely different approach has been used in a study by Baliunas and Jastrow (1990) - see also Radick *et al.* (1990). They have examined the magnetic activity of solar-type stars to try to throw some light on possible changes in irradiance associated with events like the Maunder Minimum of sunspot activity (1645-1715). The precisely-dated record of atmospheric radiocarbon measurements shows that similar periods of prolonged sunspot minima have occurred on many occasions during the past 8000 years (randomly spaced, but every 500 years on average). It has been suggested that they correspond to periods of lowered irradiance and global cooling (Eddy, 1976; Wigley and Kelly, 1990). Baliunas and Jastrow (1990) find that solar-type stars exhibit two modes of activity, a cyclic mode similar to the Sun's present condition, and a less variable mode (with lower magnetic activity) akin to

conditions thought to prevail during the Maunder Minimum. They conclude that the Sun's irradiance during the Maunder Minimum (and other similar periods) was "several tenths of a per cent" less than current levels.

A more sophisticated interpretation of these stellar data has been carried out by Lean *et al.* (1992) using knowledge of the mechanisms of irradiance variations gleaned from extant solar data. They consider two possible effects: changes in irradiance associated directly with changes in magnetic activity, and changes associated with a reduced basal emission during prolonged periods of reduced activity. The estimated irradiance reduction during a Maunder Minimum period is $0.25 \pm 0.1\%$.

It should be noted that these astronomical results do not yet prove that the Sun's irradiance was reduced during periods like the Maunder Minimum. Since no star has been observed to change mode, it is not yet known whether the observed stellar differences reflect different types of star or different modes of variation for individual stars. Nevertheless, the magnitude of the potential changes estimated by Lean *et al.* (1992) and Baliunas and Jastrow (1990) compares favourably with the empirical estimate (based on palaeoclimatic data) for a Maunder Minimum irradiance reduction of 0.22-0.55% given by Wigley and Kelly (1990). All three estimates are substantially below that of Reid's (1991) estimate of around 1%. Wigley and Kelly (1990) note that, were a similar event to begin now or in the near future, then it would partially offset the anticipated increase in forcing due to increasing greenhouse gas concentrations but only by a small amount.

References

- Baliunas, S.** and R. Jastrow, 1990: Evidence for long-term brightness changes of solar-type stars. *Nature*, **348**, 520-523.
- Blanchet, J.-P.**, 1989: Toward estimation of climate effects due to Arctic aerosols. *Atmos. Env.*, **23**, 2609-2625.
- Bojkov, R.D.**, 1987: Ozone changes at the surface and in the free troposphere. In: *Tropospheric Ozone*. Proceedings of the NATO Workshop. I.S.A. Isaksen (Ed.), D. Reidel, Boston, pp83-96.
- Chameides, W.L., S.C. Liu and R.J. Cicerone**, 1977: Possible variations in atmospheric methane. *J. Geophys. Res.*, **82**, 1795-1798.
- Charlson, R.J., J. Langner and H. Rodhe**, 1990: Sulfate aerosols and climate. *Nature*, **348**, 22-26.
- Charlson, R.J., J. Langner, H. Rodhe, C.B. Leovy and S.G. Warren**, 1991: Perturbation of the Northern Hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols. *Tellus*, **43A-B** (4), 152-163.
- Charlson, R.J., S.E. Schwartz, J.M. Hales, R.D. Cess, J.A. Coakley Jr., J.E. Hansen and D.J. Hofmann**, 1992: Climate forcing by anthropogenic aerosols. *Science*, **255**, 423-430.

