Chapter 2: Changes in Atmospheric Constituents and in Radiative Forcing

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Executive Summary

Radiative Forcing works well as a comparative estimator of the global mean surface temperature changes for the range of human and natural influences on climate.

Radiative forcings (RFs) are given for present day (2004) relative to preindustrial times (1750). Positive RFs are expected to lead to a climate warming and negative RFs to a cooling of climate.

RF is one of a number of ways to compare the effect of different climate change mechanisms. The TAR definition is retained as it continues to be a useful tool to compare the global mean surface temperature response to different forcing agents. Global mean RF is not designed as an indicator of the detailed aspects of climate response.

The concept of efficacy has been introduced as a comparative measure of the effectiveness of a given RF agent at changing the surface temperature, compared to carbon dioxide. Model studies since TAR give high confidence that efficacies for realistic anthropogenic and natural RFs lie within the 0.75–1.25 range. There is also some evidence of systematic model-independent variation in efficacies between mechanisms that may enable a future refinement of the RF concept. Therefore there is now a high confidence that global-mean RF gives a comparative estimate of the equilibrium global mean surface temperature change for realistic climate change mechanisms.

The Global Warming Potential (GWP) remains the recommended metric for comparing the potential climate impact of the emissions of different forcing agents. There are well-documented shortcomings, particularly in using GWP to assess the impact of short-lived species.

Humans have very likely contributed a net warming effect on climate. The global mean RF for combined net total of all anthropogenic effects is estimated to be 1.5 ± 1.0 W m^{-2}. A negative net RF is very unlikely. RF is estimated for the main forcing agents and, for the first time, an estimate of RF associated with each principal emission source is also made.

The RF due to long-lived greenhouse gases (LLGHGs) has the highest confidence level of any forcing agent. Their combined RF is 2.59 ± 0.26 W m^{-2} and has a high level of scientific understanding. This is a 7% increase since TAR. Its RF effect on the outgoing thermal radiation spectra has been observed directly by satellite instruments.

Carbon dioxide is increasing in the atmosphere at its fastest rate ever observed in at least the last 2000 years; it has the largest RF of any known agent and the increase in its RF since TAR is larger than the change in RF from any other agent. Since the TAR the average CO_2 concentration increase for the period 1999 to 2004 was more than 1.8 ppm yr^{-1} with a sustained growth rate of over 2ppm yr^{-1} from 2001 to 2003. Over the same period fossil fuel emissions rose from 6.5 to 7.2 Gt C yr^{-1}, representing a period of much higher rates than those considered in the TAR. Current levels of atmospheric CO_2 contribute a RF of 1.63 ± 0.16 W m^{-2}; a contribution that dominates that of all other forcing agents considered in this report. This is an increase of 0.17 W m^{-2} since the 1998 value quoted in TAR, which is much larger than the RF changes due to other agents.

The methane increase since preindustrial times makes it the second largest RF contributor to the LLGHG RF at 0.48 ± 0.05 W m^{-2}. Over the past two decades the methane growth rates in the atmosphere have generally declined. Since the TAR the growth rate of methane has continued to decline and averaged only 0.8 ppb yr^{-1} for the 5-year period from 1999 to 2004. OH measurements indicate that this is likely to be due to reductions in its emissions.

The RF from the other LLGHGs is slowly increasing.

> Nitrous oxide, the fourth most important greenhouse gas, continues to rise approximately linearly (0.26% per year) reaching 319 ppb in 2004, corresponding to a RF of 0.16 ± 0.02 W m^{-2}. Recent studies reinforce the large role of tropics emissions, and the importance of troposphere-stratosphere exchange in influencing its seasonal cycle and interhemispheric gradient.
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> The industrial Kyoto Protocol gases (hydrofluorocarbons, perfluorocarbons, SF6) have increased by large factors of 3.8 (HFC-134a), 3.6 (HFC-125), 2.5 (HFC-152a), 1.4 (HFC-23), 1.3 (PFC-116) and 1.3 (SF6) between 1998 and 2004. Their total RF in 2004 was 0.015 W m^{-2} and is rising rapidly.
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> The Montreal Protocol gases (CFCs, HCFCs, chlorocarbons) as a group contributed 0.32 W m^{-2} to RF in 2004 with CFC-12 remaining as the third most important LLGHG. Their RF peaked in 2003 and is now beginning to decline. A significant issue for future levels of these gases is their leakage from "banks" such as foams in landfills.
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> **OH has shown no net change between 1979 and 2004.** Reaction with OH is the major sink for CH_4, HFCs and HCFCs and the major producer for sulphate, nitrate, and some organic aerosols. New estimates of the global average trends of OH have come from CH_3CCl_3 and ^{14}CO measurements. OH exhibits significant interannual and interdecadal variations; notably a minimum in 1997–1999 that coincides with an El Niño event and global wildfires. This could have significant implications for the LLGHGs and their RF.

Stratospheric ozone is expected to be near its minimum level and the magnitude of its RF is expected to decrease in the future. Global stratospheric ozone may be beginning to show signs of recovery but is still ~4% below pre-1980 levels. The Antarctic ozone hole still forms every spring and at certain altitudes ozone is completely destroyed. Ozone depleting substances are at their peak in the atmosphere and are expected to slowly reduce. In addition to the chemical destruction of ozone, dynamical changes may have contributed to Northern Hemisphere midlatitude ozone depletion; the RF estimate is re-evaluated to be –0.10 ± 0.04 W m^{-2}, slightly weaker than in TAR, with a medium level of scientific understanding.

Tropospheric ozone RF is estimated to be 0.4 ± 0.2 W m^{-2} with a medium level of scientific understanding. Several new model studies of the RF due to the increase in tropospheric ozone since preindustrial time exist and have increased complexity compared to models used in TAR. Models including detailed stratospheric and tropospheric chemistry show a significant reduction in tropospheric ozone at high latitudes as a result of the decline in stratospheric ozone. The RF estimate has slightly increased since TAR due to re-evaluation.

Anthropogenic water vapour changes are likely to have contributed a positive RF. Increases in methane have likely led to an increase in stratospheric water vapour giving an indirect RF re-evaluated to be ~0.1 W m^{-2} with a factor of two uncertainty and a low confidence; this value is 5–10 times higher than suggested by TAR.

Direct Aerosol RFs are considerably better understood than in TAR. A combined total direct aerosol RF is given as –0.2 ± 0.2 W m^{-2}, with a medium level of scientific understanding.

> Satellite and surface based remote sensing retrievals have developed considerably since the TAR. These retrievals provide essential validation criteria for global model verification/validation. Atmospheric models have continued to develop, and are better constrained by remote sensing and in-situ observations. They now provide estimates of the direct RF for aerosols with a significant anthropogenic component.
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> The RF of separate aerosol species is less certain than the combined RF. The direct RF for individual species is estimated to be: sulphate –0.40 ± 0.20 W m^{-2}, fossil-fuel organic carbon ~–0.08 ± 0.05 W m^{-2}, fossil-fuel black carbon +0.30 ± 0.15 W m^{-2}, biomass burning +0.06 ± 0.08 W m^{-2}, nitrate –0.15 ± 0.15 W m^{-2}, mineral dust –0.2 to +0.1 W m^{-2}. Significant changes in the

> aerosol direct RF have occurred for biomass burning, nitrate and mineral dust aerosols. For biomass burning aerosol the direct RF is of opposite sign to that in TAR owing to better modelling of the effects of biomass burning aerosol overlying cloud. For the first time, a RF for nitrate aerosol is given. For mineral dust the range in the direct RF is reduced due to the reduction in the anthropogenic fraction.

A best estimate of –1.2 ± 0.7 W m^{-2} is now given for the RF of the cloud-albedo (also referred to as first or Twomey) indirect effect of aerosols, with a low-level of scientific understanding.

A best-estimate has become possible for warm (low-level) clouds owing to estimates being available from several models. However, scientific understanding is low owing to uncertainties in both models and observations, as several aspects associated with the mechanism are not fully understood.

The more complex aerosol-cloud interactions, involving the 'semi-direct effect' and 'cloud lifetime effect' (or referred to as second or Albrecht effect) are very uncertain owing to incomplete knowledge of and large uncertainties in both aerosol and cloud processes. This report assesses these effects as climate feedbacks rather than radiative forcing terms.

Observations and models indicate that both the direct effect of aerosols and aerosol-cloud interactions lead to a substantial reduction of radiative flux at the surface which could affect the surface heat and moisture budgets.

Changes to surface properties lead to RF and other physical alterations to the climate system.

> Global anthropogenic land cover change since 1750 has consisted of more deforestation than reforestation/afforestation, with most net deforestation occurring in temperate regions. The resulting increase in surface albedo has led to a global mean RF of –0.2 ± 0.2 W m^{-2}, with low scientific understanding. Deposition of black carbon aerosols on snow decreases surface albedo and is estimated to give a RF of +0.1 W m^{-2}, with a factor of three uncertainty and a low level of scientific understanding.

> The surface moisture flux and hence the partitioning of energy between sensible and latent heat fluxes is affected by land cover change, irrigation and the response of vegetation to increasing CO_2 concentrations. The release of heat from Human Energy Production is significant at local scales in urban areas but not globally. These effects have a very low scientific understanding.

Persistent linear contrails from global aviation contribute a small RF (0.01 W m^{-2}), which is factor of 3 to 4 smaller compared to values projected from TAR. Aviation may also alter cirrus clouds. The difference in the RF estimate of line-shaped persistent contrails compared to TAR resulted from new observations of contrail cover and revised estimates of contrail optical depth. A best estimate remains unavailable for the RF of total cloudiness changes caused by subsonic aircraft operations. Observational studies provide evidence that induced cloudiness by contrail spreading and aerosol effects on cirrus microphysics is comparable to, or as much as 8 times greater than, persistent linear contrail cover. The associated RF value depends on the optical properties of the induced cloudiness, which have not been determined. The global effect of aviation aerosol on background cloudiness remains unknown.

The direct RF due to changes in the solar output is 0.12 W m^{-2}, which is less than half of the estimate given in TAR; the estimate has a factor of two uncertainty,

> The reduced RF estimate comes from a re-evaluation of the long-term change in solar irradiance since 1610. The current estimate of 0.3–1.6 W m^{-2} is considerably smaller than the TAR estimate of 2.6 W m^{-2}. However, uncertainties remain large because of the lack of direct observations and understanding of solar variability mechanisms on long time scales. Scientific level of understanding is medium.

> New present day measurements indicate that the absolute value of total solar irradiance is ~5 W m^{-2} lower than previous values. Continuous monitoring of total solar irradiance now exists for

28 years. Levels near the peak of cycle 23 (2001) were as high as during the two prior cycles, even though sunspot numbers were not.

Ozone changes forced by the solar UV irradiance 11–year cycle are now relatively well established, and are of the order of a few percent for the global column. New studies indicate that middle atmosphere changes can affect the troposphere.

The global stratospheric aerosol concentrations are now at their lowest observed values. There have been no intense volcanic events since the 1991 Pinatubo eruption. A first-order quantitative understanding of the RF due to volcanic aerosols since 1850 exists, although there is less confidence for eruptions prior to 1960. Several models provide reasonable simulations of the observed anomalies in shortwave reflectivity due to the Pinatubo eruption (to within ~75%). Volcanic aerosols perturb the stratosphere and surface-troposphere radiative energy budgets in an episodic manner, which can result in changes in atmospheric circulation (e.g., Arctic Oscillation patterns); they also influence the depletion of stratospheric ozone.

Spatial patterns of RF remain uncertain for the non-LLGHGs species, particularly aerosols, their effects on cloud, and land-use. This is in contrast to the high confidence for LLGHGs. The Southern Hemisphere net RF is very likely larger than the Northern Hemisphere one, due to the globally very inhomogeneous aerosol RF that is more concentrated in the Northern Hemisphere.

The instantaneous radiative flux change at the surface (hereafter called "surface forcing") is a useful diagnostic tool for understanding changes in the heat and moisture surface budgets and the accompanying climate change. However, unlike RF, it does not represent a measure of the global mean surface temperature response.

The total global-mean surface forcing is very likely to have been negative whilst the total RF is positive. LLGHGs have been the principal contributor to RF, with aerosols providing some offset. In contrast, at the surface, tropospheric and stratospheric aerosols are the dominant contributors to the negative surface forcing.

The total present day surface forcing is spatially very inhomogeneous and has large negative values in regions where aerosols are present. In contrast, a relatively smooth spatial structure is seen for the total positive RF. Because of their differing spatial patterns, the RF and surface forcing differ in terms of their (a) equator-to-pole gradients and (b) northern-to-Southern Hemisphere forcing ratios.

2.1 Introduction and Scope

This chapter updates information taken from Chapters 3–6 of the IPCC WG1 Third Assessment Report (IPCC*,* 2001) (hereafter TAR). It concerns itself with trends in forcing agents and their precursors since 1750, and estimates their contribution to radiative forcing (hereafter RF). Discussion of the understanding of atmospheric composition changes will focus on what is needed to explain the *trends* in forcing agents and their precursors. Areas where significant developments have occurred since the TAR will be highlighted. The chapter will draw on various assessments since TAR, in particular WMO (2003) and IPCC (2005).

Anthropogenic greenhouse gas changes, aerosol changes, aviation induced contrails and cirrus and land-use changes are assessed. Natural solar and volcanic RFs are also assessed. As well as re-evaluating and updating the trends and RFs presented in TAR, this chapter assesses several "new" forcing mechanisms that were not extensively discussed in previous assessments. In particular, water vapour changes both from CH_4 increases (in the stratosphere) and irrigation (in the troposphere) will be discussed in Section 2.3.8. Several other mechanisms associated with the short timescale and local response of the troposphere will also be considered. These mechanisms are essentially processes that either involve the interaction of aerosols with their environs (see Section 2.4.6), involve changes to land surface properties beyond surface albedo (see Section 2.5), or comprise biogeochemical changes involving radiatively active species. These mechanisms interact with the climate very differently than say increases in the long-lived greenhouse gases (LLGHGs) and do not easily fit within the "*radiative forcing*" concept. However, as these mechanisms are not routinely or well represented in most current GCM simulations (Jacob *et al.,* 2005) they will be discussed in this chapter in conjunction with the forcing agents. The chapter will also present spatial and temporal patterns of RF and it will examine the radiative energy budget changes at the surface. These will be described as "surface forcings" presented primarily as diagnostics for understanding aspects of the climate response that is being evaluated within the other chapters. Additionally, the chapter will reassess the "radiative forcing" concept itself (Section 2.8) and present efficacies for various mechanisms (efficacies are a comparative measure of the effectiveness of a given RF agent at changing the surface temperature, compared to CO_2).

Only "bottom-up" approaches of estimating RF are considered. These rely on observations and/or modelling of the relevant forcing agent. Since TAR several studies have attempted to constrain aspects of RF using "top-down" methods. In particular, attempts have been made to constrain the aerosol RF using knowledge of the temporal and/or spatial evolution of several aspects of climate. These include temperatures over the last 100 years, other RFs, climate response, and ocean heat-uptake. These methods depend on an understanding of - and sufficiently small uncertainties in - other aspects of climate change and are consequently discussed in the detection and attribution chapter (Chapter 9), although they are briefly mentioned in Section 2.4.

Other discussions of atmospheric composition changes and their associated feedbacks are presented in Chapter 7 (Couplings between Changes in the Climate System and Biogeochemistry). RF and atmospheric composition changes before 1750 are discussed in Chapter 6 (Paleoclimate). Future RF scenarios that were presented in Ramaswamy *et al.* (2001) are not updated in this report; however, there is some discussion of these in Chapter 10 (Climate Projections).

2.2 Concept of Radiative Forcing (RF)

This chapter assesses climate change agents through RF. RF is intended to be a simple measure for both quantifying and ranking the many different climate change mechanisms. It quantifies mechanisms in terms of a W m^{-2} change in the radiative energy budget. Despite many aspects of climate response being qualitatively well understood climate sensitivity and other aspects of climate response are poorly quantified. The RF approach is used to avoid uncertainties associated with modelling the actual climate response. Figure 2.2.1 shows how the RF concept fits within a general understanding of climate change.

[INSERT FIGURE 2.2.1 HERE]

The definition of *radiative forcing* from the TAR and earlier IPCC climate assessment reports is retained. Ramaswamy *et al.* (2001) define it as "*the change in net (down minus up) irradiance (solar plus long-wave; in W m^{-2}) at the tropopause AFTER allowing for stratospheric temperatures to readjust to radiative equilibrium, but with surface and tropospheric temperatures and state held fixed at the unperturbed values*".

The concept arose from early climate studies of the climate response to changes in solar insolation and CO_2, using simple radiative-convective models. However, it has proven to be particularly applicable for the assessment of the climate impact of LLGHGs (Ramaswamy *et al.,* 2001). Radiative forcing (F) can be related through a linear relationship to the global mean equilibrium temperature change at the surface (ΔT_s) : $\Delta T_s = \lambda F$, where λ is the climate sensitivity parameter. The RF as defined by TAR is labelled Fa – the stratospherically adjusted RF – to distinguish it from alternative definitions discussed in Section 2.8 (see Figure 2.2.2).

[INSERT FIGURE 2.2.2 HERE]

RF has been applied to many types of forcing mechanisms beyond changes in the LLGHGs and its applicability to these other mechanisms has been a subject of ongoing research. For the LLGHGs it is typically calculated in offline detailed radiative transfer schemes. For the other RFs it is often estimated using general circulation model (GCM) data, using a variety of methodologies (Ramaswamy *et al.,* 2001; Stuber *et al.,* 2001a; Tett *et al.,* 2002).

Since TAR a number of studies have investigated the relationship between RF and climate response, assessing the limitations of the RF concept; related to this there has been considerable debate whether some climate change mechanisms are better considered as a forcing or a feedback (Hansen *et al.,* 2005; Jacob *et al.,* 2005). The adoption of Fa assumes that the forcing is purely radiative: the stratospheric temperature adjustment included in its calculation considers only radiative effects. The response is considered to be the climate state change and, in particular the surface temperature change, that results from the initial *radiative* perturbation (see Section 2.8).

Climate can also be altered by non-radiative effects (such as changes in the availability of moisture for evaporation at the surface). It may be possible to characterise these in terms of a W m^{-2} RF but studies have not yet attempted to do this. These forcings could be significant on local scales and for the hydrological cycle (Chapter 7); they are briefly discussed in this chapter, although the science is not sufficiently mature to quantify their forcing role.

RF is one of a number of ways of quantifying and ranking climate change agents. RF is indicative of both past and future global mean climate change associated with the specific agent, but the relationship is not straightforward. To evaluate the climate response associated with a forcing its time evolution and its spatial and vertical pattern need to be taken into account. Global Warming Potentials (GWPs, Section 2.10) compare the integrated RF over a specified period (e.g., 100 years) from a unit mass pulse emission and are a way of comparing the potential climate change associated with emissions of different agents. Different approaches can give different insights and different emphasis to the various mechanisms. A few of these alternative approaches are discussed in Sections 2.8 and 2.10.

2.3 Chemically and Radiatively Important Gases

2.3.1 Atmospheric Carbon Dioxide (CO_2)

Carbon dioxide (CO_2) has the largest RF on any LLGHG (TAR). It is also a key influence on the plant physiological processes of photosynthesis and transpiration, which are key components of the global carbon and water cycles and surface energy and moisture budgets. A wide range of direct and indirect measurements shows that its atmospheric concentration has increased globally by about 100 ppm (35%) over the last 200 years from a range of 275–285 in the preindustrial to almost 380 ppm in 2004 (Keeling and Whorf*,* 2005). During this period the growth rate of CO_2 in the atmosphere increased substantially: the first 50 ppm above the preindustrial value was reached in the 1970s after more than 200 years, whereas the second 50 ppm was achieved in about 30 years. In the 10 years from 1994 to 2004 atmospheric CO_2 increased by about 19 ppm.

In the TAR the mean global CO_2 concentration for 1999 was reported as 367 ppm and its average growth rate over the period 1990 to 1999 was documented as 1.5 ppm yr^{-1} with fluctuations ranging from 0.9 to 2.8 ppm yr^{-1}. The TAR did not report the highest global annual growth rate ever recorded for CO_2 of more than 3 ppm yr^{-1} observed in 1998. Since then the global mean of data from a series of 40 remote sites in both hemispheres shows that the average CO_2 concentration increase for the 5 year period 1999 to 2004 was more than 1.8 ppm yr^{-1} with a sustained growth rate of over 2ppm yr^{-1} from 2001 to 2003 (see Figures 2.3.1 and 2.3.2). The global mean CO_2 concentration in 2004 reported from observation networks run by two different laboratories was 377 ppm.

As reported in the TAR the 1990s showed relatively low CO_2 growth rates in the first half of the decade followed by higher growth rates later in the decade. These higher rates have continued through to 2004 and the last decade has the highest average growth rate, 1.9 ppm yr^{-1} recorded for any decade since atmospheric CO_2 measurements began in the 1950s (see Figure 2.3.2 for recent data).

From 1990 to 1999, a period reported in the TAR, emissions due to fossil fuel burning, cement production and gas flaring increased from 6.1 to 6.5 GtC yr^{-1} or about 0.7% yr^{-1}. From 1999 to 2004 however, emissions rose from 6.5 to 7.2 Gt C yr^{-1} representing a period of much higher emission rates than those considered in the TAR. Emissions in 2004 were about 10% lower than the FAR business as usual (BAU) scenario. The growth in emissions from 1999 to 2004 however, exceeds the predictions of the FAR and if continued the growth rate of the FAR BAU scenario will be reached by 2010.

Current levels of atmospheric CO_2 contribute a RF of 1.63 ± 0.16 W m^{-2} when compared to preindustrial levels; a contribution that dominates that of all other forcing agents considered in this chapter. This is an increase of 12% since the value of 1.46 reported for 1998 in the TAR and is also much larger than the RF changes due to other agents. In the decade 1994 to 2004 the RF due to CO_2 increased by about 0.28 W m^{-2} an increase far greater than observed for any decade since the beginning of the industrial era, see also Table 2.3.1, which summarizes the present day concentrations and RF for the LLGHGs, and indicates changes since TAR.

[INSERT TABLE 2.3.1 HERE]

In this section we discuss the direct atmospheric measurements documenting recent changes in atmospheric CO_2 concentrations needed for the RF calculations presented later in the chapter. In addition we provide data for of the preindustrial levels of CO_2 required as the base level for these calculations. Indirect measurements of atmospheric CO_2 covering time spans of up to 500,000 years in the past are usually determined from analyses of air bubbles trapped in polar ice cores and are considered in chapter 6 of this report.

High-precision measurements of atmospheric CO_2 are essential to the understanding of the global carbon cycle and the possibility of positive climate-carbon cycle feedbacks as discussed in Chapter 7. The first in situ continuous measurements of atmospheric CO_2 made by a high-precision non-dispersive infrared gas analyser were implemented by C.D. Keeling. These began in 1958 at Mauna Loa, Hawaii, located at 19°N (Keeling *et al.,* 1995). These data documented for the first time that not only was CO_2 increasing in the atmosphere, but also that it was modulated by cycles caused by seasonal changes in photosynthesis in the terrestrial biosphere. These measurements were followed by continuous in-situ analyser programmes at other sites in both hemispheres (Conway *et al.,* 1994; Nakazawa *et al.,* 1997; Langenfelds *et al.,* 2002). In Figure 2.3.1 atmospheric CO_2 concentration data at Mauna Loa in the Northern Hemisphere are shown with contemporaneous measurements at Baring Head, New Zealand in the Southern Hemisphere (Manning *et al.,* 1997; Keeling and Whorf, 2005). These two stations provide the longest continuous analyser records of atmospheric CO_2 in the Northern and Southern Hemispheres respectively. Remote sites such as Mauna Loa, Baring Head, Cape Grim and the South Pole were chosen because air sampled at these locations is well mixed and representative of relatively large spatial regions with little risk of contamination by local sources and sinks of CO_2. These sites provided the first data from which the global increase of atmospheric CO_2 was be documented.

[INSERT FIGURE 2.3.1 HERE]

After entering the atmosphere, CO_2 is distributed on time scales of hundreds of years between the main active carbon reservoirs, namely the oceans, the terrestrial biosphere and the atmosphere. To a first order approximation the RF of CO_2 is a function of its cumulative emissions because it never leaves these reservoirs. In fact the additional CO_2 added to the active carbon reservoirs during the industrial era is only removed over many thousands of years by erosion and sedimentation (IPCC, 2001). The processes governing the movement of carbon between the active carbon reservoirs and their importance in determining the levels of CO_2 remaining in the atmosphere will be presented in Chapter 7. The other budgets for CO_2 are also discussed in Chapter 7.

In the 1980s and 1990s it was recognized that greater coverage of CO_2 measurements over continental areas was required to provide the basis for estimating sources and sinks of atmospheric CO_2 over land as well as ocean regions. Because continuous CO_2 analysers are relatively expensive to maintain and require meticulous on site calibration these records are now widely supplemented by air sample flask programmes where air is collected in glass and metal containers at a large number of continental and marine sites. After collection the filled flasks are sent to central well-calibrated laboratories for analysis. The most extensive network of international air sampling sites is operated by the National Oceanic and Atmospheric Administration's Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) in the USA. This organization collates measurements of atmospheric CO_2 from six continuous analyser locations as well as weekly flask air samples from a global network of almost 50 surface sites.

The driving forces for the increases in global atmospheric CO_2 since the industrial revolution are mainly CO_2 emissions from the combustion of cement production, gas flaring and fossil fuels. Other sources include emissions due to land-use changes such as deforestation (Houghton*,* 2003), biomass burning (Andreae and Merlet*,* 2001) and ocean warming (Barnett *et al.,* 2001). As shown in Figure 2.3.1, from 1970 to 2002 atmospheric emissions of CO_2 from cement production and the combustion of fossil fuels increased from 4 to about 7 Gt C yr^{-1} (Gt C = Gigatonnes of carbon in the form of atmospheric CO_2) (Marland *et al.,* 2005).

[INSERT FIGURE 2.3.2 HERE]

A key question is "How is the CO_2 released during fossil fuel combustion distributed amongst the atmosphere, oceans and terrestrial biosphere?" This partitioning has been investigated using a variety of techniques. Amongst the most powerful of these are measurements of the carbon isotopes in CO_2 as well as high precision measurements of atmospheric oxygen. The carbon contained in CO_2 has three naturally occurring isotopes denoted ^{12}C, ^{13}C and ^{14}C. The first of these, ^{12}C, is the most abundant isotope at about 99% followed by ^{13}C at about 1%. ^{14}C is a rare radioactive isotope present at only about 1 part in 10^{12} in atmospheric CO_2, living plants and animals in the oceans and the terrestrial biosphere. CO_2 emitted from coal, gas and oil combustion and land clearing has $^{13}C/^{12}C$ isotopic ratios that are less than those in atmospheric CO_2 and carries a signature of the particular fossil source. Hence as CO_2 from fossil fuel combustion enters the atmosphere the $^{13}C/^{12}C$ isotopic ratio in atmospheric CO_2 decreases and is a function of the mix of fossil fuels. Note that changes in the $^{13}C/^{12}C$ ratio of atmospheric CO_2 are also caused by other sources and sinks but the changing isotopic signal due to CO_2 from fossil fuel combustion can be resolved from the other components (Francey *et al.,* 1995). These changes are small, but can easily be measured using modern isotope ratio mass spectrometry which has the capability of measuring $^{13}C/^{12}C$ in atmospheric CO_2 to better than 1 part in 10^5 (Ferretti *et al.,* 2000). Data for the $^{13}C/^{12}C$ ratio of atmospheric CO_2 at Mauna Loa are presented in Figure 2.3.1, showing strong correlation with fossil fuel CO_2 emissions to the atmosphere and increasing atmospheric CO_2 concentrations (Keeling *et al.,* 2005).

Atmospheric oxygen measurements provide a powerful and independent method of determining the partitioning of CO_2 between the oceans and land (Keeling *et al.,* 1996). Oxygen and CO_2 are inversely coupled during plant respiration and photosynthesis. Also during the process of combustion oxygen is removed from the atmosphere producing a signal that decreases as atmospheric CO_2 increases on a molar basis (Figure 2.3.1). Measuring changes in atmospheric oxygen is technically challenging however, because of the difficulty of resolving changes at the ppm level in a background signal of almost 21% (209,000 ppm). These difficulties were first overcome by Keeling and Shertz (1992) who used a new technique to show that it is possible to track both seasonal cycles and the decline of oxygen in the atmosphere (Figure 2.3.1). Recent work by Manning and Keeling (2005) indicates that atmospheric oxygen is decreasing at a faster rate than CO_2 is increasing and demonstrates the importance of the oceanic carbon sink. Measurements of both the $^{13}C/^{12}C$ in atmospheric CO_2 and atmospheric oxygen levels are valuable tools used to determine the distribution of fossil fuel derived CO_2 amongst the active carbon reservoirs as discussed in Chapter 7. Here recent measurements in both hemispheres are shown to emphasize the strong linkages between atmospheric CO_2 increases, oxygen decreases, fossil fuel consumption and the $^{13}C/^{12}C$ ratio of atmospheric CO_2.

Annual growth rates of global atmospheric CO_2 doubled between the first measurements in the 1950s and the 1970s and closely tracked industrial CO_2 emissions (Keeling *et al.,* 1995). In the 1980s the linkage was not so clear, with inter-annual growth rates varying between about 1 and almost 2.5 ppm yr^{-1}. In the 1990s even larger variations were observed with a record high growth rate of over 3 ppm yr^{-1} observed in 1998. The average growth rate determined from the NOAA/CMDL network over the period 1979 through 2002 was 1.5 ppm yr^{-1} with the rate varying from 0.6 ppm yr^{-1} to over 3 ppm yr^{-1}, see Figure 2.3.2. The growth rate for the period 2001 to 2003 was over 2ppm yr^{-1} but dropped to 1.6 ppm yr^{-1} in 2004.

The relationship between increases in atmospheric CO_2 and fossil fuel emissions have been tracked using a scaling factor known as the "airborne fraction" (Keeling *et al.,* 1995) defined as the ratio of the annual increase in atmospheric CO_2 to annual fossil fuel CO_2 emissions. This fraction is always less than 100% because not all fossil fuel derived CO_2 remains in the atmosphere: it is partitioned between the oceans and the terrestrial biosphere as well as the atmosphere. The airborne fraction shows large variability on 2-year and smaller timescales, typically 30 to 80%. However longer term averages, for example 5-year means, show no significant change in the fraction over the last 30 years. This is despite higher than average annual increases in global CO_2 in several recent years (1998, 2002-2003) and observations of anomalously low annual increases between 1990 and 1994 (Figure 2.3.2). Thus long-term trends in the atmospheric CO_2 growth rate over decades and longer, reflect the CO_2 emission rates from fossil fuel burning whereas shorter term variations are due to fluctuations in other sources and sinks of CO_2. These include CO_2 released by land-use changes, biomass burning, and ocean warming as well as changes in the terrestrial biosphere and ocean sinks (House *et al.,* 2003; Rodenbeck *et al.,* 2003). In Figure 2.3.1 the cumulative emissions data from the CDIAC website are fitted to seasonally adjusted Mauna Loa CO_2 data assuming a constant airborne fraction of 57%. The excellent correlation between CO_2 emissions and concentrations demonstrates the strong long-term linkage with the emissions. Reported global annual fossil fuel CO_2 emissions reached a record high in 2002 at 6.975 Gt C up 2% on the 2001 emissions (Marland *et al.,* 2005). However, provisional figures for 2004 indicate even higher emissions at 7.21 Gt C. These figures represent an increase of about 50% over the last 30 years. With emissions at about 7 Gt C yr^{-1} and assuming a future airborne fraction of about 60%, Hansen and Sato (2004) predict that the underlying global atmospheric CO_2 growth rate will be about 1.9 ppm yr^{-1}.

In 2004, the global mean average CO_2 concentration for the SIO network of 9 sites was 376.57 ± 0.13 ppm and for the NOAA/CMDL network of 40 sites was 376.75 ± 0.05 ppm. The uncertainties statistically derived in different ways for each network. It should be noted that, although the agreement in 2004 was excellent between the two networks, due to the different locations of sampling sites the accord is not always as good. SIO sites are predominantly in the Pacific region whereas the extensive network of NOAA/CMDL sites is located in many parts of the world. Also the error for the SIO measurements of the annual average for Mauna Loa in 2004 was only ± 0.05 ppm. For both networks only sites in the remote marine boundary layer are used and high altitude or continental sites are not included. The Mauna Loa site for example is excluded due to an "altitude effect" of about 0.5 ppm. Also the 2004 values are still pending final reference gas calibrations used to measure the samples.

New ice core measurements of CO_2 (Etheridge *et al.,* 1996 updated) show a preindustrial global mean for CO_2 from 1700 to 1800 of 277 ± 1.2 ppm (278 ± 1.2 ppm at 1750). Their ice core record has now been extended back to 1 AD (McFarling Meure*,* 2004), and shows little change in concentrations before ~1800; there were slightly lower CO_2 concentrations in 1600–1800, most likely due to the little ice age (see Chapter 6). The RF calculations usually take 1750 as the preindustrial index (e.g., TAR and this report). Therefore using 1750 may slightly over-estimate the RF, as the changes in the concentrations of CO_2, CH_4 and N_2O after the end of this naturally cooler period may not be solely attributable to anthropogenic emissions. Using 1860 as an alternative start date for the RF calculations would reduce the LLGHG RF by ~10% (Figure 2.3.3). For the RF calculation the above data from Law Dome glacier in the Australian Antarctic Territory are used because they show the highest age resolution (~5 years) of any ice core records in existence and the N-S gradient for CO_2 is less than 1 ppm. In addition the high precision data from the cores are connected to direct observational records of atmospheric CO_2 from Cape Grim, Tasmania. RF timeseries from the combined concentration datasets are presented in Figure 2.3.3.

[INSERT FIGURE 2.3.3 HERE]

There are no updates to the RF calculation to report. The simple formula for RF of the LLGHG quoted in Ramaswamy *et al.* (2001) are still valid. A recent comparison of line-by-line and GCM radiation schemes found that clear sky instantaneous RF and surface forcing agreed very well (better than 10%) among the 5 line-by-line models investigated, using the same single atmospheric background profile. The GCM radiation schemes were less accurate, with ~20% errors in the CO_2 RF and their surface forcing calculations were unreliable (Collins *et al.,* 2005 and Chapter 10). Nevertheless, the current set of Atmosphere and Ocean GCMs (AOGCMs) used in Chapter 10 of this report found values for RF, for a doubling of CO_2 that ranged between 3.5 and 4.2 W m^{-2}, in good agreement with the TAR RF value of 3.7 W m^{-2} (see Chapter 10 and Forster*,* 2005). These full calculations involved the GCMs own global and seasonal climatology and incorporated the effects of cloud and stratospheric adjustment.

2.3.2 Atmospheric Methane (CH_4)

Methane has the second largest RF of the LLGHGs after CO_2 (TAR). Over the last half million years ice core records indicate that the abundance of methane in the earth's atmosphere has varied from lows of about 400 ppb during glacial periods to highs of about 700 ppb during inter-glacials (Petit *et al.,* 1999). Over the last two millennia measurements in air bubbles trapped in polar ice and firn show that methane levels remained below or near 700 ppb until about 1700. After this the concentration rose steadily through 750 ppb in 1800, 880 ppb in 1900 to 1714 ppb in 1992 (Etheridge *et al.,* 1998 updated).

In 2004 the global average abundance of methane measured at an extensive network of 40 surface air sampling sites operated by NOAA/CMDL in both hemispheres was 1777.60 ± 0.60 ppb. This network of sites is the most extensive operated by any laboratory and it is important to note that the calibration scale used by it has changed since the TAR (Dlugokencky *et al.,* 2005). The new scale (known as NOAA04) increases all previously reported methane concentrations from NOAA/CMDL by about 1%. The systematic error is estimated to be ~2 ppb (90%). The scale has been accepted by WMO and will be used by laboratories participating in the WMO's Global Atmospheric Watch Programme as a "common reference". Atmospheric methane is also monitored at 8 sites in the Northern and Southern Hemispheres by the AGAGE network. This group uses automated systems to make 36 methane measurements a day at each site and the mean for 2004 was 1774.6 ± 44.8 ppb with calibration and methods described by Cunnold *et al.* (2002). For the NOAA/CMDL network the 90% uncertainties are calculated with a Monte Carlo technique, which only accounts for the uncertainty due to the distribution of sampling sites. For both networks only sites in the remote marine boundary layer are used and continental sites are not included. [The 2004 values are still pending final reference gas calibrations used to measure the samples.]

The TAR reported an annual global average growth rate of 7 ppb yr^{-1} over the decade 1990 to 1999. In the early 1990s the growth rate was more than 10 ppb yr^{-1} but dropped to 5 ppb yr^{-1} or less after 1993 with the exception of the year 1998 which showed a growth rate of 11 ppb for the year. Since the TAR the growth rate decreased further with an average rate of only 0.8 ppb yr^{-1} for the 5 year period from 2000 to 2004. Two years, 2001 and 2004 showed negative growth rates and the mean for 2004 is 0.2 ppb lower than for 2003 (see Figure 2.3.4).

TAR reported several reasons for the drop in the growth rate of atmospheric methane including a decrease in its sources, an increase in its principal atmospheric sink (the OH radical) and a combination of changes in both sources and sinks. Since the TAR experimental work published by different laboratories using two different tracers for OH) shows no long term change in its global abundance over 25 and 13 year records respectively (see text below). It is therefore likely that only a reduction in the source strength of methane can be responsible for the decline in its growth rate over the past two decades.

Present atmospheric levels of methane are unprecedented in at least the last half million years. Direct atmospheric measurements of the gas made at a wide variety of sites in both hemispheres over the last 25 years show that, although the abundance of methane has increased by about 40%, its growth rate has decreased substantially during that time from highs of greater than 1% yr^{-1} in the late 1970s and early 1980s (Blake and Rowland*,* 1988) to lows of close to zero towards the end of the 1990s (Dlugokencky *et al.,* 1998; Simpson *et al.,* 2002). The slowdown in the growth rate began in the 1980s decreasing from 14 ppb yr^{-1} (about 1% yr^{-1}) in 1984 to close to zero during 1999–2001 (Dlugokencky *et al.,* 2003) for the network of surface sites maintained by NOAA/CMDL. Recent measurements by Lowe *et al.* (2004) for sites in the

Southern Hemisphere and Cunnold *et al.* (2002) for a network of GAGE/AGAGE sites show similar features. However as shown in Figure 2.3.4 the mean global growth rate at the sites operated by NOAA/CMDL increased from about zero in 2001 to 5ppb yr^{-1} during 2002 and 2003.

In TAR reasons for the decrease in atmospheric methane's growth rate and the implications for future changes in its atmospheric burden are not understood (Prather *et al.,* 2001). They are the subject of ongoing research and are clearly related to changes in the magnitude of the imbalance between methane sources and sinks. The sources of methane are poorly understood but as detailed in Chapter 7 are mostly biogenic and include wetlands, rice agriculture, biomass burning and ruminant animals with smaller contributions from industrial sources (Wang *et al.,* 2004). Most methane is removed from the atmosphere by reaction with the hydroxyl (OH) free radical which is produced photochemically in the atmosphere. Other minor sinks include reaction with free chlorine (Platt *et al.,* 2004; Allan *et al.,* 2005) and soil sinks (Born *et al.,* 1990).

Hansen *et al.* (2000) considered that economic incentives have led to a reduction in methane emissions whereas Dlugokencky *et al.* (1998) and Francey *et al.* (1999) suggest that the slowdown in the growth rate reflects a stabilization of methane emissions, given that the observations are consistent with stable emissions and lifetime since 1982. Others have argued that predicting future atmospheric burdens is impossible given recent decoupling of human population growth and emissions, uncertainties of the impact of global change on natural sources, and insufficient understanding of the causes of recent variations in the methane growth rate (Simpson *et al.,* 2002; Dlugokencky *et al.,* 2003).

Karlsdottir and Isaksen (2000) and Dentener *et al.* (2003) suggest that the recent slowdown in the methane growth rate may be due to the decreasing lifetime (strengthening sink) of the gas rather than a situation of constant sources and lifetime. On the basis of a 3-D model analysis Wang *et al.* (2004) attribute the slow down in methane emissions to a combination of slower growth in methane sources and an increase in its sinks. These include a significant reduction in the growth rate of emissions due to the economic downturn in the former Soviet Union and Eastern Europe in the 1990s. In addition they consider an increase in OH leading to greater removal of methane from the atmosphere. Recent measurements reported by Prinn *et al.* (2005a) based on analyses of methyl chloroform as a tracer for global OH imply no net change of OH over the period 1979 to 2003 with ± 6% inter-annual variations in 1989 and 1998 (see Section 2.3.5). Even more recent work by Manning *et al.* (2005) using atmospheric ^{14}CO as a tracer for global OH shows no significant long-term trend in OH from 1989 to 2003 but significant short term decreases (20%) following the Mt Pinatubo eruption in 1991 and extensive wildfires in Indonesia in 1997. Stable long-term OH concentrations indicate that the reduced source of methane is the likely cause of its decreasing long-term growth rate, shown in Figure 2.3.4. A feature of the slowdown in the growth rate of methane in the atmosphere over the last 15 years is its remarkable variability, most of which remains unexplained.

