Chapter 2: Changes in Atmospheric Constituents and in Radiative Forcing

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Executive Summary

Radiative forcing is a concept used to quantitatively compare the strengths of different agents of climate change. For the range of human and natural agents of climate change, global mean radiative forcing relates to the resultant equilibrium global mean surface temperature change.

> Radiative forcings (RFs) are calculated in various ways depending on the agent: from changes in emissions and/or changes in concentrations; from observations and other knowledge of climate change drivers. In this Chapter they are given in units of Wm^{-2} for the present day (~2004) relative to preindustrial times (~1750).The RFs represent the stratospherically adjusted radiative flux change measured at the tropopause, as defined in the IPCC WG1 Third Assessment Report (IPCC*,* 2001) (hereinafter TAR). Positive RFs lead to a global mean surface warming and negative RFs to a global mean surface cooling. Unless otherwise mentioned, RF here refers to global-mean radiative forcing.
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> General Circulation Model studies since TAR give a high measure of confidence that a given RF from most realistic anthropogenic and natural drivers will have an equilibrium global mean surface temperature change comparable (to within ~25%) to that for the same RF from a CO_2 change; the factor relating this surface temperature change to that from CO_2 is termed the efficacy. RF is not, however, designed as an indicator of the detailed aspects of climate response.
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> **The Global Warming Potential (GWP) remains an appropriate metric for comparing the potential climate impact of the emissions of different forcing agents**. There are well-documented shortcomings of the GWP concept, particularly in using it to assess the impact of short-lived species. While different metrics have been proposed, their adequacy for describing impacts is dependent on the emission pathways.

Increasing concentrations of the long-lived greenhouse gases (CO_2, CH_4, N_2O, halocarbons and SF_6 hereinafter LLGHGs) have led to a combined RF of 2.59 ± 0.26 W m^{-2}. Their RF has a high level of scientific understanding[1]. The 7% increase in this RF since TAR is the result of concentration changes since 1998.

> **The current growth rate of CO_2 in the atmosphere for the decade from 1996 to 2005 is the fastest observed in any decade since instrumental records began in the 1950s; 2005 global mean concentrations were around 380 ppm. Changes in CO_2 contribute a RF of 1.63 ± 0.16 W m^{-2}, which dominates that of all other forcing agents considered in this report. This is an increase of 0.17 W m^{-2} since the RF value for 1998 quoted in TAR.** CO_2 emissions from land use change have contributed 0.36–0.71 W m^{-2} of this RF, with greater likelihood towards the lower estimate. The contribution of land use emissions to the overall CO_2 growth rate has decreased over the 20th century. The average CO_2 mixing ratio growth rate for the period 1999 to 2004 was more than 1.8 ppm yr^{-1}. Over the same period emission rates from fossil fuels, cement production and gas flaring rose from 6.5 to 7.2 Gt C yr^{-1}, representing a period of much higher emissions than considered in the TAR

The methane increase since preindustrial times to the 2004 concentration of 1776 ppb results in the second largest contribution, 0.48 ± 0.05 W m^{-2}, to the LLGHG RF. Over the past two decades methane growth rates in the atmosphere have generally declined. For the 5-year period from 1999 to 2004 the growth rate of methane declined to an average of 0.8 ppb yr^{-1} from an average of 7 ppb yr^{-1} in the preceding decade .For two of the 5 most recent years growth rates were negative. Reasons for the change in growth rate are not

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[1] Estimates of RF are accompanied by both an uncertainty range (value uncertainty) and a level of scientific understanding (structural uncertainty). The value uncertainties are 1σ, and are based on available published studies; the level of scientific understanding is a subjective measure of structural uncertainty and represents how well understood the underlying processes are. RF agents with a high level of scientific understanding will very likely have a RF that falls within the uncertainty range (see Section 2.9.2 and TS box of uncertainty for more information).

well understood. However the small and decreasing methane growth rate, combined with the small inferred trends in the main methane sink (OH), imply that methane emissions are not increasing.

> **The RF from the other LLGHGs is slowly increasing**.
>
> 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 largest contributor to the LLGHG RF. 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 buildings and landfills.
>
> Nitrous oxide, the fourth largest contributor to the LLGHG RF, continues to rise approximately linearly (0.26% per year) reaching a concentration of 319 ppb in 2004, which corresponds to a RF of 0.16 ± 0.02 W m^{-2}. Recent studies reinforce the large role of emissions from tropical regions.
>
> Many of the Kyoto Protocol gases (hydrofluorocarbons, perfluorocarbons, SF_6) have increased by large factors between 1998 and 2004. These factors are: 3.8 (HFC-134a), 3.6 (HFC-125), 2.5 (HFC-152a), 1.4 (HFC-23), 1.3 (PFC-116) and 1.3 (SF_6). Their total RF in 2004 was 0.015 W m^{-2} and this is rising rapidly, increasing by ~10% year.
>
> **OH, a key reactive chemical species that influences the lifetimes and thus RF of certain tropospheric constituents, has shown no detectable net change between 1979 and 2004.** Reaction with OH is the major sink for CH_4, HFCs and HCFCs, and the major producer for some 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 an inferred minimum in 1997–1999 that coincides with an El Niño event and several large wildfires. This could have significant implications for many of the LLGHGs and their RF.

Stratospheric ozone is near its minimum level in the satellite observations era, and the magnitude of its RF is expected to decrease in the future. The RF is evaluated to be –0.03 ± 0.07 W m^{-2}, weaker than quoted in TAR, with a medium level of scientific understanding. The total concentration of ozone depleting substances has already peaked in the atmosphere and 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. In addition to the chemical destruction of ozone, dynamical changes may have contributed to Northern Hemisphere midlatitude ozone reduction. The RF and uncertainty estimate are based on chemical transport model studies of ozone changes since preindustrial times and not on observed ozone changes since 1980 (as in TAR). (by final draft RF computations may be performed based on recent, updated observations of stratospheric ozone)

Tropospheric ozone RF is estimated to be 0.35 (+0.15/–0.1) W m^{-2} with a medium level of scientific understanding. Observations show that trends in tropospheric ozone during the last few decades vary both in terms of sign and magnitude. There are indications of significant upward trends at low latitudes. Several new chemical transport 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. The best-estimate of RF has not changed since the TAR.

Stratospheric water vapour has likely contributed a positive RF of 0.07 ± 0.05 W m^{-2}, with a low level of scientific understanding. This estimate is based on chemical transport model studies of the effect of increases in methane, which leads to an indirect RF by increasing stratospheric water vapour. The level of scientific understanding is low because the vertical profile of water vapour change is not well known – and the RF is very dependent of this. Other potential human causes of stratospheric water change have a very low level of scientific undersanding and remain unquantified. While water vapour absorbs more longwave radiation than any other greenhouse gas in the climate system, most of its change can be associated with climate feedback mechanisms, and it is therefore considered as part of the climate response, rather than a RF.

A combined total direct aerosol RF is given as –0.5 ± 0.4 W m^{-2}, with a low level of scientific understanding.

> In-situ aerosol measurements, and satellite and surface-based remote sensing retrievals have developed considerably since the TAR. These measurements and retrievals provide essential criteria for global model verification and validation. Atmospheric models have also undergone considerable developments, and are now better constrained by remote sensing and in-situ observations. The best estimate and uncertainty range of the total direct aerosol RF are based on a combination of the modelling and observational based studies.
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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 from models only to be: sulphate –0.4 ± 0.2 W m^{-2}, fossil-fuel organic carbon –0.1 ± 0.1 W m^{-2}, fossil-fuel black carbon +0.2 ± 0.1 W m^{-2}, biomass burning 0.0 ± 0.1 W m^{-2}, nitrate –0.1 ± 0.1 W m^{-2}, mineral dust –0.1 ± 0.2 W m^{-2}. Significant changes in the estimates of the aerosol direct RF have occurred since TAR for biomass burning, nitrate and mineral dust aerosols. For biomass burning aerosol the direct RF is changed from strongly negative to approximately neutral owing to better modelling of the effects of biomass burning aerosol overlying clouds. For the first time, a RF for nitrate aerosol is given. For mineral dust, the range in the direct RF is reduced due to a reduction in the estimate of its anthropogenic fraction; a best estimate is given for the first time.

The cloud-albedo RF due to aerosols (also referred to as first indirect or Twomey effect) is estimated to be –0.9 ± 0.5 Wm^{-2}, with a very low level of scientific understanding. Other processes related to aerosol-cloud interactions remain highly uncertain and there is a very low level of scientific understanding of these processes.

> The number of GCM estimates of the albedo effect for warm (low-level) clouds has increased substantially since TAR. Recent studies with models that incorporate more aerosol species and processes that describe aerosol-cloud interactions in greater detail comprise the estimate for the RF due to the albedo effect. There is now a best estimate for this RF.
>
> Observational studies, and model simulations constrained by observations, give a less negative estimate of the RF due to the albedo effect. Thus, the actual RF is *likely* to be less negative than the model-only estimate above. However, there are only a few studies based on satellite datasets. This and other limitations introduce significant uncertainties in the RF estimate, therefore the estimate of RF is associated with a very low level of scientific understanding. Other aerosol-cloud processes such as cloud lifetime and semi-direct effects remain very uncertain and also have a very low level of scientific understanding. In this report, these other effects are accounted for in the evaluation of climate response rather than RF.

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

Land-cover changes have increased the surface albedo, whilst black carbon aerosol deposited on snow surfaces has decreased the surface albedo. The net effect of these land surface changes has led to a RF of –0.1 ± 0.3 W m^{-2}, with a very low level of scientific understanding. Other surface property changes lead to both RF and other physical alterations to the climate system; these processes have a very low level of scientific understanding

> Global anthropogenic land cover change since 1750 has consisted of more deforestation than tree planting, with most net deforestation occurring in temperate regions. The resulting increase in surface albedo has led to a global mean RF of –0.2 ± 0.3 W m^{-2}, with a low level of 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 very low level of scientific understanding. There may also a very small positive RF from tropospheric water vapour increases associated with irrigation.

> 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. These effects have a very low level of scientific understanding. The release of heat from human energy production is significant at local scales in urban areas but not globally.

Persistent linear contrails from global aviation contribute a small RF of 0.01W m^{-2}, with a factor of two uncertainty and a low level of scientific understanding. Aviation may also alter cirrus clouds. This best-estimate is a factor of 3 to 4 smaller than the estimate projected from TAR for current global aviation operations. This difference results from new observations of contrail cover and reduced estimates of contrail optical depth. Observational studies provide evidence that the net RF from spreading contrails and their effects on nearby cirrus cloudiness may be from 2–10 times greater than the RF from persistent linear contrail cover. The global effect of aviation aerosol on background cloudiness remains unknown and a best estimate remains unavailable for the RF of total cloudiness changes (contrails, induced cirrus cloudiness, and aerosol effects) caused by subsonic aircraft operations, these other effects have a very low level of scientific understanding.

Since 1750 humans have *very likely*[2] exerted a net warming influence on climate. Global mean net RF for the better understood anthropogenic RF agents (LLGHGs and ozone) is estimated to be 2.9 ± 0.3 W m^{-2}. Anthropogenic RF drivers that are less well understood (e.g., aerosol direct and cloud-albedo RFs; land-use albedo RF) are likely to contribute a negative global mean RF. However, the net RF for all anthropogenic drivers taken together and weighted by level of scientific understanding is very likely to be positive. RF is estimated for the main forcing agents and, for the first time, an estimate of RF associated with each emission agent is also made.

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 and a low level of scientific understanding.

> A re-evaluation of the long-term change in solar irradiance viz., an irradiance increase of 0.08% from the Maunder Minimum to the present-day cycle mean compared to 0.2% in TAR, has resulted in a considerably smaller RF estimate. Uncertainties remain large because of the lack of direct observations and understanding of solar variability mechanisms on long time scales.

> Total solar irradiance has been monitored from space for only the last three decades. An irradiance cycle of 0.08% (peak to peak) is well established, but no significant trend has yet been detected. UV radiation below 310 nm contributes 15% of the total solar irradiance 11-year cycle. Difficulty in estimating past variations in facular brightness contributes a large uncertainty to historical solar forcing estimates.

> Changes in global column ozone forced by the solar UV irradiance 11–year cycle are now relatively well understood on an empirical basis, and are of the order of a few percent, comparable to other known influences e.g., CFC-induced depletion and natural QBO-driven fluctuations; however, ozone profile changes are less certain. Empirical associations, between globally-averaged low-level cloud cover and solar-modulated cosmic ray ionization of the atmosphere, remain controversial because of uncertainties about the reality of the decadal signal itself, the phasing or anti-phasing with solar activity, its separate dependence for low, middle and high clouds, and alternative explanations such as ENSO. Cloud cover anomalies could result from modulation of atmospheric circulation through variations in solar-UV-induced-ozone heating, but quantitative details are not well established.

The global stratospheric aerosol concentrations are now at their lowest observed values in the satellite era. There has been no explosive volcanic eruption since Mt. Pinatubo in 1991, which remains the best documented event in terms of reliable and accurate observations of the aerosols formed in the aftermath, in

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[2] The use of "very likely" is an example of the calibrated language used in this document, it represents a 90% confidence level or higher (see box TS-1).

turn leading to good quantitative estimates of the RF. There is less confidence in the forcing estimates for prior episodic and explosive volcanic events. Volcanic aerosols exert a transitory but substantial perturbation of the radiative energy budget, affect circulation patterns, and influence the chemical depletion of stratospheric ozone under enhanced anthropogenic halogen loading conditions.

The spatial pattern of RFs for ozone, aerosol direct effects, aerosol-cloud interactions and land-use have considerable uncertainties. This is in contrast to the relatively high confidence in the spatial pattern of RF for the LLGHGs. The positive net RF in the Southern Hemisphere very likely exceeds that in the Northern Hemisphere as a result of the negative aerosol RF, which is concentrated more in the Northern Hemisphere.

The instantaneous radiative flux change at the surface (hereinafter 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 cannot be used to quantitatively compare the effects of different agents on the equilibrium global-mean surface temperature change**.**

> **The total global-mean surface forcing is *very likely* negative.** By reducing the shortwave radiative flux at the surface**, t**ropospheric and stratospheric aerosols are principally responsible for the negative surface forcing. This is to be contrasted with the LLGHGs which are the principal contributors to the positive global-mean RF.
>
> **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 more uniform spatial structure exists 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) (hereinafter TAR). It concerns itself with trends in forcing agents and their precursors since 1750, and estimates their contribution to radiative forcing (hereinafter RF). Discussion of the understanding of atmospheric composition changes focuses on explaining the *trends* in forcing agents and their precursors. Areas where significant developments have occurred since the TAR will be highlighted. The chapter draws on various assessments since TAR, in particular the 2003 WMO UNEP Ozone assessment report (WMO, 2003) and the IPCC-TEAP Special Report on Safeguarding the Ozone Layer and the Global Climate System (IPCC/TEAP, 2005).

The chapter assess anthropogenic greenhouse gas changes, aerosol changes and their impact on clouds, aviation induced contrails and cirrus changes, land-use changes and natural, solar and volcanic, mechanisms. As well as re-evaluating and updating the trends and RFs presented in TAR, this report assesses several "new" forcing mechanisms that were not extensively discussed in previous assessments. It considers several mechanisms associated with the short timescale and local response of the troposphere. These mechanisms are essentially processes that either involve the interaction of aerosols with their environs (see Section 2.4.6 and Chapter 7, Section 7.5), involve changes to land surface properties beyond surface albedo (see Section 2.5). Water vapour is the strongest greenhouse gas in the atmosphere and as most of its change can be considered part of the climate response, rather than a forcing, its main effect is as a climate feedback, which Chapter 8 assesses (see Box 8.1). However, water vapour changes from CH_4 increases (in the stratosphere) and irrigation (in the troposphere) can be considered forcings and these are discussed in Section 2.3.8 and 2.5.6 respectively. The chapter also presents spatial and temporal patterns of RF and it examines the radiative energy budget changes at the surface. These "surface forcings" are presented primarily as diagnostics for understanding aspects of the climate response that is being evaluated within the other chapters. Additionally, the chapter reassess the "radiative forcing" concept itself (Section 2.8).

For the long-lived-greenhouse-gases (CO_2, CH_4, N_2O, CFCs, HCFCs, HFCs, PFCs and SF_6, hereinafter collectively referred to as the LLGHGs, Section 2.3) the chapter makes use of new global measurement capabilities and combines long term measurements from various networks to update trends through 2005. Emissions and budgets are also discussed in so far as necessary to explain the observed trends. The processes involved and further discussion of budgets occurs in Chapter 7 (Sections 7.3 and 7.4). Compared to other RF agents, their trends are considerably better understood and quantified; because of this, the chapter does not devote as much page space to them as previous assessments. Nevertheless they remain the largest and most important driver of climate change and evaluation of their trends is one of the fundamental tasks of both this chapter and the assessment report.

The chapter considers only "forward calculation" methods of estimating RF. These rely on observations and/or modelling of the relevant forcing agent. Since TAR several studies have attempted to constrain aspects of RF using "inverse calculation" 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 (see Chapter 9, Section 9.2).

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, they are briefly discussed in Chapter 10 (Climate Projections).

2.2 Concept of Radiative Forcing (RF)

The definition of radiative forcing (RF) 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). RF can be related through a linear relationship to the global mean equilibrium temperature change at the surface (ΔT_s): $\Delta T_s = \lambda RF$, where λ is the climate sensitivity parameter (e.g., Ramaswamy et al., 2001). This equation, developed from these early climate studies, represents a linear view of global mean climate change between two *equilibrium* climate states. RF is a simple measure for both quantifying and ranking the many different influences on climate change. It quantifies these influences in terms of changes in components of the Earth's radiative energy budget. RF is conventionally measured in units of W m^{-2}. RF provides a limited measure of climate change as it does not attempt to represent the overall climate response. However, as climate sensitivity and other aspects of the climate response to external forcings remain relatively poorly quantified, it has the advantage of being more readily calculable and comparable than estimates of the climate response. Figure 2.1 shows how the RF concept fits within a general understanding of climate change.