- Coakley, J.A. Jr., R.D. Cess and F.B. Yurevich, 1983:** The effect of tropospheric aerosols on the Earth's radiation budget: a parametrization for climate models. *J. Atmos. Sci.*, **40**, 116-138.
- Dickinson, R. E., 1974:** Climate effects of stratospheric chemistry. *Can. J. Chem.*, **52**, 1616-1624.
- Eddy, J.A., 1976:** The Maunder Minimum. *Science*, **192**, 1189-1202.
- Fels, S.B., J.D. Mahlman, M.D. Schwartzkopf and R.W. Sinclair, 1980:** Stratospheric sensitivity to perturbations in ozone and carbon dioxide: Radiative and dynamical response. *J. Atmos. Sci.*, **37**, 2266-2297.
- Fisher, D.A., C.H. Hales, W.-C. Wang, M.K.W. Ko and N.D. Sze, 1990:** Model calculations of the relative effects of CFCs and their replacements on global warming. *Nature*, **344**, 513-516.
- Foukal, P. and J. Lean, 1988:** Magnetic modulation of solar luminosity by photospheric activity. *Astrophysical Journal*, **328**, 347-357.
- Foukal, P. and J. Lean, 1990:** An empirical model of total solar irradiance variation between 1874 and 1988. *Science*, **247**, 556-558.
- Friis-Christensen, E. and K. Lassen, 1991:** Length of the solar cycle: an indicator of solar activity closely associated with climate. *Science*, **254**, 698-700.
- Fröhlich, C., 1987:** Variability of the solar "constant" on time scales of minutes to years. *J. Geophys. Res.*, **92**, 796-800.
- Grassl, H., 1988:** What are the radiative and climatic consequences of the changing concentration of atmospheric aerosol particles? In: *The Changing Atmosphere*. F.S. Rowland and I.S.A. Isaksen (Eds.), Wiley and Sons, Chichester, pp187-199.
- Hansen, J., A. Lacis, R. Ruedy and M. Sato, 1992:** Potential climate impact of Mount Pinatubo eruption. *Geophys. Res. Lett.*, **19**, 215-218.
- Hofmann, D.J., 1990:** Increase in the stratospheric background sulfuric acid aerosol mass in the past 10 years. *Science*, **248**, 996-1000.
- IPCC, 1990:** *Climate Change: The Scientific Assessment*. J.T. Houghton, G.J. Jenkins and J.J. Ephraums (Eds.), Cambridge University Press, Cambridge, UK, pp365.
- Isaksen, I.S.A. and Ø.Hov, 1987:** Calculation of trends in the tropospheric concentration of O₃, OH, CO, CH₄ and NO_x. *Tellus, Ser. B.*, **39**, 271-285.
- Isaksen, I.S.A., 1988:** Is the oxidizing capacity of the atmosphere changing? In: *The Changing Atmosphere*. F.S. Rowland and I.S.A. Isaksen (Eds.), Wiley-Interscience, pp141-157.
- Johnson, C., J. Henshaw and G. McInnes, 1992:** Impact of aircraft and surface emissions of nitrogen oxides on tropospheric ozone and global warming. *Nature*, **355**, 69-71.
- Kaufman, Y.J., R.S. Fraser and R.L. Mahoney, 1991:** Fossil fuel and biomass burning effect on climate - heating or cooling? *J. Clim.*, **4**, 578-588.
- Kelly, P.M. and T.M.L. Wigley, 1990:** The influence of solar forcing trends on global mean temperature since 1861. *Nature*, **347**, 460-462.
- Kiehl, J.T., B.A. Boville and B.P. Briegleb, 1988:** Response of a general circulation model to a prescribed Antarctic ozone hole. *Nature*, **332**, 501-504.
- Kiehl, J.T. and B.A. Boville, 1988:** The radiative-dynamical response of a stratospheric-tropospheric general circulation model to changes in ozone. *J. Atmos. Sci.*, **45**, 1798-1817.
- Labitzke, K. and M.P. McCormick, 1992:** Stratospheric temperature increases due to Pinatubo aerosols. *Geophys. Res. Lett.*, **19**, 207-210.
- Lacis, A.A., D.J. Wuebbles and J.A. Logan, 1990:** Radiative Forcing of Climate by Changes in the Vertical Distribution of Ozone. *J. Geophys. Res.*, **95**, D7, 9971-9981.
- Langner, J. and H. Rodhe, 1991:** A global three-dimensional model of the tropospheric sulfur cycle. *J. Atmos. Chem.* **13**, 225-263.
- Lean, J., 1989:** Contribution of ultraviolet irradiance variations to changes in the Sun's total irradiance. *Science*, **244**, 197-200.
- Lean, J., 1991:** Variations in the Sun's radiative output. *Rev. Geophys.*, **29**, 505-535.
- Lean, J., W. Livingston, A. Skumanich and O. White, 1992:** Estimating the Sun's radiative output during the Maunder Minimum. (Unpublished manuscript).
- Lelieveld, J. and P.J. Crutzen, 1992:** Indirect effects of methane in climate warming. *Nature*, **355**, 339-342.
- Liu, S.C., M. Trainer, F.C. Fehsenfeld, D.D. Parrish, E.J. Williams, D.W. Fahey, G. Hubler and P.C. Murphy, 1987:** Ozone production in the rural troposphere and the implications for regional and global ozone distribution. *J. Geophys. Res.*, **92** (4), 191-207.
- Logan, J.A., 1985:** Tropospheric ozone: seasonal behavior, trends, and anthropogenic influence. *J. Geophys. Res.*, **90**, 10463-10482.
- McCormick, M.P. and C.R. Trepte, 1987:** Polar stratospheric optical depth observed between 1978 and 1985. *J. Geophys. Res.*, **92**, 4297-4307.
- McCormick, M.P., R.E. Veiga and W.P. Chu, 1992:** Stratospheric ozone profile and total ozone trends derived from the SAGE I and SAGE II data. *Geophys. Res. Lett.*, **19**, 269-272.
- Manabe, S. and R.F. Strickler, 1964:** Thermal equilibrium of an atmosphere with a convective adjustment. *J. Atmos. Sci.*, **21**, 361-385.
- Miller, A.J., R.M. Nagatani, G.C. Tiao, X.F. Niu, G.C. Reinsel, D.J. Wuebbles and K. Grant, 1992:** Comparisons of observed ozone and temperature trends in the lower stratosphere. *Geophys. Res. Lett.* (Submitted).
- Penkett, S.A., 1988:** Indications and causes of ozone increases in the troposphere. In: *The Changing Atmosphere*. F.S. Rowland and I.S.A. Isaksen (Eds.), Wiley-Interscience, New York, pp91-103.
- Prinn, R., D. Cunnold, P. Simmonds, F. Alyea, R. Boldi, A. Crawford, P. Fraser, D. Gutzler, D. Hartley, R. Rosen and R. Rasmussen, 1992:** Global average concentration and trend for hydroxyl radicals deduced from ALE/GAGE trichloroethane (methyl chloroform) data: 1978-1990. *J. Geophys. Res.* (In press).
- Radick, R.R., G.W. Lockwood and S.L. Baliunas, 1990:** Stellar activity and brightness variations: A glimpse at the Sun's history. *Science*, **247**, 39-44.
- Ramanathan, V. and R.E. Dickinson, 1979:** The role of stratospheric ozone in the zonal and seasonal radiative energy balance of the earth-troposphere system. *J. Atmos. Sci.*, **36**, 1084-1104.
- Ramanathan, V., R.J. Cicerone, H.B. Singh and J.T. Kiehl, 1985:** Trace gas trends and their potential role in climate change. *J. Geophys. Res.*, **90**, 5547-5566.