[INSERT FIGURE 2.3.4 HERE]

Relatively large anomalies occurred in the growth rate during 1991 and 1998 with peak values reaching 15 and 14 ppb yr^{-1} respectively (about 1% yr^{-1}). The anomaly in 1991 was followed by a dramatic drop in the growth rate in 1992 and has been linked with the Pinatubo volcanic eruption in June 1991 which injected large amounts of ash and SO_2 into the lower stratosphere of the tropics with subsequent impacts on tropical photochemistry and the removal of methane by the atmospheric OH radical (Bekki *et al.,* 1994; Dlugokencky *et al.,* 1996). More recently Walter *et al.* (2001a; 2001b) proposed that lower temperatures and lower precipitation in the aftermath of the Pinatubo eruption could have suppressed methane emissions from wetlands. At this time and in parallel with the growth rate anomaly in the methane concentration an anomaly was observed in methane's $^{13}C/^{12}C$ ratio at surface sites in the Southern Hemisphere. This was attributed to a decrease in emissions from an isotopically heavy source such as biomass burning (Lowe *et al.,* 1997). However as pointed out by Dlugokencky *et al.* (2001) in the case of interannual variations in methane growth it is often difficult to deconvolve the contributing effects of sources or sinks, and the real cause or causes of the 1992 methane anomaly and linkages to the 1991 Pinatubo eruption are still undetermined.

For the relatively large increase in the methane growth rate reported for 1998 Dlugokencky *et al.* (2001) suggest that wetland and boreal biomass burning sources may have contributed to the anomaly noting that 1998 was the warmest year globally since instrumental temperature records began (see Chapter 3). Langenfelds *et al.* (2002) showed that the 1998 variability was consistent with emission pulses coinciding with large biomass burning events in 1997/1998 in tropical and boreal regions and van der Werf *et al.* (2004) attribute the entire signal to increased biomass burning during the intense El Niño event of 1997/1998. However Warwick *et al.* (2002) and Lowe *et al.* (2004) show that, in the absence of changes in source and sink terms, changes in meteorology can also have a large impact on the inter-annual growth rate of atmospheric methane at least on regional scales with increased inter-hemispheric transport during La Niña events. In addition Chen and Prinn (2005) show that both the North Atlantic Oscillation and El Niño events influence year to year methane observations at Mace Head (Ireland) and Cape Matatula (Samoa).

The model results of Wang *et al.* (2004) indicate that the present slow down in the growth rate of methane may be temporary with potential for large future increases in methane concentration with a significant impact on climate and tropospheric ozone pollution if human activities continue on a business-as–usual trajectory. Lassey *et al.* (2005) examine the evolution of the methane budget since 1990 in the context of emission estimates aggregated from country inventories reported to UNFCC and the projection of these to 2020. Lassey *et al.* show that if the projections are accurate and the methane sink remains stable (see Manning *et al.,* 2005) then atmospheric methane mixing ratios will grow again approaching the growth rates of the 1970s by 2020. However this proposition takes into account only of those mitigation measures that countries are putting in place through policy enactment and not voluntary mitigation measures.

On the basis of ice core measurements of methane (Etheridge *et al.,* 1998 updated) the preindustrial global value for methane from 1700 to 1800 was 715±4 ppb (it was also 715 ± 4 ppb in 1750). This takes into account the inter-polar difference in CH_4 as measured from Greenland and Antarctic ice cores.

The RF due to changes in methane concentrations is calculated with the simplified expression given in the TAR. The change from 715 ± 4 ppb to 1774 ± 44 ppb (the average concentration from the AGAGE and CMDL networks in 2004) in the methane concentration gives a RF of 0.48 ± 0.05 W m^{-2}, ranking methane as the second most important well-mixed greenhouse gas after CO_2 (Figure 2.3.3 and Table 2.3.1). The uncertainty range in concentrations for the present day represents intra-annual variability, which is not included in the preindustrial uncertainty estimate which is solely from ice-core sampling precision. The estimate for the RF due to methane is the same as in the TAR despite the small increase in its concentration. The absorption by methane is overlapped to some extent by N_2O (taken into account in the simplified expression). Taking this overlap into account using current N_2O concentrations instead of preindustrial concentrations (as in the TAR) reduces the current RF due to methane by 1%.

There are no updates to the methane RF calculation to report, and the simple formula for RF quoted in Ramaswamy *et al.* (2001) remains valid. Collins *et al.* (2005) confirm that line by line-models agree extremely well for the calculation of clear-sky instantaneous RF from CH_4 and N_2O when the same atmospheric background profile is used. However, as was the case for CO_2, GCM radiation schemes were not found to be in such good agreement with the line-by line models, especially for the surface forcing calculation. In addition a small effect from the absorption of solar radiation was found with the line-by-line models, which the GCMs, with one exception, did not include (see discussion in Chapter 10 and Collins *et al.,* 2005).

2.3.3 Other Kyoto Protocol Gases: N_2O, HFCs, PFCs, and SF_6

At the time of the TAR N_2O was the fourth most important greenhouse gas behind CFC-12, CH_4 and CO_2. The TAR quoted an atmospheric nitrous oxide abundance of 314 ppb in 1998, an increase of 44 ppb from its preindustrial level of around 270 ± 7 ppb (Prather *et al.,* 2001), which gave a well-quantified RF of 0.15 ± 0.02 W m^{-2} (Ramaswamy *et al.,* 2001). This RF is affected by atmospheric methane levels due to overlapping absorptions. As nitrous oxide is also the major source of ozone-depleting NO and NO_2 in the stratosphere it is routinely reviewed in the ozone assessments (Montzka *et al.,* 2003). TAR pointed out large uncertainties in the major soil, agricultural, combustion and oceanic sources of N_2O. Nevertheless, its observed rate of increase of 0.2 to 0.3% per year was consistent with its better quantified major sinks (principally stratospheric destruction) (Prather *et al.,* 2001). The primary driver for the industrial-era increase of N_2O was thought to be enhanced microbial production in expanding and fertilized agricultural lands (Prather *et al.,* 2001).

Ice-core data for N_2O is now available extending by 2000 years (McFarling Meure, 2004 and Chapter 6). These data, as for CO_2 and CH_4, show little change in concentrations over the first 1800 years of this record, and then a rapid rise (seen in RF on Figure 2.3.3). Since 1998 atmospheric N_2O levels have steadily risen to around 319 ppb in 2004 and levels have now been increasing almost linearly for the past few decades (Figure 2.3.5). A change in the N_2O concentration from 270 ± 7 ppb in 1750 to 319 ± 0.4 ppb at present results in a RF of 0.16 ± 0.02 W m^{-2}. This is calculated using the simplified expression given in TAR. This RF is only slightly larger than in TAR (Table 2.3.1).

Since the TAR, understanding of regional N_2O fluxes has evolved. Kroeze *et al.* (2005) estimate N_2O emissions from rivers and estuaries to be 1.5 Tg N_2O-N yr^{-1} or about 8.5% of the global total. These are about 18% higher than in previous budgets (Kroeze *et al.,* 1999). Using inverse methods and AGAGE Ireland measurements, Manning *et al.* (2003) have estimated European Union emissions of 0.9 ± 0.2 Tg N_2O-N yr^{-1} which agree well with the United Nations FCCC inventory (0.8 ± 0.3 Tg N_2O-N yr^{-1}). Melillo *et al.* (2001) provided evidence from Brazilian land-use sequences that the conversion of tropical forest to pasture leads to an initial increase but a later decline in emissions of N_2O relative to the original forest. They also deduced that Brazilian forest soils alone contribute about 10% of total global N_2O production. Estimates of N_2O sources and sinks using observations and inverse methods had earlier implied that a large fraction of global N_2O emissions in 1978–1988 were tropical: specifically 20–29% in 0°–30°S and 32–39% in 0°–30°N compared to 11–15% in 30°S–90°S and 22–34% in 30°N–90°N (Prinn *et al.,* 1990). These estimates were uncertain due to their significant sensitivity to assumed troposphere-stratosphere exchange rates that strongly influence interhemispheric gradients. The stratosphere is also proposed to play an important role in the seasonal cycles of N_2O (Nevison *et al.,* 2004). For example, its well-defined seasonal cycle in the Southern Hemisphere has been interpreted as resulting from the net effect of seasonal oceanic outgassing of microbially-produced N_2O, stratospheric intrusion of low-N_2O air, and other processes (Nevison *et al.,* 2005). These authors also estimated a southern ocean (30°–90°S) source of 0.9 Tg N_2O-N yr^{-1} or about 5% of the global total. The complex seasonal cycle in the Northern Hemisphere is more difficult to reconcile with seasonal variations in the northern latitude soil sources and stratospheric intrusions (Prinn *et al.,* 2000; Liao *et al.,* 2004). The destruction of N_2O in the stratosphere causes enrichment of its heavier isotopomers and isotopologues providing a potential method to differentiate stratospheric and surface flux influences on tropospheric N_2O (Morgan *et al.,* 2004). As CFC-12 levels slowly decline (see Section 2.3.4), N_2O should with its current trend take over third place in the LLGHG RF list.

[INSERT FIGURE 2.3.5 HERE]

Human-made PFCs, HFCs, and SF_6 are very effective absorbers of infrared radiation, so that even small amounts of these gases contribute significantly to the RF of the climate system. The observations and global cycles of the major HFCs, PFCs and SF_6 have recently been reviewed in the IPCC-TEAP Special Report on Safeguarding the Ozone Layer and the Global Climate System or IPCC-SROC (Velders *et al.,* 2005), and here we provide therefore only a brief review and an update for these species. Table 2.3.1 shows the present concentration and recent trends in the halocarbons; it also shows RF. Absorption spectra of most halocarbons reviewed here and in the following section are characterized by strongly overlapping spectral lines that are not resolved at tropospheric pressures and temperatures. From IPCC-SROC, the intercomparison of measured cross-sections and integrated absorption intensities performed on the same molecule by different groups shows that discrepancies between different cross-section measurements can reach 40%, but the typical uncertainties when integrated over the relevant infrared spectral range are less than 10%. Apart from the uncertainties stemming from the cross-sections themselves, differences in the radiative flux calculations can arise from the spectral resolution used, tropopause heights, vertical, spatial and seasonal distributions of the gases, cloud cover, and how stratospheric temperature adjustments are performed. IPCC-SROC concludes that the discrepancy in the RF calculation for different halocarbons, associated with uncertainties in the radiative transfer calculation and the cross-sections, can reach 40%. Studies reviewed in IPCC-SROC for the more abundant HFCs (HFC-23, HFC-32, HFC-134a, HFC-227ea) show that an agreement better than 12% can be reached for these when the calculation conditions are better constrained.

The HFCs of industrial importance have lifetimes in the range 1.4–270 years. The HFCs with the largest observed mole fractions in 1998 as reported in the TAR were, in order: HFC-23 (CHF_3), HFC-134a (CF_3CH_2F) and HFC-152a (CH_3CHF_2). According to IPCC-SROC, in 2003 the observed concentrations of the major HFCs in the atmosphere were 17.5 ppt for HFC-23, 2.7 ppt for HFC-125, 26 ppt for HFC-134a, and 2.6 ppt for HFC-152a. Within the uncertainties in calibration and emissions estimates the observed concentrations of the HFCs in the atmosphere can be explained by the anthropogenic emissions, Measurements are available from CMDL (Thompson *et al.,* 2004) and AGAGE (Prinn *et al.,* 2000; O'Doherty *et al.,* 2004; Prinn *et al.,* 2005b) networks as well as UEA measurements in Tasmania (updated from Oram *et al.,* 1998; Oram*,* 1999). These data show a continuation of positive HFC trends and increasing latitudinal gradients (larger trends in the Northern Hemisphere) due to their predominantly northern hemispheric sources. The air-conditioning refrigerant, HFC-134a, is increasing at a rapid rate, in response to its growing emissions arising from its role as a replacement for some CFC refrigerants. With a lifetime of about 14 years, its budget is determined primarily by its emissions and secondarily by its atmospheric destruction. Emissions of HFC-134a estimated from atmospheric measurements are in approximate agreement with industry (AFEAS) estimates (Huang and Prinn*,* 2002; O'Doherty *et al.,* 2004). IPCC-SROC reported that global HFC-134a emissions started rapidly increasing in the early 1990s and that in Europe, sharp increases in emissions are noted for HFC-134a over 1995–1998 and for HFC-152a over 1996–2000, with some levelling off through 2003. The concentration of the foam-blower HFC-152a, with a lifetime of only about 1.5 years, is rising approximately exponentially, with the effects of its significantly increasing emissions being only partly offset by its rapid atmospheric destruction. HFC-23 has a very long atmospheric lifetime (~260 years) and is mainly produced as a by-product of HCFC-22 (CHF_2Cl) production. Its concentrations are rising approximately linearly, driven by these emissions, with its destruction being only a minor factor in its budget. There are also smaller but rising concentrations of HFC-125 (CHF_2CF_3) and HFC-143a (CH_3CF_3) which are both refrigerants.

The PFCs, mainly CF_4 (PFC-14) and C_2F_6 (PFC-116), and SF_6 have very large radiative efficiencies and lifetimes in the range 1,000–50,000 years (see Section 2.10), and make an essentially permanent contribution to RF. SF_6 and CF_4 concentrations and RF have increased by over 20% since TAR (Table 2.3.1), CF_4 concentrations have not updated. Both anthropogenic and natural sources of CF_4 are important to explain its observed atmospheric abundance. These PFCs are produced as by-products of traditional aluminium production, among other activities. PFC-14 concentrations have been increasing linearly since about 1960 and it has a natural source that accounts for about one half of its current atmospheric content (Harnisch *et al.,* 1996). Sulphur hexafluoride (SF_6) is produced for use as an insulating fluid in electrical distribution equipment and also deliberately released as an essentially inert tracer to study atmospheric and oceanic transport processes. Its concentrations were 4.2 ppt in 1998 (TAR) and continue to increase linearly over the past decade implying that emissions may be levelling off. Its very long lifetime ensures that its emissions accumulate essentially unabated in the atmosphere.

2.3.4 Montreal Protocol Gases (CFCs and HCFCs)

The Montreal Protocol for protection of the ozone layer regulates many radiatively powerful greenhouse gases for the primary purpose of lowering stratospheric chlorine and bromine concentrations. These gases include the CFCs, HCFCs, chlorocarbons, bromocarbons, and Halons. Observations and global cycles of these gases have been recently reviewed in detail in the WMO-UNEP ozone assessment (Montzka *et al.,* 2003) and IPCC-SROC. The discussion here focuses on developments since these reviews, and on those gases that contribute most to RF, more than to halogen loading. Using observed 2004 concentrations, the Montreal Protocol gases have contributed 12% (0.315 W m^{-2}) to the direct RF of all LLGHGs and 95% to the halocarbon RF (Table 2.3.1). This contribution is dominated by the CFCs. The effect of the Montreal Protocol on these gases has been substantial. IPCC-SROC concluded that the combined CO_2-equivalent emissions of CFCs, HCFCs, and HFCs decreased from a peak of about 7 $GtCO_2$-eq yr^{-1} in the late 1980s to about 2.5 $GtCO_2$-eq yr^{-1} by the year 2000, corresponding to about 10% of that year's CO_2 emission due to global fossil fuel burning.

Measurements of the CFCs and HCFCs are available from the AGAGE network (Prinn *et al.,* 2000; Prinn *et al.,* 2005b) and the CMDL network (Montzka *et al.*, 1999 updated; Thompson *et al.*, 2004) . Certain flask measurements are also available from UCI (Blake *et al.*, 2003 updated) and UEA (Oram *et al.,* 1998; Oram*,* 1999 updated). Two of the major CFCs ($CFCl_3$ or CFC-11, CF_2ClCCl_2F or CFC-113) have both been decreasing in the atmosphere since the mid-1990s. While their emissions have decreased very substantially in response to the Montreal Protocol, their long lifetimes of around 50 and 90 years respectively mean that their sinks can reduce their levels only at about 2% and 1% per year respectively. Nevertheless the effect of the Montreal protocol has been to substantially reduce the growth of the halocarbon RF which increased rapidly from 1950 until ~1990 (Figure 2.3.3). The other major CFC (CF_2Cl_2 or CFC-12), which is the third most important LLGHG, is finally reaching a plateau in its atmospheric levels (emissions equal loss) and may have peaked in 2003. Its 100-year lifetime means that it can decrease only at about 1% per year even when emissions are zero. The levelling off for CFC-12 and quasi-linear downward trends for CFC-11 and CFC-113 continue. Latitudinal gradients for all three are very small and decreasing as expected. Note that the 1998 concentrations of CFC-11 and CFC-12 were overestimated in Table 6.1 of TAR. This means that the total halocarbon RF quoted for 2004 in Table 2.3.1 (0.33 W m^{-2}) is smaller than the 0.34 W m^{-2} quoted in TAR. In fact the measurements indicate a small 3% rise in the total halocarbon RF since TAR (Figure 2.3.3 and Table 2.3.1).

The major solvent, methyl chloroform (CH_3CCl_3) is of special importance regarding RFs, not because of its small RF (see Table 2.3.1), but because this gas is widely used to estimate concentrations of OH which is the major sink species for CH_4, HFCs, and HCFCs and a major production mechanism for sulfate, nitrate and some organic aerosols as discussed in Section 2.3.5. Global atmospheric methyl chloroform concentrations rose steadily from 1978 to reach a maximum in 1992 (Prinn *et al.,* 2001; Montzka *et al.,* 2003). Since then concentrations have decreased rapidly, driven by it's relatively short lifetime of 4.9 years and phase-out under the Protocols, to levels in 2003 less than 40% of the levels when AGAGE measurements began in 1978 (Prinn *et al.,* 2005a). Emissions of CH_3CCl_3 determined from industry data (McCulloch and Midgley*,* 2001) may be too small in recent years. The 2000–2003 emissions for Europe estimated using surface observations (Reimann *et al.,* 2005), show that 1.2–2.3 Gg yr^{-1} need to be added over this 4-year period to the above industry estimates for Europe. Estimates of European emissions in 2000 exceeding 20 Gg (Krol *et al.,* 2003) are not evident from the above extensive surface data (Reimann *et al.,* 2005). Emissions for the USA have been estimated by Li *et al.* (2005). From multi year measurements they estimate 2001–2002 USA emissions of 2.2 Gg yr^{-1} (or about half of those estimated from more limited measurements by Millet and Goldstein*,* 2004), and that 1996–1998 USA emissions may be underestimated by on average about 9.0 Gg yr^{-1} over this 3-year period. East Asian emissions deduced from aircraft data in 2001 are about 1.7 Gg above industry data (Palmer *et al.,* 2003) while recent Australian and Russian emissions are negligible (Prinn *et al.,* 2001; Hurst *et al.,* 2004).

Carbon tetrachloride (CCl_4) is the second most rapidly decreasing atmospheric chlorocarbon after CH_3CCl_3. Levels peaked in early 1990 and have decreased, essentially linearly, since then. Its major use was as a feedstock for CFC manufacturing. The fact that (unlike CH_3CCl_3) a significant interhemispheric CCl_4 gradient still exists in 2004, results from a persistence of significant northern hemispheric emissions combined with its moderately long lifetime of 25–30 years.

HCFCs of industrial importance have lifetimes in the range of 1.3-20 years. Global and regional emissions of the CFCs and HCFCs have been derived from observed concentrations and can be used to check emission inventory estimates. IPCC-SROC concluded that global emissions of HCFC-22 have risen steadily over the period 1975–2000 whilst those of HCFC-141b and HCFC-142b started increasing quickly in the early 1990s.

2.3.5 Trends in the Hydroxyl Free Radical (OH)

The hydroxyl free radical (OH) is the major oxidizing chemical in the atmosphere destroying about 3.7 Pg of trace gases each year (Ehhalt*,* 1999). It has a very significant role ameliorating the LLGHG RF (Section 2.3.6). IPCC-SROC concluded that the OH concentration might change in the 21st century by –18 to +5% depending on the emission scenario. The large-scale concentrations and long-term trends in OH can be measured indirectly using global measurements of trace gases whose emissions are well known and whose primary sink is OH. The best trace gas used to date for this purpose CH_3CCl_3 whose long-term measurements are reviewed in Section 2.3.4. Other gases which are useful OH indicators include ^{14}CO, which is produced primarily by cosmic rays (Quay *et al.,* 2000). While the accuracy of the ^{14}CO cosmic ray and other source estimates, and also the frequency and spatial coverage of its measurements, do not match those for CH_3CCl_3, its lifetime (2 months) is much shorter than that of CH_3CCl_3 (4.9 years). As a result it provides estimates of average concentrations of OH that are more regional than those estimated from CH_3CCl_3. Another useful gas is the industrial chemical HCFC-22. It yields OH concentrations similar to those derived from CH_3CCl_3 but with less accuracy due to greater uncertainties in emissions and less extensive measurements (Miller *et al.,* 1998). The industrial gases CH_2FCF_3 (HFC-134a), CH_3CCl_2F

(HCFC-141b) and CH_3CClF_2 (HCFC-142b) are potentially useful OH estimators but the accuracy of their emission estimates needs improvement (Huang and Prinn*,* 2002; O'Doherty *et al.,* 2004).

Indirect measurements of OH using CH_3CCl_3 have established that the global weighted average OH concentration in the troposphere is ~10^6 radicals cm^{-3} (Prinn *et al.,* 2001; Krol and Lelieveld*,* 2003). A similar average concentration is derived using ^{14}CO (Quay *et al.,* 2000) although the weighting here is different. While the average OH concentration appears fairly well defined by these indirect methods, the temporal trends in OH are more difficult to discern since they require long-term measurements, optimal inverse methods, and very accurate calibrations, model transports, and CH_3CCl_3 emissions data. From AGAGE CH_3CCl_3 measurements, Prinn *et al.* (2001) deduced that global OH levels grew between 1979 and 1989, but then declined between 1989 and 2000, and also exhibited significant interannual variations. They concluded that these decadal global variations were driven principally by northern hemispheric OH, with southern hemispheric OH decreasing from 1979–1989 and staying essentially constant after that. Using the same AGAGE data and identical emissions, a 3D model analysis (Krol and Lelieveld*,* 2003) confirmed qualitatively (but not quantitatively) the earlier result (Prinn *et al.,* 2001) that OH concentrations increased in the 1980s and declined in the 1990s. Prinn *et al.* (2001) also estimated the emissions required to provide a zero trend in OH. These required emissions differed substantially from industry estimates by McCulloch and Midgley (2001) particularly for 1996–2000. Krol and Lelieveld (2003) however argued that the combination of possible underestimated recent emissions, especially the >20 Gg European emissions deduced by Krol *et al.* (2003), and the recent decreasing effectiveness of the stratosphere as a sink for tropospheric CH_3CCl_3, may be sufficient to yield a zero deduced OH trend. As discussed in Section 2.3.4, estimates of European emissions by Reimann *et al.* (2005) are an order of magnitude less than those of Krol *et al.* (2003). Also, Prinn *et al.* (2005b) extend the OH estimates through 2004 and show that the Prinn *et al.* (2001) decadal and interannual OH estimates remain valid even after accounting for the additional recent CH_3CCl_3 emissions discussed in Section 2.3.4. They also reconfirm the OH maximum around 1989 and a larger OH minimum around 1998 with OH concentrations then recovering so that in 2003 they are comparable to those in 1979. They note that the 1997–1999 OH minimum coincides with, and is likely caused by, major global wildfires and an intense El Niño at this time. The 1997 Indonesian fires alone have been estimated to have lowered global late-1997 OH levels by 6% (Duncan *et al.,* 2003).

Methyl chloroform is also destroyed in the stratosphere. Because its stratospheric loss frequency is less than that in the troposphere, the stratosphere becomes a less effective sink for tropospheric CH_3CCl_3 over time (Krol and Lelieveld*,* 2003), and even becomes a small source to the troposphere beginning in 1999 in the reference case in the Prinn *et al.* (2001; 2005a) model. Loss to the ocean has usually been considered irreversible, and its rates and uncertainties have been obtained from observations (Yvon-Lewis and Butler*,* 2002). However, Wennberg *et al.* (2004) have recently proposed that the polar oceans may have effectively stored methyl chloroform during the pre-1992 years when its atmospheric levels were rising, but began re-emitting it in the subsequent years, thus lessening the overall oceanic sink. Prinn *et al.* (2005a) tried both approaches and found that their inferred interannual and decadal OH variations were present using either formulation, but inferred OH was lower in the pre-1992 years and higher after that using the Wennberg *et al.* (2004) formulation.

More recently, Bousquet *et al.* (2005) have used an inverse method with a 3D model and methyl chloroform measurements and concluded that substantial year-to-year variations occurred in global-average OH concentrations between 1980 and 2000. This conclusion was previously reached by Prinn *et al.* (2001), but subsequently challenged by Krol and Lelieveld (2003) who argued that these variations are caused by model shortcomings and that models need, in particular, to include observationally-based, interannually-varying meteorology to provide accurate annual OH estimates. However, the Bousquet *et al.* (2005) analysis, which uses observationally-based meteorology and estimates OH on monthly time scales, yields interannual OH variations that agree remarkably well with the Prinn *et al.* (2001) and equivalent Krol and Lelieveld (2003) estimates (see Figure 2.3.6). But neither the 2D Prinn *et al.* (2001) or the 3D Krol *et al.* (2003) inversion models used interannually-varying circulation. This implies that these interannual OH variations are in fact real, and from the Bousquet *et al.* (2005) sensitivity studies, their phasing in particular appears robust. Bousquet *et al.* (2005) also deduced that OH in the Southern Hemisphere shows a zero to small negative OH trend in qualitative agreement with Prinn *et al.* (2001).

[INSERT FIGURE 2.3.6 HERE]

Short-term variations in OH have also recently been deduced by Manning *et al.* (2005) using 13 years of ^{14}CO measurements in New Zealand and Antarctica. They find no significant long-term trend between 1989 and 2003 in southern hemispheric OH but provide evidence for recurring multi-month OH variations of around 10%. They also deduce even larger (20%) OH decreases in 1991 and 1997, perhaps triggered by the 1991 Pinatubo eruption and the 1997 Indonesian fires. The similarity of many of these results to those from CH_3CCl_3 discussed above is very important, given the independence of the two approaches. This evidence for substantial OH variability obtained from both CH_3CCl_3 and ^{14}CO is not mirrored in current global atmospheric chemistry models.

2.3.6 Annually Input Radiative Forcing

The RF from a LLGHG and its time evolution are determined by both the emission and the sink terms for the gas (see Sections 2.3.1–2.3.5). Manning (2005) introduced the concept of Annually Input Radiative Forcing for decomposing a RF timeseries of a LLGHG into two terms: the RF associated with emissions and the RF associated with the destruction of the LLGHG each year (Figure 2.3.7). This method provides useful insights into the causes of trends of the LLGHG RF and enables a useful comparison of the various positive and negative LLGHG RF contributions.

[INSERT FIGURE 2.3.7 HERE]

Input Radiative Forcing values in Figure 2.3.7 clearly show that CO_2 emissions are adding more and more RF each year; it also shows that CH_4 adds a comparable amount of RF to the climate system every year. However, the net RF change for CH_4 is much smaller than for CO_2 because CH_4 is almost completely removed by its sink terms, principally OH. These CH_4 sinks have been growing faster than the growth in emissions (Section 2.3.5) and, thus the CH_4 RF currently exhibits a very small net growth (see also Section 2.3.2). For all the LLGHGs a large fraction of the input radiative forcing introduced each year is removed by the sink terms. Manning (2005) also uses the Input Radiative Forcing approach to show the long-term RF commitment for individual gases based on past emissions.

2.3.7 Ozone

2.3.7.1 Stratospheric ozone

A number of recent reports have assessed changes in stratospheric ozone and the research into its causes; Chapter 3 and 4 of the 2003 WMO assessment (WMO*,* 2003) and Chapter 1 of the IPCC special report on ozone and climate (Pyle *et al.,* 2005). This section summarizes the material from these reports and updates the key results using more recent research.

An updated time series of deseasonalized global mean column ozone anomalies from is displayed in Figure 2.3.8. The changes in ozone derived from ground-based measurements (1964–2003) as well as satellite data sets (1979–2003) are in good agreement and we therefore have reasonable confidence in their quality for describing the spatial and temporal characteristics of past ozone changes. Furthermore, it has been shown (Shindell and Faluvegi*,* 2002) that stratospheric ozone during 1957 to 1975 was lower by 7.2 ± 2.3 DU relative to the first half of the 20th century as a result of water vapour increases. In the Northern Hemisphere these stratospheric ozone decreases are offset by tropospheric increases of 8.2 ± 2.1 DU so that total column ozone values are essentially unaffected, as supported by long-term measurements at Arosa, Switzerland (Staehelin *et al.,* 1998).

The global ozone amounts decrease between the late 1970s and early 1990s, show lowest value during 1992–1993 (6% below the 1964–1980 average), and slightly increasing values thereafter. Global ozone for the period 2000 to 2003 was approximately 4% below the 1964–1980 average values. Whether or not recently observed changes in ozone trends (Newchurch *et al.,* 2003) are already indicative of recovery of the global ozone layer is not yet clear and requires more detailed attribution of the drivers of the changes (Steinbrecht *et al.,* 2004; Hadjinicolaou *et al.,* 2005).

[INSERT FIGURE 2.3.8]

The stratospheric changes have been spatially, temporally and seasonally non-uniform, which has important implications for resulting RF. Ozone decreases have been driven primarily by changes in the lower stratospheric extratropics. Total column ozone changes over the midlatitudes of the Southern Hemisphere are significantly larger than over the midlatitudes of the Northern Hemisphere. Averaged over the period 2000–2003, Southern Hemisphere values are 6% below pre-1980 values, while Northern Hemisphere values are 3% lower. There is also significant seasonality in the Northern Hemisphere ozone changes with 4% decreases in winter to spring and 2% decreases in summer, while long-term Southern Hemisphere changes are similar at ~6% year round (WMO*,* 2003). Southern hemisphere mid-latitude ozone shows significant decreases during the mid-1980s and essentially no response to the Mt. Pinatubo volcanic eruption in June 1991; both of these features remain unexplained. Pyle *et al.* (2005) and Chipperfield *et al.* (2003) assess several studies which show that a substantial fraction (~30%) of Northern Hemisphere midlatitude ozone trends are not directly attributable to anthropogenic chemistry, but are related to dynamical effects, such as tropopause height changes. These dynamical effects are likely to have contributed a larger fraction of the ozone forcing in the Northern Hemisphere midlatitudes. The only study to assess this finds that 50% of the RF between 20ºN–60ºN is attributable to dynamics (Forster and Tourpali*,* 2001). These dynamical feedbacks may well have an anthropogenic cause and could even be partly caused by stratospheric ozone depletion itself (Chipperfield *et al.,* 2003; Santer *et al.,* 2004) but are not directly related to chemistry. Stratospheric ozone forcing estimates based on observed trends thus become more of an upper bound to the stratospheric ozone RF. One study since TAR has estimated a global stratospheric ozone RF. Hansen *et al.* (2005) calculate both total ozone and tropospheric ozone RF. The difference between these two RFs is small and would imply a smaller stratospheric ozone RF than indicated in TAR roughly –0.06 W m^{-2}. Their ozone changes were based on Randel and Wu (1999). TAR quotes a RF of -0.10 W m^{-2} from a different model using the same trend dataset. In light of this study and the accepted veracity of their employed trend data the central estimate of stratospheric ozone forcing has been slightly revised to a weaker value of –0.1 ± 0.04 W m^{-2}; trends in RF since 1998 are not known.

The largest ozone changes since 1980 have occurred during the late winter and spring over Antarctica where average total column ozone in September and October is about 40–50% below pre-1980 values, with up to a local 70% for periods of a week or so (WMO*,* 2003). This seasonal phenomenon removes approximately 8×10^{10} kg of ozone from the Antarctic stratosphere and is known as the Antarctic ozone hole. The ozone hole now forms every spring, although with some interannual variability driven primarily by midlatitude dynamical activity (Bodeker and Scourfield*,* 1995; Newman and Nash*,* 2004). In recent years ozone concentrations have dropped close to zero between 12 and 20 km altitude (Kröger *et al.,* 2003).

Ozone decreases over the Arctic have been less severe than those over the Antarctic. Artic stratospheric ozone levels are more variable due to interannual variability in chemical loss and dynamical transport. Dynamical variability in the winter stratosphere changes the transport of ozone to high latitudes. The same processes driving this transport affect Arctic polar stratospheric temperatures – when transport is weak the stratosphere is colder. Lower temperatures accelerate the ozone depletion chemistry. As a result, in recent decades, halogen induced polar ozone chemistry has acted in concert with dynamically induced ozone variability, and has led to column ozone losses of up to 30% in particularly cold winters (WMO*,* 2003). In dynamically active warm winters, the estimated chemical ozone loss is very small.

2.3.7.2 Tropospheric ozone

The TAR report identified large regional differences in observed trends in tropospheric ozone from ozonesondes (Logan *et al.,* 1999) and surface observations (Oltmans *et al.,* 1998). Based on the CTM model results driven by emission changes since preindustrial times TAR gave a best estimate of RF from tropospheric ozone change of 0.35 ± 0.15 W m^{-2}. The results from the OXCOMP model exercise performed for the TAR have been published, more detail in Gauss *et al.* (2003).

Trends in anthropogenic emissions of ozone precursors for the period 1990–2000 have been compiled by the Emission Database for Global Atmospheric Research (EDGAR) consortium (Olivier and Berdowski*,* 2001 updated). For specific regions there is a significant variability over the period due to variations in the emissions from open biomass burning sources. For all components (NOx, CO and VOCs) industrialized regions like USA and OECD Europe show reductions in the emissions, while regions dominated by developing countries show significant growth in the emissions.

Trends in tropospheric ozone at northern mid- and high latitudes have been estimated based on ozonesonde data by WMO (2003), Naja *et al.* (2003), Naja and Akimoto (2004), and Tarasick *et al.* (2005). The observed trends depend strongly on the region. For the European stations a growth in free tropospheric ozone was observed until the late 1980s, while in the later period the trend has levelled off or been slightly negative (Naja *et al.,* 2003; WMO*,* 2003). Naja and Akimoto (2004) analysed 33 years of ozonesonde data from Japanese stations showing an increase in ozone in the lower troposphere (750–550 hPa) between the periods 1970–1985 and 1986–2002 of 12–15% at Sapporo and Tsukuba (43° and 36°N) and 35% at Kagoshima (32ºN). Trajectory analysis indicates that the more southerly located station Kagoshima is significantly more influenced by air originating over China, while Sapporo and Tsukuba is more influenced by air from Eurasia. Tarasick *et al.* (2005) used ozonesonde data for the period 1980–2001 for Canadian stations and show negative trends of tropospheric ozone between 1980 and 1990, and a rebound with positive trends during 1991–2001. Analysis of stratosphere-troposphere exchange (STE) processes indicates that the rebound during the 1990s may be partly a result of small changes in the atmospheric circulation.

In the tropics very few long-term ozonesonde measurements are available. Thompson *et al.* (2001) have published ozone trends based on TOMS satellite data finding no significant trend in tropical tropospheric ozone (between 12ºN and 12ºS) during 1979–1992.

Trend analysis of ozone concentrations from surface observations shows just as much regional variation as the sonde data. Jaffe *et al.* (2003), derived a positive trend of 0.54 ppb yr^{-1} (about 1.4% yr^{-1}) using a 15 year record from Lassen Volcanic Park in northern California (1750 m ASL) and a consistent 0.51 ppb yr^{-1} comparing two aircraft campaigns CITE-1C (1984) and ITCT-2K2 (2002). At Mace Head (Ireland) Simmonds *et al.*(2004) found a positive trend of 0.49 ± 0.19 ppb yr^{-1} for the period 1987–2003. Trends over the Atlantic Ocean have been derived from ship observations for the period 1977–2002 by Lelieveld *et al.*(2004). Significant upward trends were found between 40ºS and 40ºN (0.12–0.68 ppbv yr^{-1}, depending on the region). Between 40ºN and 60ºN a much lower trend (0.05 ppb yr^{-1}) was found than the trend observed at Mace Head.

Since TAR there have been major developments of the models. The new model generations include several chemical tracer models (CTM) which couple stratospheric and tropospheric chemistry, as well as GCMs with online chemistry (both tropospheric and stratospheric). Inclusion of stratospheric chemistry in the models means that tropospheric ozone is affected by the reduction of the STE estimates, as the stratospheric ozone concentrations are reduced due to emissions of ozone depleting substances (Gauss *et al.,* 2005).

Changes in tropospheric ozone and the corresponding RF have been estimated by a number of recent model studies (Mickley *et al.,* 2001; Shindell *et al.,* 2003a; Mickley *et al.,* 2004; Wong *et al.,* 2004; Liao and Seinfeld*,* 2005; Shindell *et al.,* 2005). In addition, a modelling exercise was organized through the ACCENT Network (Gauss *et al.,* 2005). In the ACCENT calculations the preindustrial emissions were specified while the models used their best estimates for the current emissions. Adjusted RF for all models was calculated by the same radiative transfer model. Four of the ten models in the model exercise have detailed stratospheric chemistry included. Six GCMs with on-line chemistry participated in an additional experiment where the effect of climate change was included in the calculations.

A general feature with almost all models is their inability to reproduce the low ozone concentrations observed during the late 19th century (Mickley *et al.,* 2001; Wong *et al.,* 2004; Lamarque *et al.,* 2005). Mickley *et al.* (2001) performed sensitivity tests reducing the lightning and soil sources of NOx and increasing natural non-methane volatile organic compound (NMVOC) emissions to obtain better agreement with 19th century ozone observations. The resulting RF of tropospheric ozone then increased by 50–80% compared to the standard assumptions about preindustrial emissions. However, there are several aspects of the early observations that were not captured by the model tests, so the uncertainty of the large-scale ozone distribution during the preindustrial time remains large.

RF estimates of tropospheric ozone increase since 1750 are given in Figure 2.3.9. For most of the calculations the same set of assumptions about preindustrial emission inventories have been used. All anthropogenic emissions of ozone precursors are set to zero and emissions from biomass burning sources were reduced by 90%. Emissions of NOx from soils and biogenic hydrocarbons are assumed to be natural and are thus not changed, although there probably has been some increase in these sources, due to increased use of fertilizers in the case of soil NOx and changes in biogenic HC due to land use changes.

[INSERT FIGURE 2.3.9 HERE]

The uncertainties in the estimated RF by tropospheric ozone, originates from several factors: The models used (CTM/GCM model formulation, radiative transfer models), lack of consistency between observed and calculated preindustrial ozone levels, and potential impact of climate change on tropospheric ozone. In addition the models that include stratospheric chemistry a significant reduction in the tropospheric ozone has been modelled at high latitudes as a result of decline in the stratospheric ozone and this affected the range of results. We assume that the range of results, for the model simulations using the standard emissions and neglecting the effects of climate change, represents the model uncertainty. This range was 0.26–0.53 W m^{-2} (with a mean of 0.37 and a standard deviation of 0.06 W m^{-2}).

The uncertainty due to our lack of understanding of the photochemical state of the preindustrial troposphere is more difficult to quantify, but the results of Mickley *et al.* (2001) indicate that this could be substantial (0.3 W m^{-2}). Mickley *et al.* (2001) tune their model to reproduce preindustrial observations by reducing the pre industrial lightning source of NOx. The observations are all from the ground, although the Pic du Midi data (Marenco *et al.,* 1994) are from 3000 m altitude, while NOx from lightning does increase ozone at higher altitudes where it has a larger effect on the RF. It is also possible that part of this is not strictly a forcing but represents a climate feedback. An uncertainty which can increases the upper RF estimate by 0.2 W m^{-2} is assumed from this. The effect of climate change appears to be relatively small, enhancing the RF between 0 and 0.11 W m^{-2} compared to the standard case (see Figure 2.3.9). A 90% value uncertainty of 0.05 W m^{-2} is assumed. However, the impact of climate change on tropospheric ozone is likely to be more feedback mechanism than RF. Allowing for this structural uncertainty, RF from tropospheric ozone is taken to be 0.4 ± 0.15 W m^{-2}.

2.3.8 Anthropogenic Water Vapour

2.3.8.1 Stratospheric water vapour

TAR noted that several studies had indicated long-term increases in stratospheric water vapour and acknowledged that these trends would contribute a significant radiative impact. However, it only considered the stratospheric water vapour increase expected from methane increases as a “radiative forcing” and this was estimated to contribute only 2–5% of the total CH_4 RF (~0.02 W m^{-2}).

There remains limited evidence for an increase in global stratospheric water vapour since 1980, trends generally remain uncertain and are poorly understood; it is also likely that there are several contributing factors to the long-term change. The stratospheric water vapour RF associated with methane changes is estimated by two studies to be ~0.1 W m^{-2}, which is 5–10 times higher than that quoted in TAR. Direct injection of water vapour form aviation has an insignificant RF. RF from other proposed mechanisms are not sufficiently understood to be evaluated. Therefore the estimate of stratospheric water vapour RF is still only from CH_4 oxidation: 0.1 W m^{-2} is given as a best estimate, with a factor of three uncertainty.