[INSERT FIGURE 2.1 HERE]

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 drivers are better considered as a forcing or a feedback (Hansen et al., 2005; Jacob et al., 2005, Section 2.8). Emissions of forcing precursors, such as aerosol, ozone, and LLGHGs, are the more fundamental drivers of climate change and these emissions can be used in state-of-the-art climate models to interactively evolve forcing-agent fields along with their associated climate change. In such models some "climate response" is necessary to evaluate the RF. This "response" is most significant for aerosol-related cloud changes, where the tropospheric state needs to significantly alter in order to create radiative perturbation to the climate system (Jacob et al., 2005).

Over paleo timescales discussed in Chapter 6 long-term changes in forcing agents arise due to so-called "boundary condition" changes to the Earth's climate system (such as changes in orbital parameters, ice sheets and continents), for the purposes of this chapter these "boundary conditions" are assumed to be invariant and forcing-agent changes are considered to be *external* to the climate system. The natural RFs considered are solar changes, cosmic rays and volcanoes, the other RF agents are all attributed to humans. For the LLGHGs it is appropriate to assume that there has been little or no biogeochemical feedback (see Chapter 7, Sections 7.3 and 7.4) and RF is typically calculated in offline radiative transfer schemes, using observed changes in concentration; i.e., humans are considered solely responsible for their increase. For the other climate change drivers, RF is often estimated using general circulation model (GCM) data employing a variety of methodologies (Ramaswamy et al., 2001; Stuber et al., 2001a; Tett et al., 2002; Hansen et al., 2005; Shine et al., 2003 and see Section 2.8.3). Often alternative RF calculation methodologies to the TAR definition are used, the most important are illustrated in Figure 2.2. For most aerosol constituents (see Section 2.4) stratospheric adjustment has little effect on the RF, and the instantaneous RF at either the top of the atmosphere or tropopause can be substituted for the stratospheric-adjusted RF. For the climate change drivers, discussed in Chapter 7, Section 7.5 and Section 2.5, that are not initially radiative in nature, a RF-like quantity can be evaluated allowing the tropospheric state to change: this is the zero-surface-temperature-change RF in Figure 2.2 (see Shine et al., 2003, Hansen et al., 2005 and Section 2.8.3).

[INSERT FIGURE 2.2 HERE]

Processes which necessitate changes in the tropospheric state prior to a radiative perturbation are aerosol-cloud lifetime effects, aerosol semi-direct effects and some surface-change effects. They need to be accounted for when evaluating the overall effect of humans on climate and their radiative effects as discussed in Chapter 7 (Sections 7.1 and 7.5) . However, in both this chapter and the assessment report they are not considered to be RFs, although the RF definition could be altered to accommodate them. Reasons for this are twofold and concern the need to be simple and pragmatic. Firstly, many GCMs have some representation of these effects inherent in their climate response and evaluation of variation in climate sensitivity between mechanisms already accounts for them (see "Efficacy", Section 2.8.5). Secondly, the evaluation of these tropospheric state changes rely on some of the most uncertain aspects of a climate models response e.g., hydrologic cycle; their radiative effects are very climate model dependent and such a dependence on climate models is what the RF concept was designed to avoid. In practise these effects can also be excluded on practical grounds – they are simply too uncertain to adequately quantify (see Chapter 7, Section 7.5 and Sections 2.4.6 and 2.5.6).

The RF relationship to transient climate change is not straightforward. To evaluate the overall climate response associated with a forcing agent its time evolution and its spatial and vertical structure need to be taken into account. Further, RF alone cannot be used to assess the potential climate change associated with emissions, as it does not take into account the different atmospheric lifetimes of the forcing agents. Global Warming Potentials (GWPs) are one way to assess these emissions, they compare the integrated RF over a specified period (e.g., 100 years) from a unit mass pulse emission relative to CO_2 (see Section 2.10).

2.3 Chemically and Radiatively Important Gases

2.3.1 Atmospheric Carbon Dioxide (CO_2)

In this section we discuss the instrumental measurements documenting recent changes in atmospheric CO_2 concentrations needed for the RF calculations presented later in the section. In addition we provide data for the preindustrial levels of CO_2 required as the accepted reference level for the RF calculations. Earlier and indirect measurements of atmospheric CO_2 covering time spans of up to 740,000 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.

Carbon dioxide continues to show the largest RF since 1750 of any forcing agent. A wide range of direct and indirect measurements confirm that its atmospheric concentration has increased globally by about 100 ppm (35%) over the last 250 years, from a range of 275–285 ppm in the preindustrial era to almost 380 ppm in 2004 (Keeling and Whorf, 2005). During this period the absolute 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 1995 to 2004 atmospheric CO_2 increased by about 19 ppm; the highest average growth rate recorded for any decade since direct atmospheric CO_2 measurements began in the 1950s.

High-precision measurements of atmospheric CO_2 are essential to the understanding of the carbon cycle budgets discussed in Chapter 7, Section 7.3. 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 from Scripps Institution of Oceanography (SIO) (see Chapter 1, Section 1.5). These began in 1958 at Mauna Loa, Hawaii, located at 19°N (Keeling et al., 1995). The 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 analysis programmes at other sites in both hemispheres (Conway et al., 1994; Nakazawa et al., 1997; Langenfelds et al., 2002). In Figure 2.3 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 (Keeling and Whorf, 2005; Manning et al., 1997). These two stations provide the longest continuous records of atmospheric CO_2 in the Northern and Southern Hemispheres respectively. Remote sites such as Mauna Loa, Baring Head, Cape Grim (Tasmania) and the South Pole were chosen because air sampled at these locations has little 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 documented.

[INSERT FIGURE 2.3 HERE]

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 Earth System Research Laboratory (NOAA/ESRL) in the USA; formerly NOAA/CMDL.

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. World wide databases of atmospheric CO_2 measurements are maintained by the Carbon Dioxide Information Analysis Center (http://cdiac.esd.ornl.gov/) and by the WMO Global Atmospheric Watch programme (http://gaw.kishou.go.jp/wdcgg.html).

The driving forces for the increases in global atmospheric CO_2 since the industrial revolution are mainly CO_2 emissions from the combustion of fossil fuels, gas flaring and cement production. Other sources include emissions due to land-use changes such as deforestation (Houghton, 2003) and biomass burning (Andreae and Merlet, 2001; van der Werf, 2004). Also as oceans warm, less atmospheric CO_2 is absorbed by them with the net result that more CO_2 remains in the atmosphere. Note that the latter is not strictly an emission but a climate feedback process (Joos et al., 1999; Barnett et al., 2001).

After entering the atmosphere, CO_2 exchanges rapidly with the short-lived terrestrial biosphere and surface ocean, and is redistributed on time scales of hundreds of years among all active carbon reservoirs including the long-lived biosphere and deep ocean. 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 are presented in Chapter 7, Section 7.3 where carbon cycle budgets are discussed.

The RF of CO_2 is a function of the change in CO_2 in the atmosphere over the time period under consideration. Hence a key question is "How is the CO_2 released from fossil fuel combustion, cement production and land cover change 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 O_2 content. The carbon contained in CO_2 has two naturally occurring stable isotopes denoted ^{12}C and ^{13}C. The first of these, ^{12}C, is the most abundant isotope at about 99% followed by ^{13}C at about 1%. 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 related to its source. Thus, as shown in Prentice et al., 2001, when CO_2 from fossil fuel combustion enters the atmosphere, the $^{13}C/^{12}C$ isotopic ratio in atmospheric CO_2 decreases at a rate that is consistent with emissions of CO_2 from fossil origin. 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 decreasing in line with fossil fuel CO_2 emissions to the atmosphere and atmospheric CO_2 concentrations (Keeling et al., 2005; Andres et al., 2000).

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). O_2 and CO_2 are inversely coupled during plant respiration and photosynthesis. Also during the process of combustion, O_2 is removed from the atmosphere producing a signal that decreases as atmospheric CO_2 increases on a molar basis (Figure 2.3). Measuring changes in atmospheric O_2 is technically challenging however, because of the difficulty of resolving changes at the ppm level in a background concentration of ~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). Recent work by Manning and Keeling (2005) indicates that atmospheric O_2 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 O_2 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, Section 7.3. In Figure 2.3 recent measurements in both hemispheres are shown to emphasize the strong linkages between atmospheric CO_2 increases, O_2 decreases, fossil fuel consumption and the $^{13}C/^{12}C$ ratio of atmospheric CO_2.

From 1990 to 1999, a period reported in Prentice et al. (2001), the growth rate of 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, the emissions growth rate rose from 6.5 to 7.2 Gt C yr^{-1} (Marland et al., 2005; BP World Energy, 2005) representing a period of much higher emission rates than those considered in the TAR (see

Figure 2.3). Global annual fossil fuel combustion and cement manufacture CO_2 emissions were reported at 6.975 Gt C for 2002 up 2% from the 2001 emissions (Marland et al., 2005) and emission rates have increased by 50% over the last 30 years. The relationship between increases in atmospheric CO_2 and 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 and cement manufacture CO_2 emissions. On decadal scales this fraction has averaged about 60% since the 1950s. With current emissions at about 7 Gt C yr^{-1} and assuming the airborne fraction remains at about 60%, Hansen and Sato (2004) predict that the underlying long term global atmospheric CO_2 growth rate will be about 1.9 ppm yr^{-1}.

Prentice et al. (2001) cited an inventory-based estimate that land use change resulted in net emissions of 121 Gt C between 1850 and 1990, after Houghton (1999, 2000). The estimate for this period was revised upwards to 134 Gt C by Houghton (2003), mostly due to an increase in estimated emissions prior to 1960. Houghton (2003) also extended the inventory emissions estimate to 2000 giving cumulative emissions of 156 Gt C since 1850. Brovkin et al. (2004) simulate these emissions to contribute between 22 and 43 ppm of the observed CO_2 rise since pre-industrial (i.e., 22%–43% of the total rise), with a greater consensus towards the lower end of the range (Table 2.8). This implies a contribution of 0.36–0.71 W m^{-2} to the CO_2 RF (Table 2.8). The land use contributions to the growth rate were estimated as 42%–68% in 1850–1900, falling to 5%–38% in the 1990s (with the central estimates being towards the lower end of these ranges). Historical changes in land cover are discussed in Section 2.5.2, and the CO_2 budget over the 1980s and 1990s is discussed further in Chapter 7, Section 7.3.

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/ESRL network of 40 sites was 376.75 ± 0.05 ppm yielding a global average of almost 377 ppm. The statistical derivations of uncertainties are different 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. 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 (updated from those published by Etheridge et al., 1996) show a preindustrial global mean for CO_2 from 1700 to 1800 of 277 ± 1.2 ppm (278 ± 1.2 ppm at 1750). This ice core record has now been extended back to 1 AD (McFarling Meure*,* 2004), and shows little change in concentrations before ~1800 although there were slightly lower CO_2 concentrations in 1600–1800 (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%. For the RF calculation the above data from Law Dome glacier in the Antarctic are used because they show the highest age resolution (~5 years) of any ice core records in existence. 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 2.4.

[INSERT FIGURE 2.4 HERE]

There are no updates to the RF calculation to report. The simple formulae for RF of the LLGHG quoted in Ramaswamy et al. (2001) are still valid. These formulae are based on global RF calculations where clouds, stratospheric adjustment and solar absorption were included and give a RF of 3.7 W m^{-2} for a doubling in the CO_2 concentration. A recent comparison of detailed line-by-line models and GCM radiation schemes found that clear sky instantaneous RF agreed very well (better than 10%) among the 5 line-by-line models investigated, using the same single atmospheric background vertical profile (Collins et al., 2006). The GCM radiation schemes were less accurate, with ~20% errors in the CO_2 RF (Collins et al., 2006 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 reasonable agreement with the TAR RF value of 3.7 W m^{-2} (see Chapter 10 and Forster and Taylor*,* 2006).

Using the global average value of 377 ppm for atmospheric CO_2 in 2004 provides a RF of 1.63 ± 0.16 W m^{-2}; a contribution that dominates that of all other forcing agents considered in this chapter. This is an increase of 12% over the value of 1.46 reported for 1998 in Ramaswamy et al. (2001). This change is solely due to increases in atmospheric CO_2 and is also much larger than the RF changes due to other agents. In the decade 1995 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 2.1, which summarizes the present day concentrations and RF for the LLGHGs, and indicates changes since TAR.

[INSERT TABLE 2.1 HERE]

2.3.2 Atmospheric Methane (CH_4)

Here we describe the current global measurement programmes for atmospheric methane (CH_4), which provide the data required for the calculation of its RF at the end of the section. In addition we provide data for the preindustrial levels of CH_4 required as the accepted reference level for these calculations. Detailed analyses of methane budgets and its biogeochemistry are presented in Chapter 7, Section 7.4.

Methane has the second largest RF of the LLGHGs after CO_2 (Ramaswamy et al., 2001). Over the last 650,000 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 (Spahni et al., 2005). Over the last two millennia measurements in air bubbles trapped in polar ice and firn show that CH_4 levels remained below or near 700 ppb until about 1700 (see Chapter 6). After this, the concentration rose through 750 ppb in 1800, 880 ppb in 1900 to 1714 ppb in 1992 (Etheridge et al., 1998).

In 2004 the global average abundance of CH_4 measured at the network of 40 surface air flask sampling sites operated by NOAA/ESRL in both hemispheres was 1777.60 ± 0.60 ppb. This network of sites is the most geographically 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 CH_4 concentrations from NOAA/ESRL by about 1%, bringing them into much closer agreement with the AGAGE network (see below). This scale will be used by laboratories participating in the WMO's Global Atmosphere Watch Programme as a "common reference". Atmospheric CH_4 is also monitored at 5 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/ESRL network the 90% confidence interval is 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. Global databases of atmospheric CH_4 measurements are also maintained by the Carbon Dioxide Information Analysis Center (http://cdiac.esd.ornl.gov/) and by the WMO Global Atmospheric Watch programme (http://gaw.kishou.go.jp/wdcgg.html).

Present atmospheric levels of CH_4 are unprecedented in at least the last 650,000 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 CH_4 has increased by about 30% during that time, its growth rate has decreased substantially 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/ESRL. Recent measurements by Lowe et al. (2004) for sites in the Southern Hemisphere and Cunnold et al. (2002) for the network of GAGE/AGAGE sites show similar features. However as shown in Figure 2.5 a key feature of the reduction in the global growth rate of CH_4 is its current interannual variability with growth rates ranging from a high of 14 ppb yr^{-1} in 1998 to less than zero in 2001 and 2004.

[INSERT FIGURE 2.5 HERE]

The 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) but are clearly related to changes in the magnitude of an imbalance between CH_4 sources and sinks. 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). The importance of OH, trends in its abundance and its role in controlling atmospheric CH_4 levels are discussed in Section 2.3.5.

The global CH_4 source is poorly known. As detailed in Chapter 7, Section 7.4 it is mostly biogenic and includes wetlands, rice agriculture, biomass burning, and ruminant animals. Methane is also emitted by various industrial sources (Wang et al., 2004). The TAR documented a large range in "bottom up" estimates for the global source in CH_4 and no significant improvements have been published since then. Keppler et al., 2006 report the discovery of large emissions of CH_4 from living vegetation contributing an estimated 10-30% of the global CH_4 source. That such a potentially large source of CH_4 could have been missed highlights the large uncertainties involved in current estimates of the global CH_4 source (see Chapter 7, Section 7.4). This work has yet to be confirmed by other laboratories but substantiates spaceborne observations of CH_4 plumes above tropical rainforests reported by Frankenberg et al., (2005).

Several wide ranging hypotheses have been put forward to explain the reduction in the growth rate of CH_4 and its variability. For example Hansen et al. (2000) considered that economic incentives have led to a reduction in anthropogenic methane emissions and Dlugokencky et al. (1998) and Francey et al. (1999) suggest that the slowdown in the growth rate reflects a stabilization of CH_4 emissions, given that the observations are consistent with stable emissions and lifetime since 1982. Measurements along the world largest natural gas transport pipeline system in Russia indicate that technological improvements have reduced CH_4 leakages (Lelieveld et al., 2005). Others have argued that predicting future atmospheric burdens is difficult 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 CH_4 growth rate (Simpson et al., 2002; Dlugokencky et al., 2003).

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 CH_4 by the atmospheric OH radical (Bekki et al., 1994; Dlugokencky et al., 1996). Lelieveld et al. (1998) and Walter et al. (2001a; 2001b) proposed that lower temperatures and lower precipitation in the aftermath of the Pinatubo eruption could have suppressed CH_4 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; Mak et al., 2000). 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 CH_4 anomaly and linkages to the 1991 Pinatubo eruption are still undetermined.

For the relatively large increase in the CH_4 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 surface instrumental temperature records began (see Chapter 3, Section 3.2). Langenfelds et al. (2002), on the basis of measurements of a suite of biogenic trace gases, showed that the 1998 variability was consistent with emission pulses coinciding with large biomass burning events in 1997/1998 in tropical and boreal regions. 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), Dentener et al. (2003) 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 CH_4 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 CH_4 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 CH_4 may be temporary with potential for large future increases in CH_4 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 CH_4 budget since 1990 in the context of emission estimates aggregated from country inventories reported to UNFCCC and the projection of these to 2020. Their work shows that, if the projections are accurate and the CH_4 sink remains stable, (see Manning et al., 2005) then atmospheric CH_4 mixing ratios will resume growth rates approaching those of the 1970s by 2020. However this proposition takes into account only 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) the preindustrial global value for CH_4 from 1700 to 1800 was 715 ± 4 ppb (it was also 715 ± 4 ppb in 1750) thus providing the reference level for the RF calculation. 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 CH_4 concentrations is calculated with the simplified expression for CH_4 given in the TAR. This simplified expression still remains valid. The change from 715 ± 4 ppb to 1776 ± 44 ppb (the average concentration from the AGAGE and CMDL networks in 2004) in the CH_4 concentration gives a RF of 0.48 ± 0.05 W m^{-2}, ranking CH_4 as the second highest RF of the LLGHGs after CO_2 (Figure 2.4 and Table 2.1). The uncertainty range in concentrations for the present day represents intra-annual variability, which is not included in the preindustrial uncertainty estimate derived solely from ice-core sampling precision. The estimate for the RF due to CH_4 is the same as in Ramaswamy et al. (2001) despite the small increase in its concentration. The spectral absorption by methane is overlapped to some extent by N_2O lines (taken into account in the simplified expression). Taking the overlapping lines into account using current N_2O concentrations instead of preindustrial concentrations (as in Ramaswamy et al., 2001) reduces the current RF due to CH_4 by 1%.