- Ramaswamy, V., M.D. Schwarzkopf and K.P. Shine, 1992:** Radiative forcing of climate from halocarbon-induced global stratospheric ozone loss. *Nature*, **355**, 810-812.
- Reid, G.C., 1991:** Total solar irradiance variations and the global sea surface temperature record. *J. Geophys. Res.*, **96**, 2835-2844.
- Rind, D., N.K. Balachandran and R. Suozzo, 1991:** Climate change and the middle atmosphere. Part II: The impact of volcanic aerosols. *J. Clim.* (In press).
- Rind, D., R. Suozzo, N.K. Balachandran and M.J. Prather, 1990:** Climate change and the middle atmosphere. Part I: The doubled CO₂ climate. *J. Atmos. Sci.*, **47**, 475-494.
- Shine, K.P., 1987:** The middle atmosphere in the absence of dynamical heat fluxes. *Quart. J. Roy. Meteor. Soc.*, **113**, 603-633.
- Siegenthaler, U., 1983:** Uptake of excess CO₂ by an outcrop-diffusion model of the ocean. *J. Geophys. Res.*, **88**, 3599-3608.
- Siegenthaler, U. and H. Oeschger, 1987:** Biospheric CO₂ emissions during the past 200 years reconstructed by deconvolution of ice core data. *Tellus*, **39B**, 140-154.
- Stahelin, J. and W. Schmid, 1991:** Trend analysis of tropospheric ozone concentrations utilizing the 20-year data set of ozone balloon soundings over Payerne (Switzerland). *Atmos. Envir.*, **25a**, 1739-1749.
- Stolarski, R.S., P. Bloomfield, R.D. McPeters and J.R. Herman, 1991:** Total ozone trends deduced from NIMBUS 7 TOMS data. *Geophys. Res. Lett.*, **18**, 1015-1018.
- Sze, N.D., 1977:** Anthropogenic CO emissions: implications for the atmospheric CO-OH-CH₄ cycle. *Science*, **195**, 673-675.
- Wang, W.-C. and N.D. Sze, 1980:** Coupled effects of atmospheric N₂O and O₃ on Earth's climate. *Nature*, **286**, 589-590.
- Wang, W.-C., M.P. Dudek, X.-Z. Liang and J.T. Kiehl, 1991:** Inadequacy of effective CO₂ as a proxy in simulating the greenhouse effect of other radiatively active gases. *Nature*, **350**, 573-577.
- Wigley, T.M.L., 1991:** Could reducing fossil-fuel emissions cause global warming? *Nature*, **349**, 503-506.
- Wigley, T.M.L., M. Hulme and T. Holt, 1990:** *An alternative approach to calculating global warming potentials*. Presented to a Workshop on the Scientific Basis of Global Warming Potential Indices, Boulder, CO, USA, Nov. 1990.
- Wigley, T.M.L. and P.M. Kelly, 1990:** Holocene climatic change, ¹⁴C wiggles and variations in solar irradiance. *Phil. Trans. Roy. Soc.*, **A330**, 547-560.
- Willson, R.C. and H.S. Hudson, 1991:** The Sun's luminosity over a complete solar cycle. *Nature*, **351**, 42-44.
- WMO, 1986:** Atmospheric Ozone 1985. WMO Global Ozone Research and Monitoring Project, Report No. 16, Geneva.
- WMO, 1992:** *Scientific Assessment of Ozone Depletion*. WMO/UNEP, WMO Global Ozone Research and Monitoring Project, Report No. 25, Geneva.