There are now 14 years of global stratospheric water vapour measurements from HALOE and continued balloon-based measurements (since 1980) at Boulder, Colorado. Several new analyses have been carried out with these data. There is some limited evidence from direct observations of a sustained long term increase in stratospheric water vapour. There is also some indirect evidence of a long-term stratospheric water vapour increase in the lower stratosphere from the observed temperature changes over the last few decades, until 2000. Increasing stratospheric water vapour would be expected to cool the lower stratosphere and the observed cooling appears slightly larger than what can be accounted for by other factors (Chipperfield *et al.,* 2003; Pyle *et al.,* 2005). However, recent observations show that water vapour concentrations in the lower stratosphere have certainly been decreasing since 2000 (Chapter 3, Section 3.4.2.4). Therefore any longer term increase may not be sustained.

Several mechanisms have been proposed to explain the long-term changes in stratospheric water vapour (see Chapter 3, Section 3.4.2.4 for more details). TAR considered the stratospheric water vapour changes associated with CH_4 as an indirect RF. Some other mechanisms can also be considered as a forcing, other mechanisms are more associated with climate feedback. It is likely that different mechanisms are affecting water vapour trends at different altitudes. Aviation gives a direct RF by emitting water vapour directly into the stratosphere. Several indirect mechanisms have been discussed, including: a) volcanic eruptions (Considine *et al.,* 2001; Joshi and Shine*,* 2003) b) biomass burning aerosol (Sherwood*,* 2002); c) tropospheric SO_2 (Notholt *et al.,* 2005) and d) changes to methane oxidation rates from changes in stratospheric chlorine, ozone and OH (Rockmann *et al.,* 2004). Other proposed mechanisms relate to changes in tropopause temperatures or circulation (Stuber *et al.,* 2001b; Fueglistaler *et al.,* 2004).

Since TAR several further calculations of the radiative balance change due to changes in stratospheric water vapour have been performed (Forster and Shine*,* 1999; Oinas *et al.,* 2001; Shindell*,* 2001; Smith *et al.,* 2001; Forster and Shine*,* 2002). Smith *et al.* (2001) used estimated a 0.12 to 0.2 W m^{-2} per decade range for the RF from the change in stratospheric water vapour, using HALOE satellite data. Shindell (2001) used a GCM to estimate the increase in water vapour in the stratosphere from oxidation of CH_4 and from an increase in greenhouse gases and estimated an RF of about 0.2 W m^{-2} in a period of two decades. Forster and Shine (2002) used a constant 0.05 ppmv yr^{-1} trend of water vapour at pressures of 100–10 hPa and estimated RF to be 0.29 W m^{-2} over 1980 to 2000. Hansen and Sato (2001) and Hansen *et al.* (2005) estimated a RF of 0.1 W m^{-2} in water vapour change from oxidation of CH_4 since preindustrial times based on a two-dimensional chemistry transport model. These two estimates are 5–10 times higher than values reported in TAR and they are taken as the best estimate of water vapour change from CH_4 oxidation. The RF from direct injection of water vapour by aircraft is believed to be insignificant ~0.002 W m^{-2} (IPCC*,* 1999).

2.3.8.2 Tropospheric water vapour from anthropogenic sources

Anthropogenic use of water is less than 1% of natural sources of water vapour and about 70% of the use of water for human activity is from irrigation (Döll*,* 2002). Several regional studies have indicated an impact of irrigation on temperature, humidity, and precipitation (Barnston and Schickedanz*,* 1984; Lohar and Pal*,* 1995; de Ridder and Gallée*,* 1998; Moore and Rojstaczer*,* 2001; Jianping *et al.,* 2002). Boucher *et al.* (2004) used a GCM to show that irrigation has a global impact on temperature and humidity. Over Asia where most of the irrigation takes place the simulations showed a change in the water vapour content in the lower troposphere by up to a 1%, resulting in an RF of 0.03 W m^{-2}. The effect of water vapour increases on evaporative cooling exceeds that of its greenhouse warming effect and a decrease in surface temperature was found. Uncertainties in the water vapour flow to the atmosphere from irrigation are significant and Gordon *et al.* (2005) give a substantially higher estimate compared to that of Boucher *et al.* (2004). The Gordon *et al.* (2005) study estimates a reduced water vapour flow to the atmosphere from deforestation, most important in tropical areas. This reduced water vapour flow is a factor of 3 larger in magnitude compared to the water vapour increase due to irrigation in Boucher *et al.* (2004), but so far no estimates exist how this change affects the water vapour content of the atmosphere and its RF. The emission of water vapour from fossil fuel combustion is significantly lower than the emission from agricultural activity.

2.3.9 Observations of Long-Lived Greenhouse Gas Radiative Forcing

Harries *et al.* (2001) analyzed spectra of the outgoing longwave radiation as measured by two satellites in 1970 and 1997 over the tropical Pacific Ocean. The reduced brightness temperature observed in the spectral regions of many of the greenhouse gases is an experimental evidence for an increase in the Earth's greenhouse effect. In particular the spectral signature was large for CO_2 and CH_4, but also the halocarbons with the largest change between 1970 and 1997 had an impact on the brightness temperature. Philipona *et al.* (2004) found an increase in the measured longwave downward radiation at the surface over the period from 1995 to 2002 at eight stations over the central Alps. A significant increase in the clear-sky longwave downward flux was found to be due to an enhanced greenhouse effect after combining the measurements with model calculations to estimate the contribution from increases in temperature and humidity.

2.4 Aerosols

2.4.1 Introduction

TAR categorised the mechanisms by which anthropogenic aerosols exert RF into the direct effect and the indirect effect.

The direct effect is the mechanism by which aerosols scatter and absorb shortwave and longwave radiation, thereby altering the radiative balance of the Earth-atmosphere system. Sulphate, fossil-fuel organic and black carbon, biomass burning, and mineral dust aerosols were all identified in TAR as significant forcing agents for the direct aerosol effect. Key parameters for determining the direct effect are the aerosol optical properties (the single scattering albedo, ω_o, specific extinction coefficient, k_e, and scattering phase function or asymmetry factor, g) which vary as a function of wavelength and relative humidity, and the geographic distribution of the aerosols in the horizontal and vertical which varies as a function of time (e.g., Haywood and Boucher, 2000; Penner *et al.,* 2001; Ramaswamy *et al.,* 2001). Scattering aerosols will exert a negative direct RF, while absorbing aerosols may exert a negative top of the atmosphere forcing over dark surfaces such as oceans or dark forest surfaces, and a positive top of the atmosphere forcing over bright surfaces such as desert, snow/ice or if the aerosol is above cloud (e.g., Chylek and Wong, 1995; e.g., Haywood and Shine, 1995). Both positive and negative top of the atmosphere forcing mechanisms reduce the irradiance at the surface thereby modifying the surface radiation budget. The long-wave direct radiative effect is generally of significant magnitude only if the aerosol particles are large and occur in substantial concentrations at higher altitudes (e.g., Tegen *et al.,* 1996).

The indirect effect is the mechanism by which aerosols modify the microphysical and hence the radiative properties, cloud amount and lifetime of clouds. Key parameters for determining the indirect effect are the effectiveness of an aerosol particle to act as a cloud condensation nucleus (CCN), which is a function of the size, chemical composition, mixing state and geographic distribution (e.g., Penner *et al.,* 2001) . TAR split the indirect effect into the first indirect effect (i.e., the microphysically induced effect on the cloud droplet number concentration, and hence the cloud droplet size with the liquid water content held fixed), and the second indirect effect (i.e., the microphysically induced effect on the liquid water content, cloud height, and lifetime of clouds). The terms “cloud albedo effect” and “cloud lifetime effects” are used throughout this report because they are more representative of the microphysical processes that occur when anthropogenic aerosols interact with clouds. The cloud albedo effect was considered in TAR to be a RF because global model calculations could be performed of the influence of increased aerosol concentration on the cloud optical properties while holding the liquid water content of the cloud fixed, i.e., in an entirely diagnostic procedure where feedback mechanisms do not occur. TAR considered the cloud albedo effect to be a key uncertainty in the RF of climate but did not assign a best estimate of the RF, and showed a range of RF between 0 and –2 W m^{-2}. The other indirect effects were not considered to be a RF because, in suppressing drizzle, increasing the cloud height, or the cloud lifetime in atmospheric models (Figure 2.4.1), the hydrological cycle is necessarily altered i.e., feedback mechanisms do occur. TAR also discussed the impact of anthropogenic aerosols in the formation and modification of the physical and radiative properties of ice clouds (Penner *et al.,* 2001), although quantification of a RF from this mechanism was not considered appropriate given the host of uncertainties and unknowns surrounding ice cloud nucleation and physics.

[INSERT FIGURE 2.4.1 HERE]

2.4.2 Advances since the Third Assessment Report

Since TAR there has been much further research into tropospheric aerosols, their physical and radiative properties and their effects on climate via direct and indirect RFs in both in-situ and remote sensing observations and modelling, and the following advances should be highlighted:

Observations:

- Improved accuracy of satellite observations via dedicated retrievals.
- Longer satellite records of aerosol.
- Improved retrievals of aerosol properties from surface-based sun-photometers and lidar.
- A greater number of field campaigns and case studies using improved instrumentation.
- Further work on emissions and trends.
- More focus on aerosol optical parameters, particularly the single scattering albedo.
- Further studies of the aerosol indirect effects in clouds other than stratocumulus clouds.
- Further studies of the effects of anthropogenic aerosol on modifications of ice clouds.

Modelling:

- Improved models that now contain all major aerosol species.

- Improved model parameterisations, e.g. the hygroscopicity and absorption of aerosol mixtures.
- Improvements in modelling of aerosol vertical profiles.
- Modelling nitrate aerosol.
- Improved modelling of the cloud albedo indirect effect leading to a global mean best estimate.
- Inclusion of the effects of aerosol chemical composition on cloud-droplet broadening.

2.4.3 Advances in Observations

As satellite observations and surface-based retrievals are capable of providing near-global coverage they are discussed in detail in this sub-section; the advances in emissions estimates, trends, and in-situ measurements of the physical and optical properties are discussed with respect to their influence on RF in Section 2.4.5. Further detailed discussions of the recent observations performed into aerosol in-situ physical and optical properties of aerosols and a measurement based assessment of the aerosol direct RF are given by Yu *et al.* (2005).

2.4.3.1 Satellite retrievals

Products of satellite retrievals such as the aerosol optical depth, τ_{aer}, Ångstrom coefficient, and direct radiative effect (i.e., natural and anthropogenic) in the absence of clouds have all been developed (Kaufman *et al.,* 2002) and are invaluable for constraining the global models used to assess the direct and indirect RF of anthropogenic aerosols (see Section 2.4.5).

2.4.3.1.1 Aerosol optical depth, τ_{aer}

Figure 2.4.2 shows an example of τ_{aer} retrieved over both land and ocean together with the geographical positions of aerosol instrumentation and dedicated field campaigns since 1996. Table 2.4.1 provides a summary of aerosol data currently available from satellite instrumentation, together with acronyms for the instruments. The spatial distribution of the τ_{aer} from the MODIS instrument for January, February and March (JFM, Figure 2.4.2a) clearly differs from that for August, September and October (ASO, Figure 2.4.2b) for 2001 (Kaufman *et al.,* 1997; Tanré *et al.,* 1997). The seasonal variability in the τ_{aer} can readily be seen; biomass-burning aerosol is most strongly evident over the Gulf of Guinea in JFM but shifts to southern Africa in ASO. Likewise the biomass burning in South America is most evident in Figure 2.4.2b. In Figure 2.4.2a the transport of mineral dust from Africa to the South American continent is clearly discernible while in Figure 2.4.2b the mineral dust is transported over the West Indies and Central America. Industrial aerosol which consists of a mixture of sulphates, organic and black carbon, nitrates, and industrial dust is clearly visible over many continental regions of the Northern Hemisphere. Sea-salt aerosol is visible in regions where the windspeed is high (e.g., south of 45ºS). There are several regions where the MODIS instrument cannot perform retrievals; these areas include high latitudes when the solar insolation is insufficient, and highly reflectant areas such as deserts and snow surfaces.

[INSERT TABLE 2.4.1 HERE]

[INSERT FIGURE 2.4.2 HERE]

Early retrievals for estimating τ_{aer} include the single channel retrieval of the AVHRR) (e.g., Ignatov and Stowe*,* 2002), and the UV based retrieval from the TOMS (e.g., Torres *et al.,* 2002). A dual-channel AVHRR retrieval has also been developed (e.g., Mishchenko *et al.,* 1999; Geogdzhayev *et al.,* 2002). The AVHRR retrievals are generally only performed over ocean surfaces where the surface reflectance characteristics are relatively well known, although retrievals are also possible over dark land surfaces such as boreal forests and lakes (Soufflet *et al.,* 1997). The OCTS retrieval has a similar basis to the dual wavelength retrieval from AVHRR and uses wavelengths over the range 0.41–0.86 µm to derive τ_{aer} and, Å, over oceans (e.g., Higurashi *et al.,* 2000) using a bi-modal aerosol size distribution. The TOMS retrieval is essentially independent of surface reflectance thereby allowing retrievals over both land and ocean (Torres *et al.,* 2002), but only works for UV absorbing aerosols and is sensitive to the altitude of the aerosol. While these retrievals only use a limited number of spectral bands and lack sophistication compared to those from those from dedicated satellite instruments, they have the advantage of offering continuous long-term data sets (e.g., Geogdzhayev *et al.,* 2002).

These early retrievals have been superseded by those from dedicated aerosol instruments (e.g., Kaufman *et al.,* 2002). The first instrument designed specifically for aerosol measurements, POLDER, uses a combination of spectral channels (0.44–0.91 μm) with several viewing angles, and measures polarization of radiation. τ_{aer} and Ångstrom coefficients over ocean (Deuzé *et al.,* 2000), τ_{aer} over land (Deuzé *et al.,* 2001), and the direct radiative effect of aerosols (Boucher and Tanre*,* 2000; Bellouin *et al.,* 2003) have all been developed. Algorithms for aerosol retrievals using MODIS have been developed and validated over both ocean (Tanré *et al.,* 1997), and land surfaces (Kaufman *et al.,* 1997). The uncertainty in these retrievals of τ_{aer} is necessarily higher over land than over oceans owing to uncertainties in the land surface reflectance characteristics and is estimated to be $\pm 0.05 \pm 0.20\ \tau_{aer}$ over the land (Chu *et al.,* 2002) and $\pm 0.03 \pm 0.05\ \tau_{aer}$ over the ocean (Remer *et al.,* 2002). In addition, new algorithms have been developed for discriminating between sea-salt/dust/biomass burning and industrial pollution over oceans (Bellouin *et al.,* 2003; Bellouin *et al.,* 2005; Kaufman *et al.,* 2005) which allows for a more comprehensive comparison of aerosol models against satellite observations. The lack of spectral contrast means that the MODIS algorithm fails over bright surfaces such as deserts or snow surfaces. MISR retrievals have been developed that use the multiple viewing capability to determine aerosol parameters over ocean (Kahn *et al.,* 2001) and land surfaces including highly reflectant surfaces such as desert (Martonchik *et al.,* 2004). Five typical aerosol climatologies each containing four aerosol components are used in the retrievals and the optimum radiance signature is determined for 9 viewing geometries and 2 different radiances. The results have been validated against those from AERONET (Aerosol RObotic NETwork; see Section 2.4.3.2) ATSR (Holzer-Popp *et al.,* 2002) uses a relatively wide spectral range (0.56–1.65 μm), but also uses two viewing directions in aerosol retrievals and aerosol climatologies from the Optical Parameters of Aerosols and Clouds (OPAC) database (Hess *et al.,* 1998).

Despite the increased sophistication and realism of the aerosol retrieval algorithms, discrepancies do exist between retrievals of τ_{aer} even over ocean regions (e.g., Myhre *et al.,* 2004; Myhre *et al.,* 2005). These discrepancies are due to different assumptions in the aerosol models, different wavelengths, and view geometries used in the retrievals, different parameterisations of ocean surface reflectance etc. Satellite retrievals have been extensively validated and generally show good agreement on a case by case basis; thus it is difficult to comment objectively on the accuracy of a particular retrieval relative to the other (e.g., Myhre *et al.,* 2005).

2.4.3.1.2 Direct radiative effect, DRE.

We introduce the direct radiative effect (DRE) which is the sum of the effects due to anthropogenic and natural aerosol species. This is to be distinguished from the definition of RF which considers the anthropogenic components only. In addition to retrievals of τ_{aer}, satellite estimates of the global clear-sky direct radiative effect over oceans have also been made as summarised by Yu *et al.* (2005) (see Table 2.4.2). Retrievals of DRE have improved since TAR owing to the development of dedicated aerosol retrieval algorithms. Table 2.4.2 suggests a fair degree of agreement of the clear-sky DRE from various studies, on the order of -5 W m^{-2}. Model studies have obtained clear-sky DRE estimates that appear consistent with such observational estimates (e.g., Haywood *et al.,* 1999). Recent studies (Bellouin *et al.,* 2005; Kaufman *et al.,* 2005; Loeb and Manalo-Smith*,* 2005) have attempted to derive more than just the clear-sky direct radiative effect. Loeb and Manalo-Smith (2005) assume that there is no contribution to the direct RF from cloudy regions over oceans and derive an all sky direct radiative effect over oceans of -1.6 to -2.0 W m^{-2}. Kaufman *et al.* (2005) estimate the anthropogenic component of the aerosol fine mode fraction from the MODIS product to estimate a clear sky direct RF over ocean of -1.4 W m^{-2}. Bellouin *et al.* (2005) assume no RF from cloudy regions and use a combination of MODIS τ_{aer} and data from AEROCOM (Section 2.4.4) to determine a direct RF of aerosols over both land and ocean of -1.0 W m^{-2}. The uncertainty in these estimates is necessarily larger than in the estimates of the global mean DRE over oceans because of assumptions that either the contribution to the RF from aerosol above cloud is not significant and/or in estimating the anthropogenic fraction of aerosols.

[INSERT TABLE 2.4.2]

Furthermore, use of a combination of sensors on the same satellite offer the possibility of concurrently deriving the τ_{aer} and the direct radiative effect (e.g., Zhang and Christopher*,* 2003; Zhang *et al.,* 2005) which enables estimation of the RF efficiency, i.e. W $m^{-2}\ \tau_{aer}^{-1}$. Because the forcing efficiency removes the dependence on the retrieved τ_{aer} it is a useful parameter for comparison of models against observations (e.g., Anderson *et al.,* 2005), although the RF efficiency is non-linear at high aerosol optical depths such as those associated with large mineral dust events.

2.4.3.2 Surface-based retrievals

Surface based measurements of in-situ properties such as size distribution, chemical composition, scattering and absorption continue to be performed at a number of sites either as long-term monitoring site (e.g., the Interagency Monitoring of Protected Visual Environments - IMPROVE), or specifically as part of intensive field campaigns. These in-situ measurements again provide essential validation for global models e.g., by constraining the concentration of atmospheric aerosol constituents at the surface and also by providing high-quality information about local trends in aerosol concentration. In addition, they provide key information on variability on various timescales. However, comparisons of in situ measurements against those generated by global atmospheric models are complicated by the effects of local meteorology and the fact that the in-situ measurements are representative of conditions at the surface while the direct and indirect RF will depend on the vertical profile of the aerosol. For example the spatial resolution of global model grid-boxes are typically of the order of a few degrees of latitude and longitude and the time-steps for the atmospheric dynamics and radiation may be minutes to hours depending on the process to be studied.

A significant advance since TAR is in the continued deployment and development of surface based remote sensing sun-photometer sites such as AERONET (Holben *et al.,* 1998), and the establishment of networks of aerosol lidar systems such as the European Aerosol Research Lidar Network (EARLINET, Matthias *et al.,* 2004), the Asian Dust Network (ADNET, Murayama *et al.,* 2001), and the Micro-Pulse Lidar Network (MPLNET, Welton *et al.,* 2001). The distribution of AERONET sites since 1993 is also marked on Figure 2.4.2a. Currently there are approximately 150 sites operating at any one time, many of which are permanent, thereby enabling representative monthly and seasonal means to be determined. In addition to an expanding network, and standard measurements of τ_{aer} as a function of wavelength, new algorithms have been developed that measure the sky radiance as a function of scattering angle (Nakajima *et al.,* 1996; Dubovik and King*,* 2000). From these measurements the size distribution and, if the τaer is high enough, the aerosol single scattering albedo and refractive indices may be determined (Dubovik *et al.,* 2000), allowing partitioning between scattering and absorption. Whilst these inversion products have not been comprehensively validated a number of studies show encouraging agreement when compared against in-situ measurements from aircraft measurement campaigns for different aerosol species (e.g., Dubovik *et al.,* 2002; Haywood *et al.,* 2003a; e.g., Reid and et al*.,* 2003; Osborne *et al.,* 2004). Sato *et al.* (2003) determined the aerosol absorption optical depth from AERONET measurements and suggested that aerosol absorption simulated by global aerosol models is underestimated by a factor of between 2–4. Schuster *et al.*(2005) estimate the black carbon loading over continental scale regions. Prima facia the results suggest that the model concentrations and absorption optical depths of black carbon from models are lower than those derived from AERONET. Some of this difference in concentrations could be explained by the assumption that all aerosol absorption is due to black carbon (Schuster *et al.,* 2005), while a significant fraction may be due to absorption by organic aerosol and mineral dust (see Sections 2.4.5.2, and 2.4.5.6). Furthermore, Reddy *et al.* (2005a) show that comparison of the aerosol absorption optical depth from models against those from AERONET must be performed very carefully, reducing the discrepancy between their model and AERONET derived aerosol absorption optical depths from a factor of 4 to a factor of 1.2 by careful co-sampling of AERONET and model data.

The MPLNET Lidar network currently consists of eight lidars worldwide co-located with AERONET sites thereby providing complementary vertical distributions of aerosol extinction coefficients. EARLINET was a European-wide lidar network which currently has fifteen aerosol lidars making routine retrievals of vertical profiles of aerosol extinction. ADNET is a network of twelve lidars making routine measurements in Asia which have been used to assess the vertical profiles of Asian dust and pollution events (e.g., Husar *et al.,* 2001; e.g., Murayama *et al.,* 2001).

2.4.4 Advances in Modelling

Global atmospheric models are able to provide convenient estimates of the RF at the top of the atmosphere and at the surface. The RF may readily be diagnosed if the contributions to emissions from natural and anthropogenic sources are known. Such models now include all of the major anthropogenic species and natural species see Table 2.4.3 and 2.4.4 for references to studies published since TAR. Because all the major aerosol species are now included in these global models, a comparison of key model output parameters such as the total τ_{aer} is possible against both those obtained from satellite retrievals, and surface based sunphotometer and lidar sites (Section 2.4.3).

Major progress over the results presented in TAR has been made in the number and quality of aerosol models that have been used to derive the direct RF. Currently, 16 groups have participated in the AEROCOM initiative. Several models have used a relatively high resolution, e.g., 1º×1º degrees horizontal resolution and up to 40 vertical levels; this represents a considerable enhancement over the models used in TAR The model outputs are available via a dedicated website (Schulz*,* 2004). Three model experiments (named *A*, *B*, *PRE*) were analyzed:-

Experiment A: models simulate the years 1996, 1997, 2000 and 2001 or a five year mean encompassing these years. The model emissions and parameterisations are those determined by each research group, but the models are driven by observed meteorological fields to allow detailed comparisons with observations including those from MODIS, MISR and the AERONET sun photometer network.
Experiment B: uses prescribed aerosol emissions for the years 2000 (Schulz *et al.,* 2005).
Experiment C: uses prescribed aerosol emissions for the years 1750 (Schulz *et al.,* 2005).

The model diagnostics included information on emission and deposition fluxes, vertical distribution and sizes, thus enabling a better understanding of the differences in life times of the various aerosol components in the models (e.g., Textor *et al.,* 2005).

A wide range in several of the diagnostic parameters is found, especially in the case of natural aerosol species dust and sea salt. The emission contributions of differently defined coarse aerosol fractions of these natural compounds are responsible for the high scatter of emission fluxes. Consequently, the dry deposition fluxes of these two coarse mode aerosol species vary considerably. The higher order dependence of the source strength on wind speed adds to the problems in computing natural aerosol emissions. Dust emissions have been found to vary by a factor of two due to the difference in the high-end tail of the wind distribution e.g., this occurs for two operation modes (nudged and climatological) of the same climate model, ECHAM4 (Timmreck and Schulz*,* 2004). The major reason for the larger difference in dust emissions as compared to the fine mode components is, however, the range in source strength maps established by the different groups (Balkanski *et al.,* 2004). With respect to anthropogenic emissions it may be noted that modelling groups tend to make use of similar best guess information, e.g., recently revised emissions information available via the Global Emissions Inventory Activity (GEIA).

The variability in the reported dry deposition fluxes is larger than that in emissions and wet deposition for all five aerosol components. This is because both size distribution and vertical mixing vary between models considerably. Linked to dry deposition and vertical mixing is the aerosol burden remaining in the planetary boundary layer (PBL, here defined as the layer below 800 hPa). We note that the PBL-burden of the fine mode aerosol species varies more than the total burden. Since humidification takes place mainly in the PBL, this source of variation will have an impact on understanding differences in aerosol RF.

However, when normalised to overall burden, the variation of the upper troposphere mass fraction is much more important then that of the PBL and mid troposphere. Differences in the process description of the wet deposition become more pronounced in the upper troposphere. Some models are found to have a tendency to accumulate insoluble aerosol mass (dust, BC and POM) at higher altitudes, while others have rather efficient wet removal schemes. The soluble species – sea salt and sulphate – differ in that sulphur sources (gas-phase oxidation and in-cloud production as well as volcanic contributions) exist at higher altitudes than is the case for the other components, hence sulphate dominates the upper troposphere aerosol concentrations in all models

Tropospheric residence times, defined here as the ratio of burden over sinks established for an equilibrated one-year simulation, vary by 20–30% for the fine mode aerosol species and up to 80% for sea salt. These variations are of interest, since they express how linearly the models relate emissions to aerosol burden and eventually to forcing. Organic matter and black carbon emissions are 56% and 75%, respectively, more effective then sulphur emissions in increasing the overall aerosol burden. This is partly due to their relatively insoluble character and partly due to co-variations of the spatial emission distribution with spatial differences in longevity. Although black carbon is less soluble than particulate organic matter, and wet removal is parameterised taking into account this difference, it appears that residence times of soot in some models are slightly longer then those of organic matter. This is a result of different emission patterns with more organic matter produced in biomass burning regions. According to these model simulations, aerosol emissions from these regions have a relatively larger impact on global aerosol burdens then fossil fuel derived organic matter.

The independent model simulations may be viewed as expert realisations of an ensemble of realistic aerosol distributions. Here, they are taken to be independent, and a mean result constructed from the AEROCOM models affords one way to arrive at a central value of the direct RF (Schulz *et al.,* 2005). However, this does not preclude other modelling or observational efforts or other means to categorize forcing estimates.

Aerosol RF depends on anthropogenic emissions and the resulting burdens. Their computation remains uncertain. The AEROCOM compilation of model allows evaluation of whether the global dispersion of the aerosol is model dependent. As an example, the fraction of aerosol mass above 5 km varies between 10–60% for the anthropogenic aerosol compounds black carbon, particulate organic matter and sulphate (Textor *et al.,* 2005). An analysis of the AEROCOM results indicates that the fraction of aerosol mass above 5 km in the AEROCOM experiments A and B resemble each other for a given model. To first order the spatial (vertical and horizontal) dispersivity of the models explains the differences in lifetime and thus burdens. It appears that the models have an inherent specific dispersion property, with dispersion of different aerosol components being similar in a given model, but differing amongst the various models. This suggests that a combined aerosol effect is a more characteristic and pragmatic result for models than putting together forcing estimates due to the component species. The AEROCOM analyses also reveal that vertical distribution may have a large impact on the diagnosed direct RF owing to the complexities of absorbing aerosol such as biomass burning aerosol or fossil fuel black carbon aerosol existing above clouds (Sections 2.4.5.3, and 2.4.5.4). Verification of model simulations against reliable observations has yet to be comprehensively performed.

2.4.5 Direct Radiative Forcing

The direct RF due to different aerosol species is discussed in turn. Where possible, statistics from model results are used to assess the uncertainty in the direct RF. While this uncertainty includes the structural uncertainty associated with the direct RF, it does not include the full range of parametric uncertainty as the model results are essentially best estimates that are constrained by observations of e.g., emissions, wet and dry deposition, size distributions, optical parameters, hygroscopicity, etc (Pan *et al.,* 1997). We report the uncertainty as approximately ±1 standard deviation where sufficient model estimates of the direct RF are available. While not fully rigorous from a statistical point of view it does allow the relative uncertainty in the direct RF of each species of aerosol to be inter-compared in a more quantitative manner.

2.4.5.1 Sulphate aerosol

Pure atmospheric sulphate aerosol consists mainly of the chemical compounds H_2SO_4, NH_4HSO_4, and $(NH_4)_2SO_4$ with associated water of hydration and are formed by oxidation of SO_2 via gaseous phase reactions with the hydroxyl radical and aqueous phase reactions within cloud droplets (e.g., Penner *et al.,* 2001). The main source of anthropogenic sulphate aerosol is via sulphur dioxide emissions from fossil-fuel burning (~72%), with a small contribution from biomass burning (~2%) while natural sources of sulphate aerosol are from dimethyl emissions by marine phytoplankton (~19%) and by SO_2 emissions from volcanoes (~7%). Global SO_2 mean emissions range from 66.8 to 92.4 TgS yr^{-1} for anthropogenic emissions and from 91.7 to 125.5 TgS yr^{-1} for total emissions. Emissions of sulphur dioxide from 25 countries in Europe have reduced from approximately 18TgS yr^{-1} in 1980 to 4TgS yr^{-1} in 2002 (Vestreng and al*,* 2004). In the USA, the emissions have been reduced by some 33% in the period 1982-2001 (U. S. EPA, 2004: http://www.epa.gov). However, over the same period SO_2 emissions have been increasing significantly in Asia and developing countries. The net result of these combined regional reductions and increases leads to uncertainty in whether the global SO_2 have risen or fallen since the 1980s (Lefohn *et al.,* 1999; Van Aardenne *et al.,* 2001; Boucher and Pham*,* 2002). However, the regional shift in the emissions of sulphur dioxide from U.S., Europe, Russia, Northern Atlantic Ocean and parts of Africa to South-East Asia and the Indian and Pacific Oceans areas will lead to subsequent shifts in the pattern of the direct RF (e.g., Boucher and Pham*,* 2002).

The optical parameters of pure sulphate aerosol have been well documented (see Penner *et al.,* 2001 and references therein). Pure sulphate is essentially an entirely scattering aerosol across the solar spectrum (ω_o = 1), with theoretical and experimental data available on the relative humidity dependence of the specific extinction coefficient, f_{RH} (e.g., Tang *et al.,* 1995). Measurement campaigns concentrating on industrial pollution such as TARFOX (Russell *et al.,* 1999), ACE-2 (Raes *et al.,* 2000), INDOEX (Ramanathan *et al.,* 2001b) continue to show that sulphate contributes a significant fraction of the accumulation mode mass, and therefore contributes significantly to the anthropogenic aerosol optical depth and direct RF (e.g., Hegg *et al.,* 1997; Russell and Heintzenberg*,* 2000; Ramanathan *et al.,* 2001b; Quinn and Bates*,* 2005). However, sulphate is invariably internally/externally mixed to varying degrees with other compounds such as biomass burning aerosol (e.g., Formenti *et al.,* 2003), fossil-fuel black carbon (e.g., Russell and Heintzenberg*,* 2000), organic carbon (Novakov *et al.,* 1997; Brock *et al.,* 2004), mineral dust (e.g., Huebert *et al.,* 2003), and nitrate aerosol (e.g., Schaap *et al.,* 2003), which results in a composite effective refractive index, size distribution, and hygroscopicity and optical properties.

TAR suggested a direct RF due to sulphate aerosol of –0.40 W m^{-2} with an uncertainty of a factor of two, results that were based on global modelling studies that were available at that time. Since TAR a number of further modelling studies have been performed. The results from these model studies are summarised in Table 2.4.3 for both non-AEROCOM and AEROCOM studies. For non-AEROCOM studies, the top of the atmosphere direct RF from the models ranges from approximately –0.21 W m^{-2} (Takemura *et al.,* 2005) to –0.96 W m^{-2} (Adams *et al.,* 2001) with a mean of –0.46 W m^{-2} and a standard deviation of –0.20 W m^{-2}. As in TAR, the range in the normalised direct RF (NDRF) is significant. The reason for these differences is unclear, but may be due to different representation of aerosol optical properties, cloud, surface reflectance, hygroscopic growth etc (Ramaswamy *et al.,* 2001).The direct RF from the models participating in the AEROCOM project is slightly weaker than that obtained from those shown in Table 2.4.3 with a mean of ~–0.34 W m^{-2} and a standard deviation of 0.09 W m^{-2}; the standard deviation is reduced for the AEROCOM models owing to constraints on aerosol emissions, based on updated emission inventories. The direct RF at the surface will be similar or marginally stronger at the surface than at the top of the atmosphere as the aerosol is considered to be almost entirely scattering although some models include a little absorption. The uncertainty in this estimate of the direct RF remains relatively large compared to that for well mixed greenhouse gases.
On the basis of these results, a top of the atmosphere direct RF of ~–0.40 W m^{-2} ± 0.20 W m^{-2} is suggested for the direct RF due to sulphate aerosol.

[INSERT TABLE 2.4.3]

2.4.5.2 Organic carbon from fossil fuels

Organic aerosols are a complex mixture of chemical compounds produced from fossil-fuel burning and natural biogenic emissions either as primary aerosol particles or as secondary aerosol particles from condensation of low and semi-volatile organic gases. Hundreds of different atmospheric organic compounds have been detected in significant quantities in the atmosphere (e.g. Hamilton *et al.,* 2004; Murphy*,* 2005) which makes definitive modelling of the direct and indirect effects extremely challenging (McFiggans *et al.,* 2005). Emissions of organic carbon from fossil fuel burning have been estimated to be 10 to 30TgCyr-1 (Liousse *et al.,* 1996; Cooke *et al.,* 1999; Scholes and Andreae*,* 2000). Trends in emissions of fossil fuel organic carbon may be inferred from emission inventories for non-methyl volatile organic compounds (NMVOCs). Emissions of fossil-fuel NMVOCs from 25 countries in Europe have reduced by 60% in the period 1980 to 2002 (Vestreng and al*,* 2004). Thus the reduction in organic carbon is less dramatic than that of sulphur dioxide. Mass concentrations of fossil fuel organic aerosol are frequently similar to those for industrial sulphate aerosol. Novakov *et al.* (1997) and Hegg *et al.* (1997) measured organic carbon in pollution off the East coast of the USA during the TARFOX campaign and found that organic carbon primarily from fossil fuel burning contributed up to 40% of the total submicron aerosol mass and was frequently the most significant contributor to the total aerosol optical depth. During INDOEX which studied the industrial plume in the Indian Ocean, Ramanathan *et al.* (2001b) found that organic carbon was the second largest contributor to the aerosol optical depth behind sulphate aerosol.

Observational evidence suggests that some organic aerosol compounds from fossil fuels are relatively weakly absorbing but do absorb solar radiation at some wavelengths (e.g., Bond *et al.,* 1999; Bond*,* 2001) although organic aerosol from high temperature combustion such as fossil-fuel burning (Dubovik *et al.,* 1998; Kirchstetter *et al.,* 2004) appears less absorbing than from low temperature combustion such as biomass burning. Observations of the hygroscopicity of organic carbon suggest that a significant fraction of organic carbon is soluble to some degree; whilst at low relative humidity more water is often associated with the organic fraction than inorganic material, at higher relative humidities the hygroscopicity of organic carbon is considerably less than that of sulphate aerosol (Kotchenruther and Hobbs*,* 1998; Kotchenruther *et al.,* 1999).

Based on observations and fundamental chemical kinetic principles, attempts have been made to formulate groups of organic carbon particles into those with similar characteristics in terms of e.g., refractive indices, hygroscopicity, and cloud activation properties which would help facilitate their implementation in climate models (e.g., Decesari *et al.,* 2000; Decesari *et al.,* 2001; Maria *et al.,* 2002; e.g., Ming and Russell*,* 2002).

Organic carbon aerosol from fossil fuel sources is invariably internally/externally mixed to some degree with other combustion products such as sulphate and black carbon (e.g., Novakov *et al.,* 1997; Ramanathan *et al.,* 2001b). Theoretically, coatings of essentially non-absorbing components such as organic carbon on strongly absorbing core components such as black carbon can increase the absorption of the composite aerosol (e.g., Fuller *et al.,* 1999; Jacobson*,* 2001) However coatings of organic carbon aerosol on hygroscopic aerosol such as sulphate may lead to a suppression in the rate of water uptake during cloud activation (Xiong *et al.,* 1998; Chuang*,* 2003).

Current global models generally treat organic carbon using one or two tracers (e.g., water insoluble tracer, water soluble tracer) and highly parameterised schemes have been developed to represent the direct radiative effects. Considerable uncertainties still exist in representing the refractive indices and the water of hydration associated with the particles because the aerosol properties will invariably differ depending on the combustion process, time since emission, mixing with the ambient aerosol etc (e.g., McFiggans *et al.,* 2005).

TAR suggested a direct RF of organic carbon aerosols from fossil fuel burning of ~–0.10 W m^{-2} with a factor of three uncertainty. Many of the modelling studies that have been performed since TAR have investigated the direct RF of organic carbon aerosols from both fossil-fuel and biomass burning aerosols and the combined direct RF of both components. These studies are summarised in Table 2.4.4. The direct radiative effect from total organic carbon from both biomass burning and fossil-fuel emissions from the non-AEROCOM and AEROCOM models suggest direct RFs of –0.25 ± 0.08 W m^{-2} and –0.40 ± 0.25 W m^{-2} respectively. Where the direct RF due to organic carbon from fossil fuels is not explicitly accounted for in the studies an approximate scaling based on the source apportionment of 0.25:0.75 is applied for fossil-fuel organic carbon: biomass burning organic carbon. The mean direct radiative effect from the fossil-fuel component of organic carbon from those studies other than those in AEROCOM is –0.06 W m^{-2} while those from AEROCOM produce a RF of –0.10 W m^{-2} with a range of –0.01 W m^{-2} to –0.19 W m^{-2} and a standard deviation of around 0.05 W m^{-2}. These studies all use optical properties for organic carbon that are either entirely scattering or only weakly absorbing and hence the surface RF is only slightly stronger than that at the top of the atmosphere. Based on these results, the direct RF due to fossil fuel sources of organic carbon is therefore estimated to be in the region of ~–0.08 ± 0.05 W m^{-2} owing to difficulties in constraining the total column burden, the absorption, mixing, and hygroscopicity of the aerosol.

[INSERT TABLE 2.4.4 HERE]

2.4.5.3 Black carbon from fossil fuels

Black carbon is a primary aerosol emitted directly at source from incomplete combustion processes such as fossil-fuel and biomass burning and therefore much atmospheric BC is of anthropogenic origin. The trends in emission of fossil-fuel black carbon have been investigated in industrial areas by Novakov *et al.* (2003). Significant decreases have been recorded in the UK, Germany, the former Soviet Union, and the USA over the period 1950–2000, while significant increases were reported in India and China. Globally, Novakov *et al.* (2003) suggest that emissions of fossil-fuel black carbon increased by a factor of three between 1950 and 1990 (2.2 to 6.7 TgBC yr^{-1}) owing to the rapid expansion of the Chinese and Indian economies (e.g. Streets and al*,* 2001), and has since fallen to around 5.6 TgC yr^{-1} owing to further emission controls. Electron microscope images of BC particles show that BC particles are emitted as complex chain structure (e.g. Posfai *et al.,* 2003), which tend to collapse down as the particles age.

Black carbon aerosol strongly absorbs solar radiation. The Indian Ocean Experiment (INDOEX, Ramanathan *et al.,* 2001b and references therein) focussed on emissions of aerosol from the Indian sub-continent, and showed the importance of absorption by aerosol in the atmospheric column. Their observations showed that the local surface forcing (–23 W m^{-2}) was significantly stronger than the RF at the top of the atmosphere (–7 W m^{-2}). In this instance 16 ± 2 W m^{-2} of solar radiation was absorbed in the atmosphere, which significantly altered the atmospheric temperature and humidity structure thereby changing the cloud amount via the 'semi-direct effect' (Figure 2.4.1 and Section 2.4.6). Additionally, the presence of black carbon in the atmosphere above highly reflectant surfaces such as snow/ice or clouds is sufficient to cause a significant positive direct RF (Ramaswamy *et al.,* 2001). The vertical profile of black carbon is therefore important as black carbon aerosols or mixtures of aerosols containing a relatively large fraction of black carbon will exert a positive RF when above underlying clouds. Both microphysical (e.g., hydrophilic-to-hydrophobic nature of emissions into the atmosphere, aging of the aerosols, wet deposition) and meteorological aspects govern the horizontal and vertical pattern of distribution of the black carbon aerosols, and the residence time of these aerosols is thus sensitive to these factors (Cooke *et al.,* 2002).