Collins et al. (2006) 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. (see Chapter 10). 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. 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., 2006).

2.3.3 Other Kyoto Protocol Gases (N_2O, HFCs, PFCs, and SF_6)

At the time of the TAR, N_2O had the fourth largest RF among the LLGHGs behind CFC-12, CH_4 and CO_2. The TAR quoted an atmospheric N_2O 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; the most recent assessment (Montzka et al., 2003) recommended a lifetime of 114 years for N_2O. TAR pointed out large uncertainties in the major soil, agricultural, combustion and oceanic sources of N_2O.Given these emission uncertainties, its observed rate of increase of 0.2 to 0.3% per year was not inconsistent 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 concluded to be enhanced microbial production in expanding and fertilized agricultural lands (Prather et al., 2001).

Ice-core data for N_2O are now available extending back 2000 years before present (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 exhibit a relatively rapid rise (seen in RF on Figure 2.4). 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 2.6). 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 Ramaswamy et al. (2001). This RF is only slightly larger than in Ramaswamy et al. (2001) (Table 2.1).

Since the TAR, understanding of regional N_2O fluxes has evolved. The results of various studies which quantified the global N_2O emissions from coastal upwelling areas, continental shelves, estuaries and rivers suggest that these coastal areas contribute 0.3–6.6 TgN(in N_2O) yr^{-1} or 7–61% of the total oceanic emissions (Bange et al., 1996; Nevison et al., 2004b; Kroeze et al., 2005). (see also Chapter 7, Section 7.4) 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., 2004a). 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 2.6 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 Chapter 2 of IPCC/TEAP (2005): Velders et al. (2005), and here we provide therefore only a brief review and an update for these species. Table 2.1 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, and there is significant uncertainty in cross-section measurements. 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/TEAP (2005) 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/TEAP (2005) for the more abundant HFCs show that an agreement better than 12% can be reached for these when the calculation conditions are better constrained (see Section 2.10.1).

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/TEAP (2005), 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 GMD (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, summarized in Figure 2.7, 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 current trends are 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/TEAP (2005) 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 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.

[INSERT FIGURE 2.7 HERE]

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, Table 2.14), and make an essentially permanent contribution to RF. The SF_6 and C_2F_6 concentrations and RF have increased by over 20% since TAR (Table 2.1, Figure 2.7), but CF_4 concentrations have not been updated since TAR. 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. The CF_4 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 have continued to increase linearly over the past decade implying that emissions are approximately constant. 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 Chapter 1 of the 2003 WMO-UNEP ozone assessment (Montzka et al., 2003) and IPCC/TEAP (2005). 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 2.1). This contribution is dominated by the CFCs. The effect of the Montreal Protocol on these gases has been substantial. IPCC/TEAP (2005) concluded that the combined CO_2-equivalent emissions of CFCs, HCFCs, and HFCs decreased from a peak of about 7.5 $GtCO_2$-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, summarized in Figure 2.7, are available from the AGAGE network (Prinn et al., 2000; Prinn et al., 2005b) and the GMD 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 (CFC-11, 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 2.4). 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 Ramaswamy et al. (2001). This means that the total halocarbon RF quoted for 2004 in Table 2.1 (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 2.4 and Table 2.1).

The major solvent, methyl chloroform (CH_3CCl_3) is of special importance regarding RFs, not because of its small RF (see Table 2.1, Figure 2.7), 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 its 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 temporally but less geographically 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 (Figure 2.7). 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, in spite of its moderately long lifetime of 20–30 years, results from a persistence of significant northern hemispheric emissions. .

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. Montzka et al (2003) and IPCC/TEAP (2005) 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.

To provide a direct comparison of the effects on global warming of the annual changes in each of the non-CO_2 greenhouse gases (discussed in Sections 2.3.2, 2.3.3 and 2.3.4) relative to CO_2, we show in Figure 2.8 these annual changes in atmospheric mass multiplied by the GWP (100 year horizon) for each gas (see e.g., Prinn, 2004). By expressing them in this way, the observed changes in all non-CO_2 gases in $GtCO_2$ equivalents, the significant roles of CH_4, N_2O, and many halocarbons are very evident. This highlights the importance of considering the full suite of greenhouse gases for RF calculations.

[INSERT FIGURE 2.8. HERE]

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 Gt of trace gases each year (Ehhalt, 1999). It has a very significant role in limiting the LLGHG RF. IPCC/TEAP (2005) 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 inferred 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 is 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 (Lowe and Allan.,2002). While the accuracy of the ^{14}CO cosmic ray and other ^{14}CO 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, and capable of resolving shorter time scales, than those estimated from CH_3CCl_3. The ^{14}CO source variability is better defined than its absolute magnitude so it is better for inferring relative than absolute trends. 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 HFC-134a, HCFC-141b and 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 spatial weighting here is different. Note that methods to infer global or hemispheric average OH concentrations may be insensitive to compensating regional OH changes such as OH increases over continents and decreases over oceans (Lelieveld et al., 2002). Also, the quoted absolute OH concentrations (but not their relative trends) depend on the choice of weighting (see e.g., Lawrence et al., 2001). While the global 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) inferred 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) supported 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. (2005a) 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% due to CO enhancements (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. Neither the 2D Prinn et al. (2001) nor the 3D Krol et al. (2003) inversion models used interannually-varying circulation. 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 2.9). However, when Bousquet et al (2005) estimated both OH and methyl chloroform emissions (constrained by their uncertainties as reported by McCulloch and Midgley, 2000), the OH variations are reduced by 65% (dashed line in Figure 2.9). The error bars on the Prinn et al (2001, 2005a) OH estimates, which account for these emission uncertainties using Monte Carlo ensembles of inversions, also easily allow such a reduction in OH variability (thin vertical bars in Figure 2.9). This implies that these interannual OH variations are real, but only their phasing, and not their amplitude, is well defined. Bousquet et al. (2005) also deduced that OH in the Southern Hemisphere shows a zero to small negative trend in qualitative agreement with Prinn et al. (2001).

[INSERT FIGURE 2.9 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.

RF calculations of the LLGHGs are calculated from observed trends in the LLGHG concentrations and therefore OH concentrations do not directly affect them. Nevertheless OH trends are needed to quantify LLGHG budgets (see Chapter 7, Section 7.4) and for understanding future trends in the LLGHGs and tropospheric ozone.

2.3.6 Ozone

2.3.6.1 Stratospheric ozone

TAR reported that the ozone destruction in the stratosphere had caused a negative RF of -0.15 W m^{-2} as a best estimate over the period since 1750. A number of recent reports have assessed changes in stratospheric ozone and the research into its causes; Chapters 3 and 4 of the 2003 WMO assessment (WMO*,* 2003) and Chapter 1 of IPCC/TEAP (2005): Pyle et al. (2005). This section summarizes the material from these reports and updates the key results using more recent research. Over the satellite era (since ~1980) stratospheric ozone trends have been primarily affected by the Montreal Protocol gases (see Section 2.3.4). However, it has been suggested that stratospheric water vapour increases and tropospheric ozone changes may also have influenced stratospheric ozone values prior to 1980 (see below). Nevertheless, as the causes of change in the stratosphere and troposphere are largely associated with different emission agents they are treated separately in this report.

An updated time series of deseasonalized global mean column ozone anomalies is displayed in Figure 2.10, updated from Fioletov et al. (2002). The changes in ozone derived from ground-based measurements (1964–2003) as well as satellite data sets (1979–2003) are in good agreement (to within 1% of total column ozone) and we therefore have reasonable confidence in their quality for describing the spatial and temporal characteristics of past ozone changes (WMO, 2003; Pyle et al., 2005). The global ozone amounts decrease between the late 1970s and early 1990s, with the 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., 2004a (see also comment and reply: Cunnold et al., 2004 and Steinbrecht et al., 2004b); Hadjinicolaou et al., 2005; Krizan and Lastovicka, 2005).

[INSERT FIGURE 2.10 HERE]

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). Ozone decreases over the Arctic have been less severe than those over the Antarctic. Arctic 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 in the polar lower stratosphere 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.

The temporally and seasonally non-uniform nature of stratospheric ozone trends has important implications for resulting RF. Global ozone decreases come 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 20°N–60°N is attributable to dynamics (Forster and Tourpali, 2001). These dynamical feedbacks may well have an anthropogenic origin and could even be partly caused by stratospheric ozone changes themselves (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. At the time of writing no study has utilised ozone trend observations after 1998 to update the RF values presented in the Ramaswamy et al. (2001). However, Hansen et al. (2005) repeat the RF calculation based on the same trend datasets employed in TAR and find a RF of roughly –0.06 W m^{-2}, approximately half of the value quoted in Ramaswamy et al. (2001).

Gauss et al. (2006) compared results from six chemical transport models which included changes in ozone precursors to simulate both the increase in the ozone in the troposphere and the ozone reduction in the stratosphere over the industrial era. The 1850–2000 stratospheric column reduction for these models was 21 ± 5 DU and ranged between 14 DU and 29 DU.The overall pattern of the ozone change from the models were similar but the magnitude of the ozone change differ The models show a reduction in the ozone at high latitudes ranging from around 20 to 40% in the Southern Hemisphere and smaller change in the Northern Hemisphere but similar magnitude of the inter-model differences. All models have a maximum ozone reduction around 15 km at high latitudes in the Southern Hemisphere. Differences between the models were also found in the tropics, with some models giving around 10% increase in the lower stratosphere, wheras other models give reduction in the stratospheric ozone. These differences were especially related to the altitude where the ozone trend switched from an increase in the troposphere to a decrease in the stratosphere ranging from close to the tropopause to around 27 km. Several studies have shown that ozone changes in the tropical lower stratosphere are very important for the magnitude and sign of the ozone radiative forcing (Ramaswamy et al., 2001). The resulting stratospheric ozone RF was calculated to be –0.03 ± 0.07 W m^{-2} and range between –0.12 W m^{-2} and +0.07 W m^{-2}. Note that the models with either a small negative or a positive RF also had a small increase in tropical lower stratospheric ozone, resulting from ozone increases in the troposphere; most of this increase would have occurred before the era with human use of ozone destruction compounds. Because of this tropical increase in ozone, the mean RF from these models is smaller in magnitude than the RF from observed stratospheric ozone changes between 1980 and 1998. Additionally, these RF calculations do not include any negative radiative forcing that may have resulted from stratospheric wate vapour increases - it has been suggested (Shindell and Faluvegi, 2002) that stratospheric ozone during 1957 to 1975 was lower by ~7 DU relative to the first half of the 20th century as a result of possible stratopspheric water vapour increases; however, these increases in stratospheric water vapour may not be justified (see Section 2.3).

As no updated RF calculations from observations are available, the RF of –0.03 ± 0.07 W m^{-2} from the chemical transport modelling studies is taken as our best estimate of the stratospheric ozone RF since preindustrial times. Note though that the stratospheric ozone change solely due to the Montreal Protocol gases is very likely to be more negative than this and lie somewhere between –0.03 W m^{-2} and the value of −0.15 W m^{-2}, quoted in Ramaswamy et al. (2001).

2.3.6.2 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). The estimate in TAR of RF from tropospheric ozone change was 0.35 ± 0.15 W m^{-2}. Due to limited spatial and temporal coverage of observations of tropospheric ozone the RF estimate has to be based on model simulations. In the simulations used for TAR, the models considered only changes due to changes in the tropospheric photochemical system, driven by estimated emission changes (NOx, CO, NMVOC, and CH_4) since preindustrial times. Since the TAR there have been major developments of the models. The new-generation models 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 reduced influx of ozone from the stratosphere, due to stratospheric ozone depletion (Gauss et al., 2006). This shift in modelling capabilities and to be consistent with how the RF due to changes in stratospheric ozone is derived (based on observed ozone changes) have lead to a change in the definition of RF due to tropospheric ozone compared with TAR. It now includes changes in tropospheric ozone due to changes in transport of ozone across the tropopause due to changes in stratospheric ozone.

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. Recently, the tropospheric burden of CO and NO_2 (Edwards et al., 2004; Richter et al., 2005) have been estimated from satellite providing much needed data for model evaluation and very valuable constraints for emission estimates.

Assessment of long-term trends in tropospheric ozone is difficult due to the scarcity of representative observing locations with long records. Oltmans et al. (2006) show that the picture of long-term tropospheric ozone changes is a varied on both in terms of sign and magnitude of trends and in the possible causes for the change. 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), Tarasick et al. (2005), and Oltmans et al. (2006). Over Europe, ozone in the free tropospheric ozone increased 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 (32°N). 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. At Naha (26°N) a positive trend (5% decade^{-1}) is found between 700 and 300 hPa (1990–2004), while between the surface and 700 hPa a slightly negative trend is observed (Oltmans et al., 2006). 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. Trends are also derived from surface observations. Jaffe et al. (2003) derived a positive trend 1.4% yr^{-1} using a 15 year record from Lassen Volcanic Park in northern California (1750 m above sea level) and a consistent trend comparing two aircraft campaigns. A number of other sites show, however, no sign of significant changes over the United States over the last 15 years (Oltmans et al., 2006). Over Europe and North America, observations from Whiteface Mountain, Wallops Island, Hohenpeisenberg, Zugspitze, and Mace Head (flow from the European sector) show small trends or reductions during summer months, while there is an increase during winter months (Oltmans et al., 2006). These observations are consistent with reduced NOx emissions (Jonson et al., 2005). North Atlantic stations (Mace Head, Izana, and Bermuda) indicate increased ozone (Oltmans et al., 2006). Over the North Atlantic measurements from ships (Lelieveld et al., 2004) show small trends in ozone, however, at Mace Head a positive trend of 0.49 ± 0.19

ppb yr^{-1} for the period 1987–2003 is found, with the largest contribution from air coming from the Atlantic sector (Simmonds et al., 2004).

In the tropics very few long-term ozonesonde measurements are available. Thompson et al. (2001) found, based on TOMS satellite data, no significant trend in tropical tropospheric ozone during 1979–1992. More recent observations (1994–2003, in-situ data from the MOZAIC program) show significant upward trends in free-tropospheric ozone (7.7–11.3 km altitude) in tropical regions of both hemispheres, 1.12 ± 0.05 ppbv yr^{-1} and 1.03 ± 0.08 ppbv yr^{-1} in NH and SH respectively (Bortz and Prather, 2006)

At southern mid-latitudes surface observations from Cape Point, Cape Grim, Atlantic Ocean (from ship) and from sondes at Lauder (850–700 hPa) show positive trends in ozone concentrations, in particular during the biomass burning season in the SH (Oltmans et al., 2006). However, the trend is not accompanied by a similar trend in CO, which would have been expected if biomass burning had increased. The increase is largest at Cape Point, reaching 20% decade^{-1} (in September). At Lauder the ozone sondes shows that the increase is confined to the lower troposphere, below 500 hPa)

Changes in tropospheric ozone and the corresponding RF have been estimated by a number of recent model studies (Hauglustaine and Brasseur, 2001; 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 multi-model experiment including 10 global models was organized through the ACCENT Network (Gauss et al., 2006). In the ACCENT calculations the preindustrial emissions were specified while the models used their best estimates for the current emissions. Four of the ten models in the model exercise have detailed stratospheric chemistry included. The adjusted RF for all models was calculated by the same radiative transfer model. The normalized forcing for the models in the ACCENT study was 0.032 ± 0.004 W m^{-2} DU^{-1}, which is significantly lower than the TAR estimate of 0.042 W m^{-2} DU^{-1}.

RF estimates of tropospheric ozone increase since 1750 are given in Figure 2.11. For most of the calculations the same set of assumptions about preindustrial emission inventories have been used (zero anthropogenic emissions and biomass burning sources reduced by 90%). Emissions of NOx from soils and biogenic hydrocarbons were generally assumed to be natural and were thus not changed (cf. Chapter 7, Section 7.4). In one study (Hauglustaine and Brasseur, 2001) applied reduced soil NOx emissions based on changes in the use of fertilizers. Six models in the ACCENT experiment also made coupled climate-chemistry simulation for pre-industrial and present conditions, including climate change since pre-industrial times. The difference between the RF in the coupled climate-chemistry and the chemistry-only simulations, which indicate the possible climate feedback on tropospheric ozone, was positive in all models but generally small (Figure 2.11).

[INSERT FIGURE 2.11 HERE]

A general feature with the models is their inability to reproduce the low ozone concentrations indicated by the very uncertain semi-quantitative measurements (e.g., Pavelin et al., 1999) during the late 19th century. Mickley et al. (2001) tuned their model by reducing pre-industrial lightning and soil sources of NOx and increasing natural non-methane volatile organic compound (NMVOC) emissions to obtain close agreement with the observations. The RF then increased by 50–80% compared to calculations with the standard emissions. However, there are still several aspects of the early observations that were not captured by the tuned model tests.

The uncertainties in the estimated RF by tropospheric ozone, originates from two factors: The models used (CTM/GCM model formulation, radiative transfer models), and the potential overestimation of preindustrial ozone levels in the models. We assume that the range of results, for the model simulations using the standard emissions, represents the model uncertainty. This range was 0.25–0.45 W m^{-2} (with a mean of 0.34 W m^{-2} and a standard deviation of 0.07 W m^{-2}).

The uncertainty due to the limited understanding of the photochemical state of the preindustrial troposphere is difficult to quantify, but the results of Mickley et al. (2001) indicate that this could be substantial (0.3 W m^{-2}). To account for this uncertainty that only contribute to the upper range of our RF estimate, we have assigned a log-normal distribution to the ozone to assess the uncertainty. The best estimate for the RF of tropospheric ozone is 0.35 W m^{-2}, unchanged from the TAR, with an uncertainty of +0.15 and –0.1 W m^{-2}. The uncertainty range is ±1 standard deviation of the log-normal distribution.

2.3.7 Stratospheric Water Vapour

Water vapour in the atmosphere absorbs more radiation than any other greenhouse gas. Changes in the troposphere are almost entirely due to climate feedbacks and/or circulation changes (see Chapter 3, Section 3.2 and Chapter 8, Box 8.1), therefore these are not considered a RF: the water vapour flux from irrigation is one small exception to this and is evaluated as a forcing in Section 2.5.6. Stratospheric water vapour changes have somewhat different causes and can, at least in part, be considered a RF. 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 RF and this was estimated to contribute an extra 2–5% of the total CH_4 RF (~0.02 W m^{-2}).

Chapter 3, Section 3.4, discusses the evidence for stratospheric water vapour trends and presents current understanding as to their possible causes. There are now 14 years of global stratospheric water vapour measurements from HALOE and continued balloon-based measurements (since 1980) at Boulder, Colorado. There is some limited evidence from of a sustained long term increase in stratospheric water vapour of around 0.05 ppmv $year^{-1}$. However, recent observations show that water vapour concentrations in the lower stratosphere have been decreasing since 2000 (see Chapter 3, Section 3.4 for details and references). As well as CH_4 increases, several other indirect forcing mechanisms have been proposed, 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). These are mechanisms which can be linked to an external forcing agent. Other proposed mechanisms are more associated with climate feedbacks and are related to changes in tropopause temperatures or circulation (Stuber et al., 2001b; Fueglistaler et al., 2004). From these studies there is little consensus as to the cause(s) of stratospheric water vapour change and it is likely that different mechanisms are affecting water vapour trends at different altitudes.

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) 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) estimated an RF of about 0.2 W m^{-2} in a period of two decades, using a GCM to estimate the increase in water vapour in the stratosphere from oxidation of CH_4 and including climate feedback changes associated with an increase in greenhouse gases. 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} for 1980 to 2000. GCM radiation codes can have factor of two uncertainties in their modelling of this RF (Oinas et al., 2001). For the purposes of this chapter the above RF estimates are not readily attributable to forcing agent(s) and uncertainty as to the causes of the observed change preclude all but the component due to CH_4 increases being considered a forcing. Two related chemical transport model studies have calculated the RF associated with increases in CH_4 since preindustrial times (Hansen and Sato, 2001 and Hansen et al., 2005), no dynamical feedbacks were included in these estimates. Hansen et al. (2005) estimate a RF of 0.07 ± 0.01 W m^{-2} for the stratospheric water vapour changes over 1750–2000, which is somewhat larger than the TAR value. The RF from direct injection of water vapour by aircraft is believed to be an order of magnitude smaller than this ~0.002 W m^{-2} (IPCC, 1999). There has been little trend in CH_4 concentration since 2000 (see Section 2.3.2), therefore we base our best estimate (0.07 W m^{-2}) of the stratospheric water vapour RF on the Hansen et al., (2005) calculation and estimate a 0.05 W m^{-2} 1σ uncertainty, from the range of the RF studies which included other effects. However, we have a low confidence in this estimate as we only partially understand the vertical profile of methane induced stratospheric water vapour change. Other human causes of stratospheric water vapoue changeare unqiauntified and have a very low level of scientific understanding.

2.3.8 Observations of Long-Lived Greenhouse Gas Radiative Effects

Observations of the clear-sky radiation emerging at the atmospheric boundaries (TOA, surface) have been conducted. Such observations, by their nature, do not measure RF as defined here. Instead they yield a perspective on the influence of various species on the transfer of radiation in the atmosphere. Most importantly, the conditions involved with these observations involve varying/changing thermal and moisture profiles in the atmosphere such that they do not conform to the RF definition (see Section 2.2).

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. While both types of observations attest to the radiative influences of the gases, they should not be interpreted as having a linkage to the value of RFs in Section 2.3.

2.4 Aerosols

2.4.1 Introduction and Summary of the TAR

TAR categorised the mechanisms by which anthropogenic aerosols exert RF and how these can be separated into the direct and indirect effects. 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 having a significant anthropogenic component and therefore exerting a significant direct RF. Key parameters for determining the direct RF are the aerosol optical properties (the single scattering albedo, ω_o, specific extinction coefficient, k_e, and scattering phase function) 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 exert a net negative direct RF integrated over the solar spectrum, while partially absorbing aerosols may exert a negative top-of-the-atmosphere (TOA) direct RF over dark surfaces such as oceans or dark forest surfaces, and a positive TOA RF over bright surfaces such as desert, snow/ice or if the aerosol is above cloud (e.g., Chylek and Wong*,* 1995; Haywood and Shine*,* 1995). Both positive and negative TOA direct RF mechanisms reduce the shortwave irradiance at the surface thereby modifying the surface radiation budget. The longwave direct RF is generally of substantial magnitude only if the aerosol particles are large and occur in considerable concentrations at higher altitudes (e.g., Tegen et al., 1996). The direct RF due to tropospheric aerosols is most frequently derived at TOA rather than at the tropopause because shortwave radiative transfer calculations have shown a negligible difference between the two (e.g., Haywood and Shine, 1997 and Section 2.2). The surface forcing will be approximately the same as the direct RF at the TOA for scattering aerosols, but for partially absorbing aerosols the surface forcing maybe many times stronger than the TOA direct RF (e.g., Ramanathan et al., 2001b and references therein).

The indirect effect is the mechanism by which aerosols modify the microphysical and hence the radiative properties, 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 ambient environment 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). These effects are denoted as "cloud albedo effect" and "cloud lifetime effect", respectively. These terms are used throughout this report because they are 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 2.12), the hydrological cycle is invariably 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 2.12 HERE]

TAR did not include any assessment of the semi-direct effect (e.g., Jacobson, 2002; Menon et al., 2003; Cook and Highwood, 2004; Johnson et al., 2004; Johnson, 2005) which is the mechanism by which absorption of shortwave radiation by tropospheric aerosols leads to heating of the troposphere which changes the relative humidity and the stability of the troposphere and thereby influences cloud formation and lifetime. As with the cloud lifetime effect, in this report, the semi-direct effect is not strictly considered to be a RF because of modifications to the hydrological cycle, discussed in Chapter 7, Section 7.5 (see also Sections 2.2, 2.8 and 2.4.6).

Since TAR there has been a substantial degree of research into observations and modelling of tropospheric aerosols; these are discussed in turn in the following sections.

2.4.2 Observational Developments.

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 sites, or specifically as part of intensive field campaigns. These in-situ measurements 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, and the chemical composition. In addition, they provide key information on variability on various timescales. Comparisons of in situ measurements against those generated by global atmospheric models are complicated by differences in meteorological conditions and because in-situ measurements are representative of conditions mostly at or near the surface while the direct and indirect RF depend on the aerosol vertical profile. For example, the spatial resolution of global model grid-boxes are typically 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; this poses limitations when comparing with observations conducted over smaller spatial extent and shorter time duration.

Combinations of satellite and surface-based observations are capable of providing near-global retrievals of aerosol properties. These are discussed in detail in this sub-section; the emissions estimates, trends, and in-situ measurements of the physical and optical properties are discussed with respect to their influence on RF in Section 2.4.4. Further detailed discussions of the recent satellite observations of aerosol properties and a satellite measurement based assessment of the aerosol direct RF are given by Yu et al. (2006).

2.4.2.1 Satellite retrievals

Satellite retrievals of aerosol optical depth in cloud-free regions have been improved via new-generation sensors (Kaufman et al., 2002) and an expanded global validation program (Holben et al., 2001). Advanced aerosol retrieval products such as aerosol fine-mode fraction and effective particle radius have been developed and offer potential for improving knowledge of the aerosol direct radiative effect. Additionally, efforts have been made to determine the anthropogenic component of aerosol and associated direct RF, as discussed by Kaufman et al. (2002) and implemented by Bellouin et al. (2005) and Chung et al. (2005). However, validation programs for these advanced products have yet to be developed and initial assessments indicate some systematic errors (Levy et al., 2003; Chu et al., 2005; Anderson et al., 2005a), suggesting that the routine differentiation between natural and anthropogenic aerosols from satellite retrievals remains very challenging.

2.4.2.1.1 Aerosol optical depth, τ_{aer}

Figure 2.13 shows an example of τ_{aer} retrieved over both land and ocean together with the geographical positions of aerosol instrumentation. Table 2.2 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 (Figure 2.13a) clearly differs from that for August, September and October (Figure 2.13b) 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 Figure 2.13a but shifts to southern Africa in Figure 2.13b. Likewise the biomass burning in South America is most evident in Figure 2.13b. In Figure 2.13a the transport of mineral dust from Africa to the South American continent is clearly discernible while in Figure 2.13b 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 evident over many continental regions of the Northern Hemisphere. Sea-salt aerosol is visible in regions where the wind speed is high (e.g., south of 45ºS). The MODIS aerosol algorithm is currently unable to make routine retrievals over highly reflective surfaces such as deserts, snow cover, ice, and ocean glint, as well as over high latitude regions when the solar insolation is insufficient.

[INSERT TABLE 2.2 HERE]

[INSERT FIGURE 2.13 HERE]

Early retrievals for estimating τ_{aer} include the single channel retrieval of the AVHRR) (e.g., Husar et al., 1997, 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 TOMS retrieval is essentially independent of surface reflectance thereby allowing retrievals over both land and ocean (Torres et al., 2002), but is sensitive to the altitude of the aerosol, and has a relatively low spatial resolution. While these retrievals only use a limited number of spectral bands and lack sophistication compared to 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). 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 exponent, Å, over ocean (Deuzé et al., 2000), τ_{aer} over land (Deuzé et al., 2001), and the direct radiative effect of aerosols (Boucher and Tanré, 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 (Chu et al., 2002) than over oceans (Remer et al., 2002) owing to uncertainties in the land surface reflectance characteristics and can be minimised by careful selection of the viewing geometry (Chylek et al., 2003). 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., 2005a) which allow for a more comprehensive comparison of aerosol models against satellite observations. 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 deserts (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 nine viewing geometries and two different radiances. The results have been validated against those from AERONET (Aerosol RObotic NETwork; see Section 2.4.3.2). ATSR and ATSR-2 (Holzer-Popp et al., 2002; Veefkind et al., 1998) 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) to make τ_{aer} retrievals over both ocean and land (Robles-Gonzalez et al., 2000). 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. SeaWiFs uses 0.765 μm and 0.856 μm radiances to provide $\tau_{\lambda=0.856}$ and Å over ocean using a bi-modal aerosol size distribution (Wang et al., 2005).

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., Penner et al., 2002; Myhre et al., 2004a, 2005; Kinne et al, 2005, Jeong et al., 2005). These discrepancies are due to different assumptions in the cloud clearing algorithms, aerosol models, different wavelengths, and view geometries used in the retrievals, different parameterisations of ocean surface reflectance etc. Comparison of these satellite aerosol retrievals with the surface AERONET observations provide an opportunity to objectively evaluate as well as improve the accuracy of these satellite retrievals. Myhre et al. (2005) show that dedicated instruments using multi-channel and multi-view algorithms perform better when compared against AERONET than the simple algorithms that they have replaced, and Zhao et al. (2005) shows retrievals based on dynamic aerosol models perform better than those based on globally fixed aerosol models. While some systematic biases in specific satellite products exist (e.g., Remer et al., 2005), these can be corrected for (e.g., Bellouin et al., 2005), which then enables an assessment of the direct radiative effect and the direct RF from an observational perspective as detailed below.

2.4.2.1.2 Direct radiative effect, DRE.

The direct radiative effect (DRE) is the sum of the effects due to anthropogenic and natural aerosol species while the direct RF only considers the anthropogenic components. Satellite estimates of the global clear-sky DRE over oceans have advanced since TAR owing to the development of dedicated aerosol instruments and algorithms as summarised by Yu et al. (2006) (see Table 2.3). Table 2.3 suggests a reasonable agreement of the global mean, diurnally averaged clear-sky DRE from various studies, with a mean of -5.3 W m^{-2} and a standard deviation of 0.8 W m^{-2}. The clear-sky DRE is converted to an all-sky DRE by Loeb and Manalo-Smith (2005) who derive an all sky direct radiative effect over oceans of -1.6 to -2.0 W m^{-2} but assume that no aerosol contribution to the direct RF from cloudy regions; such an assumption is not valid if partially absorbing aerosol exists above cloud (see Section 2.4.5.4).

[INSERT TABLE 2.3 HERE]

Furthermore, use of a combination of sensors on the same satellite offers the possibility of concurrently deriving τ_{aer} and DRE (e.g., Zhang and Christopher, 2003; Zhang et al., 2005) which enables estimation of the RF efficiency viz., the DRE divided by τ_{aer} (W m^{-2} τ_{aer}^{-1}). Because the RF efficiency removes the dependence on the geographic distribution of τ_{aer} it is a useful parameter for comparison of models against observations (e.g., Anderson et al., 2005b), although the RF efficiency thus derived is not a linear function of τ_{aer} at high τ_{aer} such as those associated with intense mineral dust, biomass burning or pollution events.

2.4.2.1.3. Direct radiative forcing, RF.

Kaufman et al. (2005a) estimate the anthropogenic only component of the aerosol fine mode fraction from the MODIS product to deduce a clear sky RF over ocean of -1.4 W m^{-2}. Bellouin et al. (2005) use a combination of MODIS τ_{aer} and fine mode fraction together with data from AeroCom (see Section 2.4.4) to determine a RF of aerosols over both land and ocean of -0.8 ± 0.2 W m^{-2}, but this estimate does not include the contribution to the RF and associated uncertainty from cloudy skies. Chung et al. (2005) perform a similar satellite/AERONET/model analysis, but include the contribution from cloudy areas to deduce a RF of -0.35 W m^{-2} and -0.50 W m^{-2} depending upon whether the anthropogenic fraction is determined from a model or from the MODIS fine mode fraction and suggest an overall uncertainty in the RF due to aerosols of -0.1 to -0.6 W m^{-2}. Yu et al. (2006) used several measurements to estimate a RF of -0.5 ± 0.33 W m^{-2}. These estimates of the RF are compared to those obtained from modelling studies in Section 2.4.5.7.

2.4.3 Surface-Based Retrievals

A significant advancement since TAR is 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 2.13a. Currently there are approximately 150 sites operating at any one time, many of which are permanent, thereby enabling representative climatological and interannual column-averaged 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 column averaged size distribution and, if the τ_{aer} is high enough ($\tau_{aer}>0.5$), 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 for both the derived size distribution and ω_o when compared against in-situ measurement by instrumented aircraft for different aerosol species (e.g., Dubovik et al., 2002; Haywood et al., 2003a; Reid et al*.,* 2003; Osborne et al., 2004).

The MPLNET Lidar network currently consists of eleven lidars worldwide, nine are co-located with AERONET sites and provide complementary vertical distributions of aerosol backscatter and extinction. Additional temporary MPLNET sites have supported major aerosol field campaigns (e.g., Campbell et al. 2003; Reid et al. 2003). EARLINET was a European-wide lidar network which currently has fifteen aerosol lidars making routine retrievals of vertical profiles of aerosol extinction (Mathias et al., 2004). 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; Murayama et al., 2001).

2.4.4 Advances in Modelling

Since TAR, more complete aerosol modules in a larger number of global atmospheric models now provide estimates of the direct RF. Several models have resolutions better than 2° × 2° degrees in the horizontal and more than 20 to 30 vertical levels; this represents a considerable enhancement over the models used in TAR. Such models now include the most important anthropogenic species and natural species. Tables 2.4, 2.5 and 2.6 summarise studies published since TAR. Because the most important aerosol species are now included, a comparison of key model output parameters such as the total τ_{aer} is possible against satellite retrievals, and surface-based sun photometer and lidar sites (see Sections 2.4.3 and 2.4.5). Progress with respect to the modelling of the indirect effects due aerosol-cloud interactions is detailed in Section 2.4.6 and Chapter 7, Section 7.5. Several studies have explored the sensitivity of direct aerosol RF to current parameterisation uncertainties. These are assessed in the following sections.

Major progress over TAR has been made in the documentation of the diversity of current aerosol model simulations. Currently, 16 groups have participated in the Global Aerosol Model Intercomparison (AeroCom) initiative (Kinne et al., 2005). Extensive model outputs are available via a dedicated website (Schulz*,* 2004). Three model experiments (named *A*, *B*, *PRE*) were analysed: *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 AeroCom aerosol emissions for the year 2000; *Experiment PRE:* uses prescribed aerosol emissions for the year 1750. The model diagnostics included information on emission and deposition fluxes, vertical distribution and sizes, thus enabling a better understanding of the differences in lifetimes of the various aerosol components in the models.