Those models since TAR that explicitly model and separate out the direct RF due to black carbon from fossil fuels include those from Takemura *et al.* (2000), Reddy *et al.* (2005a), and Hansen *et al.* (2005) as summarised in Table 2.4.4. A number of studies continue to group the RF from fossil-fuel with those from biomass burning as also shown. Non-AEROCOM and AEROCOM studies suggest a combined direct RF from both sources of +0.45 ± 0.13 W m^{-2}, and 0.75 ± 0.46 W m^{-2} respectively. The stronger RF estimates from the AEROCOM models do not appear to be due to stronger sources and column loadings, but may be due to the method of internal mixing the BC aerosols with other components which should increase the absorption (e.g.. Stier *et al.,* 2005), or could involve a larger fraction of aerosols located above clouds (see earlier discussion) such that there is an enhancement of the radiative effect. Source emission inventories continue to suggest a split of approximately 50:50 between emissions from biomass burning sources and fossil-fuel burning sources. This split is applied to those estimates where the black carbon emissions are not explicitly separated into emission sources to provide an estimate of the direct RF due to fossil-fuel black carbon. The direct RF ranges from +0.11 W m^{-2} (AEROCOM submission using the model of Myhre *et al.,* 2003) to +0.72 W m^{-2} (AEROCOM submission using the model of Takemura *et al.*, (2000) with a mean of +0.26 W m^{-2} for the non-AEROCOM submissions and a mean of +0.38 W m^{-2} for the AEROCOM submissions. Grouping all the model results together as equally likely results in a direct RF due to black carbon of +0.30 W m^{-2} ± 0.15 W m^{-2} which is used as our best estimate.

2.4.5.4 Biomass burning aerosols

TAR suggested a contribution to the direct RF of roughly –0.4 W m^{-2} from the scattering components (mainly organic carbon and inorganic compounds) and +0.2 W m^{-2} from the absorbing components (BC) leading to an estimate of the RF of biomass burning aerosols of –0.20 W m^{-2} with a factor of three uncertainty. Note that the estimates of the black carbon direct RF from Hansen and Sato (2001), Hansen (2002), and Hansen and Nazarenko (2004) and Jacobson (2001) include the direct RF component from BC from biomass burning aerosol in their estimates of the total RF due to BC. We continue to group the RF due to biomass burning (i.e., primarily organic carbon, black carbon, and inorganic compounds such as nitrate and sulphate) into a single RF. This is because while each of the components of fossil-fuel emissions of e.g., sulphate, black carbon, and organic carbon can be effectively reduced using different technologies, it is unlikely that any reduction in the ratio of emissions of OC, BC and inorganic compounds from biomass burning sources can be achieved through technological emission control as biomass burning emissions are essentially uncontrolled.

Since TAR, there have been a number of measurement campaigns and modelling efforts relating to biomass burning aerosols at different geographic locations (e.g., SAFARI-2000, SMOCC). The Southern African Regional Science Initiative (SAFARI 2000,Swap *et al.,* 2002; Swap *et al.,* 2003) took place in 2000 and 2001. The main objectives of the aerosol research in the dry season were to investigate pyrogenic and biogenic emissions of aerosol in southern Africa (Eatough *et al.,* 2003; Formenti *et al,* 2003; Hély *et al.,* 2003), validate the aerosol retrievals from satellite and surface based instruments (Haywood *et al.,* 2003b; Ichoku *et al.,* 2003) and to study the influence of aerosol and trace gases on the radiation budget through the direct effect and through the ability of biomass burning aerosols to act as efficient CCN (e.g., Bergstrom *et al.,* 2003; e.g., Keil and Haywood*,* 2003; Myhre *et al.,* 2003; Ross *et al.,* 2003). Considerable efforts were made to characterise the physical and optical properties of fresh and aged regional haze rich in biomass burning aerosol by making intensive observations of aerosol size distributions, optical properties, and radiative effects through in-situ aircraft measurements (e.g., Abel *et al.,* 2003; e.g., Formenti *et al.,* 2003; Haywood *et al.,* 2003b; Magi and Hobbs*,* 2003; Kirchstetter *et al.,* 2004), and radiometric measurements (e.g., Bergstrom *et al.,* 2003; Eck *et al.,* 2003). The ω_o at 0.55 μm derived from AERONET sites was found to range between 0.85 to 0.89 (Eck *et al.,* 2003), while more aged aerosol off the west coast of Africa was slightly less absorbing with ω_o at 0.55 μm averaging approximately 0.91 (Haywood *et al.,* 2003b). Abel *et al.* (2003) showed evidence that ω_o at 0.55 μm increased from approximately 0.85 to 0.90 over a time period of approximately two hours subsequent to emission, and attributes the result to the condensation of organic gases to form aerosol particles rather than the collapse of black carbon chains as they age. However, absorption artefacts caused by the preferential orientation of chain BC particles on filter-based absorption measurements (Fuller *et al.,* 1999) cannot be ruled out. As for industrial aerosol that contains a significant amount of absorbing black carbon aerosol, biomass burning aerosol exerts a RF that is larger at the surface and in the atmospheric column than at the top of the atmosphere (see Figure 2.4.3).

Modelling efforts have used data from measurement campaigns to improve the representation of the physical and optical properties of biomass burning aerosol and the vertical profile of biomass burning aerosol (Myhre *et al.,* 2003 and Section 2.4.6; Penner *et al.,* 2003).These modifications have had important consequences on estimates of the direct RF due to biomass burning aerosols because the RF is significantly more positive when biomass burning aerosol overlies cloud than previously estimated (Keil and Haywood*,* 2003; Myhre *et al.,* 2003; Abel *et al.,* 2005). While the RF due to biomass burning aerosols in clear skies is certainly negative the overall RF of biomass burning aerosols may therefore be positive. In addition, to modelling studies, observations of this effect have been made possible via satellite measurements. Hsu *et al.* (2003) used SeaWiFs, TOMS and CERES data to show that biomass burning aerosol emitted from S.E. Asia is frequently lifted above cloud leading to a reduction in outgoing solar radiation over cloudy areas by up to 100 W m^{-2} and points out that this effect could be due to a combination of direct and indirect effects. Similarly, Haywood *et al.* (2003a) showed that remote sensing of cloud liquid water and effective radius underlying biomass burning aerosol off the coast of Africa are subject to potentially large systematic biases. This may have important consequences for studies that use correlations of aerosol optical depth and cloud effective radius in estimating the indirect radiative effect of aerosols.

That the biomass burning direct RF can exert a significant positive direct RF when above cloud is documented by the non-AEROCOM and AEROCOM models in Table 2.4.4. Both AEROCOM and non-AEROCOM models suggest an average global mean direct RF from biomass burning aerosols of +0.06 W m^{-2}, and the combined standard deviation is 0.08 W m^{-2}, hence even the sign of the direct RF due to biomass burning aerosols is in question. The most negative direct RF of –0.06 W m^{-2} is from the modelling study of Takemura *et al.* (2001), while the most positive of +0.22 W m^{-2} comes from Jacobson (2001). Thus, these recent studies yield a direct RF for biomass burning aerosols that is significantly different to that reported by TAR and is now suggested to be +0.06 ± 0.08 W m^{-2}.

2.4.5.5 Nitrate aerosol

TAR did not quantify the RF due to nitrate aerosol owing to the large discrepancies in the studies available at that time. Van Dorland (1997) and Jacobson (2001) suggested relatively minor global mean RFs of –0.03 W m^{-2} and –0.02 W m^{-2} respectively while Adams *et al.* (2001) suggested a global mean RF as strong as –0.22 W m^{-2}. Subsequent studies include those of Schaap *et al.* (2003), who estimate that the RF of nitrate over Europe is some 25% of that due to sulphate aerosol. Atmospheric nitrate is essentially non-absorbing in the visible spectrum, and laboratory studies have been performed to determine the hygroscopicity of the aerosols (e.g., Tang *et al.,* 1995). None of the models participating in AEROCOM have estimated the direct RF due to nitrate aerosol making an initial global estimate possible. The mean direct RF for nitrate is estimated to be ~–0.15W m^{-2} at the top of the atmosphere, and the conservative scattering nature means a similar bottom of the atmosphere direct RF. However, the uncertainty in this estimate in necessarily large owing to the relatively small number of studies that have been performed and the considerable uncertainty in estimates of e.g. the nitrate aerosol optical depth. Thus we tentatively adopt a direct RF of –0.15 ± 0.15 W m^{-2} but acknowledge that the number of studies performed is insufficient for accurate characterization of the magnitude and uncertainty of the RF.

2.4.5.6 Mineral dust

Mineral dust from anthropogenic sources originates mainly from agricultural practices (harvesting, ploughing, desertification due to over-grazing etc) and industrial practices (e.g., cement production, transport etc.). TAR suggested that the direct RF due to anthropogenic mineral dust lies in the range +0.4 to –0.6 W m^{-2}, and did not assign a best estimate because of the difficulties in determining the contribution to the total dust loading from anthropogenic activities and in evaluating the competing RF of the shortwave and longwave RF.

Tegen and Fung (1995) estimated the anthropogenic contribution to mineral dust to be 30–50% of the total dust burden in the atmosphere. Tegen *et al.* (2004) provided a more detailed estimate by comparing observations of visibility as a proxy for dust events from over 2000 surface stations with model results and suggest that only 5–7% of mineral dust comes from anthropogenic agricultural sources. However, Mahowald (2004) uses the same observational data and contests these finding suggesting that up to 0–50% of dust may be of anthropogenic origin. However, Tegen *et al.* (2005) perform some further sensitivity studies by reducing the model threshold friction velocity for dust production and suggest that the model produces too many mineral dust storms if the anthropogenic fraction exceeds 15% and suggest therefore that this is an upper limit. Long-term aerosol size distributions retrieved by AERONET (Dubovik *et al.,* 2002) suggest that there is a distinct coarse mode observable at a limited number of industrial sites, but the contribution to the aerosol optical depth from this mode is negligible compared to that from the more optically active sulphate, black carbon, organic carbon and nitrate that make up the accumulation mode. Thus there remains considerable uncertainty with respect to the anthropogenic component of mineral dust but we revise the estimate to 10%.

In-situ measurements of local Saharan dust (e.g., Haywood *et al.,* 2003a; Tanré *et al.,* 2003); transported Saharan mineral dust (e.g., Myhre *et al.,* 2003) and Asian mineral dust (Clarke *et al.,* 2001; Conant *et al.,* 2003; Huebert *et al.,* 2003; Clarke *et al.,* 2004) using aircraft instrumentation reveal that dust is considerably less absorbing than suggested by previous dust models such as that of WMO (1986). In addition, satellite retrievals of ω_o (Kaufman *et al.,* 2001) suggest that dust is indeed less absorbing than using the dust models of WMO (1986). Analyses of ω_0 from long-term AERONET sites influenced by Saharan dust reveal that the refractive indices of mineral dust are comparable to those refractive indices determined from the recent in-situ measurements (Dubovik *et al.,* 2002), confirming the reduced absorption by mineral dust.

Measurements of the direct radiative effect of mineral dust over ocean regions suggest that the local direct radiative effect may be extremely strong; Haywood *et al.* (2003b) made aircraft-based measurements of the local instantaneous direct shortwave radiative effect of as strong as –130 W m^{-2} off the coast of West Africa. Hsu *et al.* (2000) used ERBE and TOMS data to determine a peak monthly mean shortwave radiative effect of around –45 W m^{-2} for July 1985. Interferometer measurements from aircraft and the surface have now measured the spectral signature of mineral dust for a number of cases (e.g., Highwood *et al.,* 2003) indicating an absorption peak in the centre of the 8–13 µm atmospheric window. Hsu *et al.* (2000) determined a longwave radiative effect over land areas of North Africa of up to +25 W m^{-2} for July 1985; similar results were presented by Haywood *et al.* (2005) who determined a peak longwave direct radiative effect of up to +50 W m^{-2} at the top of the atmosphere for July 2003. However, while these studies show that the direct radiative effect due to mineral dust may be locally very significant, they cannot distinguish the contribution to mineral dust from anthropogenic sources and therefore a direct RF cannot be determined.

In some areas, the complex state of internal/external mixing of mineral dust with sulphate/organics/BC (e.g., over East Asia) appears to result in an increase in the specific absorption efficiency but a decrease in the specific scattering efficiency which will tend to reduce the single scattering albedo of the composite particles (Chuang *et al.,* 2003; Clarke *et al.,* 2004); thus even natural sources of mineral dust may play an important role in modulating the RF from anthropogenic emissions of sulphate, organics, and BC. The degree of this effect may be size-dependent.

Given the reduction in the anthropogenic component of the mineral dust since TAR and that the majority of the models in the AEROCOM project produce estimates of the direct RF due to anthropogenic mineral dust that are close to zero, the range of the direct RF due to mineral dust from Ramaswamy *et al.* (2001) of –0.6 to +0.4 W m^{-2} is reduced considerably to –0.2 to +0.1 W m^{-2}.

2.4.5.7 Combined aerosol species

Why combine the aerosol component forcings? TAR reported RF values associated to several components of the aerosol but did not provide an estimate of the overall aerosol direct effect. However, the associated uncertainties for each individual component suggested that by combining them the overall uncertainty of aerosol forcing would be very large through uncertainty propagation (e.g. Boucher and Haywood*,* 2001). Improved and intensified in-situ observations and remote sensing of aerosols suggest that the range of combined aerosol forcing is now better constrained because validation criteria do exist for combined aerosol properties representing the whole vertical column of the atmosphere. This results in less uncertainty than that obtained through an uncertainty propagation procedure based on forcing uncertainties from individual components. Note also that individual aerosol component forcings are discussed in detail above.

It would be desirable to identify the RF contribution from major aerosol components attributable to individual sources (Section 2.9.3 investigates this). Such a presentation would be advantageous over the split of aerosol forcing into contributions from black carbon, biomass burning, particulate organic matter and sulphate, since emissions from any given source are specific mixtures. Specific composition and degree of mixing for a given emission source determine its related radiative effect. Internally mixed black carbon present in biomass burning aerosols is shown to be a less efficient absorber than primary soot particles emitted in industrial and traffic combustion processes.

However, few models have separated out the RF from specific emission sources. This is partly due to recent emphasis on complex aerosol model development, which takes into account the internally mixed nature of aerosol particles (Kirkevag and Iversen*,* 2002; Liao and Seinfeld*,* 2005; Stier *et al.,* 2005; Takemura *et al.,* 2005). The increased complexity of aerosol simulations has the advantage of providing more realistic overall aerosol distributions to climate model simulations. Such complex models take into account mixing state and interactions in the presence of several aerosol species at the same time. Mixing of aerosol particle populations influences the radiative properties of the aerosol, which involve changes of size, chemical composition, mixing state, shape and feedbacks into the aerosol removal and formation processes itself. Although the source processes for anthropogenic aerosols favour their submicron nature, natural aerosols intervene by providing a condensation surface for aerosol precursor gases. Several sensitivity studies show the degree of non-linearity in estimating the overall aerosol effect from multicomponent modelling. The presence of heterogeneous reactions on sea salt and dust can reduce the fine mode sulphate load by 28% (Liao *et al.,* 2004). In their model, a doubling of SO_2 emissions over present day conditions corresponds to 45% more sulphate, 14% more ammonium and 44% less nitrate. Assuming external mixing of black carbon can reduce the associated RF by up to 0.5 W m^{-2} (see compilation in Chuang *et al.,* 2002).

The model specific treatment of transport and removal processes is responsible for that the RF from the major aerosol components are not independent from each other in a given model. As discussed in 2.4.4 the transport efficiency in a given model results in more or less dispersed aerosols. A less dispersive model with smaller burdens necessarily has both less scattering and absorbing aerosols interacting with the radiation field. Using the combined forcing to assess aerosol forcing from several models removes some of the variance which would appear when estimating the combined aerosol RF uncertainty from error propagation analysis based on uncertainties of individual aerosol components.

The reason for the remaining large range of RF results is to be found in the important scatter of possibilities to account for the opposing scattering and absorbing aerosols:

- Aerosols that predominantly scatter radiation (sulphate, organic carbon, nitrate) exert a negative RF and these can be conveniently grouped together and the magnitude of each aerosol RF readily intercompared.
- Aerosols that absorb radiation (primarily black carbon aerosol) exert a positive RF and the relative contribution of the fossil fuel component and the biomass burning component and mixing assumptions make this component the more uncertain and variable component.

Aerosol optical depth and absorption measurements report bulk quantities, which enable to constrain the range of useful model realisations. Bulk aerosol properties such as total aerosol optical depth are reconstructed from models. The AEROCOM compilation suggests low understanding of the local and regional composition of the global aerosol, but overall reproduction of the total aerosol optical depth variability (Kinne *et al.,* 2005).This suggests that the different model realisations test for the effect of different composition on the resulting RF. The combined RF taken together from several models is more robust then an analysis per component or by just one model.

Finally, to provide independent evaluation of the models, measurement based estimates of total and anthropogenic radiative perturbation using satellites and in-situ measurements of relevant aerosol characteristics over cloud-free ocean have become recently available (Bellouin *et al.,* 2005; Kaufman *et al.,* 2005).

When comparing the total radiative perturbation in oceanic clear-sky conditions models appear to underestimate the negative aerosol forcing by 20–50% (Yu *et al.,* 2005). The reason for the discrepancy is not clear. A measurement based estimate of the anthropogenic compound allows to confirm model forcing results more directly by excluding consideration of rather uncertain but large contributions from natural aerosols. For this purpose satellite observed fine mode aerosol optical depth can be used to estimate the anthropogenic aerosol optical depth. Kaufmann *et al.* (2005) have used information from regions where one of the major aerosol types dominates to establish a consistent correction for fine aerosol contributions from dust and sea salt. Subtracting these from observed fine mode aerosol optical depth by MODIS they find 21% of the aerosol optical depth. Table 2.4.5 suggests that the anthropogenic aerosol optical depth estimated from models is slightly higher (27% as derived from AEROCOM models). Both the modelled underestimation of total aerosol radiative perturbation in clear sky conditions over ocean and the modelled overestimation of anthropogenic aerosol optical depth could be explained by deficiencies in our understanding of the predominating natural aerosol components.

Regional aspects are of importance to assess the climate impact of the aerosol. The partly absorbing nature of the aerosol is responsible for a heating of the lower tropospheric column. Surface forcing is considerably higher than top-of-the atmosphere forcing. This is now confirmed through several experimental and observational studies as discussed in earlier subchapters. Table 2.4.5 summarises the surface forcing obtained in the different models. Figure 2.4.3 depicts the regional distribution of several important parameters when assessing the regional impact of aerosol forcing. The results are based on a mean model constructed from AEROCOM simulation results B and PRE. Anthropogenic aerosol optical depth (Figure 2.4.3a) is shown to have important maxima in industrialised regions and above biomass burning dominated areas. The difference of simulated to observed aerosol optical depth shows that regionally τ_{aer} can be up to 0.1(Figure 2.4.3b). Figures 2.4.3c) shows the regions off Southern Africa where the biomass burning aerosol above clouds leads to an overall heating. Figure 2.4.3d) reports the local diversity as standard deviation from 9 models of the overall RF. Largest uncertainties of ±3 W m^{-2} are to be found in East Asia and in the African biomass burning regions. Figure 2.4.3e) reveals that on the average 0.9 W m^{-2} heating can be expected to happen in the atmospheric column as a consequence of absorption by aerosols. Regionally this can reach annually averaged values of more than 5 W m^{-2}.

[INSERT TABLE 2.4.5]

Since TAR, a greater number of detailed aerosol models have computed the combined direct RF from all aerosol components by using consistent microphysical and meteorological fields. Recent coordinated validation attempts with AEROCOM have shown that all of the models capture major features of the global aerosol distribution (Kinne *et al.,* 2005). It is difficult to assess the performance of each of the models as the performance depends on the observational dataset against which the models are evaluated. This implies that currently no preference or weighting to individual model may be rigorously performed. The results summarised in Table 2.4.5 are suggested to provide a new estimate for the combined aerosol RF. These estimates are suggested to better represent the combined role of aerosol direct RF among the other RF agents. It is estimated to range between +0.1 and -0.6 Wm^{-2}. From both the Non-AEROCOM and the AEROCOM model groups, using the standard deviation of the model estimates as a measure of the uncertainty, we quote the best estimate of the combined direct RF due to aerosols as –0.2 ± 0.2 W m^{-2}. [Note that this value is subject to revision. Currently there is a mismatch with summing the aerosol species, some would be expected, but it is currently outside of uncertainty range – this will be investigated for the second draft.]

2.4.6 Cloud-Aerosol Interaction

Aerosol particles are needed for the formation of clouds. Only a subset of the aerosol particle population act as cloud condensation nuclei (CCN) and ice nuclei (IN). Increases in ambient concentrations of CCN and IN due to anthropogenic activities can modify the microphysical properties of clouds, thereby affecting the climate system (Penner *et al.,* 2001); (Ramanathan *et al.,* 2001a). Aerosols can increase the albedo of clouds (Twomey, 1977), referred to for convenience and simplicity here as the "albedo" effect. Aerosols can also increase the lifetime (Albrecht, 1989), referred to simply here as the "lifetime" effect. The modification of the microphysical characteristics of clouds and the related changes in their optical properties are referred to as the indirect climatic effect of aerosols (Penner *et al.,* 2001; Jacob *et al.,* 2005). This term involves several mechanisms as presented schematically in Figure 2.4.1.

The interactions between aerosol particles and clouds are complex and can be non-linear (Ramaswamy *et al,* 2001). The chemical composition of the initial nuclei (e.g., anthropogenic sulphates, nitrates, organic and black carbon) is important in the activation and early growth of the cloud droplets, particularly the water-soluble fraction and the presence of compounds that affect surface tension (Tang, 1997; McInnes *et al.,* 1998; Ming and Russell, 2002). Cloud optical properties are a function of wavelength and depend on the characteristics of the droplet size distributions, ice crystal concentrations and shapes in the various cloud types occurring in the climate system.

The interactions of aerosol particles with water (stratocumulus and shallow cumulus) and deep convective clouds (with mixed phase) are discussed in this subsection, while the impacts of contrails and aviation-induced cirrus are discussed in Section 2.6 and the indirect impact of aerosol on surface properties is discussed in Section 2.5

2.4.6.1 New observational evidence

2.4.6.1.1 New evidence related to enhanced reflectance and regional variability

The evidence concerning potential aerosol modification of clouds provided by the shiptrack observations in TAR has been confirmed, to a large extent qualitatively, by results from a number of studies using in situ aircraft and satellite data, covering continental cases and several regional studies. Feingold *et al.* (2003), Kim *et al*. (2003) and Penner *et al*. (2004) present evidence of an increase in the reflectance in continental stratocumulus cases, utilizing remote sensing techniques at specific field sites. The estimates in Feingold *et al.* (2003) confirm that the relationship between aerosol and cloud droplet number concentrations is non-linear, e.g., $N_d \sim (N_a)^b$ where N_d is the cloud drop number density and N_a is the aerosol number concentration. The parameter *b* in this relationship can vary widely, with values ranging from 0.06 to 0.48, highlighting the difference in aerosol characteristics (low values of *b* correspond to low hygroscopicity). Penner *et al.* (2004) use a simple parcel model to represent cloud droplet growth in stratocumulus clouds and suggest that the different relationship observed between cloud optical depth and liquid water path in clean and polluted clouds can be explained by the difference in sub-cloud aerosol particle distributions.

Data obtained from different field experiments in regions of high aerosol loading give ambiguous results. Some studies indicate an increase in cloud reflectance for enhanced ambient aerosol concentrations (Brenguier *et al.,* 2000b; Brenguier *et al.,* 2000a; Rosenfeld *et al.,* 2002). In contrast, the study of (Jiang *et al.,* 2002) shows that high pollution entrained into clouds led to a decrease in the liquid water path and a reduction in the observed cloud reflectance, also suggested in the results of (Brenguier *et al.,* 2003). Han *et al.* (2002) analyze the cloud climatology derived from AVHRR, for warm clouds during daytime to examine the premise that enhanced aerosol concentrations will lead to larger number concentrations and smaller droplets while maintaining the liquid water content constant. The liquid cloud sensitivity can be defined as the change in liquid water path (LWP) as a function of the column-averaged droplet number concentration (N_c). The results indicate that for warm clouds with optical depths between 1 and 15, the sensitivity is close to zero in only 1/3 of the cases, which corresponds to decreasing effective radius for increasing N_c, independent of LWP. The results highlight the difficulty of devising observational studies that can isolate the processes in the real world. The feedbacks between aerosol and clouds make it difficult to determine a clear enhancement in reflectance under high pollution conditions.

Satellite retrievals combined with a chemical transport model in the case of two pollution episodes over the Atlantic indicate the brightening of clouds over a timescale of a few days in cases of comparable liquid water path (Hashvardhan *et al.,* 2002; Schwartz *et al.,* 2002), suggesting that the Twomey effect may be present in these cases.

Other observational studies using satellite data effects (e.g. Rosenfeld and Woodley*,* 2000) made no attempt to separate the effect of enhanced reflectance from a reduction in precipitation, providing an estimate of the combined cloud albedo and lifetime effects. Observations by the POLDER and AVHRR satellite instruments (Kaufman *et al.,* 2002) have also shown that the reflectance of low level clouds increases with aerosol concentration.

Smoke from sugarcane and forest fires was shown to reduce cloud droplet sizes in early case studies utilising in situ aircraft observations (Warner and Twomey*,* 1967; Eagan *et al.,* 1974). More recently and on a regional scale, studies have shown that heavy smoke from forest fires in the Amazon basin led to increased cloud droplet number concentrations and to reduced cloud droplet sizes (Reid *et al.,* 1999; Andreae *et al.,* 2004). Satellite observations of ice crystal effective diameter and water vapour content from HALOE (Halogen Occultation Experiment) in the lower stratosphere have shown that there is a strong negative statistical correlation in biomass burning regions (Sherwood*,* 2002). The enhanced aerosol concentrations associated with biomass burning are likely causing smaller ice crystals in the convective clouds that develop and those smaller crystals evaporate more readily and lead to the observed increase in relative humidity in the lower stratosphere.

Some satellite data suggest much larger effective radii in remote oceanic regions than in highly polluted continental areas, varying from 14 down to 6 μm (Bréon *et al.,* 2002; Quaas *et al.,* 2004). The rather low spatial and temporal resolution of the satellite data could mask in a single measurement, aerosol with different properties. Together with the absence of liquid water path measurements, it handicaps the inferences from such studies and hinders an accurate analysis and estimate of the indirect cloud albedo RF.

2.4.6.1.2 New evidence related to modified cloud cover

Observations from MODIS and MISR have been used to conclude that the aerosol indirect effect is likely primarily due to an increase in cloud cover, rather than an increase in cloud albedo (Kaufman *et al.,* 2005).

Cloud cover has been shown to decrease when biomass burning aerosols inhibit the formation of low clouds (Koren *et al.,* 2004). For values of the aerosol optical depth above 1.2, very few low-lying clouds were observed in a large area with high biomass burning aerosol loading. Likewise increasing emissions of absorbing aerosols from the late 1980s to the late 1990s in China caused a reduction in cloud amount which resulted in a decrease of the local planetary albedo, as deduced from satellite data (Krüger and Graßl*,* 2004).

2.4.6.1.3 New evidence related to basic physics and chemistry of CCN and IN

The last few years have witnessed a large increase of studies highlighting the importance of aerosol particle composition in the activation process, particularly the organic fraction and soot content. The study by Feingold and Chuang (2002) indicates the occurrence of cloud droplet spectral broadening due to organic coating on CCN, delaying activation. Nevertheless, earlier observations of fog water (Facchini *et al.,* 2000), suggest that the presence of organic aerosols may reduce surface tension (Ming *et al.,* 2005a) and lead to an increase in the cloud droplet number concentration. In a modelling study performed using observations from the Monterey Area Ship Track experiment, Erlick *et al.* (2001) demonstrate the importance of the drop-size spectrum and the chemical composition associated with this spectrum on the resulting cloud droplet distribution and albedo. The presence of internal mixtures (e.g., sea-salt and organic compounds) can affect the uptake of water and the resulting optical properties compared to a pure sea-salt particle (Randles *et al.,* 2004). The review study of cloud droplet activation by McFiggans *et al.* (2005) highlights the importance of the aerosol organic component in cloud droplet activation, and also point outs the difficulty in explaining a quantitative relationship between aerosol size/composition and drop size distribution.

Airborne aerosol mass spectrometers, whose use has become widespread recently, provide firm evidence that ambient aerosols consist mostly of internal mixtures e.g., biomass-burning components, organics and soot are mixed with other aerosols (Cziczo *et al.,* 2004a). These studies are necessarily limited in space and time but are nevertheless quite illustrative of the complexity of particle composition and their relation to cloud droplet and ice crystal formation. The presence of sea salt, desert dust and meteoritic material inside small ice crystals, as well as sulphates, nitrates and organic compounds in haze and cloud particles, highlights a new perspective in viewing the atmospheric aerosol (e.g., Maria *et al.,* 2004). A question that arises is the degree of internal mixing amongst ambient aerosols in the various sizes – nucleation, accumulation and coarse modes.

The chemical composition of the interstitial particles differs from the nuclei in ice crystals, even though homogeneous freezing was the likely ice formation mechanism. Cziczo *et al.* (2004b) provide laboratory results of homogenous freezing which suggest that organic compounds do not partition equally to the ice and aqueous phases, with organic-rich particles remaining unfrozen, and potentially having an impact on mixed phase clouds developing under anthropogenic influence.

The presence of insoluble particles within the ice crystals will affect the radiation transfer through such crystals. The inclusions of scattering and absorbing particles within large ice crystals has been studied (Macke *et al.,* 1996), suggesting a significant effect when soot particles are embedded leading to an increase in the asymmetry parameter. Inclusions of ammonium sulphate or even air bubbles lead to a decrease in the asymmetry parameter. Given the recent observations of partially insoluble nuclei in ice crystals, there is a need to further develop the theoretical aspects of the radiative transfer and to consider also the transfer in small crystals not included in the referred study.

2.4.6.1.4 Cloud lifetime effect

This effect involves increased concentrations of smaller droplets that lead to a decreased drizzle production and reduced precipitation efficiency, longer lived clouds (Albrecht*,* 1989), increased cloud cover, thickness and cloud height (Pincus and Baker*,* 1994, see Section 2.4.1 and Figure 2.4.1). It has proven difficult to devise observational studies that can separate the cloud lifetime from the cloud albedo effects; thus, observational studies in most instances provide estimates of the combined effects. Similarly, climate modelling studies cannot easily separate the cloud lifetime indirect effect once the aerosol scheme is fully coupled to the cloud microphysics scheme. In this report the cloud lifetime effect is classified as a first-response climate feedback (see Section 2.8).

2.4.6.1.5 Semi-direct effect

This effect relates to the absorption of solar radiation by aerosols that modifies the atmospheric temperature and humidity structure, thereby changing the cloud amount (Hansen *et al.,* 1997; Ackerman *et al.,* 2000; Ramanathan *et al.,* 2001a and see Section 2.8.5 and Figure 2.4.1) It has been modelled both by GCMs and high-resolution cloud resolving models, since it is implicitly included whenever absorbing aerosols are modelled (see Section 2.8). Direct aerosol heating modifies clouds in all GCMs analysed (Hansen *et al.,* 1997; Lohmann and Feichter*,* 2001; Jacobson*,* 2002; Menon *et al.,* 2003; Penner *et al.,* 2003; Cook and Highwood*,* 2004; Hansen *et al.,* 2005). Aerosol heating within cloud layers reduced cloud fractions, whereas aerosol heating above the cloud layer tended to increase cloud fractions. When diagnosed within a GCM framework, the semi-direct effect can also include cloud changes due to circulation effects and/or surface albedo effects. Moreover, the semi-direct effect is not exclusive to absorbing aerosol, as potentially any radiative heating of the mid-troposphere can produce a similar response in a GCM (Hansen *et al.,* 2005 see also Section 2.8). Cloud resolving models of cumulus and stratocumulus case-studies also diagnose semi-direct effects and they indicate a similar relationship between the height of the aerosol layer relative to the cloud and the sign of the semi-direct effect (Ackerman *et al.,* 2000; Ramanathan *et al.,* 2001a; Johnson *et al.,* 2004; Johnson*,* 2005). Johnson (2005) points out that the necessarily coarse resolution of GCM cloud schemes means that they may incorrectly model such effects, thus global estimates of the semi-direct effect should be viewed with caution. In this report the semi-direct effect is classified as a first-response climate feedback (see Section 2.8).

2.4.6.2 Estimates of the radiative perturbations from aerosol-cloud interactions

2.4.6.2.1 Cloud albedo indirect effect

General circulation models are used to estimate the RF associated with the indirect effect of aerosols on a global scale, considering preindustrial and present-day top of the atmosphere radiation budgets. Since the TAR, the effect has been estimated in a more systematic and rigorous way, and many more modelling results are available. These modelling studies are limited partially due to the underlying uncertainties in aerosol emissions (e.g. emission rates of primary particles, of secondary particle precursors). The main problem in the comparison between model results still resides in the formulation of the relationships between aerosol particle concentrations and cloud droplet or ice crystal populations, and the impact of microphysical changes on the optical properties of clouds (McFiggans *et al.,* 2005).

The global mean RF due to the cloud albedo indirect effect is estimated from the climate models by the difference in the results from simulations under preindustrial conditions (including only natural particle sources) and current emissions of gas-precursors and primary particles.

The range of model estimates for the cloud albedo indirect RF varies from –0.5 to –1.9 W m^{-2}. All models indicate a negative RF, which on average is –1.18 W m^{-2}, with 0.45 W m^{-2} as standard deviation from the mean. Figure 2.4.4 presents the range of results from several climate model simulations.

There are considerable differences in the treatment of aerosol processes, cloud processes and the aerosol-cloud interactions in these models, that should be mentioned, in order to understand some of the differences in the mean RF estimates shown. Most models include an interactive sulphur cycle and anthropogenic aerosol particles composed of sulphate, as well as naturally producing sea-salt, dust and continuously outgassing volcanic sulphate aerosols (the main effect of volcanic aerosol emission is disposition of aerosols into higher levels of the atmosphere). Only the study by Lohmann *et al.* (2000) includes internally mixed sulphate, black and organic carbon, sea salt and dust aerosols, resulting in the smallest estimate of the cloud albedo indirect effect. Table 2.4.6 describes some of the details of the different models, including the aerosol species included and the parameterized treatment of the aerosol-cloud interaction to determine the albedo indirect effect. The table also shows (when available), the combined indirect effect (albedo and lifetime).

[INSERT TABLE 2.4.6 HERE]

Figure 2.4.4 present the results of the albedo effect according to the type of aerosol species included. Note that the studies with only sulfate and sea-salt (and one that also includes OC from biomass, (Menon *et al.,* 2002b), show much less scatter resulting in an average of –1.436 ± 0.078. In contrast, the studies that include more species, show larger variability (–0.9683 ± 0.5345). Note however that the two studies with most negative RFs use the NCAR CCM, while the others are from four different model results. [Other models will be added.]

[INSERT FIGURE 2.4.4 HERE]

Modelling results also indicate that the mean RF due to the indirect effect is on average somewhat larger over land than over oceans, but over oceans there is a more consistent response from the different models, resulting in a smaller standard deviation.

2.4.6.2.2 Cloud lifetime indirect effect

The cloud lifetime effect varies considerably between the different models (–0.3 and –1.4 W m^{-2}), resulting in an average forcing of –0.7 W m^{-2} and a standard deviation of 0.5 W m^{-2} (Lohmann and Feichter*,* 2005). This effect is included in the efficacy term as part of the climate response (see Section 2.8).

2.4.6.2.3 Semi-direct effect

Estimates for the semi-direct effect range from +0.1 to –0.5 W m^{-2}. The variations arise from different locations of black carbon with respect to the cloud (see Section 2.4.6.1.5).

The estimates of these indirect effects in terms of changes in the radiative fluxes at the top of the atmosphere (F_TOA) and at the surface (F_SFC), from the results of the climate simulations discussed in the context of Figure 2.4.4 are summarised in Table 2.4.6. The significance of reporting the radiative fluxes at the surface resides in the fact that it is possible to obtain near zero TOA radiative fluxes that are the result of a slight imbalance between surface and tropospheric fluxes. It is particularly important for the case of absorbing aerosols in the troposphere (such as over the Indian Ocean and in large biomass burning areas). The vertical distribution of the heating cannot be accounted for by the TOA radiative fluxes alone as the surface temperature changes may not respond directly to those TOA fluxes in certain cases and the vertical stratification may be modified by the presence of absorbing aerosols. Chapter 7 further discusses these concepts.

It is not possible to obtain a best estimate of the total indirect aerosol effect from preindustrial times to present-day solely from observations. The satellite record is not long enough and other existing long-term records do not provide the aerosol and cloud microphysical properties needed for such an assessment. Climate models by themselves have weaknesses that could bias the indirect effect. Thus, to obtain a best estimate of the indirect aerosol effect, measurements and models should be combined. In a first approach, using this method, Lohmann and Lesins (2002) obtained a total indirect effect of –0.85 W m^{-2}, which falls within the range of the indirect effect as estimated from inverse simulations (Anderson *et al.,* 2003). By applying the cloud droplet number – fine mode aerosol optical depth relationship from MODIS in two GCMs the total indirect effect is reduced even further to –0.3 to –0.5 W m^{-2} (Quaas *et al.,* 2005).

2.4.6.4 Uncertainties

Modelling the cloud albedo indirect and cloud lifetime effects from first principles is difficult because the representation of aerosol-cloud interactions and of clouds themselves in climate models is still somewhat crude (Lohmann and Feichter*,* 2005). Model intercomparisons (e.g., Lohmann *et al.,* 2001; Menon *et al.,* 2003) suggest that the predicted cloud distributions vary significantly, particularly their horizontal and vertical extents, since the vertical resolution and parameterization of convective and stratiform clouds can be very different between models.

Even though the spread in the magnitude of the RF due to the cloud albedo indirect effect has been reduced substantially since the TAR, it is still somewhat difficult to compare directly the results from the different models as uncertainties are not well understood. Uncertainties may be underestimates as all models could be suffering from similar biases. Another uncertainty is that models do not often quote the statistical significance of the RF estimates. Ming *et al.* (2005b), for example, demonstrate that it is only in the midlatitude Northern Hemisphere that their model yields a statistically significant result at the 95% confidence level when compared to the unforced model variability. There are also large differences in the way that the different models treat the appearance and evolution of aerosol particles and the subsequent cloud droplet formation. Further, these models have considerable differences in the horizontal and vertical resolution, which introduce uncertainties in their ability to accurately represent the shallow warm cloud layers over the oceans most susceptible to show the changes due to anthropogenic aerosol particles. Chemical composition and size distribution spectrum are also two factors that likely are insufficiently understood on a fundamental microphysical level. Above all, comparisons with observations have not reached the same degree of testing as, say, for the direct RF estimates; this is not just due to model limitations, for also the observational basis has not yet reached a sound footing.

The observational evidence indicates that aerosol particles in nature tend to be composed of several compounds and are typically internally mixed. Such conditions are difficult to simulate and may lead to differences in the results obtained from the different climate models. The calculation of the cloud albedo indirect effect mostly ignores nuances arising from the particle chemical composition and state of the mixture (external vs. internal). The relationship between ambient aerosol particle concentrations and the resulting cloud droplet size distribution is important during the activation process, which has to be parameterised in the climate models. It is treated in different ways in the different models, ranging from simple empirical functions (Menon *et al.,* 2002b), to more physical parameterisations that tend to be more costly computationally (Abdul-Razzak and Ghan*,* 2002; Nenes and Seinfeld*,* 2003; Ming *et al.,* 2005a; Storelvmo *et al.,* 2005).

All climate models discussed above include sulphate particles; some models produce them from gaseous precursors over oceans, where ambient concentrations are low; some models only condense mass onto pre-existing particles over the continents. Some other climate models also include sea-salt and dust particles produced naturally, typically parameter zing particle production in terms of wind speed. Some models include anthropogenic nitrate, black carbon and organic compounds, which in turn affect activation. So far, no climate model includes natural biogenic particles. Even without considering biases in the modelled-generated clouds, these differences in the aerosol chemical composition and the subsequent treatment of activation lead to uncertainties that are difficult to quantify. The presence of organic carbon owing to is distinct hygroscopic and absorption properties can be particularly important for the indirect effect in the tropics (Ming *et al.,* 2005b).

Further uncertainties may be due to changes in the droplet spectral shape, typically considered invariant in climate models under clean and polluted conditions but which can be substantially different in typical atmospheric conditions (e.g., Erlick *et al.,* 2001; Liu and Daum*,* 2002). Liu and Daum (2002) estimated that a 15% increase in the droplet number concentration can lead to a reduction of between 10 and 80% in the estimated RF of the cloud albedo indirect effect. Peng and Lohmann (2003) and Rotstayn and Liu (2003) studied the sensitivity of their estimates to this dispersion effect, confirming that their estimates of the cloud albedo effect without taking the droplet spectra change into account had overestimated the RF by 15% and 15–35%, respectively.