The AeroCom results from Textor et al., (2005) are assessed in the remainder of this paragraph. The model comparison study finds a wide range in several of the diagnostic parameters; these, in turn, indicate which aerosol parameterisations are poorly constrained and/or understood. For example, coarse aerosol fractions are responsible for a large range in the natural aerosol emission fluxes (dust: ±49%, sea salt: ±200%, where uncertainty is 1σ standard deviation of inter-model range), and consequently in the dry deposition fluxes. The complex dependence of the source strength on wind speed adds to the problem of computing natural aerosol emissions. Dust emissions for the same time period can vary by a factor of 2 or more depending on details of the dust parameterization (Luo et al., 2003; Timmreck and Schulz, 2004; Balkanski et al., 2004; Zender et al., 2004), and even depends on the reanalysis meteorological dataset used (Luo et al., 2003). With respect to anthropogenic emissions, 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 vertical aerosol distribution is shown to vary considerably, which is a consequence of important differences in removal and vertical mixing parameterisations. The inter-model range for the fraction of sulphate mass below 2.5 km to that of total sulphate is 45% ± 23%. Since humidification takes place mainly in the boundary layer, this source of variation increases the range of modelled direct RF. Additionally, differences in the parameterization of the wet deposition/vertical mixing process becomes more pronounced above 5 km altitude. Some models have a tendency to accumulate insoluble aerosol mass (dust and carbonaceous aerosols) at higher altitudes, while others have much more efficient wet removal schemes. 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. These variations are of interest, since they express the linearity of modelled emissions to aerosol burden and eventually to RF.

Considerable progress has been made in the systematic evaluation of global model results (see references in Tables 2.4–2.6). The simulated global τ_{aer} in models ranges from 0.11 to 0.14 as compared to that from remote sensing from ground (AERONET ca. 0.135) and space (satellite composite ca. 0.15) (Kinne et al., 2003 and 2005), but significant differences exist in regional and temporal distributions. Modelled absorption optical thickness has been suggested to be underestimated by a factor of 2–4 when compared to observations (Sato et al., 2003) and RF efficiencies have been shown to be lower in models both for the global average and regionally (Yu et al., 2006) (see Section 2.4.5.7). The merging of modelled and observed fields of aerosol parameters through assimilation methods of different degree of complexity have also been performed since TAR (e.g., Chung et al., 2005). Model results are constrained to obtain present day aerosol fields consistent with observed ones. Collins et al. (2001) showed that assimilation of satellite derived fields of τ_{aer} can reduce the model bias down to 10% with respect to daily mean τ_{aer} measured with a sun photometer at the INDOEX station Kaashidhoo. Liu et al. (2005) demonstrate similar efficient reduction in errors τ_{aer}. The magnitude of the global dust cycle has been suggested to range between 1500 and 2600 TG y^{-1} by minimizing the bias between model and multiple dust observations (Cakmur et al. 2006). Bates et al., (2006) focused on three regions downwind of major urban/population centres and performed radiative transfer calculations constrained by extensive observational inputs to derive 24 hour average clear sky direct radiative effects of –3.3 ± 0.47, –14 ± 2.6, –6.4 ± 2.1 W m^{-2} for the North Indian Ocean, the North West Pacific, and the North West Atlantic, respectively. By constraining aerosol models with extensive observations, the uncertainty associated with the direct radiative effect was reduced by approximately a factor of two.

2.4.5 Direct Radiative Forcing

Unless otherwise stated, we discuss the TOA direct RF of different aerosol types as a global-mean quantity integrated over 24 hours and inclusive of the effects of clouds. Where possible, statistics from model results are used to assess the uncertainty in the RF. Both AeroCom and non-AeroCom results are assessed. Because AeroCom-(B-PRE) results are from prescribed emissions, the uncertainty from these results is multiplied by the estimated uncertainty in the emissions. The quoted uncertainties therefore include the structural uncertainty associated with the RF, but do not include the full range of parametric uncertainty as the model results are essentially best estimates constrained by observations of emissions, wet and dry deposition, size distributions, optical parameters, hygroscopicity, etc (Pan et al., 1997). We report the uncertainty as approximately ±1 standard deviation to allow the uncertainty in the RF of each species of aerosol to be inter-compared in a more quantitative manner.

2.4.5.1 Sulphate aerosol

Atmospheric sulphate aerosol consists mainly of the chemical compounds H_2SO_4, NH_4HSO_4, and $(NH_4)_2SO_4$ in either the aqueous or crystalline form. Sulphate is formed by aqueous phase reactions within cloud droplets and oxidation of SO_2 via gaseous phase reactions with the hydroxyl radical (e.g., Penner et al., 2001). Emission estimates are summarised by Haywood and Boucher (2000). 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 sulphide emissions by marine phytoplankton (~19%) and by SO_2 emissions from volcanoes (~7%). Current estimates of global SO_2 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 et al, 2004). In the USA, the emissions have been reduced from about 10TgSyr^{-1} to 7TgSyr^{-1} (U. S. EPA, 2004: http://www.epa.gov) in the period 1982-2001. However, over the same period SO_2 emissions have been increasing significantly from Asia which is estimated to currently emit 17TgSyr^{-1} (Streets et al., 2003) and from developing countries (e.g., Boucher and Pham, 2002). The net result of these combined regional reductions and increases leads to uncertainty in whether the global SO_2 has increased or decreased 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 Ocean areas will lead to subsequent shifts in the pattern of the RF (e.g., Boucher and Pham*,* 2002, Pham et al., 2005).

The optical parameters of sulphate aerosol have been well documented (see Penner et al., 2001 and references therein). Sulphate is essentially an entirely scattering aerosol virtually across the solar spectrum ($\omega_o = 1$) but with a small degree of absorption in the near-infrared spectrum, Theoretical and experimental data are available on the relative humidity dependence of the specific scattering 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), MINOS (2001 campaign), ACE-Asia (2001), APEX (2000-2003), NEAQS (2003), and CLAMS (Smith et al., 2005) continue to show that sulphate contributes a significant fraction of the accumulation mode mass, anthropogenic τ_{aer} and RF (e.g., Hegg et al., 1997; Russell and Heintzenberg*,* 2000; Ramanathan et al., 2001b; Magi et al., 2005; 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., 2004). This results in a composite aerosol state in terms of effective refractive indices, size distributions, physical state, morphology, hygroscopicity and optical properties.

TAR suggested a 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. Results from model studies since TAR are summarised in Table 2.4 for both non-AeroCom and AeroCom studies. For non-AeroCom studies, the 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 RF per unit τ_{aer} is substantial. These differences are due to differing representation of aerosol optical properties, cloud, surface reflectance, hygroscopic growth, sub-gridscale effects, radiative transfer codes etc (Ramaswamy et al., 2001). Myhre et al. (2004b) performed several sensitivity studies finding the uncertainty was particularly linked to the hygroscopic growth and that differences in the model relative humidity fields could cause differences of up to 60% in the RF. The RF from the models participating in the AeroCom project is slightly weaker than that obtained from the other studies with a mean of ~–0.35 W m^{-2} and a standard deviation of 0.15 W m^{-2}; the standard deviation is reduced for the AeroCom models owing to constraints on aerosol emissions, based on updated emission inventories (see Table 2.4). Including the uncertainty in the emissions reported in Haywood and Boucher (2000) increases the standard deviation to 0.2 W m^{-2}. As the sulphate aerosol is considered to be almost entirely scattering, the surface forcing will be similar or marginally stronger than the RF diagnosed at the top of the atmosphere The uncertainty in the RF estimate relative to the mean value remains relatively large compared to the situation for LLGHGs.

On the basis of these results, a top of the atmosphere RF of –0.40 ± 0.20 W m^{-2} is suggested for the RF due to sulphate aerosol.

[INSERT TABLE 2.4 HERE]

2.4.5.2 Organic carbon from fossil fuels

Organic aerosols are a complex mixture of chemical compounds produced from fossil-fuel and biofuel 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 primary organic carbon from fossil fuel burning have been estimated to be 10 to 30 Tg C yr^{-1} (Liousse et al., 1996; Cooke et al., 1999; Scholes and Andreae*,* 2000). More recently, Bond et al. (2004) provide a detailed analysis of primary organic carbon emissions from fossil-fuels, biofuels and open burning, and suggest that contained burning (approximately the sum of fossil-fuel and biofuel) emissions are in the range 5-17 Tg C yr^{-1}, with fossil-fuel contributing only 2.4 Tg C yr^{-1}. Ito and Penner (2005) estimated global fossil-fuel OC emissions of around 2.2 Tg C yr^{-1}, and global biofuel OC emissions of around 7.5 Tg C yr^{-1}. Trends in emissions of fossil/bio fuel organic carbon have been estimated for the period 1870–2000 by Ito and Penner (2005) to have increased by a factor of three. Mass concentrations of 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 τ_{aer}. During INDOEX which studied the industrial plume over the Indian Ocean, Ramanathan et al. (2001b) found organic carbon was the second largest contributor to the τ_{aer} after 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 open biomass burning. Observations of the hygroscopicity of organic carbon suggest that a considerable 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 organic carbon composition by functional group analysis in some main classes of organic species (e.g., Decesari et al., 2000; Decesari et al., 2001; Maria et al., 2002; Ming and Russell, 2002) capturing some general characteristics in terms of e.g., refractive indices, hygroscopicity, and cloud activation properties. This facilitates improved implementation in global models (e.g., Fuzzi et al., 2001; Ming et al., 2005; Kanakidou et al., 2005).

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), results that are backed up by laboratory studies (e.g., Schnaiter et al., 2003). However coatings of organic carbon aerosol on hygroscopic aerosol such as sulphate may lead to suppression of 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 RF. Secondary organic carbon is highly simplified in the global models and in many cases treated as an additional source similar to primary organic carbon. 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 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 RF of organic carbon aerosols from both fossil-fuel and biomass burning aerosols and the combined RF of both components. These studies are summarised in Table 2.5. The RF from total organic carbon from both biomass burning and fossil-fuel emissions from the non-AeroCom and AeroCom models are –0.24 ± 0.08 W m^{-2} and –0.18 ± 0.10 W m^{-2} respectively. Where the 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 (Bond et al., 2004). The mean RF of the fossil-fuel component of organic carbon from those studies other than in AeroCom is –0.06 W m^{-2} , while those from AeroCom produce a RF of –0.04 W m^{-2} with a range of –0.01 W m^{-2} to –0.07 W m^{-2} and a standard deviation of around 0.02 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 forcing is only slightly stronger than that at the top of the atmosphere. Based on these results, the RF due to fossil fuel sources of organic carbon is estimated to –0.10 ± 0.10 W m^{-2} the relatively large uncertainty being due to difficulties in constraining the total column burden, the absorption, mixing, and hygroscopicity of the aerosol.

[INSERT TABLE 2.5 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. Global current day fossil-fuel emissions range form 5.8 $TgCyr^{-1}$ to 8.0 $TgCyr^{-1}$ (Haywood and Boucher, 2000 and references therein). Bond et al. (2004) estimate the total current global emission of BC to be approximately 8 $TgCyr^{-1}$ with contributions of 4.6 $TgCyr^{-1}$ from fossil-fuel and biofuel combustion and 3.3 $Tgyr^{-1}$ from open biomass burning and estimate an uncertainty of about a factor of two. Ito and Penner (2005) suggest fossil-fuel black carbon emissions for 2000 of around 2.8 $TgCyr^{-1}$. The trends in emission of fossil-fuel black carbon have been investigated in industrial areas by Novakov et al. (2003) and Ito and Penner (2005). 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 TgC yr^{-1}) owing to the rapid expansion of the USA, European and Asian economies (e.g., Streets et al*,* 2001, 2003), and has since fallen to around 5.6 TgC yr^{-1} owing to further emission controls. Ito and Penner (2005) determine a similar trend in emissions over the period 1950–2000 of approximately a factor of three, but the absolute emissions are smaller than in Novakov et al. (2003) by approximately a factor of 1.7.

Black carbon aerosol strongly absorbs solar radiation. 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 as the particles age modifying the optical properties (e.g., Abel et al., 2003). 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 local RF at the top of the atmosphere (–7 W m^{-2}). Additionally, the presence of black carbon in the atmosphere above highly reflectant surfaces such as snow/ice or clouds is may cause a significant positive 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).

The TAR assessed the RF due to fossil-fuel black carbon as being +0.2 W m^{-2} with an uncertainty of a factor of two. Those models since TAR that explicitly model and separate out the 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 2.5. 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 RF from both sources of 0.44 ± 0.13 W m^{-2} and 0.29 ± 0.15 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 are probably due to the method of internal mixing the BC aerosols with other components which increases the absorption (e.g., Stier et al., 2005), or a larger fraction of aerosols located above clouds (see earlier discussion) which enhances the RF. Carbonaceous aerosols emission inventories suggest a split of approximately 34–38% between emissions from biomass burning sources and fossil-fuel burning sources. Models which separate fossil fuel from biomass burning suggest a split in RF of 50:50, which is applied to those estimates where the black carbon emissions are not explicitly separated into emission sources to provide an estimate of the RF due to fossil-fuel black carbon. For the AeroCom results, the fossil-fuel black carbon RF ranges from +0.08 W m^{-2} to +0.18 W m^{-2} (model of Iversen and Seland, 2002) with a mean of +0.13 W m^{-2} and a standard deviation of +0.03 W m^{-2}. For the non-AeroCom results, the RF ranges from +0.15 W m^{-2} to approximately +0.27 W m^{-2} with a standard deviation of 0.11 W m^{-2}. Grouping all the model results together as equally likely results in a RF due to black carbon from fossil-fuels of +0.20 W m^{-2} ± 0.10 W m^{-2} which is used as our best estimate. This estimate does not include the semi-direct effect or the BC impact on snow and ice surface albedo.

2.4.5.4 Biomass burning aerosols

TAR suggested a contribution to the 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 RF from Hansen and Sato (2001), Hansen et al. (2002), and Hansen and Nazarenko (2004) and Jacobson (2001) include the RF component of 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. Emission inventories show more significant differences for biomass burning aerosols than for aerosols of from fossil fuel origin (Kasischke and Penner, 2004). Furthermore, the preindustrial levels of biomass burning aerosols are also difficult to quantify (Mouillot et al., 2006; Ito and Penner, 2005).

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; 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; 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 which contains a significant amount of absorbing black carbon particles, 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 2.14).

[INSERT FIGURE 2.14 HERE]

Modelling efforts have used data from measurement campaigns to improve the representation of the physical and optical properties as well as the vertical profile of biomass burning aerosol (Myhre et al., 2003, ; Penner et al., 2003 and Section 2.4.6).These modifications have had important consequences on estimates of the 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 the clouds leading to a reduction in reflected 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 τ_{aer} and cloud effective radius in estimating the indirect radiative effect of aerosols.

The biomass burning aerosols may exert a significant positive RF when above clouds. This is documented by the non-AeroCom and AeroCom models in Table 2.5. The AeroCom and non-AeroCom models suggest an average global mean RF from biomass burning aerosols of +0.05 W m^{-2} and +0.07 W m^{-2}, respectively and the combined standard deviation is 0.08 W m^{-2}. Hence even the sign of the RF due to biomass burning aerosols is in question. The most negative RF of –0.05 W m^{-2} is from the model of Koch. (2001) and from the Myhre et al. (2003) AeroCom submission, while the most positive of +0.23 W m^{-2} comes from the Stier et al. (2005) AeroCom submission. Thus, these recent studies yield a RF for biomass burning aerosols that is significantly different than reported by TAR and is now suggested to be +0.0 ± 0.10 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. (2004), who estimate that the RF of nitrate over Europe is some 25% of that due to sulphate aerosol, and of Martin et al. (2004), who report –0.04 to –0.08 W m^{-2} for global mean RF by nitrate. Further, Liao and Seinfeld (2005) estimated a global mean RF due to nitrate of –0.16 W m^{-2}. In this study, heterogeneous chemistry reactions on particles were included and this strengthens the RF due to nitrate and accounts for 25% of its RF. Surface observations of fine mode nitrate particles show that it is mainly in highly industrialized regions where high concentrations are found, while low concentrations are found in rural areas (Malm et al., 2004; Putaud et al., 2004). 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). In the AeroCom exercise, nitrate aerosols were not included so fewer estimates of this compound exist compared to the other aerosol species considered. The mean RF for nitrate is estimated to be –0.10 W m^{-2} at the top of the atmosphere, and the conservative scattering nature means a similar flux change at the surface. However, the uncertainty in this estimate is 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 τ_{aer}. Thus we tentatively adopt a RF of –0.10 ± 0.10 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, over-grazing), changes in surface water (e.g., Caspian and Aral Sea, Owens Lake), and industrial practices (e.g., cement production, transport) (Prospero et al. 2002). TAR suggested that the RF due to anthropogenic mineral dust lies in the range of +0.4 to –0.6 W m^{-2}, and did not assign a best estimate because of the difficulties in determining the anthropogenic contribution to the total dust loading, the uncertainty in the optical properties of dust and in evaluating the competing shortwave and longwave radiative effects. For the sign and magnitude of the mineral dust RF the most important factor for the solar RF is the single scattering albedo whereas the longwave RF is dependent on the vertical profile of the dust.

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 an updated, alternative 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. Yoshioka et al. (2005) suggests that a model simulation best reproduces the North African TOMS aerosol index observations when the cultivation source in the Sahel region contributes 0–15% to the total dust emissions in North Africa. A 35-year dust record established from Barbados surface dust and TOMS plus Meteosat satellite observations shows the importance of climate control and Sahel drought for interannual and decadal dust variability, with no overall trend yet documented (Chiapello et al. 2005). As further detailed in Chapter 7, Section 7.3, climate change and carbon dioxide variations on various timescales can change vegetation cover in semi-arid regions. Such processes dominate over land-use changes as defined above, which would give rise to anthropogenic dust emissions (Mahowald and Luo, 2003; Moulin and Chiapello, 2004, Tegen et al., 2004). A best guess of 0–20% anthropogenic dust burden from these works is used here, but it is acknowledged that a very large uncertainty remains, because the methods used can not exclude either a reduction of 24% in present day dust nor a large anthropogenic contribution of up to 50% (Mahowald and Luo, 2003; Mahowald et al., 2004; Tegen et al., 2005). The RF efficiency of anthropogenic dust has not been well differentiated from that of natural dust and we assume them to be equal.

In-situ measurements of the optical properties of local Saharan dust (e.g., Haywood et al., 2003c; Tanré et al., 2003); transported Saharan mineral dust (e.g., Kaufman et al., 2001; Moulin et al., 2001; Coen et al., 2004) and Asian mineral dust (Huebert et al., 2003; Clarke et al., 2004) reveal that dust is considerably less absorbing in the solar spectrum than suggested by previous dust models such as that of WMO (1986). These new simultaneous spectral remote and in-situ observations suggest that the single scattering albedo (ω_o) of pure dust at a wavelength of 670 nm is predominantly in the range 0.90–0.99, with a central global estimate of 0.96. This is in accordance with the bottom-up modelling of ω_o based on the hematite content in desert dust sources (Claquin et al., 1999). Analyses of ω_o from long-term AERONET sites influenced by Saharan dust suggest an average ω_o of 0.95 (Dubovik et al., 2002). These high ω_o-values suggest that a positive RF by dust in the solar region of the spectrum is unlikely. However, absorption by particles from source regions with variable mineralogical distributions is generally not represented by global models.

Measurements of the direct radiative effect of mineral dust over ocean regions, where natural and anthropogenic contributions are indistinguishably mixed, 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 Wm^{-2} at the top of the atmosphere for July 2003.

Recent model simulations report the total anthropogenic and natural dust direct radiative effect (shortwave/longwave/net TOA) [in W m^{-2}]: Liao et al. (2004): –0.21/+0.31/+0.1; Reddy et al. (2005a): –0.28/+0.14/–0.14; Jacobson 2001: –/–/–0.13; Myhre and Stordal (2001a) (sensitivity experiments, except case 6): –1.4 – +0.2/+0.0 – +0.8/–0.7 – +0.6; and from AeroCom database: models UIO-CTM*: –0.56/–/–; LSCE*: –0.6/+0.3/–0.3; UMI*: –0.56/–/–; GISS: –0.75/–/–. Models using a higher single scattering albedo as described above are marked with a star (*) and show more negative shortwave effects. The scatter of dust radiative effect estimates reflects that dust burden and τ_{aer} show diversities of ±40% and ±44% respectively among 16 AeroCom A model simulations (Textor et al., 2005; Kinne et al., 2005). Dust emissions from different studies range between 1000 and 2150 TG yr^{-1} (Zender, 2004). Finally, a major effect of dust may be in reducing the burden of anthropogenic species in the accumulation mode and reducing their residence time (Bauer and Koch, 2005), see Section 2.4.5.7.

The range of the reported total dust radiative effect, the revised anthropogenic contribution to dust radiative effect of 0–20% and the revised absorption properties of dust suggest a small negative anthropogenic RF of -0.1 W m^{-2}. The uncertainty for the dust anthropogenic RF is estimated to be ± 0.2 W m^{-2}, reflecting the uncertainty in total dust emissions and burdens and the range of possible anthropogenic dust contribution values, which can not be excluded at this point. At the upper and lower ends of this uncertainty range anthropogenic dust RF could be as negative as –0.3 W m^{-2} and even slightly positive up to +0.1 W m^{-2}, a range that includes almost all recent estimates.

2.4.5.7 Combined aerosol species

TAR reported RF values associated with several components of the aerosol but did not provide an estimate of the overall aerosol RF. Improved and intensified in-situ observations and remote sensing of aerosols suggest that the range of combined aerosol RF is now better constrained. For model results extensive validation now exists for combined aerosol properties, representing the whole vertical column of the atmosphere, such as τ_{aer}. Using the combined estimates provides an alternative procedure to estimate the RF uncertainty. This may be more robust than that obtained through uncertainty propagation calculation based on RF uncertainties from all individual aerosol components. Furthermore, a RF estimate accounts for non-linear processes due to aerosol dynamics and interactions between radiation field and aerosols. The role of non-linear processes of aerosol dynamics for RF has been recently studied in global aerosol models, which account for the internally mixed nature of aerosol particles (Kirkevag and Iversen*,* 2002; Liao and Seinfeld*,* 2005; Stier et al., 2005; Takemura et al., 2005). Mixing of aerosol particle populations influences the radiative properties of the aerosol, which involves changes of size, chemical composition, mixing state, shape and feedbacks into the aerosol removal and formation processes itself. Assuming external or internal mixing of black carbon in organic matter changes the associated RF from –0.78 to –0.48 W m^{-2} (see compilation in Chung and Seinfeld, 2002). Although the source-related processes for anthropogenic aerosols favour their submicron nature, natural aerosols enter the picture by providing a condensation surface for aerosol precursor gases. 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. Bauer and Koch, (2005) estimate the sulphate RF to weaken from –0.25 to –0.18 W m^{-2} when dust is allowed to interfere with the sulphur cycle. It would be even more desirable from a policy perspective to identify the RF contribution attributable to source categories (Section 2.9.3 investigates this). However, few models have separated out the RF from specific emission source categories. Estimating the combined aerosol RF is a first step to quantify the anthropogenic perturbation brought to the aerosol and climate system by individual source categories.

A central model-derived estimate for the aerosol RF is based here on a compilation of recent simulation results using multi-component global aerosol models (see Table 2.6). This is a robust method for several reasons: The complexity of multi-component aerosol simulations captures non-linear effects as described above. Combining model results removes part of the errors in individual model formulations. As shown by Textor et al. (2005), the model specific treatment of transport and removal processes is partly responsible for correlated dispersion of the different aerosol components. A less dispersive model with smaller burdens necessarily has both less scattering and absorbing aerosols interacting with the radiation field. An error in accounting for cloud cover would affect the all-sky RF from all aerosol components. Such errors result in correlated RF efficiencies for major aerosol components within a given model and directly combining aerosol RF results will give a more realistic aerosol RF uncertainty estimate. The AeroCom compilation suggests significant differences in the modelled local and regional composition of the aerosol (see also Figure 2.14), but overall reproduction of the total τ_{aer} variability (Kinne et al., 2005). The scatter in performance of the models suggests that currently no preference or weighting of individual model results can be used (Kinne et al., 2005). The aerosol RF taken together from several models is more robust than an analysis per component or by just one model. The central estimate from Table 2.6 of the aerosol RF is –0.2 W m^{-2} with a standard deviation of ±0.2 W m^{-2}. This is a low-end estimate for both the aerosol RF and uncertainty because nitrate and anthropogenic mineral dust are missing in most of the model simulations. Adding their contribution would suggest an overall model-derived aerosol RF of –0.4 W m^{-2} and increased standard deviation of approximately ±0.3 W m^{-2}, according to results from Sections 2.4.5.6 and 2.4.5.5.

[INSERT TABLE 2.6]

Three measurement-based estimates of the aerosol RF have become available, which all suggest a more negative aerosol RF than the model studies (see Section 2.4.2.1.3.). Bellouin et al. (2005) compute a top of the atmosphere aerosol RF of –0.8 ± 0.1 W m^{-2}. Chung et al. (2005) based upon similarly extensive calculations estimate the value to be –0.35 ± 0.25 W m^{-2} and Yu et al. (2006) –0.5 ± 0.33 W m^{-2}. A central measurement based estimate would suggest a direct aerosol RF of –0.55 W m^{-2}. Figure 2.15 shows the observational based direct aerosol RF estimates together with the published model estimates since TAR.

[INSERT FIGURE 2.15]

The discrepancy between measurements and models is also apparent in oceanic clear-sky conditions where the measurement-based estimate of the combined aerosol radiative effect including natural aerosols is considered unbiased. In these areas models underestimate the negative aerosol DRE by 20–40% (Yu et al., 2006). The anthropogenic fraction of τ_{aer} is less different and rather underestimated by measurement based methods. Kaufman et al. (2005a) have used satellite observed fine mode τ_{aer} to estimate the anthropogenic τ_{aer}. Correcting for fine mode τ_{aer} contributions from dust and sea salt, they find 21% of the total τ_{aer} to be anthropogenic. Table 2.6 suggests that the fraction of anthropogenic τ_{aer} estimated from models is slightly higher (29%). Finally, cloud contamination of satellite products, aerosol absorption above clouds, not accounted for in some of the measurement-based estimates, and the complex assumptions on aerosol properties in both methods can contribute to the present discrepancy and increase uncertainty in aerosol RF.

A large source of uncertainty in the aerosol RF estimates is associated with aerosol absorption. Sato et al. (2003) determined the absorption τ_{aer} from AERONET measurements and suggested that aerosol absorption simulated by global aerosol models is underestimated by a factor of 2–4. Schuster et al. (2005) estimate the black carbon loading over continental scale regions. *Prima facie* the results suggest that the model concentrations and absorption τ_{aer} 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 τ_{aer} from models against those from AERONET must be performed very carefully, reducing the discrepancy between their model and AERONET derived aerosol absorption τ_{aer} from a factor of 4 to a factor of 1.2 by careful co-sampling of AERONET and model data. As mentioned above, uncertainty in the vertical position relative of absorbing aerosol relative to clouds can lead to large uncertainty in the top of the atmosphere aerosol RF.

The partly absorbing nature of the aerosol is responsible for a heating of the lower tropospheric column and also results in the surface forcing being considerably more negative than top-of-the atmosphere RF, results that have been confirmed through several experimental and observational studies as discussed in earlier sections. Table 2.6 summarises the surface forcing obtained in the different models. Figure 2.14 depicts the regional distribution of several important parameters when assessing the regional impact of aerosol RF. The results are based on a mean model constructed from AeroCom simulation results B and PRE. Anthropogenic τ_{aer} (Figure 2.14a) is shown to have local maxima in industrialised regions and above biomass burning dominated areas. The difference in simulated to observed τ_{aer} shows that regionally τ_{aer} can be up to 0.1 (Figure 2.14b). Figures 2.14c suggests the regions off Southern Africa where the biomass burning aerosol above clouds leads to an overall heating. Figure 2.14d shows the local diversity as standard deviation from 9 models of the overall RF. The largest uncertainties of ±3 W m^{-2} are to be found in East Asia and in the African biomass burning regions. Figure 2.14e) reveals that on the average 0.9 W m^{-2} heating can be expected in the atmospheric column as a consequence of absorption by anthropogenic aerosols. Regionally this can reach annually averaged values of more than 5 W m^{-2}. These regional effects and the negative surface forcing in the shortwave is expected to exert an important effect on climate since it alters the hydrological cycle as further detailed in Chapter 7.

An uncertainty estimate of the model derived aerosol RF can be based upon two alternative error analyses:
1) An error propagation analysis using the errors given in the chapters on sulphate, fossil fuel BC and OC, biomass burning aerosol, nitrate and anthropogenic mineral dust. Assuming linear additivity of the errors this results in an overall uncertainty of 0.3 W m-2.
2) The standard deviation of the aerosol RF results in Table 2.6 suggests a minimum uncertainty of 0.2 W m^2.

Thus the two methods for deriving the uncertainty provide roughly comparable results. However, both error estimates are suggested as being too low, given the still limited number of measurement studies and model simulations available and the harmonised emissions used in AeroCom, therefore the overall uncertainty for the aerosol RF is assessed to be 0.4 W m^{-2}.

Since TAR, a greater number of detailed global aerosol models have computed the combined direct aerosol RF. The results are summarised in Table 2.6, together with the estimates of nitrate and mineral dust RF and with the measurement based estimates provide an estimate for the combined direct aerosol RF, which is assessed to be –0.5 ± 0.4 W m^{-2}, with a low level of scientific understanding.

2.4.6 Cloud-Aerosol Interaction

As pointed out in Section 2.4.1, aerosol particles affect the formation and properties of clouds. Only a subset of the aerosol population act as cloud condensation nuclei (CCN) and/or 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). As noted in

Ramaswamy et al. (2001), enhanced aerosol concentrations can lead to an increase in the albedo of clouds under the assumption of fixed liquid water content (Junge, 1975; Twomey, 1977); this is referred to in this report as the "albedo effect". The aerosol enhancements can also lead to an increase in the lifetime of clouds (Albrecht, 1989), referred to in this report as the "lifetime" effect. The modification of the microphysical characteristics of clouds and the related changes in their optical properties are recognized as the indirect effects of aerosols on the climate system (Penner et al., 2001; Jacob et al., 2005). This terminology involves several mechanisms as presented schematically in Figure 2.12.

The interactions between aerosol particles (natural and anthropogenic in origin) 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, dust, 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 (Mc Figganns et al., 2005 and references herein). Cloud optical properties are a function of wavelength and depend on the characteristics of the droplet size distributions and ice crystal concentrations, and on the morphology of the various cloud types occurring in the climate system.

The interactions of anthropogenic enhancements in the concentrations of aerosol particles with shallow (stratocumulus and shallow cumulus) and deep convective clouds (with mixed phase) are discussed in this subsection. We present new observations and model estimates of the albedo effect and its associated radiative forcing on the climate system. In-depth discussion of the induced changes that are considered non-radiative forcings (e.g., semi-direct and cloud cover and lifetime effects, thermodynamic response, changes in precipitation development) are presented in Chapter 7. The impacts of contrails and aviation-induced cirrus are discussed in Section 2.6 and the indirect impact of aerosol on surface energy budgets and snow albedo is discussed in Section 2.5

2.4.6.1 Link between aerosol particles and cloud microphysics

The evidence concerning potential aerosol modification of clouds provided by the shiptrack observations in TAR has been further 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, utilising 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 (low values of b correspond to low hygroscopicity). Such range highlights the sensitivity to aerosol characteristics (primarily size distribution), updraught velocity, and the usage of aerosol extinction as a proxy for CCN (Feingold, 2003). Disparity in the estimates of b (or equivalent) based on satellite studies (Nakajima et al. 2001; Breon et al. 2002) also suggests that a quantitative estimate of the albedo effect from remote sensors is problematic (Rosenfeld and Feingold 2003), particularly since measurements are not considered for similar liquid water paths.

The local impact of anthropogenic aerosols has been known for a long time, e.g., smoke from sugarcane and forest fires had been 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 have led to increased cloud droplet number concentrations and to reduced cloud droplet sizes (Reid et al., 1999; Andreae et al., 2004).

Many recent studies highlight the importance of aerosol particle composition in the activation process and droplet spectral evolution (indicated in the early laboratory work of Gunn and Philips, 1957), but the picture that emerges is not completely certain. Airborne aerosol mass spectrometers provide firm evidence that ambient aerosols consist mostly of internal mixtures, e.g., biomass-burning components, organics and soot are mixed with other aerosol components (Cziczo et al., 2004a). 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). Furthermore, the varying contents of soluble and insoluble substances in internally mixed particles and their effect on cloud droplet sizes makes the situation even more complex. Earlier observations of fog water (Facchini et al., 1999; Facchini et al., 2000), had suggested that the presence of organic aerosols would reduce surface tension and lead to an increase in the cloud droplet number concentration (Nenes et al. 2002; Ming et al., 2005a; Mc Figgans et al., 2005). On the other hand, Feingold and Chuang (2002) and Shantz et al. (2003) indicate that organic coating on CCN delayed activation, leading to a broadening of the cloud droplet spectrum, which had not been previously considered. Ervens et al. (2005) address numerous composition effects in unison to show that the effect of composition on droplet number concentration is much less than suggested by studies that address individual composition effects, such as surface tension. The different relationships observed between cloud optical depth and liquid water path in clean and polluted stratocumulus clouds (Penner et al., 2004) have been explained by differences in sub-cloud aerosol particle distributions, while some contribution can be attributed to CCN composition (e.g., internally mixed insoluble dust, Asano et al., 2002). Nevertheless, the review study of cloud droplet activation by McFiggans et al. (2005) points to the remaining difficulty in explaining quantitatively the relationship between aerosol size and composition, and the resulting droplet size distribution.

The complexity of the aerosol-cloud interactions and the local atmospheric conditions in the environment where the clouds are developing are possibly responsible for the ambiguous results. Advances have been made in our understanding of the regional/global impact based on observational studies, particularly for low-level stratiform clouds that constitute a simpler cloud system to study. Observations of column aerosol number concentration and column cloud droplet concentration over the oceans from the Advanced Very High Resolution Radiometer, AVHRR, (Nakajima et al., 2001) indicate a positive correlation and an increase in shortwave reflectance of low-level, warm clouds with increasing cloud optical thickness, while the LWP showed insignificant changes. Note that, while these results are only applicable over the oceans and are based on data for only four months, the positive correlation between an increase in cloud reflectance and an enhanced ambient aerosol concentrations has been confirmed by other studies (Brenguier et al., 2000a; Brenguier et al., 2000b; Rosenfeld et al., 2002). Nevertheless, other studies highlight the sensitivity to the liquid water path (LWP), linking high pollution entrained into clouds to a decrease in LWP and a reduction in the observed cloud reflectance (Jiang et al., 2002; Brenguier et al., 2003). While still others (Han et al., 2002) have reported an absence of LWP changes in response to increases of the column-averaged droplet number concentration (N_c) for clouds with optical depths between 1 and 15 from AVHRR observations in 1/3 of the cases studied. Results of large eddy simulations of stratocumulus clouds (Jiang et al., 2002; Ackerman et al., 2004; Lu and Seinfeld, 2005) and of cumulus clouds (Xue and Feingold 2005; Jiang and Feingold, 2005), seem to confirm the lack of increases in LWP due to increases in aerosols. These studies highlight the difficulty of devising observational studies that can isolate the albedo effect from other effects related to changing meteorology and cloud dynamics that affect LWP and therefore cloud RF, even for relatively simple cloud systems.

The global scale impact of fine aerosol on cloud properties is indicated by a recent study, that includes data obtained from the POLarization and Directionality of the Earth's Reflectances (POLDER) instrument, which is well suited to retrieve both the aerosol loading due to submicron particles and the cloud droplet size. The results suggest much larger cloud effective radii in remote oceanic regions than in highly polluted continental areas and in their adjacent oceanic areas downwind from the continental sources, from a maximum of 14 down to 6 μm (Bréon et al., 2002), confirming earlier studies of hemispheric differences using AVHRR. Sekiguchi et al. (2003) investigated the correlations between aerosol and cloud parameters derived from AVHRR and POLDER data, concluding that the data were consistent with an aerosol indirect effect. These results suggest that the impact of aerosols on cloud microphysics is global, but note that the estimates of aerosol loading and cloud droplet size cannot be made coincident with each other, since an aerosol index is not determined in the presence of clouds, and no discussion of the LWP is presented.