2.5 Surface Changes

2.5.1 Introduction

Anthropogenic changes to the physical properties of the land surface can perturb the climate, both by exerting a RF and by modifying other processes such as the fluxes of latent and sensible heat and the transfer of momentum from the atmosphere. In addition to contributing to changes in greenhouse gas concentrations and aerosol loading, anthropogenic changes in the large-scale character of the vegetation covering the landscape (“land cover”) can affect the physical properties such as surface albedo. The albedo of agricultural land can be very different to that of a natural landscape, especially if the latter is forest. The albedo of forested land is generally lower than that of open land because the greater leaf area of a forest canopy and multiple reflections within the canopy result in a higher fraction of incident radiation being absorbed. Changes in surface albedo change induce a RF of climate by perturbing the shortwave radiation budget (Ramaswamy *et al.,* 2001). The effect is particularly accentuated when snow is present (Betts*,* 2000), because open land can become entirely snow-covered and hence highly reflective whilst trees can remain exposed above the lying snow. Even a snow-covered canopy exhibits a relatively low albedo as a result of multiple reflections within the canopy (Harding and Pomeroy*,* 1996). Surface albedo change may therefore provide the dominant influence of mid- and high-latitude land cover change on climate (Betts*,* 2001; Bounoua *et al.,* 2002). The TAR cited two estimates of RF due to anthropogenic land cover-induced albedo change relative to potential natural vegetation (PNV) of -0.4 W m^{-2} and -0.2 W m^{-2}, and assumed that the RF relative to 1750 was half of that relative to PNV, so gave a central estimate of the RF due to surface albedo change of -0.2 Wm^{-2} ± 0.2 W m^{-2}.

Surface albedo can also be modified by the settling of anthropogenic aerosols on the ground, especially in the case of black carbon on snow (Hansen and Nazarenko*,* 2004). This mechanism may be considered to be a RF mechanism because diagnostic calculations may be performed under the strict definition of RF (Section 2.8).

Land cover change can also affect other physical properties such as surface emissivity, the flux of moisture through evaporation, the ratio of latent to sensible heat fluxes (the Bowen ratio) and the aerodynamic roughness which exerts frictional drag on the atmosphere and also affects turbulent transfer of heat and moisture. All these processes can affect the air temperature near the ground and also modify humidity, precipitation and windspeed. Direct human perturbations to the water cycle, such as irrigation, can affect surface moisture fluxes and hence the surface energy balance. Changes in vegetation cover can affect the production of dust, which then exerts a RF. Changes in certain gases, particularly CO_2 and O_3, can also exert an additional RF of climate through their effects on the Bowen ratio through plant responses which affect transpiration. While such processes will act as anthropogenic perturbations to the climate system (Pielke Sr. *et al.,* 2002) and will fall at least partly within the “forcing” component of the forcing-feedback-response conceptual model, it is difficult to unequivocally quantify the pure forcing component as distinct from feedbacks and responses. The term “non-radiative forcing” has been proposed (Jacob *et al.,* 2005) and this report adopts the same term, but no quantitative metric separating forcing from feedback and response has yet been implemented for climatic perturbation processes which do not act directly on the radiation budget.

Energy consumption by human activities, such as heating of buildings, powering of electrical appliances and combustion of fuel by vehicles, can directly release heat into the environment. This was not discussed in the TAR. Anthropogenic heat release is not a RF in that it does not directly perturb the radiation budget and was not discussed in the TAR, so is here referred to as an “effect”. It can, however, be quantified as a direct input of energy to the system in terms of W m^{-2}.

2.5.2 Changes in Land Cover Since 1750

In 1750, 12.52 million km^2 (0.096% of the global land surface) were under cultivation or pasture (Figure 2.5.1), mainly in Europe, the Indo-Gangetic Plains and China (Ramankutty and Foley, 1999; Klein Goldewijk, 2001).Over the next hundred years, croplands and pasture expanded and intensified in these areas, and new agricultural areas emerged in North America. The period 1850–1950 saw a more rapid rate of increase of cropland and pasture areas. In the last 50 years, several regions of the world have seen cropland areas stabilize, and even decrease (Figure 2.5.1) In the U.S., as cultivation shifted from the east to the Midwest, croplands were abandoned along the eastern seaboard around the turn of the century, the eastern forests have undergone a regeneration over the last century. Similarly, croplands areas have decreased in China and Europe. Overall, global cropland and pasture expansion was slower since 1950 than before. However, deforestation is occurring more rapidly in the tropics. Latin America, Africa, and South and Southeast Asia experienced slow cropland expansion until the 20th century, but have seen exponential increases in the last 50 years. China had a steady expansion of croplands throughout most of the last three centuries. By 1990, croplands and pasture covered 49.31 million km^2 (37.8% of global land), and forest cover had decreased by 10.5 million km^2 (Ramankutty and Foley, 1999; Klein Goldewijk, 2001).

[INSERT FIGURE 2.5.1]

Overall, most deforestation until the mid-20th Century had occurred in the temperate regions (Figure 2.5.1). However, in more recent decades, land abandonment in western Europe and North America is leading to reforestation while deforestation is now progressing rapidly in the tropics.

2.5.3 Radiative Forcing by Anthropogenic Surface Albedo Change

Since the TAR, a number of estimates of the RF over the industrial era have been made. Matthews *et al.* (2003) and Brovkin *et al.* (2005) estimated the global mean RF relative to 1700 to be –0.15 W m^{-2} and –0.14 W m^{-2} respectively, considering only cropland changes (Ramankutty and Foley, 1999) and not pastures. Hansen *et al.* (2000) also considered only cropland changes (Ramankutty and Foley, 1999) and simulated the forcing relative to 1880 to be –0.09 W m^{-2}. Using historical reconstructions of both croplands (Ramankutty and Foley, 1999) and pasturelands (Klein Goldewijk, 2001), Betts *et al.* (2005) simulated RFs of and –0.18 W m^{-2} since 1750. This study also estimated the RF relative to PNV to be 0.24 W m^{-2}.

Other studies since the TAR have also estimated the RF at the present day relative to PNV. Govindasamy *et al.* (2001) estimated the RF as –0.08 W m^{-2}. Myhre *et al.* (2005) used land cover and albedo data from MODIS (Friedl *et al.,* 2002) and estimated the RF as –0.03 W m^{-2}. The results of Betts *et al.* (2005) suggest that the RF relative to 1750 is approximately 0.75 of that relative to PNV. Therefore by employing this factor published RFs relative to PNV can be used to estimate the RF relative to 1750.

In all the published studies, the RF showed a very high degree of spatial variability, with some areas showing no RF at 1990 relative to 1750 while values more negative than –5 W m^{-2} are typically seen in the major agricultural areas of North America and Eurasia. In historical simulations, the spatial patterns of RF relative to the potential natural vegetation remain generally similar over time, with the regional RFs at 1750 intensifying and expanding in the area covered. The major new areas of land cover change since 1750 are North America and central and eastern Russia.

Changes in the underlying surface albedo could affect the RF due to aerosols if such changes took place in same regions. Similarly, surface albedo forcing may depend on aerosol concentrations. Estimates of time evolution of aerosol forcings and surface albedo forcings may need to consider changes in each other.

2.5.3.1 Uncertainties

Uncertainties in estimates of RF due to anthropogenic surface albedo change arise from several factors.

(i) Land use change are due to the characterisation of both the present-day vegetation and the reference historical state. The forcing estimates reported in the TAR used atlas-based datasets for present-day vegetation (Matthews, 1983; Wilson and A.Henderson-Sellers, 1985). More recent datasets of land cover have been obtained from satellite remote sensing. Data from the Advanced Very High Resolution Radiometer (AVHRR) in 1992–1993 were used to generate two global land cover datasets at 1km resolution using different methodologies (Hansen and Reed, 2000; Loveland *et al.,* 2000) The IGBP-DIS dataset is used as the bases for global cropland maps (Ramankutty and Foley, 1999) and historical reconstructions of croplands, pasture and other vegetation types (Ramankutty and Foley, 1999; Klein Goldewijk, 2001). The Moderate Resolution Imaging Spectrometer (MODIS – Friedl *et al.,* 2002) provides another product. The two interpretations of the AVHRR data agree on the classification of vegetation as either tall (forest and woody savannah) or short (all other land cover) over 84% of the land surface (Hansen and Reed, 2000). However, some of the key disagreements are in regions subject to anthropogenic land cover change so may be important for the estimation of anthropogenic RF. In the HadAM3 GCM, the estimate of RF relative to PNV varied from –0.2 W m^{-2} with the Wilson and Henderson-Sellers (1985) atlas-based land use dataset to –0.24 W m^{-2} with a version of the Wilson and Henderson-Sellers (1985) dataset adjusted to agree with the cropland data of Ramankutty and Foley (1999) (Betts *et al.,* 2005). Myhre and Myhre (2003) found the RF relative to PNV to vary from –0.66 W m^{-2} to 0.29 W m^{-2} according to whether the present-day land cover was from Wilson and Henderson-Sellers (1985), Ramankutty and Foley (1999) or other sources.

(ii) Reconstructions of historical land use states require information or assumptions regarding the nature and extent of land under human use and also the nature of the PNV. Ramankutty and Foley (1999) reconstructed fraction of land under crops at 0.5º resolution from 1700 to 1990 by combining the IGBP Global Land Cover Dataset with historical inventory data, assuming that all areas of past vegetation occur within areas of current vegetation. Klein Goldewijk (2001) reconstructed all land cover types from 1700 to 1990, combining cropland and pasture inventory data with historical population density maps and PNV. Klein Goldewijk used a Boolean approach which meant that crops, for example, covered either 100% or 0% of a 0.5º grid box. The total global cropland of Klein Goldewijk is generally 25% less than that reconstructed by Ramankutty and Foley (1999) throughout 1700 to 1990. At local scales the disagreement is greater due to the high spatial heterogeneity in both datasets. Large-scale PNV is reconstructed either with models or by assuming that small-scale examples of currently-undisturbed vegetation are representative of the PNV at the large scale.

(iii) Parameterizations of the surface radiation processes are subject a number of uncertainties. The albedo for a given land surface or vegetation type may either be prescribed or simulated on the basis of more fundamental characteristics such as vegetation leaf area. But either way, model parameters are set on the basis of observational data which may come from a number of conflicting sources. Both the AVHRR and MODIS instruments have been used to quantify surface albedo for the IGBP vegetation classes in different regions and different seasons, and in some cases the albedo for a given vegetation type derived from one source can be twice that from the other (e.g., Strugnell *et al.,* 2001; Myhre *et al.,* 2005). Myhre and Myhre (2003) examined the implications of varying the albedo of different vegetation types either together or separately, and found the RF relative to PNV to vary from –0.65 W m^{-2} to positive 0.47 W m^{-2}; however, the positive RFs occurred in only a few cases and resulted from large reductions in surface albedo in semi-arid regions on conversion to pasture, so were considered unrealistic by the study's authors. The single most important factor for the uncertainty in the study by Myhre and Myhre (2003) was found to be the surface albedo for cropland. In simulations where only the cropland surface albedo was varied between 0.15, 0.18, and 0.20 it resulted in radiative forcing of –0.06, –0.20, and –0.29 W m^{-2}, respectively. Similar result was found in (Matthews *et al.,* 2003) where simulations were performed for cropland surface albedo of 0.17 to 0.20 with corresponding radiative forcing of –0.15 and –0.28 W m^{-2}, respectively.

(iv) When climate models are used to estimate RF, uncertainties in other parts of the model also affect the estimates. In particular, the simulation of snow cover affects the extent to which land cover changes affect surface albedo. Betts (2000) estimated that the systematic biases in snow cover in HadAM3 introduce errors of up to approximately 10% in the simulation of local RF due to conversion between forest and open land. Such uncertainties may be reduced by the use of an observational snow climatology in a model which just treats the radiative transfer (Myhre and Myhre, 2003). The simulation of cloud cover affects the extent to which the simulated surface albedo changes impact on planetary albedo – too much cloud cover could diminish the contribution of surface albedo changes to the planetary albedo change.

On the basis of the 9 studies assessed here, which together present 24 new estimates since the TAR, our assessment is that the central estimate of RF relative to 1750 due to land-use related surface albedo change should remain at –0.2 W m^{-2}. However, the uncertainty bounds have now expanded to ±0.2 W m^{-2}, with a slight possibility of a positive RF, although very unlikely.

2.5.4 The Radiative Forcing of Black Carbon in Snow Ice

The presence of soot particles in snow could cause a decrease in the albedo of snow and affect snowmelt. Initial estimates by Hansen *et al.* (2000) suggested that black carbon could thereby exert a positive RF of +0.2 W m^{-2}. This estimate was refined by Hansen and Nazarenko (2004) who used measured BC concentrations within snow/ice at a wide range of geographic locations to deduce the perturbation to the surface and planetary albedo deriving a global mean adjusted RF of +0.15 W m^{-2}. The uncertainty in this estimate is substantial owing to whether BC and snow particles are internally or externally mixed, to uncertainties in BC and snow particle shapes and sizes, to voids within BC particles, and to uncertainties in the BC imaginary refractive index and is estimated as a factor of three. Jacobson (2004) developed a global model that allows the BC aerosol to enter snow via precipitation and dry deposition thereby modifying the snow albedo and emissivity and found modelled concentrations of BC within snow in reasonable agreement with those from many observations. Jacobson (2004) modelled a decrease in the global albedo by 0.4% globally and 1% in the Northern hemisphere, which would imply a significant positive global RF estimated at around +0.25 W m^{-2}. Hansen *et al.* (2005) allowed the albedo change to be proportional to local BC deposition according to Koch (2001) and presented a further revised estimate of 0.08 W m^{-2}. They also suggest that this RF mechanism produces a greater temperature response by a factor of 1.7 than an equivalent CO2 RF i.e. the 'efficacy' may be higher for this RF mechanism (Section 2.8.5.7). This report adopts a best estimate of +0.10 W m^{-2} and a factor of three uncertainty, primarily based on the previous Hansen *et al.* results.

2.5.5 Other Effects of Anthropogenic Changes in Land Cover

Anthropogenic land use and land cover change can also modify climate through other mechanisms, some directly perturbing the Earth radiation budget and some perturbing other processes. Land use can modify the emissions of mineral dust which exerts a RF (Section 2.4.5.6), and is also often accompanied by irrigation (Section 2.3.8.2). Land cover change itself can also modify the surface energy budget through changes in the fluxes of latent and sensible heat, and Gordon *et al.* (2005) suggest that net global deforestation has decreased evaporative fluxes by more than the increase caused by irrigation. Model results suggest that the combined effects of past tropical deforestation may have exerted regional warmings of approximately 0.2 K relative to PNV, and may have perturbed the global atmospheric circulation affecting on regional climates remote from the land cover change (Chase *et al.,* 2000; Zhao *et al.,* 2001; Pielke Sr. *et al.,* 2002)

Since the dominant aspect of land cover since 1750 has been deforestation in temperate regions, the overall effect of anthropogenic land cover change on global temperature will depend largely on the relative importance of increased surface albedo in winter and spring (exerting a cooling) and reduced evaporation in summer and in the tropics (exerting a warming) (Bounoua *et al.,* 2002). Estimates of global temperature responses from past deforestation vary from 0.01 K (Zhao *et al.,* 2001) to –0.25 K (B Govindasamy *et al.,* 2001; Brovkin *et al.,* 2005). If cooling by increased surface albedo dominates, then the historical effect of land cover change may still be adequately represented by RF. With tropical deforestation becoming more significant in recent decades, RF will be less useful as a metric of climate change induced by land cover change recently and in the future.

2.5.6 Anthropogenic Heat Release

Urban heat islands result partly from the physical properties of the urban landscape and partly from the release of heat into the environment by the production of energy by human activities such heating of buildings and the powering of appliances and vehicles (Human Energy Production, HEP). The global total HEP heat flux is estimated as 0.03 W m^{-2} (Nakicenovic*,* 1998). If this energy release were concentrated in cities, which are estimated to cover 0.046% of the Earth's surface (Loveland *et al.,* 2000) the mean local heat flux in a city would be 54 W m^{-2}. Daytime values in central Tokyo typically exceed 400 W m^{-2} with a maximum of 1590 W m^{-2} in winter (Ichinose *et al.,* 1999). Although HEP is a small influence at the global scale, it may be very important for local climate changes in cities (Betts and Best*,* 2004; Crutzen*,* 2004).

2.5.7 Effects of CO_2 changes on Plant Physiology

As well as exerting a RF on the climate system, increasing concentration of atmospheric CO_2 may also perturb climate through direct effects on plant physiology. A number of studies have shown that plant stomata open less under higher CO_2 concentrations (Field *et al.,* 1995), which directly reduces the flux of moisture from the surface to the atmosphere through transpiration (Sellers *et al.,* 1996). The occurrence of this on a large scale could have a significant impact on the surface water balance of the landscape, affecting runoff and the supply of moisture to the atmosphere. A decrease in moisture flux modifies the surface energy balance, increasing the ratio of sensible heat flux to latent heat flux and therefore warming the air near the surface (Sellers *et al.,* 1996; Betts *et al.,* 1997). It would also be expected to reduce atmospheric water vapour causing a negative RF, but no estimates of this have been made. (Sellers *et al.,* 1996) propose the term "physiological forcing" for this mechanism, but such changes in the surface energy budget are subject to the same difficulties of quantification as those arising from irrigation (Section 2.3.8.2).

Although no studies have yet explicitly quantified the temperature response to physiological forcing at the present-day, Gedney *et al.* (2005) find that a perturbation to the global hydrological cycle through this mechanism is detectable in river flow records so a perturbation to surface temperatures may have occurred through this mechanism. Modelling studies suggest that doubling CO_2 would lead to a warming of 0.4 K–0.7 K over land due to CO_2-induced stomatal closure. With CO_2 having risen by approximately 35% relative to preindustrial, this may suggest that this process may have contributed 0.1 K to 0.2 K to the temperature rise over land. Further details of this process and its roles in the climate system are given in Chapter 7.

2.6 Contrails and Aircraft-Induced Cloudiness

2.6.1 Introduction

The IPCC separately evaluated the RF (RF) of climate by subsonic and supersonic aircraft in the Special Report on Aviation and the Global Atmosphere (IPCC*,* 1999), hereafter designated as IPCC-1999. Like many other industrial sectors, subsonic aircraft operations around the globe contribute directly and indirectly to the RF of climate change. Here we only assess the RFs that are unique to the aviation sector, namely the formation of persistent condensation trails (contrails) and the impact on cirrus cloudiness. The other effects associated with aviation emissions are included in the atmospheric changes discussed in Sections 2.3 and 2.4. Persistent contrail formation and induced cloudiness are indirect RF effects from aircraft operations that depend on humidity and temperature conditions along aircraft flight tracks. Thus, future changes in atmospheric humidity and temperature distributions in the free troposphere will have consequences for aviation cloudiness. Aviation aerosol also can potentially alter the properties of clouds that form later in air containing aircraft emissions.

2.6.2 Radiative-Forcing Estimates for Persistent Line-Shaped Contrails

Aircraft produce condensation trails (contrails) in the upper troposphere where humidity and temperature conditions are suitable. Contrails are thin cirrus clouds, which reflect solar radiation and trap outgoing long-wave radiation. The latter effect is expected to dominate for thin cirrus (Hartmann *et al.,* 1992), thereby resulting in a net positive RF value for contrails. Persistent contrail cover can be calculated globally from atmospheric data (e.g., Sausen *et al.,* 1998) or by using a modified cirrus cloud parameterization in a GCM (Ponater *et al.,* 2002). The associated contrail RF follows from choosing an optical depth for contrails. The global RF values for contrail and induced cloudiness are assumed to vary linearly with fuel use if aircraft flight tracks remain unchanged. The best estimate for the RF of persistent linear contrails for aircraft operations in 2000 is 0.010 W m^{-2} (Table 2.6.1). The value is based on two independent estimates (Myhre and Stordal*,* 2001; Marquart *et al.,* 2003), which have been rescaled for the year 2000 in Sausen *et al.* (2005) to give RFs of 0.006 W m^{-2} and 0.015 W m^{-2}. The two values also serve to set the uncertainty range. This new best estimate is significantly lower than the scaled IPCC-1999 value of 0.034 W m^{-2}. The change results from reassessments of persistent contrail cover using observations, lower optical depth estimates calculated interactively, and including the interaction with other clouds (Marquart and Mayer*,* 2002; Meyer *et al.,* 2002; Ponater *et al.,* 2002; Marquart *et al.,* 2003). The new estimates include diurnal changes in the shortwave solar forcing, which decreases net forcing for a given contrail cover by about 20% (Myhre and Stordal*,* 2001).

[INSERT TABLE 2.6.1 HERE]

2.6.3 Radiative-Forcing Estimates for Aviation-Induced Cloudiness

Individual persistent contrails are routinely observed to shear and spread, covering large additional areas with cirrus cloud (Minnis *et al.,* 1998). Aviation aerosol could also lead to changes in cirrus cloud (see Section 2.6.4). Aviation-induced cloudiness (AIC) is defined to be the sum of all changes in cloudiness associated with aviation operations. Thus, an AIC estimate includes persistent contrail cover. A component of AIC necessarily does not have the characteristic linear shape of a persistent contrail and, hence, is indistinguishable from background cirrus. This basic ambiguity, which prevented the formulation of a best estimate of AIC amounts and the associated RF in IPCC-1999, still exists for this assessment. Estimates of the ratio of induced cloudiness cover to that of persistent linear contrails range from 1.8 to 10 (Minnis *et al.,* 2004; Mannstein and Schumann*,* 2005), indicating the uncertainty in estimating AIC amounts. First attempts to quantify AIC used trend differences in cirrus cloudiness between regions of high aviation fuel consumption and low consumption (Boucher*,* 1999). Since IPCC-1999, two studies have confirmed significant positive trends in cirrus cloudiness in regions of high air traffic and found lower to negative trends outside air traffic regions (Zerefos *et al.,* 2003; Stordal *et al.,* 2005). Using the International Satellite Cloud Climatology Project (ISCCP) database, these studies derived cirrus cover trends for Europe of 1 to 2% per decade over the last 1–2 decades. Cirrus trends that could arise from natural variability or climate change could not be accounted for in these studies.

Regional cirrus trends were used as a basis to compute a global mean RF value of 0.030 W m^{-2} for AIC in 2000 (Stordal *et al.,* 2005). This value is not considered a best estimate because of the uncertainty in the optical properties of AIC and in the assumptions used to derive AIC cover. However, this value is in good agreement with an AIC estimate for 1992 of 0.006 to 0.026 W m^{-2} derived from surface and satellite cloudiness observations (Minnis *et al.,* 2004). Without an AIC best estimate, the best estimate of the total RF value for aviation cloudiness (Table 2.9.1 and Figure 2.9.1) includes only that due to persistent linear contrails.

Minnis *et al.* (2004) use their RF estimates for total aviation cloudiness over the USA in an empirical model to show that the surface temperature response for the period 1973–1994 could be as large as the observed surface warming over the USA (~0.3 K per decade). This unexpectedly large impact has not been confirmed in two climate model studies (Hansen *et al.,* 2005; Ponater *et al.,* 2005; Shine*,* 2005). One explanation is that the Minnis *et al.* study used a forcing-response relation derived for the global mean for calculating a regional response (Shine*,* 2005).

Aviation-induced cloudiness has been linked to increases in the diurnal temperature range (DTR) by using surface observations made during the period when all USA air traffic was grounded for several days starting on 11 September 2001 (Travis *et al.,* 2002; Travis *et al.,* 2004). The Travis *et al.* studies show that during this period: (i) DTR was enhanced across the conterminous USA, with increases in the maximum temperatures that were not matched by increases of similar magnitude in the minimum temperatures, and (ii) the largest DTR changes corresponded to regions with the greatest contrail cover. The Travis *et al.* conclusions are based on a correlation rather than a physical model and rely (necessarily) on very limited data. Unusually clear weather across the USA during the shutdown period also has been proposed to account for the observed DTR changes (Kalkstein and Balling Jr.*,* 2004). Thus, more evidence and a physical model are needed before this unexpected relationship between regional contrail cover and DTR can be considered accurate.

2.6.4 Aviation Aerosols

Global aviation operations emit aerosols and aerosol precursors into the upper troposphere and lower stratosphere (IPCC*,* 1999; Hendricks *et al.,* 2004). As a result, aerosol number and/or mass are enhanced in these regions. AIC includes the possible affects of aviation aerosol on cirrus cloudiness amounts. The most important aerosols are those composed of sulphate and black carbon (soot). Sulphate aerosols arise from the emissions of fuel sulphur and black carbon aerosol results from incomplete combustion of aviation fuel. Aviation operations cause enhancements of sulphate and black carbon in the background atmosphere (IPCC*,* 1999; Hendricks *et al.,* 2004). Of concern is that aviation aerosol can act as nuclei in ice cloud formation, thereby altering the microphysical properties of clouds (Jensen and Toon*,* 1997; Kärcher*,* 1999; Lohmann *et al.,* 2004) and perhaps cloud cover. A study by Hendricks *et al.* (2005) shows the potential for significant cirrus modifications by aviation caused by increased numbers of black carbon particles. The modifications would occur in flight corridors and in regions far away from flight corridors. Aviation aerosols either increase or decrease ice nuclei in background cirrus clouds, depending on assumptions about the cloud formation process. Changes in ice nuclei number can alter the radiative properties of cirrus clouds and, hence, their radiative impact on the climate system, similar to the aerosol indirect effects discussed in Section 2.4.6. No estimates are yet available for the global or regional RF changes caused by the effect of aviation aerosol on background cloudiness, although some of the RF from AIC, determined by correlation studies (Section 2.6.3), may be associated with these aerosol effects.

2.7 Solar Variability and Volcanic Activity

2.7.1 Solar Variability

2.7.1.1 Direct observations of solar irradiance

2.7.1.1.1 Satellite measurements of total solar irradiance

Four independent space-based instruments directly measure total solar irradiance (TSI) at the present time, continuing the extant database that is uninterrupted since November 1978 (Fröhlich and Lean*,* 2004). The Variability of Irradiance and Gravity Oscillations (VIRGO) experiment on the Solar Heliospheric Observatory (SOHO) has been operating since 1996, the ACRIM III on the Active Cavity Radiometer Irradiance Monitor Satellite (ACRIMSAT) since 1999, and the Earth Radiation Budget Satellite (ERBS) since 1984. Most recent are the measurements made by the Solar Radiation and Climate Experiment (SORCE) since 2003 (Rottman*,* 2005). From February 2003 to June 2004 SORCE measured an average total solar irradiance of 1361 W m^{-2}, which is 5.2 W m^{-2} lower than measured by the other radiometers (see e.g., the comparisons made by (Lean *et al.,* 2005). This significant difference exceeds the claimed accuracies of the TSI measurements (±0.01% for SORCE). SORCE's solar radiometers employ a new approach of phase sensitive detection in which measurements are made in the frequency, rather than time, domain (Kopp *et al.,* 2005). The US National Institute of Standards and Technology (NIST) is leading an investigation of causes of uncertainty in absolute irradiance values arising from different measurement techniques.

2.7.1.1.2 Observed decadal trends and variability

Three composite records of total solar irradiance, shown in Figure 2.7.1, have been constructed from different combinations of the direct radiometric measurements. The PMOD composite (Fröhlich and Lean*,* 2004) combines the observations by the ACRIM I on the Solar Maximum Mission (SMM), the Hickey-Friedan radiometer on Nimbus 7, ACRIM II on the Upper Atmosphere Research Satellite (UARS) and VIRGO on SOHO by analyzing the sensitivity drifts in each radiometer prior to determining radiometric offsets. In contrast, the ACRIM composite (Willson and Mordvinov*,* 2003), which utilizes ACRIMSAT rather than VIRGO observations in recent times, cross calibrates the reported data assuming that radiometric sensitivity drifts have already been fully accounted for. The Space Absolute Radiometric Reference (SARR) composite uses individual absolute irradiance measurements from the shuttle to cross calibrate satellite records (Dewitte *et al.,* 2005). The gross temporal features of the composite irradiance records are clearly very similar. Evident in each are day-to-week variations associated with the Sun's rotation on its axis, and decadal fluctuations arising from the 11-year solar activity cycle. But the linear slopes (determined using common data between Nov 1978 and Jun 2004) differ among the three different composite records, as do levels at solar activity minima (1986 and 1996). These differences are the result of different cross calibrations and drift adjustments applied to individual radiometric sensitivities when constructing the composites. That residual instrumental drifts are present in the composites is also evident when they are compared with the independent SORCE measurements. From February 2003 to June 2004, a time of overall decreasing solar activity with the approach of solar minimum, TIM's downward slope is 1.55 times that of the PMOD composite, but 0.57 times that of the ACRIM composite (Lean *et al.,* 2005).

[INSERT FIGURE 2.7.1]

Solar irradiance levels are likely comparable in the two most recent cycle minima when absolute uncertainties and sensitivity drifts in the measurements are assessed (Fröhlich and Lean*,* 2004). An upward secular trend in excess of 0.04% over the 27-year period of the irradiance database is likely of instrumental rather than solar origin. This trend, proposed by Willson and Mordvinov (2003), is absent in the PMOD composite, in which total irradiance between successive solar minima is constant to better than 0.01%. Although a long-term trend is present in the SARR composite, the increase of 0.15 W m^{-2} between successive solar activity minima (in 1986 and 1996) is not significant because the uncertainty is ±0.35 W m^{-2} (Dewitte *et al.,* 2005). Furthermore, the irradiance "trend" in the ACRIM and SARR composites is not a slow secular increase but the result of a single episodic increase between 1989 and 1992 that was measured by the Nimbus 7 instrument. Independent, overlapping ERBS observations do not show this increase; nor do they have a significant secular trend (Lee *et al.,* 1995).

Current understanding of solar activity and the known sources of irradiance variability is similarly consistent with comparable irradiance levels during the past two solar minima, and does not support an upward trend in irradiance in the past 25 years. The model of irradiance variability in Figure 2.7.1 illustrates the net effect of sunspot darkening (which decreases irradiance) and facular brightening (which increases irradiance) by combining a record of the global sunspot darkening calculated directly from white light images and the Mg index as a proxy for the facular signal. Neither the sunspot blocking nor the Mg index exhibits a significant secular trend during activity minima. From a theoretical view, changes in surface emissivity by magnetic sunspot and facular regions are the most effective in altering irradiance (Spruit*,* 2000), but other mechanisms have also been proposed. Of these, changes in solar diameter have been considered a likely candidate (e.g.. Sofia and Li*,* 2001). But recent analysis of solar imagery, primarily from the MDI instrument on SOHO, indicate that solar diameter changes are no more than a few km per year during the solar cycle (Dziembowski *et al.,* 2001), for which associated irradiance changes are 0.001%, two orders of magnitude less than the measured solar irradiance cycle.

Since TAR, irradiance time series such as the PMOD composite in Figure 2.7.1 and the 10.7 cm radio flux have been used to advance empirical knowledge of climate responses to forcing by solar variability on annual to decadal time scales. A number of independent analysis have identified tropospheric changes associated with the solar cycle (van Loon and Shea*,* 2000; Douglass and Clader*,* 2002; Gleisner and Thejll*,* 2003; Haigh*,* 2003; White *et al.,* 2003; Coughlin and Tung*,* 2004; Labitzke*,* 2004; Crooks and Gray*,* 2005). Peak-to-peak amplitudes are of order 0.1°C near the surface, increasing to 0.3°C near 10 km. Further analyses have explored the meridional dependence of the atmospheric response to solar forcing (with additional variance related to the NAO and QBO). Overall, the troposphere is warmer and moister during solar maximum, and thickens in response to solar variability with a distinct zonal signature. The strongest response occurs at mid latitudes (40–50°) and near the equator, with sub tropical minima. The primary surface temperature expression of these changes is warming in two mid-latitude bands (increases of 0.5 K at 20–60° N and S) that extend vertically downwards from the lower stratosphere where they expand equatorward (Haigh*,* 2003). The patterns suggest that solar forcing invokes dynamical responses in the troposphere, involving the Hadley, Walker and Ferrel circulation cells, which subsequently impact tropical rainfall (Kodera*,* 2004; van Loon *et al.,* 2004).

2.7.1.1.3 Measurements of solar spectral irradiance

The solar UV spectrum from 120 to 400 nm continues to be monitored from space, with SORCE observations extending those made since 1991 by two instruments on the Upper Atmosphere Research Satellite (Woods *et al.,* 1996). SORCE also monitors, for the first time from space, solar spectral irradiance in the visible and near IR spectrum, providing unprecedented spectral coverage that promises a detailed characterization of solar spectral irradiance variability. Initial results (Harder *et al.,* 2005; Lean *et al.,* 2005) indicate that, as expected, variations occur at all wavelengths, primarily in response to changes in sunspots and faculae. UV spectral irradiance variability in the extended database is consistent with that seen in the UARS observations since 1991, as described in TAR.

Radiation in the visible and IR spectrum has a notably different temporal character during solar rotation than the spectrum below 300 nm. Maximum energy changes occur at wavelengths from 400 to 500 nm. Fractional changes are greatest at UV wavelengths but the actual energy change is considerably smaller than in the visible spectrum. Relative spectral irradiance changes modelled by incorporating the spectral dependence of the theoretical sunspot and facular contrasts (Lean*,* 2000) show overall good agreement with initial SORCE observations but distinct differences are nevertheless evident. During a major episode of solar activity from 17 to 30 October 2003, the model predicts energy changes that are smaller than observed at wavelengths from 400 to 500 nm, and larger than observed at wavelengths from 700 to 1000 nm. Nor are the variations of the spectral features in the region 300 to 400 nm modelled exactly (Lean *et al.,* 2005). The measured and modelled spectral irradiance changes during this time are shown in Figure 2.7.2. As yet the SORCE observations are too short to provide reliable information about the amplitude of solar spectral irradiance changes during the solar cycle. A particular deficiency of the models may be in the spectral region near 1.6 μm where they underestimate facular brightness and predict anti-phase solar cycle changes (Fontenla *et al.,* 2004). Whereas sunspot blocking often dominates facular brightening during episodes of activity such as in October, 2003, and causes decreased irradiance at most wavelengths (Figure 2.7.2), over the longer time scale of the 11-year solar cycle, facular brightness exceeds sunspot blocking by about a factor of two, so that there is an increase in spectral irradiance at most, if not all, wavelengths from the minimum to the maximum of the solar cycle. The cycle estimates from the model (ratio of 1989 to 1986) are 1.3% at 200–300 nm, 0.2% at 315–400 nm, 0.08% at 400–700 nm, 0.04% at 700–1000 nm and 0.025% at 1000–1600 nm.

[INSERT FIGURE2.7.2 HERE]

2.7.1.2 Estimating past solar radiative forcing

2.7.1.2.1 Reconstructions of past variations in solar irradiance

New studies (Lean *et al.,* 2002; Foster*,* 2004; Foukal *et al.,* 2004; Wang *et al.,* 2005) suggest that long-term irradiance changes are notably less over the past four hundred years than in the reconstructions of Hoyt and Schatten (1993), Lean *et al.* (1995), Lean (2000), Lockwood and Stamper (1999) and Fligge and Solanki (2000) that were employed in a number of TAR climate change simulations. In addition to the known 11-year cycle, the latter irradiance reconstructions assume the existence of a long-term variability component such that during the seventeenth century Maunder Minimum total irradiance was reduced from 0.15% to 0.4% (2 to 5 W m^{-2}) below contemporary solar minima. The temporal structure of this long-term component, typically associated with facular evolution, was assumed to track either the smoothed amplitude of the solar activity cycle or its instantaneous period (cycle length). The motivation for adopting a long-term irradiance component was three-fold. Long-term trends in, firstly, the *aa* index and, secondly, the cosmogenic isotopes, combined with, thirdly, the range of variability in Sun-like stars (Baliunas and Jastrow*,* 1990) suggested that the Sun is capable of a broader range of activity than witnessed during recent solar cycles (i.e., the observational record in Figure. 2.7.1). Compared in Table 2.7.1 are different estimates of the reduction in total solar irradiance from current activity minima to the seventeenth century Maunder Minimum.

[INSERT TABLE 2.7.1]

Recent work questions each of the three assumptions and points to long-term total solar irradiance variations a factor of 3 to 4 less than those in TAR. A reassessment of the stellar data has been unable to recover the original bimodal separation of (lower) Ca emission in non-cycling stars (assumed to be in Maunder Minimum type states) compared with (higher) emission in cycling stars (Hall and Lockwood*,* 2004) which underpins the Lean *et al.* (1995) and Lean (2000) irradiance reconstructions. Rather, the current Sun is thought to have "typical" (rather than high) activity relative to other stars. Plausible lowest brightness levels inferred from stellar observations are higher than the peak of the lower mode of the initial distribution of Baliunas and Jastrow (1990). Other studies raise the possibility of long-term instrumental drifts in the *aa* index (Svalgaard *et al.,* 2004), which would reduce somewhat the long-term trend in the current aa index on which the Lockwood and Stamper (1999) irradiance reconstruction is based. Furthermore, simulations of the transport of magnetic flux on the Sun and propagation of open flux into the heliosphere indicate that long-term trends in the *aa* index and cosmogenic isotopes (generated by open flux) do not necessarily imply equivalent long-term trends in solar irradiance (which track closed flux) (Lean *et al.,* 2002; Wang *et al.,* 2005). Conceptually, the excess cosmogenic isotopes in the Maunder and other solar minima relative to the present is associated with reduced and less structured heliospheric magnetic fields as a result of altered open flux but Wang and Sheeley (2003) suggest that with the reduced solar activity the modulation of the interplanetary magnetic field derived from the open flux associated with the very low sunspot numbers is too small to account for the significant fluctuations of ^{10}Be during the Maunder Minimum (Beer *et al.,* 1998), including apparent cyclicity.

Two new reconstructions of solar irradiance (Foster*,* 2004; Wang *et al.,* 2005) derive from solar considerations alone, without invoking geomagnetic, cosmogenic or stellar proxies. From the identification of bright faculae in MDI images Foster (2004) estimates that removing this component would reduce solar irradiance by 1.6 W m^{-2} (model #1 in Table 2.7.1). This estimate of the irradiance of the "non-magnetic" Sun is consistent with an earlier estimate of Lean *et al.* (1992), who inferred a reduction of 1.5 W m^{-2} from a similar analysis of solar Ca K images and fluxes (removal of all network but no alteration of basal cell centre brightness). Both the Foster (2004) and Lean *et al.* (Lean *et al.,* 1992) approaches suggest that if the Maunder Minimum irradiance were equivalent to the "non-magnetic" Sun, then the irradiance reduction from the present would be about half that of earlier estimates, which were adopted for the long-term irradiance reconstructions in TAR.

A quite different approach also suggests that the amplitude of the background component is significantly less than has been assumed, specifically 0.27 times that of Lean (2000). This estimate is the result of simulations of the eruption, transport and accumulation of magnetic flux during the past 300 years using a flux transport model with variable meridional flow (Wang *et al.,* 2005). Variations in both the total flux, and in just the flux that extends into the heliosphere (the "open" flux) are estimated, arising from the deposition of bipolar magnetic regions (active regions) and smaller-scale ephemeral regions on the Sun's surface, in strengths and numbers proportional to the sunspot number. The open flux compares reasonably well with the geomagnetic and cosmogenic isotopes whose variations arise, in part, from heliospheric modulation. This gives confidence that the approach is plausible. A small accumulation of total flux (and possibly ephemeral regions) produces a net increase in facular brightness which, in combination with sunspot blocking, permits the reconstruction of total solar irradiance, shown in Figure 2.7.3. The increase from the Maunder Minimum to the present quiet Sun is ~0.5 W m^{-2} (see Table 2.7.1), i.e., about one third the reduction estimated for the 'non-magnetic" Sun. Compared with this reconstruction in Figure 2.7.3 is a reconstruction by Foster (2004) based on extrapolation of an identified long-term trend in the 11-year smoothed total solar irradiance composite.

In contrast with the Foster (2004) and Wang *et al.* (2005) reconstructions, that of Solanki and Krivova (2005) varies very similarly to that of Lean (2000) because of an adopted ephemeral region contribution of considerable magnitude in addition to the active regions. In their reconstruction, Solanki and Krivova use the sunspot amplitude and cycle length to parameterize, respectively, the sunspot and active region irradiance sources, and the assumed ephemeral region changes. Ephemeral regions cause the upward secular trend and are also invoked as an additional source of open flux to explain why ^{10}Be levels fluctuated during the seventeenth century (Solanki *et al.,* 2002). But evidence for long-term changes in ephemeral regions is highly uncertain. Although postulated to vary in a significant way on long-term scales from a comparison of current solar activity with the distribution of Ca brightness in Sun-like stars (White *et al.,* 1992), Foukal and Milano (2001) did not detect in a few selected Ca K solar images long-term changes in the network, where ephemeral regions mainly reside.