Fewer number of studies exist on aerosol-cloud relationship under more complex meteorological conditions e.g., presence of different types of clouds simultaneously, which makes an assessment difficult. An indication of the albedo effect can be concluded from studies that combined satellite retrievals with a chemical transport model in the case of two pollution episodes over the mid-latitude Atlantic Ocean. Results indicate the brightening of clouds over a timescale of a few days in instances when liquid water path did not undergo any significant changes (Hashvardhan et al., 2002; Schwartz et al., 2002; Krüger and Grassl, 2002).

Even fewer studies have addressed the changes observed in mixed phase convective and cirrus clouds due to the influence of anthropogenic enhancements of aerosol concentrations. There are still several microphysical aspects not completely understood in the aerosol-crystal interactions, some of which are beginning to get addressed in laboratory studies. Even though homogeneous freezing is the likely ice formation mechanism at low temperatures, Cziczo et al. (2004b) provide laboratory results of homogenous freezing which suggest that organic compounds are not partitioned equally into the ice and aqueous phases, with organic-rich particles remaining unfrozen and potentially having an impact on mixed phase clouds developing under anthropogenic influence. Moreover, the chemical composition of the interstitial particles differs from the nuclei in ice crystals. In particular, the insoluble fraction (e.g., dust, soot) affects heterogeneous ice formation in mixed phase clouds as indicated by a combination of laboratory and modelling results (Diehl and Wurzler, 2005; Lohman and Diehl, 2006).

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 and thus the forward-to-backward scattering property. 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 the transfer in small crystals not included in the above study.

Satellite observations from HALOE (Halogen Occultation Experiment) in the lower stratosphere have shown that there is a negative statistical correlation in biomass burning regions between ice crystal effective diameter and relative humidity (Sherwood*,* 2002). The hypothesis that, enhanced aerosol concentrations associated with biomass burning would result in smaller ice crystals in the developing convective clouds and those smaller crystals would evaporate more readily, leading to an increase in relative humidity in the upper troposphere and lower stratosphere, should be further explored.

2.4.6.2 Estimates of the RF due to the albedo effect from GCMs

General circulation models are used to estimate the global mean RF associated with the cloud albedo effect of anthropogenic aerosols, considering pre-industrial and present-day times, and top of the atmosphere radiation budgets. These model estimates are based on forward calculations, considering emissions of anthropogenic primary particles and secondary particle production from anthropogenic gases, to estimate the changes in cloud reflectance. Since the TAR, the cloud albedo effect has been estimated in a more systematic and rigorous way, and more modelling results are now available. Most climate models use parameterisations to relate the cloud droplet number concentration to the aerosol concentration; these parameterisations vary in complexity from simple empirical fits to more physically-based relationships. Some models are run under an increasing greenhouse gas concentration scenario, and include estimates of present day aerosol loadings (both primary and secondary aerosol production from anthropogenic sources). These global modelling studies (Table 2.7) have limitations in part due to the underlying uncertainties in aerosol emissions (e.g., emission rates of primary particles and of secondary particle precursors). Another limitation resides in the different formulations of the relationships between aerosol particle concentrations and cloud droplet or ice crystal populations, and the resulting impact of microphysical changes on the optical properties of clouds, which makes it generally difficult to perform a meaningful comparison between the various model results. Further, even when the relationships used in different models are similar, there are noticeable differences in the spatial distributions of the simulated low-level clouds between models. Table 2.7 describes some of the details of the different models, including the aerosol species considered and the parameterised treatment of the aerosol-cloud interaction to determine the RF due to the albedo indirect effect. Some studies present results obtained a few years ago; models have undergone continued evolution since then and it is difficult to clearly identify all the changes in the three-dimensional models as they evolve in complexity. Note the differences in the treatment of the aerosol mixtures (internal vs. external, with the latter being the more frequently employed method). Case studies of droplet activation indicate a clear sensitivity to the aerosol composition; also, radiative transfer is sensitive to the aerosol composition and particularly to the insoluble fraction present in the cloud droplets.

[INSERT TABLE 2.7 HERE]

Note that the model results in the recent studies do not indicate the larger negative values presented in TAR, and none of the models show positive or zero RF associated with the albedo effect. All models indicate a negative RF, with the range of model estimates varying widely, from –0.3 to –1.8 W m^{-2}. If all models are considered equally valid and all results given the same weight, then the average RF from all models in Table

2.7 is –1.09 W m^{-2}, with 0.47 W m^{-2} as the standard deviation. A straightforward average, calculated from the estimate of each model, is given, since there are no clear indications whether one model can be considered better or worse than the others, and a judicious weight for each model cannot be determined at this time. Nevertheless, a closer inspection of the treatment of aerosol species in the models leads to a broad separation into two groups: models with only a few aerosol species versus those that include a more complex mixture of aerosols of different composition. There are also considerable differences in the treatment of aerosol processes, cloud processes and the aerosol-cloud interactions in these models. These are mentioned in order to understand some of the differences in the mean RF estimates shown. Several 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 studies by Lohmann et al. (2000) and Chuang et al. (2002) include internally mixed sulphate, black and organic carbon, sea salt and dust aerosols, resulting in the smallest estimate of the cloud albedo indirect effect. More recently, Takemura et al. (2005) use a global aerosol transport-radiation model coupled to a general circulation model, to estimate the direct and indirect effects of aerosols and their associated radiative forcing. A microphysical parameterisation diagnosing the cloud droplet number concentration based on the Köhler theory is introduced into the model, which depends not only on the aerosol particle number concentration but also on the updraft velocity, size distributions, and chemical properties of each aerosol species and saturation. The results indicate a global decrease in cloud droplet effective radius by anthropogenic aerosols, with the global mean RF at the tropopause due to the indirect effect by anthropogenic aerosols calculated to be –0.52 W m^{-2}, ; the land and oceanic contributions are –1.14 and –0.28 W m^{-2}, respectively. Other modelling results also indicate that the mean RF due to the cloud albedo 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 inter-model standard deviation (Lohmann and Feichter, 2005).

In Figure 2.16 the estimates of the RF due to the albedo effect from the different models are grouped according to the type of aerosol species included in the simulations. In the top panel of Figure 2.16, where estimates from models that mainly include anthropogenic sulphate are shown, there seems to be an indication that the results are converging, even though a range of models is presented from studies published between 2001 and 2005. These studies show much less scatter than in TAR and result in a mean and standard deviation of –1.37 ± 0.14 W m^{-2}. In contrast, in the btoom panel of Figure 2.16 where the studies that include more species are presented, a much larger variability is observed. As shown in Table 2.7, these models include the “state of the art” in the parameterisations of droplet activation concerning a variety of aerosols which includes both internal and external mixtures. Note however that the two studies with most negative RFs use the NCAR CCM model. The mean and standard deviation from these 8 estimates are (–0.84 ± 0.52 W m^{-2}), which are presented here as the best estimate for the RF due to the albedo effect. This estimate is derived from the models that represent, to the best possible extent, the complexity of the aerosol-cloud interactions, in terms of the species included, the state of the mixtures, and the process of droplet spectral broadening.

[INSERT FIGURE 2.16 HERE]

Some studies have commented about inconsistencies between some of the earlier estimates of RF due to the albedo effect from forward and inverse calculations (Anderson et al., 2003b). Nevertheless, the more recent results from forward calculations presented here have reduced estimates of the RF, appear to be within range of the estimates from inverse calculations.

2.4.6.3 Estimates of the RF due to the albedo effect from observations and constrained models

It is difficult to obtain a best estimate of the total indirect aerosol effect from pre-industrial times to present-day solely from observations. The satellite record is not long enough and other existing long-term records do not provide the pre-industrial aerosol and cloud microphysical properties needed for such an assessment. Nevertheless, some studies have attempted to estimate the RF due to the aerosol indirect effect utilising satellite observations. Furthermore, some studies have incorporated the empirical relationships derived from satellite observations to estimate the RF due to the albedo effect in climate simulations.

Nakajima et al (2001) retrieve aerosol optical thickness and, by assuming a bimodal log-normal size distribution, determine the Angstrom exponent from AVHRR data over the oceans (for a period of four months), together with cloud properties, optical thickness and effective radii. The non-linear relationship obtained is consistent with Twomey's hypothesis and formula; however, when plotted on a log-log graph, the slope obtained is smaller than previous estimates (0.5 versus 0.7–0.8; Kaufman et al, 1991), but larger than the 0.26 value obtained by Martin et al (1994). Using this relationship, they provide an estimate of the RF in the range between –0.7 and –1.7 W m^{-2}, with a global average of –1.3 W m^{-2}. Lohmann and Lesins (2002) used POLDER data to estimate aerosol index and cloud droplet radius; they then scaled the results of the simulations with ECHAM4, with and without aerosol indirect effects. The results show that the simulated clouds are more susceptible than the observations, particularly over land, leading to an overestimate of the indirect effect. The scaled values using the constraint from POLDER lead to a globally-averaged total indirect effect of –0.85 W m^{-2}, an almost 40% reduction from their previous estimate. It should be noted that indirect effects were noted in the pre-industrial simulations for clouds over land and ocean, which is not related to anthropogenic activities. The study of Sekiguchi et al. (2003) presents results from the analysis of AVHRR data over the oceans and of POLDER data both over land and ocean. Assuming that the aerosol column number concentration increased by 30% from the pre-industrial era, they estimate the aerosol indirect effect as the difference between the cloud radiative forcing in the present condition and that in the pre-industrial condition. They estimate a global mean forcing due to the total aerosol indirect effect to be between –0.6 and –1.2 W m^{-2}, somewhat lower than the estimate that Nakajima et al. (2001) obtained over the ocean. When the assumption is made that the liquid water content is constant, the RF due to the cloud albedo effect is estimated from AVHRR data to be –0.64 ± 0.16 W m^{-2}. The equivalent estimate when using POLDER data is –0.37 ± 0.09 W m^{-2}. The results from these two studies are very sensitive to the magnitude of the increase in the aerosol concentration from pre-industrial to current conditions, and the spatial distributions; these factors constitute a large uncertainty in the estimates.

Data from the POLDER and MODIS instruments were used by Quaas and Boucher (2005) to evaluate the relationship between cloud properties and aerosol concentrations on a global scale in order to incorporate it in a general circulation model (GCM). In particular, they derive relationships, which show a decreasing effective radius as the aerosol optical depth increases. They estimate relationships that correspond to marine stratiform clouds as well as convective clouds over land. All these retrievals involve a variety of assumptions that introduce uncertainties in the relationships, in particular, the fact that the retrievals for aerosol and cloud properties are not coincident and the assumption that the aerosol optical depth can be linked to the subcloud aerosol concentration. When these empirical parameterisations are included in a climate model, the simulated RF due to the cloud albedo effect is reduced by 50% from their baseline simulation. Quaas et al (2005) also utilise satellite data to establish a relationship between cloud droplet number concentration and fine mode aerosol optical depth, minimising the dependence on cloud liquid water content. This relationship is implemented in the ECHAM4 and LMDZ climate models and the results indicate that the original parameterisations used in both models overestimated the strength of the aerosol indirect effect. Even though both models show a consistent decrease of the RF, it should be noted that the original estimates of their respective RFs are very different (by almost a factor of two) and the amount of the reduction was 37 % in LMDZ and 81% in ECHAM4. The two models have highly different spatial distributions of low clouds, and also differ in the simulated aerosol concentrations and the anthropogenic fractions. These estimates would tend to suggest that the current best estimate of the RF due to only the cloud albedo effect from the GCMs presented in Figure 2.16 could be too negative, and suggest that the albedo effect is in general likely to be less than the inference from the simulations in Section 2.4.6.2, but further studies are currently needed to confirm the results from the ECHAM4 and LMDZ models and before a general conclusion can be drawn.

When only sulphate aerosols are considered, Dufresne et al (2005) obtain a much reduced RF due to the albedo effect. Their model uses a relationship between aerosol mass concentration and cloud droplet number concentration modified from that originally proposed by Lohmann and Boucher (1995) and adjusted to POLDER data to avoid unrealistic droplet concentrations in the simulations. Their simulations give a factor of two reduction in the RF from the previous parameterisation, but the results are highly sensitive to the distribution of clouds over land, which in turn depend on the land surface model. Further investigations, such as the one performed by Dufresne et al. (2005), need to be conducted by other model to confirm the reduced RF estimate.

2.4.6.4 Uncertainties in satellite estimates

The improvements in the retrievals and the instrumentation available in satellites have provided valuable data to begin observational-motivated assessments of the global impact of the effect of aerosols on cloud properties. It should be mentioned first that satellite measurements cannot distinguish unambiguously natural from anthropogenic aerosols. Nevertheless, an obvious advantage of the satellite data is their global coverage, and the large number of datapoints that can be analysed to determine the relationships between aerosol and cloud properties. The spatial resolution of some datasets is comparable to that used in the general circulation models, and some studies (Sekiguchi et al., 2003; Quaas et al., 2004) suggest that the magnitude of the aerosol indirect effect may be to some degree resolution-dependent. The rather low spatial and temporal resolution of some of the satellite datasets could mask, in a single measurement, aerosol with different properties. This, together with the absence of liquid water path measurements in several instances, handicaps the inferences from such studies, and hinders an accurate analysis and estimate of the cloud albedo RF from low-level warm clouds.

Retrievals involve a variety of assumptions that introduce uncertainties in the relationships. In particular, the retrievals for aerosol and cloud properties are not coincident and the assumption is made that the aerosol optical depth can be linked to the subcloud aerosol concentration. Furthermore, a satellite system such as POLDER may underestimate the mean cloud top droplet radius due to uncertainties in the sampling of clouds (Rosenfeld and Feinglod, 2003). Further, the retrieval of the aerosol index over land may be less reliable and lead to an underestimate of the indirect effect over land. There is an indication of a systematic bias between MODIS-derived cloud droplet radius and that derived from POLDER that has not been fully resolved (Breon and Doutriaux-Boucher, 2005); there are also differences in the aerosol optical depth retrieved from those instruments (Myhre et al., 2004)

2.4.6.5 Uncertainties in RF due to model biases

One of the large sources of uncertainties is the poor knowledge of the amount and distribution of anthropogenic aerosols that are used in the model simulations, as well as aerosols in the pre-industrial time. Some studies show a large sensitivity in the RF to the ratio of pre-industrial to present day aerosol number concentrations.

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 relating particle production in terms of wind speed. Some models include anthropogenic nitrate, black carbon and organic compounds, which in turn affect activation. Even without considering the existing biases in the model-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 its distinct hygroscopic and absorption properties can be particularly important for the indirect effect in the tropics (Ming et al., 2006b).

Modelling the cloud albedo indirect effect from first principles has proven difficult because the representation of aerosol-cloud interactions and of clouds themselves in climate models are still crude (Lohmann and Feichter, 2005). Clouds often do not cover a complete grid box and are inhomogeneous in terms of droplet concentration, effective radii and LWP, which introduces added complications in the microphysical and radiative transfer calculations. Model intercomparisons (e.g., Lohmann et al., 2001; Menon et al., 2003) suggest that the predicted cloud distributions vary significantly between models, particularly their horizontal and vertical extents, since the vertical resolution and parameterisation of convective and stratiform clouds can be very different between models (Chen and Penner, 2005). Even high resolution models have difficulty in accurately estimating the amount of cloud liquid water and ice water content in a grid box.

Even though the spread in the magnitude of the RF due to the cloud albedo indirect effect has been reduced substantially since the TAR, the current level of scientific understanding is still very low. It is still difficult to compare directly the results from the different models, as uncertainties are not well identified and understood. Uncertainties quoted here may be underestimates, as all models could be suffering from similar biases, and modeling studies do not often quote the statistical significance of the RF estimates that are presented. Ming et al. (2006b), 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 that are most susceptible to the changes due to anthropogenic aerosol particles. A fundamental problem is that GCMs do not resolve the small scales (order of 100s of metres) at which aerosol-cloud interactions occur. Chemical composition and size distribution spectrum are also two factors that likely are insufficiently understood on a fundamental microphysical level, although some modeling studies suggest that the albedo effect is more sensitive to the size of aerosols than to aerosol composition (Feingold, 2003). 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 is a critical parameterisation element in the climate models. It is treated in different ways in different models, ranging from simple empirical functions (Menon et al., 2002b), to more complex physical parameterisations that also tend to be more costly computationally (Abdul-Razzak and Ghan*,* 2002; Nenes and Seinfeld*,* 2003; Ming et al., 2006a). And finally, comparisons with observations have not reached the same degree of testing as, for example for the direct RF estimates; this is not just due to model limitations, for the observational basis and inferences also have not yet reached a sound footing.

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 width of the size distribution 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 that depends on the shape of the size distribution. The studies confirm that their estimates of the RF due to the cloud albedo effect, without taking the droplet spectra change into account, were overestimated by 15% and 15–35%, respectively.

The effects of aerosol particles on heterogeneous ice formation are currently insufficiently understood and present another challenge for both observations and modelling. Ice crystal concentrations cannot be easily measured with present equipment because of the difficulty of detecting small particles (Hirst et al., 2001) and the fact that ice particles often shatter when hitting the probes (Korolev and Isaac, 2005). Current GCM's do not have sufficient rigor of the microphysics or subgrid scale processes to accurately predict cirrus clouds or supercooled clouds explicitly in the models. Ice particles in clouds are often represented in models by simple shapes, or even spheres, even though it is well known that few ice crystals are like that in reality. The radiative properties of ice particles in GCMs often do not effectively simulate the normally found irregular shapes, nor the inclusions of crustal or soot in the crystals, thus introducing uncertainties in the estimates.