Prior to direct telescopic measurements of sunspots, which commenced around 1610, knowledge of solar activity is inferred indirectly from the ^{14}C and ^{10}Be cosmogenic isotope records in tree-rings and ice cores, respectively. Some studies of cosmogenic isotopes (Jirikowic and Damon, 1994) and spectral analysis of the sunspot record (Rigozo *et al.,* 2001) suggest that solar activity during the twelfth century Medieval Solar Maximum was comparable to the present Modern Solar Maximum Recent work attempts to account for the chain of physical processes in which solar magnetic fields modulate the heliosphere, in turn altering the penetration of the galactic cosmic rays whose flux produces the cosmogenic isotopes which are subsequently deposited in the climate system following additional transport and chemical processes. An initial such effort reported exceptionally high levels of solar activity in the past 70 years, relative to the preceding eight thousand years (Solanki *et al.,* 2004). In contrast, a different study taking account of differences among isotopes records and correcting ^{14}C for fossil fuel burning finds that while current levels of solar activity are historically high, they are not exceptionally so (Muscheler *et al.,* 2005b; Muscheler *et al.,* 2005a).

2.7.1.2.2 Implications for solar radiative forcing

In terms of plausible physical understanding, the most likely secular increase in total irradiance from the Maunder Minimum to current cycle minima is 0.5 W m^{-2} (although in some estimates it is as large as 1.6 W m^{-2} – see Table 2.7.1). Accounting for the 11-year cycle as well, the increase is 1.1 W m^{-2}. From 1750 to the present the net increase in total solar irradiance is estimated to be 0.7 W m^{-2}, according to the 11-year smoothed total solar irradiance time series of Wang *et al.* (2005), shown in Figure 2.7.3. This corresponds to a RF of 0.12 Wm^{-2}, which is more than a factor of two less than the solar RF estimate in TAR, also from 1750 to the present. Using the Lean (2000) reconstruction as an upper limit, the irradiance increase since

1750 is 1.6 W m^{-2}, for which the RF is 0.3 W m^{-2}. The lower limit of the irradiance increase from 1750 to the present is 0.35 W m^{-2} , due to the increase in the 11-year cycle only. The corresponding lower limit of the RF is 0.06 W m^{-2}. As with solar cycle changes, long-term irradiance variations are expected to have significant spectral dependence. For example, the Wang *et al.* (2005) flux transport estimates imply decreases during the Maunder Minimum relative to contemporary activity minima of 0.43% at 200-300 nm, 0.1% at 315–400 nm, 0.05% at 400 to 700 nm, 0.03% at 700–1000 nm and 0.02% at 1000 to 1600 nm (Lean *et al.,* 2005), compared with, respectively, 1.4%, 0.32%, 0.17%, 0.1% and 0.06% in the earlier model of Lean (2000).

[INSERT FIGURE 2.7.3 HERE]

2.7.1.3 Indirect effects of solar variability

The energy changes in the near UV, visible and near IR spectrum (at wavelengths longer than ~300 nm), are considered to provide the most plausible mechanism for solar forcing of climate because they penetrate directly to the troposphere and contribute more than 85% of the total irradiance solar cycle variation. The Earth's atmosphere absorbs about 15 W m^{-2} (~1%) of the Sun's radiant energy, in the ultraviolet portion of the spectrum. Solar UV radiation is more variable than total solar irradiance by at least an order of magnitude. It contributes significantly to changes in total solar irradiance (15% of the total irradiance cycle, (Lean *et al.,* 1997), and creates the ozone layer, but is unavailable for direct forcing of climate because it does not reach the Earth's surface. Since TAR, new studies have confirmed and advanced the plausibility of indirect effects involving the modification of the stratosphere by solar UV irradiance variations, with subsequent dynamical and radiative coupling to the troposphere. Whether solar-induced heliospheric modulation of galactic cosmic rays also contributes an indirect forcing, as discussed in detail in TAR, remains ambiguous

As in the troposphere, anthropogenic effects, internal cycles (e.g., QBO) and natural influences all affect the stratosphere. It is now well established from both empirical and model studies that solar cycle changes in UV radiation alter middle atmospheric ozone concentrations (Fioletov *et al.,* 2002; Geller and Smyshlyaev*,* 2002; Hood*,* 2003), temperatures and winds (Ramaswamy *et al.,* 2001; Labitzke *et al.,* 2002; Haigh*,* 2003; Labitzke*,* 2004; Crooks and Gray*,* 2005) including the QBO (McCormack*,* 2003; Salby and Callaghan*,* 2004). In their recent survey of solar influences on climate, Gray *et al.* (2005) note that updated observational analyses have confirmed earlier 11-year cycle signals in zonally averaged stratospheric temperature, ozone and circulation with increased statistical confidence. There is a solar-cycle induced increase on global total ozone of 2–3%, accompanied by temperature responses that increase with altitude, exceeding 1 K around 50 km. However, the amplitudes and geographical and altitudinal patterns of these variations are only approximately known, and are not linked in an easily discernible manner to the forcing. For example, solar forcing appears to induce a significant and unexpected (from a modelling perspective) lower stratospheric response (Hood*,* 2003).

Increased stratospheric ozone and temperature during solar cycle maxima alters both incoming solar radiation and outgoing IR radiation, producing latitudinal and attitudinal thermal gradients that drive dynamical motions and alter circulation patterns, winds and the large scale planetary waves (Haigh*,* 2001; Rind*,* 2002; Rind *et al.,* 2004). Attendant tropospheric effects (e.g., a weakening and expansion of the Hadley cells, and poleward shift of the Ferrel cells) are manifested in, for example, tropical rainfall (Kodera*,* 2004) and Atlantic storm tracks (Haigh*,* 2001; Haigh *et al.,* 2004) Equatorial winds in the upper stratosphere appear to play an important role in this process because their impact on wind climatology results in modification of wave transport at higher latitudes and thus the structure of the polar lower stratosphere (Matthes *et al.,* 2004). In both observations and model simulations there arise questions of statistical significance but the addition of data during the most recent solar maximum period without major volcanic activity has strengthened the empirical evidence by allowing the solar signal to be more effectively distinguished from the volcanic signal (Grey *et al.*, 2005).

The energy of galactic cosmic rays is one billionth (10^{-9}) of that in the total solar irradiance. Nevertheless, various scenarios have been proposed whereby galactic cosmic rays might influence climate by altering, for example, the tropospheric electric field and cloud cover (as surveyed by Gray *et al.,* 2005). When solar activity is high, the more complex magnetic configuration of the heliosphere reduces the cosmic ray flux.

The approximate 15% modulation of cosmic ray flux by solar activity produces an energy change less than one millionth (10^{-6}) of the energy change in the 0.1% total solar irradiance cycle.

It is supposed that the galactic cosmic rays with sufficient energy to reach the troposphere alter the population of cloud condensation nuclei and hence microphysical cloud properties (droplet number and concentration). In this way, cosmic rays may induce processes analogous to the indirect effect of tropospheric aerosols (Table 1 from Carslaw *et al.,* 2002). Since the plasma produced by cosmic ray ionization in the troposphere is part of an electric circuit that extends from the Earth's surface to the ionosphere, cosmic rays may also affect thunderstorm electrification (Carslaw *et al.,* 2002). Noting the altitude dependence of cosmic ray ionization and precursor gas concentrations, Yu (2002) suggests, furthermore, that solar activity also affects high clouds, in an opposite way to low clouds, and that their respective changes in response to longer-term galactic cosmic ray flux trends may account for differences in surface and tropospheric temperature trends.

Many ambiguities are still to be resolved regarding cloud cover variations and solar activity, including the reality of the decadal signal itself, the phasing or anti-phasing with solar activity, and its separate dependence for low, mid and high clouds, and alternative explanations such as ENSO. Nevertheless, the presence of ions, such as produce by cosmic rays, is recognized as influencing several microphysical mechanisms (Harrison and Carslaw*,* 2003). Aerosols may nucleate preferentially on atmospheric cluster ions. In case of low gas-phase sulphuric acid concentrations, ion-induced nucleation may dominate over binary sulphuric acid-water nucleation (Tegen *et al.,* 2004). Also, increased ion nucleation and increased scavenging rates of aerosols in turbulent regions around clouds seem likely. Because of the difficulty in tracking the influence of one particular modification brought about by ions through the long chain of complex interacting processes, quantitative estimates of the magnitude of galactic cosmic ray-induced changes in aerosol and cloud formation have not been reached.

An unequivocal determination of specific mechanisms – whether direct or indirect - that involve solar variability and climate has yet to be accomplished. As a result, alternate explanations are often proffered for common empirical evidence. For example, an apparent relationship between solar variability and cloud cover has been interpreted as a result of 1) sea surface temperatures altered directly by changing total solar irradiance (Kristjánsson *et al.,* 2002), 2) solar induced changes in ozone (Udelhofen and Cess*,* 2001), 3) internal variability by ENSO (Kernthaler *et al.,* 1999), and 4) changing cosmic ray fluxes modulated by solar activity in the heliosphere (Usoskin *et al.,* 2004). In reality, different direct and indirect physical processes may operate simultaneously.

2.7.2 Volcanic Activity

2.7.2.1 Inferences using observations

Altitude-dependent stratospheric optical observations at a few wavelengths, together with columnar optical and physical measurements, are used to construct the time-dependent global field of aerosol size distribution. Then, using Mie scattering theory, the wavelength-dependent stratospheric aerosol single-scattering characteristics are calculated for the solar and longwave spectrum. These are deployed in a radiation model to calculate the radiative (solar and longwave) perturbations due to stratospheric aerosol variations. Significant perturbations arise in the aftermath of volcanic eruptions. In general, volcanic aerosol RF is considered to be better quantified in terms of a first order understanding, compared to tropospheric aerosol or tropospheric ozone effects. SAR and TAR have documented the episodic nature of the volcanic eruptions and RFs. As noted in TAR, the stratospheric aerosol concentrations are at the lowest concentrations since the advent of global satellite coverage, with no major volcanic eruptions having occurred since the 1991 Pinatubo eruption. The volcanic sulphate aerosols are formed as a result of oxidation of the sulphur gases emitted by explosive volcanic eruptions into the stratosphere. The process of gas-to-particle conversion has an e-folding time of ~35 days and takes typically about 2–3 months. However, there are other kinds of particulates e.g., volcanic ash that are directly emitted during the eruption which consist of siliceous material. These are particles usually larger than 2 μm and sediment out of the stratosphere due to gravity fairly rapidly (within 3 months or so). The siliceous material could also play a role in the radiative perturbations in the immediate aftermath of an eruption, even though it is short-lived and not as long-lasting as the sulphate aerosols. Most of the stratospheric aerosol data incorporated for climate change simulations and analyses tends to be that of the sulphates (Sato *et al.,* 1993; Stenchikov *et al.,* 1998; Ramachandran *et al.,* 2000; Hansen *et al.,* 2002; Ammann *et al.,* 2003).

[INSERT FIGURE 2.7.4 HERE]

Over the past ~2.5 decades several satellite instruments (e.g., SAM II, SAGE I, SAGE II, SAGE III, SME, CLAES, ISAMS, POAM and HALOE) have provided valuable aerosol measurements above the 100 hPa level (nominal tropopause level at the low latitudes). However, so far the only well documented strong volcanic event, by way of reliable and accurate observations, happens to be the Mt. Pinatubo eruption of 1991 in the Philippines. Not all the features of the aerosols following this eruption are well quantified, and extending and improving the available aerosol data sets remains an important problem. Using available satellite and ground based observations Hansen *et al.* (2002) improved a Goddard Institute for Space Studies (GISS) volcanic aerosols data set for the 1850–1999 period (Sato *et al.,* 1993). This has yielded zonal mean vertically resolved aerosol optical depths for visible wave lengths as well as column-average effective radii. Amman *et al.* (2003) developed a similar data set of total aerosol optical depth for the period since 1890. This is based on estimates of atmospheric loadings that are then globally distributed employing a seasonally varying parameterization mainly designed for paleoclimate applications. Amman *et al.* (2003) used fixed effective radius of 0.42 μm for calculating aerosol optical properties; in general, they infer higher values of optical depth than Sato *et al.* (1993). These two data sets have essentially provided the bases for the volcanic aerosol implemented in the current IPCC AR4 climate models. Figure 2.7.4 compares the global average volcanic aerosol midvisible stratospheric optical depths of Sato *et al.* (1993) and Amman *et al.* (2003). Amman *et al.* (2003) estimate yields a larger value of the optical depth in comparison with Sato *et al.*, (1993), by 20–30% in the second part of the 20th century and by 50% for eruptions at the end of 19th- and beginning of 20th century, e.g., Santa Maria in 1902. Among the models that have participated in the IPCC AR4 study of the climatic effects of volcanic aerosols, the NCAR climate model, for example, employs Amman's data while GISS, MIROC and GFDL models use the Sato dataset. Stenchikov *et al.* (2005) used UARS observations to modify the effective radii from Hansen *et al.* (2002), accounting for its variations with altitude, and implemented this feature in the GFDL model. Bingen *et al.* (2004a; 2004b) have made a better estimate of stratospheric aerosols size distribution parameters using SAGE II data. However SAGE II, because of saturation, has significant gaps that are especially severe for the shorter-wave lengths during half a year after the eruption. Other efforts represent continued efforts to refine and improve the datasets. Bauman *et al.*, (2003b; 2003a) provide a new approach for calculating aerosol optical characteristics using SAGE and UARS data. Randall *et al.* (2000; 2001) have intercompared the POAM and SAGE data extensively and normalized them combining into a consistent data set to fill in gaps in the polar regions. Note, however, that while the aerosol characteristics are better constrained for the Pinatubo eruption, and to some extent even for El Chichón and Agung eruptions, the reliability degrades for volcanic events further back in time with little by way of observational constraints on optical depth and size evolution.

Estimates of volcanic RF evolution have also been revised since TAR. Figure 2.7.5 compares instantaneous net TOA radiative forcing calculated using Sato's and Ammann's data sets, along with the approximation of Andronova *et al.*, (1999) which has been used in some climate studies. As expected, Ammann's RF is 20–30% larger than Sato's RF for the recent major eruptions viz., Agung, El Chichón, and Pinatubo. The RF approximation of Andronova *et al.* (1999) uses Sato's optical depth except for the case of Pinatubo when it uses the larger optical depth estimate of Stenchikov *et al.* (1998). Andronova's RF appears to be very close to Sato's estimate for Agung and El Chichón cases, which is expected as both of them are based on the same optical depth. Discrepancy in estimates of the maximum optical depth attained in the case of Pinatubo remains unresolved because, for about 6 months following the eruption, the aerosol cloud was too dense to be remotely sensed by SAGE II in the visible spectrum. The Sato and Amman clear-sky net RF (not shown) has a similar shape to the all-sky RF in Figure 2.7.5 but is about 30% higher, consistent with the findings of Stenchikov *et al.* (1998).

[INSERT FIGURE 2.7.5 HERE]

2.7.2.2 Indirect aspects of volcanic forcing

The radiative effects due to volcanic aerosols from major eruptions are manifest in other radiation variables too e.g. the global mean anomaly of reflected solar radiation - this variable affords a good estimate of aerosol radiative effects that can actually be tested against observations, as shown for the case of Pinatubo

(Ramachandran *et al.,* 2000; Hansen *et al.,* 2002). However, unlike RF, this variable contains the effects due to feedbacks (e.g., changes in cloud distributions) so that it is actually more a signature of the response (or forcing-plus-feedback) rather than an effect due to forcing only. It is interesting that, in the Pinatubo case, the GISS models that use Sato's data set with the low estimate of optical depth show even more solar reflection than the NCAR model which uses the larger Ammann optical depth value. This example illustrates that, at present, differences in model responses yield additional uncertainties over and above that due to limitations in the knowledge of the basic volcanic aerosol optical properties.

The model estimates of reflected solar radiation caused by Pinatubo aerosols compare reasonably well with the all-sky ERBS observations (see Figure 4.6, IPCC*,* 1994). The reflected short wave flux anomaly is about 4 W m^{-2}. However the ERBS observations were conducted for a relatively short period of time and the model-observation comparisons are likely affected by differing cloud effects in simulations and measurements.

At least three distinct mechanisms have been invoked to explain the climate response to natural RF by both solar and volcanic activity. First, these forcings can directly affect the Earth's radiative balance, which alters the tropospheric energy balance and can thereby alter surface temperature. Second, the forcings set up gradients of heating perturbations vertically and horizontally; these can alter the stratospheric circulation that affects the troposphere providing one type of indirect climate effect. Third, the forcings can interact with internal climate system variability (e.g., ENSO, NAO, QBO) and dynamical noise, thereby triggering, amplifying or shifting these modes (Yang and Schlesinger*,* 2001; Stenchikov *et al.,* 2004). (Yang and Schlesinger*,* 2001; Stenchikov *et al.,* 2004). Each of these mechanisms has its own spatial, altitudinal and temporal response pattern. And each of the mechanisms could further depend on the background state of the climate system, and thus on other forcings (e.g., due to well-mixed gases, Meehl *et al.,* 2004), or interactively with each other.

Both solar and volcanic forcings affect global stratospheric ozone distributions (Chipperfield *et al.,* 2003) so that changes in this trace gas arising due to solar variations or volcanic aerosol-induced effects also have to be accounted for in order to quantify the total RF and the resulting climate responses. A point to be considered in this regard is whether the resulting ozone anomalies are to be declared as part of the natural forcings or as part of the net stratospheric ozone RF (Section 2.3.6.1).

Each of these mechanisms has its own spatial, altitudinal and temporal response pattern. And each of the mechanisms could further depend on the background state of the climate system, and thus on other forcings (e.g., due to well-mixed gases, Meehl *et al.,* 2004), or interactively with each other.

2.7.2.2.1 Volcanic impact on atmospheric temperature and moisture

In climate studies it is important to account for volcanic aerosols to correctly attribute observed climate trend and variability. Recent studies have highlighted the effects of volcanic aerosol radiative perturbations on temperature and moisture, which bring about changes in the heat and hydrologic balance. Volcanic eruptions, such as the Mt. Pinatubo in 1991, with global visible optical depth maximizing at about 0.15, cause a large negative perturbation of the global averaged radiative balance at the top of the atmosphere reaching –3 W m^{-2} , and a global surface cooling of –0.5 K. An improved analysis of the vertical structure of the volcanic thermal impact using radiosonde data was conducted by Free and Angell (2002) which has resulted in a better quantification of tropospheric and stratospheric temperature changes caused by the strongest recent eruptions of 20th century: Agung, El Chichón, and Pinatubo. Climate responses to volcanic eruptions have been inferred in European surface temperature and tree-ring based records dating back to the 15th century (e.g., Jones *et al.,* 2003 see Chapter 6).

Different prescriptions of volcanic aerosol characteristics (optical depths, RFs) in various modelling studies, together with the inherent difference in the physics parameterizations in the respective models, can lead to varying estimates of the volcano impacts. Thus, Broccoli *et al.* (2003) show that the response to the Andronova *et al.* (1999) prescription employed in their model yields a larger surface cooling for the Pinatubo eruption than the Hansen *et al.* (2002) simulation which used the Sato *et al.* (1993) optical depths; the latter study is closer to observed. Two studies that have used the Stenchikov *et al.* (1998) prescription have resulted in tropospheric simulated responses that are comparable to observed (Yang and Schlesinger*,* 2002), while Ramachandran *et al.* (2000) show a good agreement with the observed stratospheric warming. Chapter

9 gives further details of the significance of the simulated responses and model-observation comparisons for the 20th century eruptions.

A mechanism closely linked to the optical depth perturbation and the ensuing warming of the tropical lower stratosphere is the potential increase in the cross-tropopause water vapour flux (Joshi and Shine*,* 2003 see also Section 2.3.6.1, Chapter 3 and Chapter 8). Although Joshi and Shine (2003) show that expected water vapour changes would be small and HALOE observations (Randel *et al.,* 2004) do not show a large effect for the post-Pinatubo period, models need to be thoroughly tested on this matter.

2.7.2.2.2 Volcanic effects on atmospheric circulation

Anomalies in the global radiative heating distribution caused by volcanic aerosol direct and indirect effects, both in the stratosphere and surface-troposphere, can cause significant changes in atmospheric circulation. Tropical eruptions set up a transient radiative warming of the low-latitude lower stratosphere which sets up a perturbation in the equator-to-pole heating gradient whose magnitude depends on the state of the climate system at the time of the eruption (Ramaswamy *et al.,* 2005). Further, the surface-troposphere system in the lower latitudes experiences a reduction in radiative flux relative to the middle and polar latitudes (Robock*,* 2000). Since TAR, a better understanding of the regional effects of volcanic forcing on the high-latitude temperature and circulation has been achieved. The dynamical response to the radiative perturbations can force a positive phase of the Arctic Oscillation (AO) and cause counterintuitive boreal winter warming in middle and high latitudes over Eurasia and North America (Stenchikov *et al.,* 2002; Shindell *et al.,* 2003b; Collins*,* 2004; Shindell *et al.,* 2004; Stenchikov *et al,* 2004; Miller *et al.,* 2005). Thus, strong volcanic eruptions, like those of El Chichón and Pinatubo, could contribute to the observed long-term positive trend of the AO.

Stenchikov *et al.* (2002) have examined the evolution of the circulation in the two years following the Pinatubo eruption. A positive phase of the AO was produced in the experiment with only the tropospheric effect of aerosols showing that aerosol heating in the lower tropical stratosphere is not necessary to force positive AO response, as has been previously assumed. Aerosol-induced tropospheric cooling in the subtropics decreases the meridional temperature gradient in the winter troposphere between 30°N and 60°N, with an accompanying reduction of mean zonal energy and amplitudes of planetary waves in the troposphere, a decrease of wave activity flux into the lower stratosphere, and a resulting strengthening of the polar vortex which forces a positive phase of the AO.

Oman *et al.* (2005) simulated the climate impact of the 1912 Katmai eruption in Alaska. They used a 20-member ensemble of simulations and found that a volcanic aerosol cloud spread mostly north of 30ºN cannot produce significant winter warming pattern, even if it yields a higher hemispheric optical depth than the Pinatubo eruption in 1991. This is because, in the winter season, the lower stratospheric heating due to the high-latitude volcanic cloud is too weak to produce sufficient meridional temperature gradient in the lower stratosphere to force changes in the polar vortex.

2.7.2.2.3 Volcanic aerosol effects on stratospheric chemistry

Stratospheric aerosols affect the chemical processes in the stratosphere by serving as surfaces for heterogeneous reactions that liberate anthropogenic chlorine and cause ozone depletion (Chipperfield *et al.,* 2003). Stenchikov *et al.* (2002) demonstrate a climate link between ozone depletion and AO response through volcanic impact on stratospheric chemistry, arising as a result of enhanced halogen loading and reactions on particle surfaces. As ozone change is a forcing agent, this is a secondary radiative mechanism induced by volcanic aerosols through stratospheric chemistry. To better understand chemical feedback processes Al-Saadi *et al.* (2001) used Pinatubo aerosol heating rate inputs and aerosol surfaces from HALOE in a coupled radiation-dynamics-chemistry model to study the effect of the Mt. Pinatubo eruption on the middle atmosphere. Tabazadeh *et al.* (2002) found that stratospheric cooling in polar regions associated with the stronger polar vortex caused by volcanic effect will increase the probability of formation of polar stratospheric clouds and therefore increase the rate of heterogeneous chemical destruction of stratospheric ozone especially in the Northern Hemisphere. These studies indicate effects on the stratospheric ozone layer in the wake of a volcanic eruption and in a stratosphere with enhanced halogen loading. This process would be an *anthropogenic* indirect forcing, as it requires ozone depleting substances to create the initial halogen loading.

Since TAR several studies have been conducted using interactive chemistry-climate models. Rozanov *et al.* (2002) calculated the Mt. Pinatubo eruption impact on atmospheric temperature, circulation, and ozone distribution using prescribed aerosols (Stenchikov *et al.,* 1998). They simulated a fairly realistic strengthening of the Northern Hemisphere stratospheric polar vortex and winter warming at the surface. However, they overestimated heating in the equatorial lower stratosphere and underestimated ozone losses in comparison with observations. Timmreck *et al.* (2003) utilized a middle atmosphere model to study aerosol formation and stratospheric temperature perturbations interactively. They also accounted for effects on the stratospheric chemistry caused by volcanic aerosols. Shindell (2003b) use a middle atmosphere model with parameterized ozone chemistry to simulate climate response to the Mt. Pinatubo eruption and hypothetical eruptions with optical depths two and three times larger, approximating the effect of Tambora, which was a very intense eruption. Dameris *et al.* (2005) conducted transient simulation using couple a chemistry-climate model for the period 1960 to 1999, parameterising effects of Agung and El Chichón based on calculations for Pinatubo period (Stenchikov *et al.,* 1998). They reported significant effect on stratospheric ozone. However, they overestimated lower stratospheric heating following volcanic eruptions because of their simplified implementation of volcanic radiative impact. The interactive chemistry-climate studies generally show that aerosol-induced stratospheric heating affects the dispersion of aerosol cloud, thus affecting the spatial RF. However the models usually overestimate the mixing at the tropopause level and intensity of meridional transport in the stratosphere. A simplified treatment of aerosol microphysics causes errors in calculating aerosol heating/cooling effects. This indicates that although the interactive approach is conceptually superior with respect to those that use prescribed aerosol characteristics, at the present time and for climate forcing and change studies, it is practical to utilize more simple approaches that are constrained by aerosol observations. These approaches could serve as a benchmark for future interactive fully coupled modelling studies.

2.8 Utility of Radiative Forcing

The TAR and previous IPCC assessments have concluded that stratospheric adjusted RF (Fa) is a useful tool for estimating, to a first order, the relative climate impacts of differing climate-change mechanisms (Ramaswamy *et al.,* 2001). Through a quasi-constant factor (the climate sensitivity parameter) RF can be used to estimate the relative equilibrium globally averaged surface temperature response. Modelling studies indicated that the climate sensitivity parameter was more or less constant (varying by less than 25%) between mechanisms (Ramaswamy *et al.,* 2001; Chipperfield *et al.,* 2003). However, this level of agreement was found not to hold for certain mechanisms such as some ozone changes and changes in absorbing aerosol. Because the climate response, and in particular the range of equilibrium climate sensitivities exhibited by GCMs is much more than 25% (see Chapter 8), RF still remains the preferred measure for the quantitative assessment of climate change mechanisms (see also Jacob *et al..,* 2005).

Since TAR several studies have examined the relationship between RF and response for many climate change mechanisms. Several of these studies have also examined how different forcing mechanisms lead to different spatial patterns of climate response, both for the surface and for changes in the vertical. The concept of efficacy quantifies the variation in climate sensitivity between mechanisms and may help rank RFs in terms of their expected global mean temperature response (Section 2.8.5). Importantly, global mean RFs are not necessarily a suitable metric for many other aspects of climate response, and, in particular cannot be used for comparing the complete climate response of forcing agents against each other. For a full picture of the effect of a climate-forcing agent several other aspects need to be considered (see Sections 2.2 and 2.10 as well as Chapters 7 and 9).

2.8.1 Vertical Forcing Patterns and Surface Energy Balance Changes

The vertical structure of a forcing agent is important both for efficacy (Section 2.8.5) and other aspects of climate response, particularly for evaluating regional and vertical patterns of temperature change and also changes in the hydrological cycle. For absorbing aerosol the surface forcings are arguably a more useful measure of the climate response (particularly for the hydrological cycle) than the RF (Ramanathan *et al.,* 2001a; Menon *et al.,* 2002a). It should be noted that the surface energy budget involves components due to sensible and latent heat fluxes as well as solar and longwave irradiance. It can quantitatively be very different from the RF measured at the tropopause and is *not* representative of the energy balance perturbation to the surface-troposphere (climate) system. While the surface forcing adds to the overall description of the total perturbation brought about by an agent, the two numbers should not be directly compared nor should the surface term be considered in isolation for evaluating the climate sensitivity (see e.g., the caveats expressed in Manabe and Wetherald, 1967; Ramanathan, 1981). Therefore, surface forcings are presented as an important and useful diagnostic tool that aids understanding of the climate response (see Section 2.9.4 and 2.9.5).

2.8.2 Spatial Patterns of Radiative Forcing

RF spatial patterns affect the global mean surface temperature response (see Section 2.8.5) and also the pattern of climate response. However, also note that to a first order very different RF patterns can have similar patterns of surface temperature response and the location of maximum RF is rarely coincident with the location of maximum response (Mitchell *et al.,* 2001). Identification of different patterns of response is particularly important for attributing past climate change to particular mechanisms and is also important for the prediction of regional patterns of future climate change. These aspects of the forcing-response relationship are discussed in Chapter 9. Our chapter employs RF as method of ranking a forcing agents effect on the equilibrium global temperature change, and we discuss only this aspect of the forcing-response relationship. However, we present the patterns of RF as a diagnostic (Section 2.9.5).

2.8.3 Linearity of Forcing-Response Relationship

Reporting findings from several studies the TAR concluded that responses to individual RFs could be linearly added to gauge the global mean response, but not necessarily the regional response (Ramaswamy *et al.,* 2001). Since then studies with several equilibrium and/or transient integrations of several different GCMs have found no evidence of any non-linearity for changes in greenhouse gas and sulphate aerosol (Boer and Yu, 2003b; Sexton *et al.,* 2003; Gillett *et al.,* 2004; Matthews *et al.,* 2004). Two of these studies also examined many other forcing agents without finding evidence of a non-linear response (Sexton *et al.,* 2003; Matthews *et al.,* 2004). In all four studies even the regional changes typically added linearly. Studies with one GCM (Feichter *et al.,* 2004; see also Lohmann and Feichter, 2005) exhibits nonlinearities. However, their model may be linear with respect to RF but not linear with respect to emissions, because of the impact of the climate response on their interactive aerosol cycle. One study does find marked non linearity for large negative RFs (Hansen *et al.,* 2005); they suggest this is due static stability change in the upper troposphere affecting the climate feedbacks in their model. For the magnitude and range of realistic RFs discussed it this chapter we have high confidence of a linear relationship between global mean RF and global mean surface temperature response.

2.8.4 Alternative Methods of Calculating Radiative Forcing

RFs are increasingly being diagnosed from GCM integrations where the calculation is more complicated (Stuber *et al.,* 2001a; Tett *et al.,* 2002; Gregory *et al.,* 2004). This chapter also discusses several mechanisms that allow some response in the troposphere, such as cloud changes. These mechanisms are not initially radiative in nature, but will eventually lead to a radiative perturbation of the surface-troposphere system, that could conceivably be measured at the top of the atmosphere.

Alternatives to the standard RF definition have been proposed that may help account for these difficulties (see Figure 2.2.2). Since TAR several studies have employed equilibrium integrations of GCMs to diagnose a fixed sea-surface temperature forcing (Fs) and/or fixed global temperature forcing (Fg).

Hansen *et al.* (2002) concluded that use of Fs was not particularly beneficial for diagnosing global mean equilibrium surface temperature response. Hansen *et al.* (2005) added an extra term to Fs to simulate an Fg type forcing. They then found that this climate forcing was a better predictor of the resulting response (i.e., efficacies were closer to 1.0) than either Fa or Fs. Similarly, Shine *et al.* (2003) found that Fg was near-perfect predictor of the global mean surface temperature response in their model and suggested that it was a particularly useful diagnostic for absorbing aerosol and ozone changes, where the Fa failed as a predictor of the surface temperature response. In addition, Hansen *et al.* (2005) has compared a regression method (see also Gregory *et al.,* 2004) and a fixed sea-surface temperature method of estimating Fg. They found that the regression method gives a reasonable approximation to the fixed surface temperature method for a wide range of RFs but is more uncertain. Sokolov (2005) evaluated Fg a different way by splitting climate feedbacks into there surface and atmospheric only components. Modifying Fa by the atmospheric-only component of climate feedback gave an estimate of Fg, which also had efficacies closer to 1.0. For most forcing agents Fg appears close in magnitude to Fa. The difference essentially can be interpreted as a semi-direct effect and for most mechanisms, aside from absorbing aerosol, it appears small (Shine *et al.,* 2003; Hansen *et al.,* 2005; Sokolov, 2005).

In summary there is modest confidence that Fg is more representative of the equilibrium global mean surface temperature change than Fa. The Fg and Fs calculation also remove problems associated with defining the tropopause in the Fa definition (Ramaswamy *et al.,* 2001; Shine *et al.,* 2003; Hansen *et al.,* 2005). However, their calculation also can be somewhat intricate and Fa has the advantage that it does not depend on relatively uncertain components of a GCMs response, such as cloud change. For the LLGHGs Fa also has the advantage that is also readily calculated in detailed off-line radiation codes. All the methods assessed can provide useful diagnostic tools for understanding climate response.

2.8.5 Efficacy

Efficacy is defined as the ratio of the climate sensitivity parameter for a given forcing agent to the climate sensitivity parameter for CO_2 changes (Joshi *et al.,* 2003; Hansen and Nazarenko, 2004) Preliminary studies found that efficacy values for a number of forcings show less model dependency than the climate sensitivity values (Joshi *et al.,* 2003). Efficacies have been used as weightings for individual RF to get one step closer to an estimator of the likely surface temperature response than can be achieved by using RF alone (Sausen and Schumann, 2000; Hansen *et al.,* 2005; Lohmann and Feichter, 2005). Adopting a different definition of RF which has efficacies close to unity may be another way of achieving similar goals (see Sections 2.8.4).

Each type of RF (F) multiplied by its appropriate efficacy (E) would give you the effective RF (Fe=FE) that can be directly compared to a RF from CO_2. For this effective forcing the climate sensitivity parameter is independent of mechanism, so comparing this forcing is equivalent to comparing the equilibrium global mean surface temperature change. That is, $\Delta T_s= \lambda_{CO2}Fe$, where λ_{CO2} is the climate sensitivity parameter for a carbon dioxide change.

This section assesses the efficacy associated with Fa – the stratospherically adjusted RF, as defined in Ramaswamy *et al.* (2001) and employed in the rest of this chapter. The findings presented in this section are from an assessment of all the studies referenced in the caption of Figure 2.8.1, which presents a synthesis of efficacy results. As space is limited not all these studies are explicitly discussed in the main text.

2.8.5.1 Generic understanding

Since the TAR more GCM climate modelling studies have calculated efficacies and a general understanding is beginning to emerge as to how and why efficacies vary between mechanisms. The initial climate state, and the sign and magnitude of the RF have less importance but can still affect efficacy (Boer and Yu, 2003a; Joshi *et al.,* 2003; Hansen *et al.,* 2005). These papers have also developed useful conceptual models to help explain variations in efficacy with forcing mechanism. The efficacy primarily depends on the spatial structure of the forcings and the way they project onto the various different feedback mechanisms (Boer and Yu, 2003b). Therefore different patterns of RF and any non-linearities in the forcing response relationship affects the efficacy (Boer and Yu, 2003b; Joshi *et al.,* 2003; Hansen *et al.,* 2005; Sokolov, 2005; Stuber *et al.,* 2005). Many of the studies presented in Figure 2.8.1 find that both the geographical and vertical distribution of the forcing can have the most significant effect on efficacy (in particular see Boer and Yu, 2003b; Joshi *et al.,* 2003; Sokolov, 2005; Stuber *et al.,* 2005). Nearly all studies that examine it find that high latitude forcings have higher efficacies than tropical forcings. Even without any albedo feedback, some high latitude amplification is likely to remain (Stuber *et al.,* 2001b; Joshi *et al.,* 2003; Stuber *et al.,* 2005). Some studies break down the regional analysis of efficacy still further (Forster *et al.,* 2000; Boer and Yu, 2003b; Joshi *et al.,* 2003; Stuber *et al.,* 2005). Efficacy has also been shown to vary with the vertical distribution of an applied forcing (Hansen *et al.,* 1997; Christiansen, 1999; Joshi *et al.,* 2003; Cook and Highwood, 2004; Roberts and Jones, 2004; Forster and Joshi, 2005; Sokolov, 2005; Stuber *et al.,* 2005). Forcings which predominately affect the upper troposphere are often found to have smaller efficacies that those that affect the surface. However, this is not ubiquitous as climate feedbacks (such as cloud and water vapour) will depend on the static stability of the troposphere and hence the sign of the temperature change in the upper troposphere (B. Govindasamy *et al.,* 2001; Joshi *et al.,* 2003; Sokolov*,* 2005), it is therefore difficult to draw generic conclusions.

[INSERT FIGURE 2.8.1]

2.8.5.2 Long-lived greenhouse gases

The few models that have examined efficacy for combined LLGHG changes generally find efficacies slightly higher than 1.0 (Figure 2.8.1). Further, the most recent result with the NCAR model (B. Govindasamy *et al.,* 2001) finds an efficacy of over 1.2 with no clear reason of why this changed from earlier versions of the same model. Individual LLGHG efficacies have only been analysed in 2 or 3 models. Two GCMs suggest higher efficacies from individual components (over 30% for CFCs in Hansen *et al.,* 2005). In contrast another GCM gives efficacies for CFCs (Forster and Joshi*,* 2005) and methane (Berntsen *et al.,* 2005b) which are slightly smaller than one. Overall there is modest confidence that the observed changes in the combined LLGHG changes have an efficacy close to 1.0 (within 10%), but there are not enough studies to constrain the efficacies for individual species.

2.8.5.3 Solar

Solar changes, compared to CO_2, have less high latitude forcing with more of the forcing realized at the surface. Established but incomplete knowledge suggests that there is partial compensation between these effects, at least in some models, which leads to solar efficacies close to 1.0. All models with a positive solar forcing find efficacies of 1.0 or smaller. One study finds a smaller efficacy than other models (0.63: Gregory *et al.,* 2004). However they use a fully coupled model and employed a unique methodology for calculating climate sensitivity, (see Section 2.8.4): the slab-ocean version of the same model has an efficacy which is within the range of that from other models. These studies have only examined direct solar RF, any indirect effects (Section 2.7.1.3) are not included in this efficacy estimate. Overall there is high confidence that the direct solar efficacy excluding any indirect effects is within the 0.75–1.0 range.

2.8.5.4 Ozone

Stratospheric ozone efficacies have normally been calculated from idealised ozone perturbations. Experiments with three models (Stuber *et al.,* 2001b; Joshi *et al.,* 2003; Stuber *et al.,* 2005) find higher efficacies for idealized changes; these were due to larger than otherwise tropical tropopause temperature changes which led to a positive stratospheric water vapour feedback. However, this mechanism may not operate in the two versions of the GISS model, which found smaller efficacies. The forcing calculation for stratospheric ozone (and hence efficacy) depends heavily on the definition of tropopause height (Ramaswamy *et al.,* 2001; Chipperfield *et al.,* 2003; Hansen *et al.,* 2005). Only one study has used realistic stratospheric ozone changes (Hansen *et al.,* 2005) thus our knowledge is still incomplete. This study performed experiments applying ozone changes throughout the atmosphere an in the troposphere separately, and found the same efficacy for each experiment, implying that stratospheric ozone changes, if modelled separately, would also have the same efficacy. As this is only one model, generic conclusions are only drawn from the idealised studies where there is (1) high confidence that its efficacy is within a 0.5–2.0 range and; (2) established but incomplete physical understanding of how and why its efficacy could be larger than 1.0.

Some studies have examined efficacy variation with vertically constrained ozone changes, but there is little consensus. Two studies estimate efficacies of ~0.8 for realistic tropospheric changes using different ozone changes in different versions of the GISS model (Mickley *et al.,* 2004; Hansen *et al.,* 2005), the precise number is slightly affected by tropopause height (Hansen *et al.,* 2005). Overall we have high confidence that for realistic tropospheric ozone perturbations since preindustrial times the efficacy is within the 0.6–0.8 range.

2.8.5.5 Scattering aerosol

For idealised global perturbations the efficacy for the direct effect of scattering aerosol is very similar to that for changes in the solar constant (Cook and Highwood*,* 2004). As for ozone, realistic perturbations of scattering aerosol exhibit larger changes at higher latitudes and thus have a higher efficacy than solar changes (Hansen *et al.,* 2005). Although the number of modelling results is limited it is expected that efficacies would be similar to other solar effects, thus we can have high confidence that efficacies for scattering aerosol would be in the 0.7–1.1 range. Efficacies are similar for scattering aerosol in the troposphere and stratosphere.

Studies of the first indirect aerosol effect, where most of the forcing also comes from shortwave effects, have similar efficacies to the direct effect of scattering aerosols. More models have evaluated this than the direct effect and we have high confidence that its efficacy falls within the 0.7–1.1 range.

2.8.5.6 Absorbing aerosol

For absorbing aerosols the simple ideas of a linear forcing-response relationship and efficacy can break down (Hansen *et al.,* 1997; Cook and Highwood, 2004; Feichter *et al.,* 2004; Roberts and Jones, 2004; Hansen *et al.,* 2005). Aerosols within a particular range of single scattering albedos have negative RFs but induce a global mean warming– i.e., the efficacy can be negative. The surface albedo and height of the aerosol layer relative to the cloud also affects this relationship (Section 2.4.6, Penner *et al.,* 2003; Cook and Highwood, 2004; Feichter *et al.,* 2004; Johnson *et al.,* 2004; Roberts and Jones, 2004; Hansen *et al.,* 2005). Studies which increase black carbon in the planetary boundary layer find very high efficacies much larger than 1.0 (Cook and Highwood, 2004; Roberts and Jones, 2004; Hansen *et al.,* 2005). These studies also find that efficacies are considerably smaller than 1.0 when BC aerosol is changed above the boundary layer. These changes in efficacy can at least partly be attributable to a semi-direct effect whereby absorbing aerosol modifies the background temperature profile and tropospheric cloud (see Section 2.4.6). Another possible feedback mechanism is the modification of snow-albedo by black carbon aerosol (BC) (Menon *et al.,* 2002a; Hansen and Nazarenko, 2004; Hansen *et al.,* 2005); however this report does not classify this as part of the response, but rather as a separate RF (see Section 2.5.4 and 2.8.5.7). Most GCMs likely have some representation of the semi-direct feedback (Cook and Highwood, 2004) but its magnitude is very uncertain (Section 2.4.6.2.3) and dependant on aspects of cloud parameterizations within GCMs (Johnson, 2005). Two studies using realistic vertical and horizontal distributions of BC find that overall the efficacy is around 0.7 (Hansen *et al.,* 2005; Lohmann and Feichter, 2005). However, Hansen *et al.* (2005) acknowledge that they may have underestimated BC within the boundary layer and another study with realistic vertical distribution of BC changes finds an efficacy of 1.3 (Sokolov, 2005). There is high confidence that for realistic BC changes the efficacy falls within the 0.5–1.5 range.

2.8.5.7 Other forcing agents

Efficacies for some other effects have been evaluated (see especially Hansen *et al.,* 2005). Although not verified by multiple modelling groups the efficacies for these other effects are broadly consistent with our general understanding of how solar efficacies vary with the latitude of the applied forcing. In particular, land use changes are largest in northern-hemisphere mid-to-high latitudes; and the snow-albedo forcing from BC is largest at northern-hemisphere high latitudes. Hansen *et al.* (2005) find that that land-use albedo changes have an efficacy of ~1.0, whilst the BC snow albedo forcing has an efficacy of 1.7. Contrail forcing may have a efficacy smaller than 1.0 (Ponater *et al.,* 2005, find an efficacy of 0.6) and this agrees with a suggestion that high cloud changes should have smaller efficacies (Hansen *et al.,* 2005). Boucher *et al.* (2004) suggest a negative efficacy for water vapour increases in the boundary layer associated with irrigation. As in the case of BC and ozone changes this may be an indication that forcing applied in the boundary layer elicits a different response than more homogeneous forcings. The results of Hansen *et al.* (2005) and Forster (1999) suggest that stratospheric water vapour efficacies are roughly one.

2.8.6 Efficacy and the Forcing-Response Relationship

Although our conclusions regarding efficacy remain similar to those in the TAR, our physical understanding is now considerably more established (Section 2.8.5). We have increased confidence that RF (Fa) is a predictor of global mean temperature response, to within 25% for most RFs and a factor of 2 for any realistic RF. However, it should be noted that efficacies have only been evaluated in GCMs and the actual climate efficacies could be different than the ones quoted in Section 2.8.5.

Different forcing methodologies (Section 2.8.4) assume different forcing-response relationships and there has been considerable debate as to whether first-response effects such as cloud lifetime and semi-direct aerosol effects should be considered as a forcing or a response (Ramaswamy *et al.,* 2001; Jacob *et al.,* 2005). By adopting Fa as the chosen measure of forcing we are also choosing to describe cloud lifetime and the semi-direct effects part of the overall climate response. This chapter coins the phrase “first-response” mechanisms. Most GCMs already have at least some representation of the semi-direct effect (Section 2.8.5.6) although many may poorly represent it. By comparing Fa and Fg forcings, the semi-direct effect can be estimated (Section 2.8.4). Hansen *et al.* (2005) evaluate Fg and Fa forcings for many different mechanisms. It is clear at least in their model that all forcings exhibit some semi-direct effect but it is only appreciable (>20%) for ozone and absorbing aerosol changes.

The estimate of efficacy from Fa forcings will include any semi-direct effect exhibited by the GCM plus an additional component associated with the surface temperature response. By attaching efficacies to the Fa forcings the semi-direct effect is accounted for but it is not separated out. As a diagnostic tool to understand the overall climate response in GCMs these cloud interaction terms are quantified separately in Chapter 7. However, we chose not to adopt this methodology in the synthesis (Section 2.9), as there is still much uncertainty associated with the evaluation of a globally averaged semi-direct effect (Section 2.4.6) and very little information on the semi-direct effect for mechanisms other than absorbing aerosol. In contrast to the semi direct effect, aerosol-cloud life time interactions are not typically modelled by GCMs. However, the efficacy of the direct aerosol and/or cloud albedo aerosol RF could be modified to account for these effects, if they were sufficiently well quantified (see Chapter 7).

2.9 Synthesis

This section begins by synthesizing the discussion of the RF concept. It also presents summaries of the global mean RFs assessed in earlier sections and discusses time evolution and spatial patterns of RF. It also presents a brief synthesis of surface forcing diagnostics. It breaks down the analysis of RF several ways to aid understanding.

2.9.1 Global Mean Radiative Forcing

The RFs discussed this chapter, their uncertainty ranges, and efficacies are summarized in Figure 2.9.1 and Table 2.9.1. RFs from forcing agents have been combined into their main groupings. This is particularly useful for aerosol as its total direct RF is considerably better constrained than the RF from individual aerosol types (Section 2.4.5.7). Table 2.3.1 gives a further component breakdown of RF for the LLGHGs.

[INSERT FIGURE 2.9.1 HERE]

[INSERT TABLE 2.9.1 HERE]

In TAR because of a) uncertainties in the RFs, b) the uncertainty in the linear additivity assumption, and c) the uncertainty of efficacies, the various RFs from the different mechanisms were not added. Many of the limitations discussed in Ramaswamy *et al.* (2001) still apply. However, efficacies are now better understood and quantified (see Section 2.8.5). Secondly the linear-additivity assumption has been more thoroughly tested (Section 2.8.3). Thirdly the uncertainties in the direct aerosol and cloud-albedo aerosol RFs are substantially reduced. However, it should still be noted that the caveats discussed in Section 2.8 apply. Adding the RF values shown in the upper panel of Figure 2.9.1 and combining individual uncertainties results in the probability density function of RF shown in the bottom panel of Figure 2.9.1 (different efficacies are not accounted for). This summation gives a combined anthropogenic RF of 1.5 ± 1.0 W m^{-2}, which implies that it is very likely that humans have had a net warming effect on climate.

2.9.2 Uncertainties in Radiative Forcing

TAR assessed uncertainties in global-mean RF by attaching an error bar to each term that was *"guided by the range of published values and physical understanding"*. It also quoted a level of scientific understanding (LOSU) for each RF best estimate that was a subjective judgment of the estimate's reliability.

The concept of LOSU has been slightly modified based on the IPCC AR4 uncertainty guidelines. Only "well established" RFs are quantified. "Well established" implies that there is qualitatively both sufficient evidence and sufficient consensus from published results to estimate a central RF estimate and a range. "Evidence" is assessed by an A-C grade, with an A grade implying strong evidence and C insufficient evidence. "Consensus" is assessed by assigning a number between 1–3, where 1 implies a good deal of consensus and 3 insufficient consensus. B2 is the minimum grade required for a forcing to be sufficiently well established to be quantified. These two factors are combined to give a scientific understanding rank of very low, low, medium or high. As in TAR, the quoted range of RF is usually based on the available range of published values, giving the value uncertainty. This range has been altered to subjectively take into account any structural uncertainty caused by an incomplete sampling of the possible parameter space. For most RFs many studies have now been published and this generally makes the sampling of parameter space more complete and the error bars more realistic, compared to TAR. This is particularly true for both the direct and cloud-albedo aerosol RF (Section 2.4). The quoted uncertainty range is roughly equivalent to a 90% confidence interval and thus the true RF is "very likely" to fall within the quoted range. Table 2.9.2 summarises the key certainties/uncertainties how the range was evaluated. Aerosol and ozone RFs will have added uncertainties due to the uncertain semi-direct and cloud-lifetime effects. These uncertainties in the response to the forcing (efficacy) are discussed in Section 2.8.5.

Table 2.9.2 indicates that there is now stronger evidence for most of the RFs being discussed in this chapter. Some effects are not quantified either because they do not have enough evidence or their quantification lacks consensus. These include certain mechanisms associated with land-use, stratospheric water vapour and cosmic rays. Cloud-lifetime and the semi-direct effects are excluded on two grounds. Firstly, they are deemed to be part of the climate response (Section 2.8) and secondly there is little consensus of their effect. The RFs from the LLGHGs have both a high degree of consensus and a very large amount of evidence and, thereby, place our understanding of these effects at a considerably higher level than any other effect.

[INSERT TABLE 2.9.2 HERE]

2.9.3 Global-Mean Radiative Forcing by Emission Precursor

The RF from a single forcing agent can have contributions from several sources. Methane, for example, has a contribution from direct methane emissions, as well as NO_x emissions. The methane RF quoted in Table 2.9.1 and shown in Figure 2.9.1 is a value that combines the effects of both emissions. As an anthropogenic or natural emission can affect several forcing agents, it is useful to assess the RF associated with each primary emission. For example emission of NO_x affects methane, tropospheric ozone, and tropospheric aerosols. Based on a development, carried forward from the TAR, this chapter assessed the RF terms associated with each principal emission, with the results shown in Figure 2.9.2. Each principal component has a direct RF and one or more indirect RFs related to perturbations of other forcing agents. The following forcing mechanisms are considered.

- Fossil carbon emissions associated with non-CO_2 gaseous emissions, which eventually increase CO_2 in the atmosphere (from CO, CH_4, and NMVOCs emissions)
- Changes in stratospheric ozone (from N_2O, CFCs, and HCFCs emissions)
- Changes in tropospheric ozone (from CH_4, NO_x, CO, NMVOCs, and SO_2 emissions)
- Changes in OH affecting the lifetime of CH_4 and HFCs (from CH_4, CO, NOx, and NMVOCs emissions)
- Changes in organic carbon aerosols through changes in the O_3/OH ratio, which affects the amount of OC aerosols, produced through oxidation of natural NMVOCs.
- Changing concentrations of nitrate and sulphate aerosols through changes in NO_x and SO_2 emissions, respectively.

A number of the principal RFs (e.g., N_2O, land-use and mineral dust) do not affect other agents, thus their RFs are the same as those presented in Table 2.9.1. Table 2.4.4 gives the total aerosol RFs for black carbon and organic carbon that are used here (taking an average of the AEROCOM and non-AEROCOM models). The RFs for other agents are determined from both the various RFs assessed in this chapter and the chemical modelling results discussed in Sections 2.3.7.2 and 2.10. Uncertain indirect effects are not shown. These include ozone changes due to solar effects and cloud albedo changes caused by non-sulphate aerosols.

[INSERT FIGURE 2.9.2 HERE]

2.9.4 Time Evolution of Radiative Forcing and Surface Forcing

There is a good understanding of the time evolution for the concentrations of the LLGHGs both from flask and in-situ measurements over the last few decades and extending further back using firn and ice-core data (see Section 2.3, Figure 2.3.3 and Chapter 6). Increases in RF are clearly dominated by CO_2. Halocarbon RF has grown rapidly since 1950, but the growth of the RF was cut dramatically by the Montreal Protocol (see Section 2.3.4). CFC RF is declining; in addition the combined RF of all ODS appears to have peeked, at 0.32 W m^{-2}, during 2003. However, substitutes for ODS are growing at a slightly faster rate, so halocarbon RF growth is still positive (Table 2.3.1) - the decrease since TAR in the halocarbon RF shown in Table 2.9.1, is due to re-evaluation rather than a trend.

RF timeseries for the natural forcings are reasonably well known (Section 2.7). Determining timeseries for aerosol and ozone RF is far more difficult because a knowledge of past emissions and chemical transport modelling is required to evaluate these time histories. Several timeseries exist in the literature for these and other RFs. (e.g., TAR, Myhre *et al.,* 2001; Hansen *et al.,* 2002). GCMs also employ their own time histories of many of the forcing agents (e.g., Figure 2.9.3 and Chapter 10). Although spatial patterns and present day RFs differ greatly between the various models and RF reconstructions they typically have similar time-evolutions of their global mean RFs, as they often base their time histories on similar emission data. Aerosol and ozone RF time-histories remain too uncertain to ascertain an accurate time-evolution of RF beyond the examples given in Figure 2.9.3.

GCMs compute the climate response based on the knowledge of the forcing agents and their time evolution. While most current GCMs incorporate the trace gas RFs, aerosol direct effects, solar and volcanoes, a few have in addition incorporated land-use change and aerosol indirect effect (see Chapter 10). As an example of the timeseries of the RF and surface forcing due to the principal agents, the global-and-annual-mean time series, as implemented in the MIROC AOGCM (Takemura *et al.,* 2005), is illustrated in Figure 2.9.3. As for the present day RF, past RF is also dominated by the LLGHGs (see also Figures 2.3.3 and 2.9.1). The surface forcing, in contrast, is dominated by the negative effect of the aerosols and the LLGHGs have a much smaller positive effect. Note that quantitative values of the RFs and surface forcings by the agents differ across models in view of the differences in model physics and in the formulation of the forcing due to the short-lived species (see Chapter 10, Collins *et al.,* 2005 and; Forster*,* 2005 for further discussion on uncertainties in GCMs calculation of RF and surface forcing).

[INSERT FIGURE 2.9.3 HERE]

2.9.5 Spatial Patterns of Radiative Forcing and Surface Forcing

Figure 6.7 of TAR presented examples of the spatial patterns for most of the RF agents discussed in this chapter; these examples still hold. Many of the features seen in Figure 6.7 of TAR are generic, although additional uncertainties exist, compared to uncertainties in the global-mean RF. Spatial patterns of the aerosol RF exhibit some of the largest differences between models, depending on where source regions of aerosol types are located and whether or not the indirect aerosol effects are included. The aerosol direct and cloud-albedo RF also depends critically on the location of clouds, which differs between GCMs. An example from two GCMs of the spatial pattern of RF due to the sum of the various natural plus anthropogenic agents discussed in the chapter is illustrated in Figure 2.9.4. The MIROC AOGCM includes an aerosol cloud-albedo effect and the GFDL AOGCM does not. Generic features are that the RF over most of the globe is positive and is dominated by the LLGHGs. The regions of significant aerosol RF can be seen, although the locations of these differ in the two GCMs. The direct effect of aerosols is seen in the total RF of the GFDL model over Northern Hemisphere land regions, whereas the cloud albedo effect dominates the MIROC GCM RF in the stratocumulus low-latitude ocean regions.

[INSERT FIGURE 2.9.4 HERE]

The spatial pattern of surface forcing (also shown in Figure 2.9.4) shows a pronounced deficit relative to preindustrial times where aerosol RF dominates, particularly the Northern Hemisphere. At high latitudes and in parts of the Southern Hemisphere there are fewer anthropogenic aerosols and surface forcing has increased due to the LLGHGs, whose effect is globally more homogeneous.

These spatial patterns of RF and surface forcing imply differences in the Northern Hemisphere equator-to-pole gradients for the surface and tropopause and the amount of energy absorbed by the troposphere. These are relevant to the climate response discussed in the other chapters (see especially Chapters 8, 9 and 10).

2.10 GWPs and Other Metrics for Comparing Different Emissions

2.10.1 Definition of a Metric and the GWP

The purpose of the Global Warming Potential (GWP) or other metrics described in this section is to provide a necessary tool to operationalize the goal of UNFCCC's Article 3 (Article 3 of the UNFCCC states that policies and measures should be comprehensive and cost-effective) through a multi-gas abatement strategy in a decentralised manner – i.e., to give the multi-gas emitters (nations, industries) a tool to compare emissions of the various species according to a specified objective. A very general formulation of a metric can be given by (e.g., Kandlikar, 1995, 1996):

$$AM_i = \int_0^{\infty} \left[(I(\Delta C_{(r+i)}(t)) - I(\Delta C_r(t))) \times g(t) \right] dt$$

Where $I(\Delta C_i(t))$ is a function describing the impact of change in climate (ΔC) at time t. The expression $g(t)$ is a weighting function over time (Heal, 1997; Nordhaus, 1997)(e.g., $g(t) = e^{-kt}$ as a simple discounting). The subscript r refers to a baseline emission path. For two emission perturbations i and j the absolute metric values AM_{i} and AM_{j} can be calculated to provide a quantitative comparison of the two emission scenarios. In the special case where the emission scenarios consist of only one component (as for the assumed pulse emissions in the definition of GWP), the ratio between AM_i and AM_j can be interpreted as a relative emission index for component i versus a reference component j (as CO_2 for GWP).

There are numerous problematic issues related to defining a comprehensive metric based on the general formulation given above (cf. review article by Fuglestvedt *et al.,* 2003). The main problem is to define appropriate impact functions, although there have been some attempts to do this for a range of possible climate impacts (Hammitt *et al.,* 1996; Tol, 2002). Given that impact functions can be defined, they would need regionally resolved climate change data (temperature, precipitation, winds, etc.) which would have to be based on GCM results with its inherent uncertainties. Additional issues such as definition of the weighting function $g(t)$ and the baseline emission scenarios will also have to be resolved.

Due to these difficulties the simpler and purely physical GWP index based on the time integrated global mean RF of a pulse emission of 1 kg of some gas (i) relative to that of 1 kg of the reference gas CO_2 was developed (e.g., Ramaswamy *et al.,* 2001). The GWP of component x is given by

$$GWP_x \equiv \frac{\int_0^{TH} RF_x(t)dt}{\int_0^{TH} RF_r(t)dt} = \frac{\int_0^{TH} a_x \cdot [x(t)]dt}{\int_0^{TH} a_r \cdot [r(t)]dt} \quad 33$$

Where TH is the time horizon, RF_x is the global mean TOA RF by component x, a_x is the RF per unit mass due to a unit increase in atmospheric abundance of substance in question (often called radiative efficiency), $[x(t)]$ is the time-dependent decay in abundance of substance x, and the corresponding quantities for the reference gas in the denominator. The nominator is called the absolute global warming potential (AGWP) of x, and the denominator is the AGWP for the reference gas. CO_2 is generally used as the reference gas, and all GWPs given in this report use CO_2 as the reference gas.

The simplifications made to derive the standard GWP index include:

- Put $g(t) = 1$ (i.e., no discounting)
- The choice of a pulse emission
- Integrate over a fixed time-horizon TH
- Define the impact function, $I(\Delta C)$ as the global mean RF, and assume that the climate response is equal for all RF mechanisms.
- Evaluate the impact relative to a baseline equal to current concentrations (i.e., setting $I(\Delta C_r(t)) = 0$). This simplification means that all potential feedbacks (e.g., on the carbon cycle) are ignored.

All GWPs depends on the AGWP for CO_2 (the denominator in the definition of the GWP). The AGWP of CO_2 again depends on the radiative efficiency for a small perturbation of CO_2 from the current level of 374 ppm. The radiative efficiency per kilogram CO_2 has been calculated using the same expressions as in IPCC (2001), but with a background CO_2 mixing ratio of 374 ppm. For a small perturbation from 374 ppm the RF is 0.01513 W m^{-2} ppm^{-1} (2.3% lower than the TAR value). The CO_2 response function is based on the "Bern" carbon-cycle model and the same as have been used in previous IPCC reports and WMO assessments. The AGWP values for CO_2 for 20, 100, and 500 years time horizons are 0.2023, 0.6803, and 2.191 W m^{-2} yr $(ppm)^{-1}$ (or 2.600×10^{-14}, 8.779×10^{-14}, and 28.16×10^{-14} W m^{-2} yr $(kg(CO_2))^{-1}$, [The AGWP values will be updated when revised pulse response functions for CO_2 are available (October 2005)]. Updated radiative efficiencies for well mixed-greenhouse gases are given in Table 2.10.1. Since the TAR radiative efficiencies have been reviewed by Montzka *et al.* (Montzka *et al.,* 2003) and Velders *et al.* (2005). Gohar *et al.* (2004) and Forster *et al.* (2005) have investigated HFC compounds with up too 40% differences in earlier published results. Based on a variety of radiative transfer codes they found that uncertainties can be reduced to around 12% with well-constrained experiments. The HFC studied were HFC-23, HFC-32, HFC-134a, and HFC-227ea. Hurley *et al.* (2005) studied the IR spectrum and RF of CF_4 and derived a 30% higher GWP value than in the TAR.

The RF calculation for GWPs for CH_4, N_2O, and halogen containing well-mixed greenhouse gases employ the simplified formulas given in the TAR (Table 6.2). The lifetimes for CH_4 and N_2O are taken from Chapter 7 of this report. GWP values for 47 gases are given in Table 2.10.1 for time horizons of 20, 100 and 500 years.

[INSERT TABLE 2.10.1 HERE]

2.10.2 Indirect GWPs

Indirect radiative effects include direct effects of degradation products or radiative effects through changes in concentrations of greenhouse gases caused by the presence of the emitted gas or its degradation products. Indirect effects from the direct effects of degradation products are not considered to be important (WMO, 2003) and are not discussed further. The indirect effects discussed here are linked to ozone formation or destruction, enhancement of stratospheric water vapour, and changes in concentrations of the OH radical with the main effect of enhancing the lifetime of methane. Uncertainties for the indirect GWPs are generally much higher than for the direct GWPs, and the indirect GWP will in many cases depend on the location of the emissions since background levels of reactive species (e.g., NO_x) can affect the chemical response non-linearly. Thus their usefulness to inform policy decisions is limited. However, they are readily calculable and alternative methodologies have similar caveats and have yet to be proven or adopted (see Section 2.10.3).

2.10.2.1 Methane

Four indirect radiative effects of methane emissions have been identified (cf. Chapter 4 and Section 6.6 of the TAR). Methane enhances its own lifetime through changes in the OH concentration, it leads to changes in tropospheric ozone, it enhances stratospheric water vapour levels, and produces CO_2. Following the approach taken by the SAR and TAR, we do not include CO_2 produced from CH_4 oxidation in the GWP estimates since it is often the case that this CO_2 is included in national carbon inventories. AS in TAR this report uses a value of 23 .This is supported by an additional study (Derwent *et al.,* 2001) which found a value of 23.3 from lifetime and tropospheric ozone effects.

2.10.2.2 Carbon monoxide

CO has indirect effects similar to those of methane (except it does not cause increases in stratospheric water vapour). Since the TAR Collins *et al.* (Collins *et al.,* 2002) and Berntsen *et al.* (2005b) have calculated GWPs for CO emissions, the range between 1.6 and 2.0, depending on the location. Berntsen *et al.* (2005b) found that emissions of CO from Asia had a 25% higher GWP, compared to European emissions.

2.10.2.3 NMVOCs

Collins *et al.* (2002) have calculated indirect GWPs for 10 non-methane volatile organic compounds (NMVOCs) with a global 3-D lagrangian chemistry-transport model. Impacts on tropospheric ozone, methane, and CO_2 have been considered, using either an "anthropogenic" emission distribution or a "natural" emission distribution depending on the main sources for each gas. The indirect GWP values are given in

Table 2.10.2. Due to their short lifetimes and the non-linear chemistry involved in ozone and OH chemistry, there are significant uncertainties in the calculated GWP values. Collins *et al.* estimate an uncertainty range of –50% to +100%. As discussed in the SAR the RF due to CO_2 produced from the oxidation of the NMVOCs are not included in the GWP to avoid double counting.

[INSERT TABLE 2.10.2 HERE]

2.10.2.4 NO_x

The short lifetime and complex non-linear chemistry which cause two opposing indirect effects through ozone enhancements and methane reductions make calculations of GWP for NO_x emissions very uncertain. Due to the non-linear chemistry the net RF of NO_x emission will depend strongly on location of emission and with a strict definition of a pulse emission for the GWP, also on timing of the emissions (Fuglestvedt *et al.,* 1999; Derwent *et al.,* 2001; Wild *et al.,* 2001; Stevenson *et al.,* 2004; Berntsen *et al.,* 2005b; Berntsen *et al.,* 2005a)

2.10.2.5 Halocarbons

Chlorine and bromine containing halocarbons can lead to catalytic ozone depletion in the stratosphere when the halocarbon molecules are broken down in the stratosphere and chlorine or bromine atoms are released. Indirect GWPs for ozone depleting halocarbons are estimated in Velders *et al.* (2005 Table 2.7). These are from observed ozone depletion between 1980 and 1990 for 2005 emission using the Daniel *et al.* (1995) formalism. Velders *et al.* (2005) did not quote net GWPs pointing out that as the physical characteristics of the CFC warming effect and ozone cooling effect were very different from each other, it made such offsetting unphysical and misleading. The same caveat applies when comparing GWPs of the other indirect effects and is probably even more important for the GWP from short lived species.

2.10.2.6 Hydrogen (H_2)

Anthropogenic emissions of H_2 through leakages could be substantial in an H_2 economy (60–120 Tg yr^{-1}, Tromp *et al.,* 2003). The main loss of H_2 is believed to be through surface deposition, but about 25% is lost through oxidation by OH. In the stratosphere this enhances the water vapour concentrations (by about 20% at 30 km altitude with the source given above (Tromp *et al.,* 2003), and thus also affect the ozone concentrations. In the troposphere the chemical effects are similar to those of CO leading to ozone production and methane enhancements (Prather*,* 2003). Derwent *et al.* (2001) have calculated an indirect GWP(100) for the tropospheric effects of H_2 of 5.8, which includes the effects of methane lifetime and tropospheric ozone.

2.10.2.7 Aerosols and aerosol precursors

Previous IPCC reports have not given GWPs values for aerosols or aerosol precursors. Since the TAR significant progress has been made in the understanding of the radiative effects of aerosols (Section 2.4). Bond and Sun (2005) have calculated GWPs for the direct effect of black carbon aerosols (i.e., neglecting the semi-direct effect and surface albedo effects). Based on previously published results for the lifetime of BC and a normalized RF of 1800 W/g, they derive GWP values of 2200 and 680 for time horizons of 20 and 100 years. The uncertainty range for the GWP_{100} estimate is 210–1500, and for GWP_{20} 690–4700. The main sources for the relatively large uncertainties are model assumptions about transport and removal of particles and optical properties.

A global mean GWP for SO_2 from fossil fuel combustion (including only the direct effect of sulphate aerosols) can be estimated based on the model results from the AEROCOM experiments summarized in Tables 2.4.3, 2.4.4 and 2.4.5 in this report. Using the modelled global sulphate loading of 3.12 mg m^{-2}, and all sky RF of –0.37 W m^{-2}, and a residence time of 4.1 days, GWP values of –161, –48, and –15 are estimated for time horizons of 20, 100 and 500 years respectively.

Care should be taken when applying GWPs for BC or SO_2 since as with other short lived species the GWPs for BC could vary significantly depending on location and time of the emissions.

2.10.2.8 GWP weighted emissions

A simple method to compare future climate impacts of current emissions is to multiply current emissions of all climate agents with their GWP values to obtain equivalent CO_2 emissions. This is consistent with the

Kyoto Protocol through its adoption of GWPs with 100 years time horizon to compare emissions of different climate agents. Figure 2.10.1 shows the equivalent CO_2 emissions for all climate agents (or groups of agents) considered in this report. Uncertainties in the estimates of the equivalent CO_2 emissions originate both from uncertainties in lifetimes and optical properties (through the GWP values) as well as uncertainties in the current global emissions. It should be noted that the compounds with long lifetimes (in particular CO_2) contribute significantly more to the total with this perspective than in the frequently cited "IPCC RF bar-chart diagram" (Figure 3 of the Summary for Policymakers in the TAR, updated in Figure 2.9.1 in this report). Again strong caveats and cautions apply when comparing uncertain emissions from the short lived species to those of the LLGHGs; the Kyoto protocol only considers LLGHG species. These GWPs have small uncertainties and do not depend on the location of the emission source. Further they are all positive. Decisions on how to treat negative GWPs, GWP variation by source region and uncertain GWPs would need to be made to use these for policy decisions.

[INSERT FIGURE 2.10.1 HERE]

2.10.3 New Alternative Metrics for Assessing Emissions

While the GWP is a simple and straight-forward index to apply for policy makers to rank emissions of different greenhouse gases, it is not obvious on what basis "equivalence" between emissions of different species is obtained (Smith and Wigley, 2000; Fuglestvedt *et al.,* 2003). The GWP metric is also not well suited for handling short-lived gases or aerosols (e.g., NO_x or black carbon aerosols).

2.10.3.1 Revised GWP formulations

2.10.3.1.1 Pulse versus sustained emission changes

Many mitigation measures will lead to emission reductions over a certain period and can thus not be regarded as pulse emission changes. Some authors have calculated GWPs for a sustained (or "slab") increase in emissions (Fuglestvedt *et al.,* 1996; Johnson and Derwent, 1996; Berntsen *et al.,* 2005b). Berntsen *et al.* (2005b) show that for gases with lifetimes shorter than 10 years, the pulse GWP is about a factor of about 0.6 lower than the sustained GWP. The sustained GWP is equivalent to consider a series of pulse emissions over the time-horizon where the GWP of each pulse is evaluated with a time-horizon equal to the difference between the time of emission and the original time horizon. The effect of changing from pulse to sustained GWPs, which is genuinely a policy question, can be regarded as committing future policy makers to use ever-shorter time horizons in their metric as the time horizon is approached.

2.10.3.1.2 Including the climate efficacy in the GWP

Climate efficacy can vary considerably between different forcing agents (Section 2.8.5). Fuglestvedt *et al.* (2003) have proposed a revised GWP concept including the efficacy of a forcing agent in the GWP expression. Berntsen *et al.* (2005a) have calculated GWP values in this way for NO_x and CO emissions in Europe and in Shout East Asia. The efficacies are less uncertain than climate sensitivities. However, Berntsen *et al.* (2005a) showed that for ozone produced by NO_x emissions the climate efficacies will also depend on the location of the emissions.

2.10.3.2 New metrics

2.10.3.2.1 Global-temperature potential

Shine *et al.* (2005) have proposed a Global Temperature Potential (GTP) as a new relative emission metric. The GTP is defined as the ratio between the global mean surface temperature change at a given future time horizon following an emission (pulse or sustained) of a compound x relative to a reference gas (Shine *et al.*, use CO_2).

$$GTP_x^{TH} = \frac{\Delta T_x^H}{\Delta T_r^H}$$

where ΔT_x^H denotes the global mean surface temperature change after H years following an emission compound x. Note that while the GWP is an integral quantity over the time horizon (i.e. the RF at the beginning and end of the time horizon counts exactly equal), the GTP uses the temperature change at time H (i.e., RF closer to time H contributes more). The GTP metric requires knowledge of the same parameters as the GWP metric (radiative efficiency and lifetimes), but in addition the response times for the climate system must be known, in particular if the lifetime of component x is very different from the lifetime of the reference gas. Differences in climate efficacies can be easily be incorporated into this metric discussed. Due to the inclusion of the response times for the climate system, the GTP values for pulse emissions of gases with shorter lifetimes than the reference gas will be lower than the corresponding GWP values. As noted by Shine *et al.* (2005) there is a near equivalence between the GTP for sustained emission changes and the pulse GWP. The GTP metric has the potential advantage over GWP that it is more directly related to surface temperature change.

2.10.3.2.2 Alternatives including economic aspects

Manne and Richels (2001) propose to construct emission metrics given a binding ceiling on future climate impact (e.g., formulated as a stabilization of future global mean temperature change). Using the computable general equilibrium model (MERGE), fixed surface temperature scenarios, and assumptions about future mitigation costs for the various gases, they show that the metric value for short-lived species like methane can vary significantly over time and for all gases the metric value depend on the stabilization goal (2ºC and 3ºC are used) and assumptions about mitigation costs.

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Question 2.1: How do human activities contribute to climate change and how do they compare with natural influences?

Human activities contribute to climate change by increasing the amounts of greenhouse gases, aerosols (microscopic particles), and cloudiness in Earth's atmosphere. The largest contribution comes from the burning of fossil fuels, which releases carbon dioxide gas to the atmosphere. Greenhouse gases, aerosols, and clouds are climate change agents because they interact with incoming solar radiation and outgoing infrared radiation that are part of Earth's energy balance. Changing the atmospheric abundances of these agents therefore can lead to a warming or cooling of the climate system. Since the start of the industrial era (about 1750), the contribution of human activities to climate change is a warming effect that exceeds that due to changes in natural processes, such as solar changes and volcanic eruptions. In the following, we discuss the climate change agents and how their impact on the climate system is measured.

[START OF QUESTION 2.1, BOX 1]

What is a greenhouse gas? A greenhouse gas is any gas present in Earth's atmosphere that absorbs infrared radiation. Water vapour (H_2O) is the most important and the most abundant. Other examples are carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), the halocarbons (gases containing fluorine, chlorine, and bromine) and ozone (O_3).

What is the greenhouse effect? The greenhouse effect occurs when gases in the atmosphere, known as greenhouse gases, allow incoming solar radiation to pass through the atmosphere but absorb outgoing infrared radiation. Incoming solar radiation warms Earth's surface and the atmosphere. The Earth naturally emits infrared radiation to space, which cools the planet. The balance between incoming and outgoing radiation determines Earth's average temperature. When greenhouses gases are added to the atmosphere or are increased in abundance, more infrared radiation is trapped in the atmosphere, which is expected to lead to higher average surface temperatures.

The term *greenhouse effect* as used here is a misnomer because the walls of a true greenhouse pass both solar and infrared radiation. An actual greenhouse warms by trapping air within its boundaries so as not to lose the warming from solar radiation to the surrounding air.

[END OF QUESTION 2.1, BOX 1]

Greenhouse-Gas Emissions

Human activities result in emissions of four principal greenhouse gases: carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and the halocarbons (a group of gases containing fluorine, chlorine, and bromine). Question 2.1, Figure 1 shows how the abundances of three of these gases have significantly increased since the start of the industrial era (about 1750). Fossil fuel use in transportation, building heating and cooling, and the manufacture of cement and other goods has increased carbon dioxide. Deforestation has also increased carbon dioxide by reducing the total uptake of carbon dioxide by plants. Methane has increased as a result of human activities related to agriculture, natural gas distribution, and landfills. Methane is also released in natural processes, such as those occurring in wetlands. More than half of methane emissions are caused by human activities. Nitrous oxide has increased less than the other gases and is also emitted by both human activities and natural processes. For most halocarbons, human activities are the sole source of emissions. Principal halocarbons include the chlorofluorocarbons (e.g., CFC-11 and CFC-12), which were used extensively in refrigeration and other uses before their presence in the atmosphere was found to contribute to stratospheric ozone depletion. In certain applications CFCs have been replaced by the hydrochlorofluorocarbons (HCFCs) and hydrofluorocarbons (HFCs), these are also greenhouse gases with growing atmospheric abundances.

[INSERT QUESTION 2.1, FIGURE 1 HERE]

Ozone

Ozone is a greenhouse gas that is not emitted directly into the atmosphere. Instead, ozone is continually produced and destroyed in the atmosphere by chemical reactions. Ozone is naturally present in both the lower atmosphere (troposphere) and upper atmosphere (stratosphere). The stratosphere contains the ozone layer, which absorbs ultraviolet light from the sun that is harmful to humans and other life. The manufacture and release of certain halocarbons in human activities has led to reductions in stratospheric ozone over the globe, with the most notable example being the ozone hole over the Antarctic continent. In the troposphere, human activities release carbon monoxide, hydrocarbon and nitrogen oxide gases, which chemically react to increase ozone. Thus, human activities indirectly produce and destroy ozone in the atmosphere.

Water vapour

Water vapour is the most abundant and important greenhouse gas in the atmosphere. However, human activities have only a small direct impact on the amount of atmospheric water vapour, although a changing climate, from any cause, can have a large effect on water vapour. Methane undergoes chemical destruction in the stratosphere to produce water vapour. As a result, increases in human methane emissions are followed by increases in stratospheric water vapour.

Aerosols

Aerosols are microscopic particles present in the atmosphere with widely varying size, concentration, and chemical composition. Aerosols contain compounds that are emitted or produced via human activities or natural processes. Human activities such as fossil fuel burning and biomass burning increase atmospheric aerosols containing sulphur compounds, organic compounds, and black carbon. Natural sources include mineral dust released from the surface, biogenic emissions from the land and oceans, and volcanic eruptions. Atmospheric sulphate levels also increase temporarily for up to several years following major explosive volcanic eruptions.

[START OF QUESTION 2.1, BOX 2]

What is a radiative forcing? The impact of a climate change agent, such as a greenhouse gas, on Earth's climate is often measured using the concept of radiative forcing. Radiative forcing is a change in the energy available to the global Earth-atmosphere system due to changes in the abundances or amount of climate change agents. Radiative forcing is measured in units of 'energy per unit area of the globe' or, more specifically, 'watts per square meter' ($W\ m^{-2}$). Values are often expressed as an average global value for a particular agent. The forcing arises because an agent interacts with incoming solar radiation or outgoing thermal radiation from the surface or atmosphere and, thereby, forces a change in the net energy balance in the atmosphere. A positive radiative forcing is expected to lead to a warming of climate and a negative forcing is expected to lead to a cooling. Net radiative forcing from all human activities is related to the sum of forcings over all agents. Scientists have shown that the global temperatures are very likely to increase as net radiative forcing increases.

[END OF QUESTION 2.1, BOX 2]

Radiative Forcing of climate change agents

The contributions to radiative forcing from each of the agents influenced by human activities are shown in Question 2.1, Figure 2. The forcings for greenhouse gas changes are all positive. Carbon dioxide increases cause the largest forcing.

Aerosols cause radiative forcing *directly* through interaction with solar and thermal radiation in the atmosphere. The direct radiative forcing from all aerosol types is slightly negative. Some aerosols cause a positive forcing while others cause a negative forcing. Aerosols also cause radiative forcing *indirectly* through the changes they cause in cloud properties throughout the atmosphere. This effect is expected to lead to a significant negative forcing.

Radiative Forcing from other climate change agents

Human activities since the industrial era have increased the nature of land cover over the globe, principally through changes in croplands, pastures, and forests. They also modify the reflective properties of ice surfaces. Overall, more solar radiation is now being reflected from the global surface as a result of our activities, which results in a negative forcing.

[INSERT QUESTION 2.1, FIGURE 2 HERE]

Aircraft produce contrails at cruise altitudes in regions that have suitably low temperatures and high humidity. Contrails are a form of cirrus cloud that both reflect solar radiation and absorb infrared radiation. The effect of global aircraft operations is to increase the total amount of cirrus cloudiness and, thereby, cause a small positive radiative forcing; however with future growth in aircraft flights, this forcing is one that is expected to increase substantially in the future.

Radiative Forcing from Natural Changes

The largest known natural forcings are solar changes and explosive volcanic eruptions. In the industrial era, solar output has increased gradually with the largest changes in the 20th century, creating a positive radiative forcing. The increases in solar radiation are in addition to the cyclic changes that follow an 11-year cycle. Solar energy directly heats the climate system and also affects the atmospheric abundance of some greenhouse gases, such as ozone. Explosive volcanic eruptions can create a short-lived (2 to 3 years) negative forcing through the associated increases in sulphate aerosol in the stratosphere. The known long-term trend in natural solar and episodic volcanic forcing in the 20th century are both small compared to the trends in forcing agents due to human activities. Overall, the total radiative forcing from all human activities since the start of the industrial era is positive and exceeds the change in natural forcings over that period.

[END QUESTION 2.1]

Tables

Table 2.3.1. Present day concentrations and RF for the measured LLGHGs. The changes since TAR are also shown.

	Concentrations and their changes		Radiative Forcing	
Species	2004 (ppt)	2004–1998 (ppt)	2004 (W m^{-2})	Change since 1998
CO_2	**377 ± 1 ppm**	**+12 ppm**	**1.63**	**+12%**
CH_4	**1776 ± 44 ppb**	**+22 ppb**	**0.48**	**-**
N_2O	**319 ± 0.5 ppb**	**+5 ppb**	**0.16**	**+6%**
CFC-11	254 ± 1	-8	0.063	3%
CFC-12	540 ± 5	+6	0.17	+1%
CFC-113	79.2 ± 0.5	–3.7	0.023	–5%
CH_3CCl_3	22 ± 1	-	0.0013	-
CCl_4	94 ± 2	–6	0.012	–6%
HCFC-22	164 ± 10	+33	0.033	+25%
HCFC-141b	17 ± 1	+7.6	0.0024	+81%
HCFC-142b	15 ± 1	+5	0.0030	+50%
HFC-125	3.3 ± 0.5[a]	+2.4	0.0008	+260%
HFC-134a	30 ± 4	+22.2	0.0048	+275%
HFC-152a	3.3 ± 1.5[a]	+2.0	0.0003	+150%
HFC-23	18.2 ± 0.3[b]	+4.8	0.0034	+36%
SF_6	5.4 ± 0.1[c]	+1.3	0.0028	+32%
CF_4	73 ± 1[d]	-	0.0020	-
C_2F_6	3.0 ± 0.1[b]	+0.6	0.00077	+25%
CFCs Total			**0.263**	**–1%**
HCFC Total			**0.038**	**+30%**
Montreal Gases			**0.315**	**+1%**
Other Kyoto Gases			**0.015**	**+66%**
Halocarbons			**0.330[e]**	**+3%**
Total LLGHGs			**2.59**	**+7%**

Notes:

Errors are standard deviation of combined 2004 data, including intra-annual standard deviation, measurement and global averaging uncertainty. Percent changes are calculated relative to 1998 reference. 90% confidence ranges in RF range are not shown but are ~10% (see Section 2.10).

CO_2 are combined measurements from the CMDL and SIO networks (Section 2.3.1).

CH_4 measurements are combined data from the CMDL and AGAGE networks (Section 2.3.2).

Halocarbon measurements are average of CMDL and AGAGE networks. UEA and PSU measurements were also used (Section 2.3.3). CFC total includes a small 0.004 W m^{-2} RF from CFC-114, CFC-115 and the halons, as measurements of these were not updated.

Preindustrial values are zero except for: CO_2 (278 ppm), CH_4 (715 ppb; 700 ppb used in TAR), N_2O (270 ppb), CF_4 (40 ppt)

Radiative efficiencies are from Table 2.10.1

(a) Data available from AGAGE network only

(b) Data from UEA only

(c) Data from CMDL only

(d) 1997 data from PSU(Khalil *et al.,* 2003), (not updated) preindustrial level of 46ppt assumed for RF calculation

(e) Totals are not perfect sums, due rounding of higher precision data.

Table 2.4.1. Showing the periods of operation, spectral bands, and products available from various different satellite sensors that have been used to retrieve aerosol properties.

Satellite Instrument	Period of operation	Spectral bands	Products	Comment & Reference
AVHRR (Advanced Very High Resolution Radiometer)	1979–present	5-bands (0.63, 0.87, 3.7, 10.5, 11.5 µm)	τ_{aer}, Å	1-channel retrieval gives $\tau_{\lambda\tilde{\square}63}$ over ocean (Husar *et al.,* 1997; Ignatov and Stowe*,* 2002) 2-channel using 0. 63 µm and 0.86 µm gives $\tau_{\lambda=0.55}$ and Å over ocean assuming mono-model aerosol size distribution (Mishchenko *et al.,* 1999) 2-channel using 0. 63 µm & 0.86 µm gives $\tau_{\lambda=0.55}$ and Å over dark forests and lake surfaces (Soufflet *et al.,* 1997)
TOMS (Total Ozone Mapping Spectrometer)	1979–present	0. 33 µm, 0.36 µm	Aerosol Index, τ_{aer}	Aerosol index to τ_{aer} conversion sensitive to the altitude of the8 mono-modal aerosol models used in the retrieval (Torres *et al.* 2002).
POLDER (Polarization and Directionality of the Earth's Reflectances)	Nov 1996–June 1997; Apr 03–Oct 2003	8 bands [0.44–0.91 µm]	τ_{aer}, Å, DRE	Multiple view angles and polarization capabilities. 0.67 µm and 0.86m radiances used with 12 mono-modal aerosol models over ocean (Goloub *et al.,* 1999; Deuzé *et al.,* 2000). Polarization allows fine particle retrieval over land (Herman *et al.,* 1997; Goloub and Arino*,* 2000). DRE determined over ocean (Boucher and Tanre*,* 2000; Bellouin *et al.,* 2003).
OCTS (Ocean colour and temperature scanner)	Nov 1996–Jun 1997; Apr 2003–Oct 2003	9 bands [0.41–0.86 µm] and 3.9 µm	τ_{aer}, Å	0.67 µm and 0.86 µm retrieval gives $\tau_{\lambda=0.50}$ and Å over ocean. Bi-modal aerosol size distribution assumed (Higurashi *et al.,* 2000).
MODIS (Moderate resolution Imaging Spectrometer)	2000–present	12 bands 0.41–2.1 µm	τ_{aer}, Å , DRE	Retrievals developed over ocean surfaces using bi-modal size distributions (Tanré *et al.,* 1997; Remer *et al.,* 2002) Retrievals developed over land except bright surfaces (Kaufman *et al,* 1997; Chu *et al.,* 2002). Optical depth speciation and DRE determined over ocean and land (e.g., Bellouin *et al.,* 2005; Kaufman *et al.,* 2005).
MISR (Multi-angle Imaging Spectro-Radiometer)	2000–present	4 bands [0.47–0.86 µm]	τ_{aer}, Å	9 different viewing angles. Five climatological mixing groups composed of four component particles are used in the retrieval algorithm (Kahn *et al.,* 2001; Kahn *et al.,* 2005). Retrievals over bright surfaces are possible (Martonchik *et al.,* 2004).
CERES (Clouds and the Earth's Radiant Energy System)			DRE	DRE determined by a regression of e.g. VIRS (AVHRR-like) τ_{aer} against upwelling irradiance Loeb and Kato (2002)
GLAS Geoscience Laser Altimeter System	2003–present	Active lidar [0.53, 1.06 µm]	Aerosol vertical profile	Lidar footprint ~70m at 170 m intervals. 8-day repeat orbiting cycle.
ATSR-2/AATSR (Along Track Scanning Radiometer)	1996–present	4-bands [0.56–1.65 µm]	τ_{aer}, Å	Nadir and 52º forward viewing geometry. 40 aerosol climatological mixtures containing up to six aerosol species are used in the retrievals Holzer-Popp *et al.* (2002).

Table 2.4.2. The Direct Radiative Effect (DRE) estimated from satellite remote sensing studies (adapted and updated from Yu *et al.,* 2005).

Reference	Instrument	Data Analysed	Brief Description	Clear Sky DRE (ocean)
Bellouin *et al.* (2005)	MODIS/ TOMS/ SSMI	2002	MODIS fine and total τ_{aer} with TOMS AI and SSMI to discriminate dust from sea-salt.	–6.8
Loeb and Manalo-Smith (2005)	CERES/ MODIS	Mar 2000–Dec 2003	CERES radiances/irradiances and angular distribution models and aerosol properties from either MODIS[*1] or from NOAA-NESDIDS[*2] algorithm used to estimate the direct radiative effect.	–3.8[*2] to –5.5[*1]
Remer and Kaufman (2005)	MODIS	Aug 2001–Dec 2003	τ_{aer} from fine mode fraction.	–5.7 ± 0.4
Zhang *et al.* (2005) ; Christopher and Zhang (2004)	CERES/ MODIS		MODIS aerosol properties and CERES radiances/irradiances and angular distribution models used to estimate the direct radiative effect.	–5.3 ± 1.7
Bellouin *et al.* (2003)	POLDER		Best prescribed aerosol model fitted to POLDER data	–5.2
Loeb and Kato (2002)	CERES/ VIRS		τ_{aer} from VIRS regressed against the top of the atmosphere CERES irradiance (35°N-35°S).	–4.6 ± 1.0
Chou *et al.* (2002)	SeaWiFs			–5.4
Boucher and Tanré (2000)	POLDER		Best prescribed aerosol model fitted to POLDER data	–5 to –6
Mean				–5.3

Table 2.4.3. The direct radiative forcing for sulphate aerosol derived from models published since TAR and from the AEROCOM simulations using identical emissions. AODant; fraction of anthropogenic sulfate to total sulfate aerosol optical depth of present day.

No	**Model**	**LOAD [mg SO_4/m^2]**	**AOD**	**AODant [%]**	**DRFTOA [W/m^2]**	**NDRFM [$Wm^{-2}gSO_4^{-2}$]**	**NDRF [Wm^{-2} τ^{-1}]**	**Reference**
Published since IPCC, 2001								
A	CCM3	2.23			–0.56	–251		(Kiehl *et al.,* 2000)
B	GEOSCHEM	1.53	0.018		–0.33	–216	–18	(Martin *et al.,* 2004)
C	GISS	3.30	0.022		–0.65	206	–32	(Koch, 2001)
D	GISS	3.27			–0.96	–293		(Adams *et al.,* 2001)
E	GISS	2.12			–0.57	–269		(Liao and Seinfeld, 2005)
F	KYU	2.16	0.028		–0.21	–97	–8	(Takemura *et al.,* 2005)
G	LMD	2.76			–0.42	–152		(Boucher and Pham., 2002)
H	LOA	3.03	0.030		–0.41	–135	–14	(Reddy *et al.,* 2005b)
I	GATORG	4.29			–0.31	–72		(Jacobson, 2001)
J	PNNL	5.50	0.042		–0.44	–80	–10	(Ghan *et al.,* 2001)
K	UIO_CTM	1.79	0.019		–0.37	–207	–19	(Myhre *et al.,* 2004)
L	UIO-GCM	2.28			–0.29	–127		(Kirkevag and Iversen, 2002)
AEROCOM / identical emissions used for year 1750 and 2000								
M	UMI		0.020	58%				(Liu and Penner, 2002)
N	UIO_CTM	1.70	0.019	57%	–0.35*			(Myhre *et al.,* 2003)
O	LOA	2.87	0.028	58%	–0.39*			(Reddy and Boucher, 2004)
P	LSCE	3.01	0.023	60%	–0.40	–133	–18	
Q	MPI_HAM	2.47	0.026		–0.46	–186	–18	(Stier *et al.,* 2005)
R	GISS	1.32	0.006	39%	–0.27*			(Koch, 2001)
S	UIO_GCM	1.70	0.012	59%	–0.22*			(Iversen and Seland, 2002; Kirkevag and Iversen, 2002)
T	KYU	3.54	0.013	59%	–0.34*			(Takemura *et al.,* 2005)
U	ULAQ	1.62	0.019	41%				(Pitari *et al.,* 2002)
Average A-L		2.86	0.026		–0.46	–175	–17	
Average M-U		2.28	0.018	54%	–0.35			
Minimum A-U		1.32	0.006	39%	–0.96	–293	–32	
Maximum A-U		5.50	0.042	60%	–0.21	–72	–8	
Stddev A-L		1.13	0.009		0.20	75	9	
Stddev M-U		0.80	0.007	9%	0.08			

Notes:

Forcing efficiencies per mass sulfate (NDRFM) were used to derive the AEROCOM sulphate forcings where model results were only available for the original model.

Table 2.4.4. Recent estimates of anthropogenic carbonaceous aerosol forcing. BB=biomass burning sources included, FFBC=fossil fuel forcing, FFPOM= fossil fuel particulate organic matter.

No	Model	LOAD POM [mgPOM/m2]	AOD POM	AOD ant	LOAD BC	DRF TOA BCPOM [W m^{-2}]	DRF TOA POM	DRF TOA BC	DRF TOA FFPOM	DRF TOA FFBC	DRF TOA BB	Reference
Published since TAR												
A	KYU					0.04	–0.29	0.33	–0.06	0.17	–0.06	(Takemura *et al.,* 2001)
B	LOA	2.33	0.016		0.37	0.30	–0.25	0.55	–0.02	0.19	0.14	(Reddy *et al.,* 2005b)
C	GISS	1.86	0.017		0.29	0.35	–0.26	0.61	–0.13	0.49	0.065	(Hansen *et al.,* 2005)
D	GISS	1.86	0.015		0.29	0.05	–0.30	0.35	–0.08*	0.18*	–0.05*	(Koch, 2001)
E	GISS	2.39			0.39	0.32	–0.18	0.5	–0.05*	0.25*	0.12*	(Chuang *et al.,* 2002)
F	GISS	2.49			0.43	0.30	–0.23	0.53	–0.06*	0.27*	0.09*	(Liao and Seinfeld, 2005)
G	KYU	3.22	0.032		0.67	0.15	–0.27	0.42	–0.07*	0.21*	0.01*	(Takemura *et al.,* 2005)
H	GATORG				0.45	0.47	–0.06	0.53	–0.01*	0.27*	0.22*	(Jacobson, 2001)
I	MOZGN	3.03	0.018				–0.34					(Ming *et al.,* 2005a)
J	CCM				0.33			0.34				(Wang, 2004)
K	UIO-GCM				0.30			0.19				(Kirkevag and Iversen, 2002)
AEROCOM / identical emissions used for year 1750 and 2000												
L	UMI		0.006	53%								(Liu and Penner, 2002)
M	UIO_CTM	1.12	0.006	55%	0.19	0.03#	–0.07§	0.13§	–0.04	0.11	–0.04	(Myhre *et al.,* 2003)
N	LOA	1.35	0.008	51%	0.22	0.09#	–0.22§	0.41§	–0.05*	0.20*	0.04*	(Reddy and Boucher, 2004)
O	LSCE	1.49	0.008	46%	0.25	0.17	–0.15	0.32	–0.04*	0.16*	0.05*	
P	ECHAM	1.65	0.005		0.22	0.23	–0.54§	1.01§	–0.14*	0.50*	0.10*	(Stier *et al.,* 2005)
Q	GISS	1.21	0.006	51%	0.24	0.15#	–0.35§	0.64§	–0.09*	0.32*	0.06*	(Koch, 2001)
R	UIO_GCM	0.87	0.005	59%	0.19	0.20#	–0.47§	0.88§	–0.12*	0.44*	0.08*	(Iversen and Seland, 2002; Kirkevag and Iversen, 2002)
S	KYU	2.13	0.020	52%	0.37	0.33#	–0.78§	1.44§	–0.19*	0.72*	0.14*	(Takemura *et al.,* 2005)
T	ULAQ	1.72	0.007	58%	0.38							(Pitari *et al.,* 2002)
Average A-K		2.45	0.02		0.39	0.25	–0.24	0.44	–0.06	0.25	0.07	
Average L-T		1.44	0.01	53%	0.26	0.17	–0.37	0.69	–0.10	0.35	0.06	
Minimum A-T		0.87	0.00	46%	0.19	0.03	–0.78	0.13	–0.19	0.11	–0.06	
Maximum A-T		3.22	0.03	59%	0.67	0.47	–0.06	1.44	–0.01	0.72	0.22	
Stddev A-K		0.52	0.01		0.12	0.15	0.08	0.13	0.04	0.10	0.10	
Stddev L-T		0.39	0.00	4%	0.08	0.10	0.25	0.45	0.06	0.22	0.06	

Notes:
(b) Reddy *et al.* SW and LW
BCPOM forcing: total aerosol forcing - sulfate forcing
Models A-C are used to provide a split in sources and components where missing:
* Models D-I: source split derived from total POM and BC
[FFPOM=POM*0.25/FFBC=BC*.5/BB=(POM+BC)-(FFPOM+FFBC)]
§ Models M-T: in addition POM and BC split derived from total carbonaceous effect [POM=BCPOM*-2.3 and BC=BCPOM*4.3]

Table 2.4.5. Recent estimates of anthropogenic aerosol load (LOAD), anthropogenic aerosol optical depth (AOD), its fraction of total aerosol optical depth (AODant), cloud cover in these aerosol model versions, total aerosol direct radiative forcing at the top of the atmosphere (DRF TOA), at the surface (DRF surface) and the resultant column heating (DRF column).

No	**Model**	**LOAD**	**AOD**	**AOD ant**	**Cloud cover**	**DRF TOA clear sky**	**DRF TOA all sky**	**DRF surface all sky**	**DRF column all sky**	**Reference**
		[mg/m2]	**[ ]**	**[%]**	**[%]**		**[W/m2]**			
A	GISS	5.0			79%		–0.27			(Liao and Seinfeld, 2005)
B	LOA	6.0	0.049	34%	70%	–0.53	–0.09			(Reddy and Boucher, 2004)
C	KYU	7.4	0.060	57%	63%	–0.77	–0.04	–1.92	1.88	(Takemura *et al.,* 2005)
D	UIO-GCM	2.7			57%		–0.11			(Kirkevag and Iversen, 2002)
E	GATORG					–0.89	–0.12			(Jacobson, 2001)
F	GISS	6.7	0.049				–0.23			(Hansen *et al.,* 2005)
G	GISS	5.6	0.040				–0.63			(Koch, 2001)
AEROCOM / identical emissions used for year 1750 and 2000										
H	UMI		0.028	25%	63%	–0.80	–0.41	–1.24	0.84	(Liu and Penner, 2002)
I	UIO_CTM	3.0	0.026	20%	70%	–0.85	–0.34	–0.95	0.61	(Myhre *et al.,* 2003)
J	LOA	4.4	0.039	24%	70%	–0.60	–0.23	–1.26	1.03	(Reddy and Boucher, 2004)
K	LSCE	5.3	0.033	37%	70%	–0.87	–0.24	–0.90	0.66	
L	MPI_HAM	4.3	0.031	22%	72%	–0.50	–0.12	–1.07	0.95	(Stier *et al.,* 2005)
M	GISS	1.9	0.013	10%	79%		–0.01	–0.81	0.79	(Koch, 2001)
N	UIO_GCM	2.7	0.028	19%	57%		–0.01	-0.85	0.84	(Iversen and Seland, 2002; Kirkevag and Iversen, 2002)
O	KYU	5.3	0.035	45%	63%		0.04	–0.91	0.96	(Takemura *et al.,* 2005)
P	ULAQ	4.9	0.030	39%			0.07			(Pitari *et al.,* 2002)
Average A-G		5.6	0.049	46%	67%	–0.73	–0.21	–1.92	1.88	
Average H-P		4.0	0.029	27%	68%	–0.72	–0.14	–1.00	0.83	
Minimum A-P		1.9	0.013	10%	57%	–0.89	–0.63	–1.92	0.61	
Maximum A-P		7.4	0.060	57%	79%	–0.50	0.07	–0.81	1.88	
Stddev A-G		1.6	0.008			0.19	0.20			
Stddev H-P		1.3	0.007	11%		0.17	0.17	0.17	0.15	

Table 2.4.6. Published model studies of the cloud albedo RF with most relevant modelling details.

Model	Model type	Aerosol types[a]	Aerosol mixtures[b]	Cloud Types for Indirect Effect	Microphysics	**Cloud albedo Radiative Forcing (W/m^2)**
Lohmann *et al.* (2000)	AGCM + sulphur cycle (ECHAM4)	S, OC, BC, SS, D	I	warm and mixed phase	Droplet number conc. and LWC, Beheng (1994), Sundqvist *et al.* (1989). Also, mass and number from field observations	–1.1 (total) –0.45 (albedo)
			E			–1.5 (total)
Jones *et al.* (2001)	AGCM (SST) + sulphur cycle (Hadley)	S, SS D(crude attempt over land, no radiation)	E	stratiform and shallow Cu	Droplet number conc. and LWC Wilson and Ballard (1999), Smith (1990), Tripoli and Cotton (1980); Bower *et al.* (1994) warm and mixed phase, radiative treatment of anvil cirrus, non-spherical ice particles	–1.89 (total) **–1.34 (albedo)**
Williams *et al.* (2001b)	GCM with slab ocean + sulphur cycle (Hadley) --------------- AGCM (SST)	S, SS	E	stratiform and shallow Cu	Jones *et al.* (2001)	–1.69 (total) **–1.37 (albedo)** -------------------------- –1.62 (total) **–1.43(albedo)**
Rotstayn and Penner (2001) ------------------- Rotstayn and Liu (2003)	AGCM (CSIRO) (prescribed SST and sulphur loading)	S	-	warm	Gregory and Rowntree (1990), Tripoli and Cotton (1980); warm and mixed phase Inclusion of dispersion	–1.46 (albedo) -------------------------- (12–35% decrease)
Ghan *et al.* (2001)	AGCM (PNNL) + chemistry (MIRAGE)	S, OC, BC, SS, N, D	E (for different modes) I (within modes)		droplet number conc. and LWC, crystal concentration and IWC different processes affecting the modes	–1.7 (total) **–0.85 (albedo)**
Chuang *et al.* (2002)	CCM1 (NCAR)+ chemistry (GRANTOUR)	S, OC, BC, SS, D	E (for emitted particles) I: when growing by condensation	warm	Modified from Chuang and Penner (1995), no collision/coalescence warm and mixed phase	**–1.85 (albedo)**
Menon *et al.* (2002b)	GCM (GISS) + sulphur cycle (SST?)	S,OC, SS	E	warm	Droplet number conc. and LWC, Del Genio *et al.* (1996), Sundqvist *et al.* (1989) warm and mixed phase, improved vertical distribution of clouds (but only 9 layers) global aerosol burdens poorly constrained	–2.41 (total) **–1.55 (albedo)**
Kristjansson (2002)	CCM3 (NCAR)	S, OC, BC, SS, D	E (for natural) I (for		Detailed aerosol model included	-1.82 (total) **-1.35 (albedo)**

			anthropogenic)			
Suzuki *et al.* (2004)	AGCM Japan (SST)	S, OC, BC, SS	E	stratiform	Berry (1967), Sundqvist (1978), L&F (1997)	**–0.54 (albedo)**
Hansen *et al.* (2005)	GCM (GISS) Ocean A (SST) Ocean B Ocean C	S, OC, BC, SS, N, D (D not included in clouds)	E	warm and shallow (below 720hPa)	Schmidt *et al.* (2005), 20 vertical layers Droplet number concentration (Menon and Del Genio, 2005)	**–0.77 (albedo)**
Ming *et al.* (2005b)	AGCM (GFDL) (prescribed SST and sulphur loading)	S	-	warm	Rotstayn *et al.* (2000), Khainroutdinov and Kogan (2000), (aerosols offline)	–2.3 (total) **–1.4 (albedo)**
Storelvmo *et al.* (2005)	CCM3 (NCAR)+ sulphur and carbon cycles (slab ocean)	S, OC, BC, SS, D	E (for natural) I (for anthropogenic)	warm and mixed phase	Kristjansson (2002)	–1.15 (total, at the surface)

Notes:
(a) S: sulphate, SS: sea salt, D: mineral dust, BC: black carbon, OC: organic carbon, N: nitrate
(b) E: external, I: internal

Table 2.6.1. Radiative forcing terms for global subsonic aircraft operations

Radiative forcing ($W m^{-2}$) [a]	1992 IPCC[b]	2000 IPCC[c]	2000 [d]	2004 [e]
CO_2 [d]	0.018	0.025	0.025	0.028
Persistent contrails	0.020	0.034	0.010 (0.007–0.015)	0.011
Aviation-induced cloudiness without persistent contrails	0–0.040	–		
Aviation-induced cloudiness (with persistent contrails)			0.030 (0.010–0.080)	0.034

Notes:
(a) Values for contrails are best estimates. Values in parentheses give the uncertainty range.
(b) Values from IPCC-1999 (IPCC, 1999)
(c) Values interpolated from 1992 and 2015 estimates in IPCC-1999 (Sausen *et al.*, 2005)
(d) Sausen *et al.* (2005)
(e) Values projected from year 2000 values using a 3.2%/yr growth rate (increase of 13%) of fuel burn and the assumption that contrail and cloud effects vary linearly with fuel burn. Inventories of aviation fuel burn for 2004 are not yet available.

Table 2.7.1. Compared are estimates of the reduction in total solar irradiance and RF during the Maunder Minimum relative to contemporary solar minimum.

Reference	Assumptions and Technique	Maunder Minimum irradiance reduction (global RF) from contemporary minimum (W m^{-2})
Schatten and Orosz (1990)	11-year cycle extrapolation	~ 0.0 (0)
Lean, Skumanich, and White (1992)	no spots, plage, network in Ca images	1.5 (0.26)
Lean, Skumanich, and White (1992)	no spots, plage, network and reduced basal emission in cell centres in Ca images non-cycling stars	2.6 (0.45)
Hoyt and Schatten (1993)*	convective restructuring implied by changes in sunspot umbra/penumbra ratios	3.7 (0.65)
Lean, Beer, and Bradley (1995)	non-cycling stars	2.6 (0.45)
Fligge and Solanki (2000)*	combinations of above	4.1 (0.72)
Lean (2000)	non-cycling stars (revised solar- stellar calibration)	2.2 (0.38)
Foster (2004)Model #1	non magnetic sun estimates by removing bright features from MDI images	1.6 (0.28)
Foster (2004)Model #3	extrapolated from fit of 11-year smoothed total solar irradiance composite	0.8 (0.14)
Solanki and Krivova (2005)	accumulation of bright sources from simple parameterization of flux emergence and decay	2.2 (0.38)
Wang, Lean, and Sheeley (2005)*	flux transport simulations of total magnetic flux evolution	0.5 (0.09)

Notes:

The solar activity cycle of order 1 W m^{-2} is superimposed on this increase.

The RF is the irradiance change divided by 4 (geometry) and multiplied by 0.7 (albedo).

Reconstruction identified by * extend only to 1713, the end of the Maunder Minimum.

Table 2.9.1. Global mean radiative forcings

	Global mean radiative forcing (1750-2004) (W m^{-2}) [Uncertainty]			
	SAR	TAR	AR4	Summary comments
Long Lived Greenhouse gases {Comprising CO_2, CH_4, N_2O, and halocarbons}	**+2.45 [15%] {CO_2 (1.56); CH_4 (0.47); N_2O (0.14); Halocarbons (0.28)}**	**+2.43 [10%] {CO_2 (1.46); CH_4 (0.48); N_2O (0.15); Halocarbons (0.34)}**	**+2.59 [±0.26] {CO_2 (1.63); CH_4 (0.48); N_2O (0.15); Halocarbons (0.33)}**	**Total increase in RF, due to upward trends, particularly in CO_2. Halocarbon RF- re-evaluated to be lower (trend is positive)**
Total ozone	**Not evaluated**	**Not evaluated**	**+0.30 ± 0.20**	**Combined RF not estimated in TAR**
Stratospheric O_3	–0.1 [2X]	–0.15 [67%]	–0.10 ± 0.04	Re-evaluated to be slightly smaller
Tropospheric O_3	+0.40 [50%]	+0.35 [43%]	+0.40 ± 0.15	Better constrained
Total direct aerosol	**Not evaluated**	**Not evaluated**	**-0.20 ± 0.20**	**Re-evaluation gives smaller estimate than TAR**
Direct sulphate aerosols	*–0.40 [2X]*	*–0.40 [2X]*	*–0.40 ± 0.20*	*Better constrained*
Direct nitrate aerosols	*Not evaluated*	*Not evaluated*	*–0.15 ± 0.15*	*Newly evaluated*
Direct biomass burning aerosols	*–0.20 [3X]*	*–0.20 [3X]*	*+0.0 6± 0.08*	*Re-evaluated to be +ve. Response affected by semi-direct effects*
Direct FF aerosols (BC)	*+0.10 [3X]*	*+0.20 [3X]*	*+0.30 ± 0.15*	*Re-evaluated to be slightly larger*
Direct FF aerosols (OC)	*Not evaluated*	*–0.10 [3X]*	*-0.08 ± 0.05*	*Similar*
Direct mineral dust aerosols	*Not evaluated*	*–0.60 to +0.40*	*–0.20 to +0.10*	*Re-evaluated to have a reduced range and smaller magnitude*
Cloud albedo indirect aerosol effect	**0 to –1.5 (sulphate only)**	**0.0 to –2.0 (all aerosols)**	**–1.2 [±0.7] (all aerosols)**	**best estimate now possible**
Contrails Aviation-induced cirrus	**Not evaluated**	**0.02 [3.5 X]** 0 to +0.04	**0.01 [2X]** 0 to 0.05	Re-evaluated contrail forcing is smaller and better constrained
Anthropogenic surface albedo	**Not evaluated**	**Not evaluated**	**–0.10 [±0.3]**	**Combination of opposing RFs**
Land-use(albedo)	*Not evaluated*	*–0.20 [100%]*	*–0.20 [±0.20]*	*Additional studies*
Black-Carbon on snow (albedo)	*Not evaluated*	*Not evaluated*	*+0.10 [2X]*	*Not evaluated in TAR*
Anthro. water vapour	**Not evaluated**	**Not evaluated**	**+0.13 [2X]**	**Combined RF**
Stratospheric: from CH_4	*Not evaluated*	*+0.01 to +0.03*	*+0.10 [±0.05]*	*Re-evaluated to be 5-10x higher than TAR*
Tropospheric: from irrigation	*Not evaluated*	*Not evaluated*	*+0.03 [3X]*	*Not evaluated in TAR*
Solar (direct)	**+0.30 [67%]**	**+0.30 [67%]**	**+0.12 [2X]**	**Re-evaluated to be less than half of TAR value**

Notes:
Bold rows appear on Figure 2.9.1

Table 2.9.2. Uncertainty assessment of RFs discussed in this report.

	Evidence	Consensus	Overall	Certainties	Uncertainties	RF range
LLGHGs	A	1	High	Past and present concentrations; spectroscopy	Preindustrial concentrations of some species; vertical profile in stratosphere; spectroscopic strength of minor gases	Uncertainty assessment of measured trends from different datasets and differences between radiative transfer models
Stratospheric ozone	A	2	Medium	Measured trends and its vertical profile since 1980; cooling of stratosphere; spectroscopy	Changes prior to 1970; trends near tropopause; effect of recent trends	Range of model results weighted to calculations employing trustworthy observed ozone trend data
Tropospheric ozone	A	2	Medium	Present day concentration at surface and some knowledge of vertical and spatial structure of concentrations and emissions; spectroscopy	Preindustrial values and role of changes in lightening; vertical structure of trends near tropopause; aspects of emissions and chemistry	Range of published model results, upper-bound increased to account for anthropogenic trend in lightening
Stratospheric water vapour From methane	B	3	Low	Global trends since 1990; methane contribution to trend; Spectroscopy	Global trends prior to 1990; radiative transfer in climate models; CTM models of methane oxidation	Range based on uncertainties in methane contribution to trend and radiative transfer
Stratospheric water vapour From other	C	3	V. Low	Empirical and simple model studies suggest link; spectroscopy	Other causes of water vapour trends poorly understood	Not given
Tropospheric water vapour from irrigation	C	3	V. Low	Process understood; spectroscopy; some regional information	Global injection poorly quantified	Range based on uncertainties in estimating global sources
Direct scattering aerosols	A	2	Medium	Ground-based and satellite observations; source regions and modelling	Emission sources and their history, optical properties, mixing and separation from natural background aerosol	Range of published model results with allowances made for under sampling of parametric uncertainties.
Direct absorbing aerosols	A	2	Low	Ground-based and satellite observations; some source regions and modelling	Vertical structure of aerosol, its optical properties, mixing and separation from natural background aerosol; emission sources and their history	Range of published model results with allowances made for under sampling of parametric uncertainties.
Cloud albedo effect	B	2	Low	Observed in case studies – e.g., ship tracks; GCMs model one	Lack of observational evidence of a global forcing	Range of published model results with allowances made for under sampling of parametric uncertainties.
Cloud lifetime effect	B	3	V. Low	Some evidence from models	Little consensus, lack of observations	Not given
Semi direct	B	3	V. Low	Cloud resolving models and GCM model exhibit an effect	Lack of observations; lack of agreement on sign of global semi direct effect	Not given

Contrails and Aviation Cirrus	A	2	Med	Cirrus radiative and microphysical properties; aviation emissions; contrail coverage	Aviation's effect on cirrus clouds; transformation of contrails to cirrus	Best estimate based on recent work and range from published model results
Solar	A	2	Med	Measurements over last 25 years; proxy indictors of solar activity	Relationship between proxy data and TSI; indirect ozone effects;	Range from available reconstructions of TSI and there qualitative assessment
Cosmic rays	C	3	V. Low	Some empirical evidence and some observations microphysical models suggest link to clouds	General lack/doubt regarding physical mechanism; dependence on correlation studies	Not given
Surface albedo	B	3	Low	Some quantification of deforestation and desertification; estimates of black carbon aerosol on snow	Separation of anthropogenic changes from natural; mixing of snow and black carbon aerosol	Based on range of published estimates and published uncertainty analyses.
Non-albedo – surface effects	C	3	V. Low	Some model studies suggest link	Separation of anthropogenic changes from natural; quantification of RF	Not given
Volcanic	A	2	Med	Observed aerosol changes from Mt Pinatubo and El Chichón; proxy data for past eruptions; radiative effect of volcanic aerosol	Stratospheric aerosol concentrations from pre 1980 eruptions; atmospheric feedbacks	

Notes:
Evidence for the forcing is given a grade (A-C), with A implying strong evidence and C insufficient evidence.
The degree of consensus among forcing estimates is given a 1, 2 or 3 grade. Where grade 1 implies a good-deal of consensus and grade 3 implies an insufficient consensus.
From these two factors an understanding level is determined (quoted in the 4th Column).

Table 2.10.1. Global Warming Potentials (GWPs)

Industrial Designation or Common Name	Chemical Formula	Other Name	Lifetime[a] (years)	Radiative Efficiency[a] (W m^{-2} ppb$^{-1)}$	Global Warming Potential for Given Time Horizon (years)			
					TAR (100)	20	100	500
Carbon dioxide	CO_2				1	1	1	1
Methane	CH_4		12.0[c]	3.7x10^{-4}	23	63	23	7
Nitrous oxide	N_2O		114[c]	3.1x10^{-3}	296	275	296	156
Substances controlled by the Montreal Protocol								
CFC-11	CCl_3F	Trichlorofluoromethane	45	0.25	4680	6330	4680	1630
CFC-12	CCl_2F_2	Dichlorodifluoromethane	100	0.32	10720	10340	10720	5230
CFC-113	CCl2FCClF2	1.1.2-trichlorotrifluoroethane	85	0.3	6030	6150	6030	2700
CFC-114	CClF2CClF2	Dichlorotetrafluoroethane	300	0.31	9880	7560	9880	8780
CFC-115	CClF2CF3	Monochloropentafluoroethane	1700	0.18	7250	4990	7250	10040
Halon-1301	CBrF3	Bromotrichloroethane	65	0.32	7030	7970	7030	2780
Halon-1211	CBrClF2	Bromochlorodifluoroethane	16[d]	0.3	1860	4460	1860	578
Halon-2402	CBrF2CBrF2	Dibromotetrafluoroethane	20[d]	0.33[d]	1620[d]	3460[d]	1620[d]	505[d]
Carbon tetrachloride	CCl4	(Halon-104)	26[d]	0.13	1380[d]	2540[d]	1380[d]	437[d]
Methyl bromide	CH3Br	(Halon-1001)	0.7	0.01	5	16	5	1
Bromochloromethane	CH2BrCl	(Halon-1011)	0.37[d]					
Methyl chloroform	CH3CCl3	1,1,1-trichloroethane	5[d]	0.06	144[d]	476[d]	144[d]	45[d]
HCFC-22	CHClF2	Chlorodifluoromethane	12[d]	0.2	1780[d]	4850[d]	1780[d]	552[d]
HCFC-123	CHCl2CF3	Dichlorotrifluoroethane	1.3[d]	0.14[d]	76[d]	257[d]	76[d]	24[d]
HCFC-124	CHClFCF3	Chlorotetrafluoroethane	5.8[d]	0.22	599[d]	1950[d]	599[d]	186[d]
HCFC-141b	CH3CCl2F	Dichlorofluoroethane	9.3	0.14	713	2120	713	222
HCFC-142b	CH3CClF2	Chlorodifluoroethane	17.9[c]	0.2	2270	5170	2270	709
HCFC-225ca	CHCl2CF2CF3	Dichloropentafluoropropane	1.9[d]	0.2[d]	120[d]	404[d]	120[d]	37[d]
HCFC-225cb	CHClFCF2CClF2	Dichloropentafluoropropane	5.8[d]	0.32	586[d]	1910[d]	586[d]	182[d]
Hydrofluorocarbons								
HFC-23	CHF3	Trifluoromethane	270[d]	0.19[e]	14310[g]	11100[g]	14310[g]	12100[g]
HFC-32	CH2F2	Difluoromethane	4.9[d]	0.11[e]	670g	2220g	670g	210[g]
HFC-125	CHF2CF3	Pentafluoroethane	29	0.23	3450	5970	3450	1110
HFC-134a	CH2FCF3	1,1,1,2-Tetrafluoroethane	14[d]	0.16[e]	1410[g]	3590[g]	1410[g]	440[g]
HFC-143a	CH3CF3	1,1,1-Trifluoroethane	52	0.13	4400	5540	4400	1600
HFC-152a	CH3CHF2	1,1-Difluoroethane	1.4	0.09	122	411	122	38
HFC-227ea	CF3CHFCF3	1,1,1,2,3,3,3-heptafluoropropane	34.2[d]	0.25[e]	3140[g]	4930[g]	3140[g]	1030[g]
HFC-236fa	CF3CH2CF3	1,1,1,3,3,3-hexafluoropropane	240[d]	0.28	9500[d]	7620[d]	9500[d]	7700[g]

HFC-245fa	CHF2CH2CF3	1,1,1,3,3-pentafluoropropane	7.6[d]	0.28	1020[d]	3180[d]	1020[d]	316[g]
HFC-365mfc	CH3CF2CH2CF3	1,1,1,3,3-pentafluorobutane	8.6[d]	0.21	782[d]	2370[d]	782[d]	243[g]
HFC-43-10mee	CF3CHFCHFCF2CF3	1,1,1,2,3,4,4,5,5,5-decafluoropentane	15.9[d]	0.4	1610[d]	3890[d]	1610[d]	502[g]
Perfluorocarbons								
	SF6	Sulphur hexafluoride	3200	0.52	22450	15290	22450	32780
	NF3	Nitrogen trifluoride	740	0.13	10970	7780	10970	13240
PFC-14	CF4	Carbon tetrafluoride	50000	0.08	5820	3920	5820	9000
PFC-116	C2F6	Perfluoroethane	10000	0.26	12010	8110	12010	18280
PFC-218	C3F8	Perfluoropropane	2600	0.26	8690	5940	8690	12520
PFC-318	c-C4F8	Perfluorocyclobutane	3200	0.32	10090	6870	10090	14740
PFC-3-1-10	C4F10	Perfluorobutane	2600	0.33	8710	5950	8710	12550
PFC-5-1-14	C6F14	Perfluorohexane	3200	0.49	9140	6230	9140	13350
Fluorinated ethers								
HFE-449sl	CH3O(CF2)3CF3	(HFE-7100)	5	0.31	397	1310	397	123
HFE-569sf2	CH3CH2O(CF2)3CF3	(HFE-7200)	0.77	0.3	56	189	56	17
HFE-347pcf2 (i)	CF3CH2OCF2CHF2		7.1	0.25	540	1800	540	170
Hydrocarbons and other compounds - Direct Effects								
Dimethylether	CH3OCH3		0.015	0.02	1	1[b]	1[b]	<<1[b]
Methylene_chloride	CH2Cl2	(Freon-40) Dichloromethane	0.38[j]	0.03	10	35[b]	10[b]	3[b]
Methyl_chloride	CH3Cl	(Freon-30) Chloromethane	1.3	0.01	16	55[b]	16[b]	5[b]

Notes:
(a) From IPCC (2001, chapter 6)
(b) Values adopted under the UNFCCC for the national inventories
(c) Lifetime of methane includes feedbacks on emissions (IPCC, 2001, Chapter 6) and GWPs include indirect effects
(d) Updated in WMO (2003, chapter 1)
(e) Updated from two averaged model results in Gohar *et al.,* (2004) and rounded for constancy.
(f) Highwood *et al.* (1999)
(g) Scaled for the updated radiative efficiency in (e)
(h) Suggested as upper limit
(i) From original paper of Tokuhashi *et al.* (2000). IPCC (2001) erroneously referred to this compounds as HFE-374pcf2.
(j) Global lifetime estimated from a process lifetime with respect to tropospheric OH calculated relative to 6.1 years for CH_3CCl_3, assuming an average temperature of 272 K.
(k) Upper value reported by Taniguchi *et al.* (2003).
(l) From direct effects only. Some values from indirect effects are given in Table 2.10.2

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Table 2.10.2 Indirect GWPs (100) for 10 NMVOCs from Collins *et al.* (2002) and for NO_x emissions (on N-basis).

Organic compound/Study	GWP^{CH4}	GWP^{O3}	GWP
Ethane (C_2H_6)	2.9	2.6	5.5
Propane (C_3H_8)	2.7	0.6	3.3
Butane (C_4H_{10})	2.3	1.7	4.0
Ethylene (C_2H_4)	1.5	2.2	3.7
Propylene (C_3H_6)	–2.0	3.8	1.8
Toluene (C_7H_8)	0.2	2.5	2.7
Isoprene (C_5H_8)	1.1	1.6	2.7
Methanol (CH_3OH)	1.6	1.2	2.8
Acetaldehyde (CH_3CHO)	–0.4	1.7	1.3
Acetone (CH_3COCH_3)	0.3	0.2	0.5
Derwent *et al.* NH surface NO_x	–8.5	3.9	–4.6
Derwent *et al.* SH surface NO_x	–24	12	–12
Berntsen *et al.*, surface NO_x Asia	÷31–(÷42)[a]	55–70[a]	25–29[a]
Berntsen *et al.*, surface NO_x Europe	÷8.6–(÷11)[a]	8.1–12.7	÷2.7–(+4.1)[a]
Derwent *et al.*, Aircraft NO_x[b]	–44	75	31
Wild *et al.*, Aircraft NO_x[b]	–44	61	17
Stevenson *et al.* Aircraft NO_x[b]	–40	39	–0.9

Notes:

(a) Range from two 3-D chemistry transport models and two radiative transfer models