2.4.6.6 Combined satellite and modeling estimates

Since the TAR, the cloud albedo effect has been estimated in a more systematic and rigorous way, and more modelling results are now available. Even though large uncertainties still remain in the estimate of the RF due to the albedo effect, clear progress has been made leading towards a convergence of the estimates from the different models. No model now gives values as low as reported in TAR and none yields a value that in magnitude is larger than about -1.8 W m^{-2}.

It is possible now to present a best estimate for the RF of (-0.9 ± 0.43) W m^{-2} as mean and standard deviation, based on the results from the 8 modelling studies shown in Figure 2.16: Bottom. These models represent the complexity of the aerosol-cloud interactions, in terms of the species included, the state of the mixtures, etc., to the best current knowledge in forward calculations. The few recent studies that use satellite observations to constrain the model performance may be providing an upper limit for the value of the RF due to the albedo effect. Further studies are currently needed to confirm the results from the ECHAM4 and LMDZ models and before a general conclusion can be drawn, but the methodology represents a step forward in the estimation of the RF.

Even though the spread in the magnitude of the RF due to the cloud albedo indirect effect has been reduced substantially since the TAR, the current level of scientific understanding remains very low. It is still difficult to compare directly the results from the different models, as uncertainties are not well identified and understood.

2.5 Anthropogenic Changes in Surface Albedo and the Surface Energy Budget

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 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 W m^{-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 (see Section 2.8).

Land cover change can also affect other physical properties such as surface emissivity, the fluxes of moisture through evaporation and transpiration, 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. These processes are discussed in detail in Chapter 7. 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 a similar term (see Section 2.2), 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, the mechanisms are not well identified and so is here referred to as a non initial radiative forcing. 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, 7.9–9.2 million km^2 (6%–7% of the global land surface) were under cultivation or pasture (Figure 2.17), 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 2.17). In the U.S.A., 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, cropland 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. By 1990, croplands and pasture covered 45.7–51.3 million km^2 (35%–39% of global land), and forest cover had decreased by 11.3–12.9 million km^2 (Ramankutty and Foley, 1999; Klein Goldewijk, 2001) (Table 2.8).

[INSERT FIGURE 2.17 HERE]

Overall, most deforestation until the mid-20th Century had occurred in the temperate regions (Figure 2.17). 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. In the 1990s compared to the 1980s, net removal of tropical forest cover had slowed in the Americas but increased in Africa and Asia.

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 (Table 2.8). Matthews et al. (2003) and Brovkin et al. (2006) 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. (2005) also considered only cropland changes (Ramankutty and Foley, 1999) and simulated the forcing relative to 1750 to be –0.15 W m^{-2}. Using historical reconstructions of both croplands (Ramankutty and Foley, 1999) and pasturelands (Klein Goldewijk, 2001), Betts et al. (2006) simulated a RF of –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 (Table 2.8). Govindasamy et al. (2001a) 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; Schaaf et al., 2002) (Figure 2.18) and estimated the RF as –0.09 W m^{-2}. The results of Betts et al. (2006) and Brovkin et al. (2006) 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 (Table 2.8).

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. The local RF depends on local albedo changes, which depend on the nature of the PNV replaced by agriculture (see Biome map, top panel of Figure 2.17). 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 the 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) Uncertainties in the mapping and characterisation of present-day vegetation. 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 basis 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) (Table 2.8). The Moderate Resolution Imaging Spectrometer (MODIS – Friedl et al., 2002) and Global Land Cover 2000 (Bartholome and Belward, 2005) provide other products. 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) Uncertainties in the mapping and characterisation of the reference historical state. 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 0.5° resolution from 1700 to 1990 (Figure 2.17, Table 2.8) 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 (Figure 2.17, Table 2.8), 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 0.5° 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 (Figure 2.17) 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.

[INSERT TABLE 2.8 HERE]

(iii) Uncertainties in the parameterizations of the surface radiation processes. 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 (Schaaf et al., 2002; Gao et al., 2005) instruments have been used to quantify surface albedo for the IGBP vegetation classes in different regions and different seasons (Figure 2.18), 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 by 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.

[INSERT FIGURE 2.18 HERE]

(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 ±0.3 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 and 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 due to uncertainties in; (i) whether BC and snow particles are internally or externally mixed, (ii) BC and snow particle shapes and sizes, (iii) voids within BC particles, and (iv) uncertainties in the BC imaginary refractive index. The overall uncertainty 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 surface 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 CO_2 RF i.e., the 'efficacy' may be higher for this RF mechanism (see Section 2.8.5.7). This report adopts a best estimate of +0.10 ± 0.30 W m^{-2} , giving the Hansen et al. (2005) estimate greater weight due to closer ties to observations and more complete treatment of processes.

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. Impacts of land cover change on emissions of CO_2, CH4, biomass burning aerosols and dust aerosols are discussed in Sections 2.3 and 2.4. Land cover change itself can also modify the surface energy budget through changes in the fluxes of latent and sensible heat. 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 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 change 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 (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 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 1%, resulting in an RF of 0.03 W m^{-2}. The effect of irrigation on surface temperature was dominated by evaporative cooling rather than by the excess greenhouse effect and thus a decrease in surface temperature was found. Since irrigation impacts the temperature, humidity, clouds, and precipitation as well as the natural evaporation through change in the surface temperature this questions the strict use of RF. 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). Most of this uncertainty is likely to be linked to differences in what is actually used for irrigation purposes and the total withdrawal for irrigation (see further discussions in Boucher et al., 2004). Further, the Gordon et al. (2005) study also 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. Water vapour changes from deforestation will in a similar way as irrigation impact the surface evaporation and temperature as well as impact the water cycle in the atmosphere. We thus refrain giving a best estimate of RF from anthropogenic sources of tropospheric water vapour since these impact climate feedbacks and keeping them constant in the troposphere is impossible in realistic simulations. The emission of water vapour from fossil fuel combustion is significantly lower than the emission from changes in land use.

2.5.7 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 use of energy by human activities such as the 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).

2.5.8 Effects of CO_2 Changes on Climate via Plant Physiology: "Physiological Forcing"

As well as exerting a RF on the climate system, increasing concentration of atmospheric CO_2 also perturb the climate system through direct effects on plant physiology. Plant stomatal apertures 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). 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; Cox et al., 1999). The details of this process are discussed further in Chapter 7. Sellers et al. (1996) propose the term "physiological forcing" for this mechanism. Although no studies have yet explicitly quantified the temperature response to physiological forcing at the present-day, the presence of this forcing has been detected in global hydrological budgets (Gedney et al., 2006). This process can be considered a non-radiative forcing of climate change, as distinct from a feedback, since the mechanism involves a direct response to CO_2 rise and not a response to climate change. It is not possible to quantify this with RF; further research on appropriate metrics to compare non-radiative forcings with RF is required.

Reduced global transpiration would also be expected to reduce atmospheric water vapour causing a negative RF, but no estimates of this have been made.

Increased CO_2 concentrations also fertilize photosynthesis, which models suggest will have contributed to increased vegetation cover and leaf area over the 20th Century (Cramer et al., 2001). Leaf area has been observed to increase (Zhou et al., 2001) although other causes including climate change itself are also likely to have contributed. Increased leaf area would decrease surface albedo, which would act to oppose the increase in albedo due to deforestation. The RF due to this process has not been evaluated.

2.6 Contrails and Aircraft-Induced Cloudiness

2.6.1 Introduction

The IPCC separately evaluated the RF of climate by subsonic and supersonic aircraft in the Special Report on Aviation and the Global Atmosphere (IPCC, 1999), hereinafter 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), their impact on cirrus cloudiness, and the effects of aviation aerosols. 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 upper troposphere will have consequences for aviation-induced 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 persistent contrails in the upper troposphere in ice-supersaturated air masses (IPCC, 1999). 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; Meerkötter et al., 1999), thereby resulting in a net positive RF value for contrails. Persistent contrail cover is calculated globally from meteorological 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 determining an optical depth for the computed contrail cover. The global RF values for contrail and induced cloudiness are assumed to vary linearly with global fuel use if aircraft flight regions remain unchanged. The best estimate for the RF of persistent linear contrails for aircraft operations in 2000 is 0.010 W m^{-2} (Table 2.9; Sausen et al., 2005). The value is based on independent estimates derived from Myhre and Stordal (2001) (0.015 W m^{-2}) and Marquart et al. (2003) (0.006 W m^{-2}). The two values also serve to set the uncertainty range of a factor of 2. 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 and lower optical depth estimates (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). The level of scientific understanding of contrail radiative forcing is considered low, since important concerns remain in determination of global values. For example, regional differences found in contrail optical depths between Europe and the United States have not been fully accounted for in model calculations of contrail RF (Palikonda et al., 2005; Meyer et al., 2002; Ponater et al., 2002).

[INSERT TABLE 2.9 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. Because spreading contrails lose their characteristic linear shape, a component of AIC 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 also found significant positive trends in cirrus cloudiness in some 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. A study with the TOVS satellite provides further support for these trends (Stubenrauch and Schumann, 2005). However, cirrus trends that occurred due to natural variability, climate change, or other anthropogenic effects could not be accounted for in these studies. Cirrus trends over the United States but not over Europe were found to be consistent with changes in contrail cover and frequency (Minnis et al., 2004). Thus, significant uncertainty remains in attributing observed cirrus trends to aviation.

Regional cirrus trends were used as a basis to compute a global mean RF value for AIC in 2000 of 0.030 W m^{-2} with a range of 0.01–0.08 W m^{-2} (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 the upper limit estimate for AIC RF in 1992 of 0.026 W m^{-2} derived from surface and satellite cloudiness observations (Minnis et al., 2004). A value of 0.03 W m^{-2} is close to the upper limit estimate of 0.04 W m^{-2} derived for non-contrail cloudiness in IPCC-1999. Without an AIC best estimate, the best estimate of the total RF value for aviation-induced cloudiness (Table 2.12 and Figure 2.24) includes only that due to persistent linear contrails.

Minnis et al. (2004) use their RF estimate for total aviation cloudiness over the USA in an empirical model to conclude 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). In response to the Minnis et al. conclusion, contrail forcing was examined in two global climate modelling studies (Hansen et al., 2005; Ponater et al., 2005). Both studies concluded that the surface temperature response calculated by Minnis et al. (2004) is too large by one to two orders of magnitude. For the Minnis et al. result to be correct, the climate efficacy or climate sensitivity of contrail forcing would need to be much greater than that of other larger forcing terms, e.g., CO_2. Instead, contrail forcing is found to have a smaller efficacy than equivalent CO_2 forcings (Hansen et al., 2005; Ponater et al., 2005), which is consistent with the general ineffectiveness of high clouds in influencing diurnal surface temperatures (Hansen et al., 1995; 2005). Several substantive explanations for the enhanced response found in the Minnis et al. study have been presented (Hansen et al., 2005; Ponater et al., 2005; Shine, 2005). In reply, Minnis (2005) highlights the uncertainty in evaluating the regional response to regional forcings in GCMs.

Aviation-induced cloudiness has been proposed as the cause of the increased diurnal temperature range (DTR) found in surface observations made during the period when all USA air traffic was grounded 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 weak because they are based on a correlation rather than a quantitative model and rely (necessarily) on very limited data (Schumann, 2005). 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 quantitative physical model are needed before the proposed relationship between regional contrail cover and DTR can be considered valid.

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 above background values in these regions. AIC includes the possible influence 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). An important 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. In the study, aviation aerosols either increase or decrease ice nuclei in background cirrus clouds, depending on assumptions about the cloud formation process. Results from a cloud chamber experiment showed that a sulphate coating on soot particles altered their nucleation properties (Möhler et al., 2005). Changes in ice nuclei number or nucleation properties of aerosols 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 Sections 2.4.6 and Chapter 7, Section 7.5. 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 (see Section 2.6.3), may be associated with these aerosol effects.

2.7 Natural Forcings

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) (intermittently) since 1984. Most recent are the measurements made by the Total Solar Irradiance Monitor (TIM) on the Solar Radiation and Climate Experiment (SORCE) since 2003 (Rottman, 2005).

2.7.1.1.2 Observed decadal trends and variability

Different composite records of total solar irradiance have been constructed from different combinations of the direct radiometric measurements. The Physikalisch-Meteorologisches Observatorium Davos (PMOD) composite (Fröhlich and Lean, 2004), shown in Figure 2.19, 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), also shown in Figure 2.19, utilizes ACRIMSAT rather than VIRGO observations in recent times and cross calibrates the reported data assuming that radiometric sensitivity drifts have already been fully accounted for. A third composite, 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 very similar, each showing 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 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 (Fröhlich and Lean, 2004).

[INSERT FIGURE 2.19 HERE]

Solar irradiance levels are comparable in the two most recent cycle minima when absolute uncertainties and sensitivity drifts in the measurements are assessed (Fröhlich and Lean, 2004). The purported upward "secular" trend in excess of 0.04% over the 27-year period of the ACRIM irradiance composite (Willson and Mordvinov, 2003) is thought to be of instrumental rather than solar origin. As Figure 2.19 shows, the irradiance "trend" in the ACRIM composite is not a slow secular increase but rather a single episodic increase between 1989 and 1992 that is present in the Nimbus 7 data. Independent, overlapping ERBS observations do not show this increase; nor do they have a significant secular trend (Lee et al., 1995). Nor is such a trend present in the PMOD composite, in which total irradiance between successive solar minima is constant to better than 0.01%. Although a long-term trend of order 0.01% is present in the SARR composite between successive solar activity minima (in 1986 and 1996) it is not significant because the uncertainty is ±0.026% (Dewitte et al., 2005).

Current understanding of solar activity and of the known sources of irradiance variability also suggests comparable irradiance levels during the past two solar minima, and does not support an upward trend in irradiance in the past 25 years. The primary known sources of irradiance variability are sunspots (compact, dark features on the solar disk where radiation is locally depleted) and faculae (extended bright features on the solar disk where radiation is locally enhanced). Models that combine records of the global sunspot darkening calculated directly from white light images and the Mg irradiance index as a proxy for the facular signal do not exhibit a significant secular trend during activity minima. While changes in surface emissivity by magnetic sunspot and facular regions are, from a theoretical view, the most effective in altering irradiance (Spruit, 2000), other mechanisms have also been proposed that may cause additional, possible secular, irradiance changes. 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 Michelson Doppler Imager (MDI) instrument on SOHO, indicates 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, solar time series such as the irradiance composites in Figure 2.19 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 analyses 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 globally 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 in two mid-latitude bands that extend vertically from the lower stratosphere to the surface. Increases of up to 0.5K are observed at the surface near 40–50 N and S (Haigh, 2003). The associated response in zonal wind reveals that the sub-tropical jets are weaker and further poleward when the sun is more active (Haigh et al, 2005). 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 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. Over the time scale of the 11-year solar cycle, bolometric facular brightness exceeds sunspot blocking by about a factor of two, and there is an increase in spectral irradiance at most, if not all, wavelengths from the minimum to the maximum of the solar cycle. Estimated solar cycle irradiance changes (ratio of 1989 to 1986), shown in Figure 2.20, 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. However, during episodes of strong solar activity sunspot blocking can dominate facular brightening, causing decreased irradiance at most wavelengths. Spectral irradiance changes measured on these shorter time scales provide tests of the wavelength-dependent sunspot and facular parameterizations in solar irradiance variability models. The modelled spectral irradiance changes are seen in Figure 2.20 to be in good agreement with initial SORCE observations during the major episode of solar activity from 17 to 30 October 2003. Distinct differences are nevertheless evident. 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). 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).

[INSERT FIGURE 2.20 HERE]

2.7.1.2 Estimating past solar radiative forcing

2.7.1.2.1 Reconstructions of past variations in solar irradiance

Long-term irradiance changes may be less over the past four hundred years than in the reconstructions employed by TAR for climate change simulations. The irradiance reconstructions of Hoyt and Schatten (1993), Lean et al. (1995), Lean (2000), Lockwood and Stamper (1999) and Fligge and Solanki (2000), used in TAR, assumed the existence of a long-term variability component in addition to the known 11-year cycle, such that during the seventeenth century Maunder Minimum total irradiance was reduced in the range of 0.15% to 0.4% 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 geomagnetic activity and cosmogenic isotopes, together with 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. 2.19). Various estimates of the reduction in total solar irradiance from current activity minima to the seventeenth century Maunder Minimum from these irradiance reconstructions are compared in Table 2.10 with recent results.

[INSERT TABLE 2.10 HERE]

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 historical indices of geomagnetic activity (Svalgaard et al., 2004), which would reduce somewhat the long-term trend in the Lockwood and Stamper (1999) irradiance reconstruction. Furthermore, the relationship between solar irradiance and geomagnetic and cosmogenic indices is complex, and not necessarily linear. Simulations of the transport of magnetic flux on the Sun and propagation of open flux into the heliosphere indicate that "open" magnetic flux (which modulates geomagnetic activity and cosmogenic isotopes) can accumulate on inter-cycle time scales even when closed flux (such as in sunspots and faculae) does not (Lean et al., 2002; Wang et al., 2005).

A new reconstruction of solar irradiance based on solar considerations alone, that does not invoke geomagnetic, cosmogenic or stellar proxies, suggests that the amplitude of the background component is significantly less than previously assumed, specifically 0.27 times that of Lean (2000). This estimate results from 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 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 2.21. There is a 0.037% increase from the Maunder Minimum to the present-day quiet Sun.

In contrast to the Wang et al. (2005) reconstruction, that of Solanki and Krivova (2005) varies similarly to that of Lean (2000) (the lower envelope of the shaded curve in Figure 2.21) 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, which exhibit solar-related cycles near 90–, 200– and 2300 years. 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 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, when differences among isotopes records are taken into account and the ^{14}C record corrected for fossil fuel burning, current levels of solar activity are found to be historically high, but not exceptionally so (Muscheler et al., 2005).

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 0.037% (an irradiance increase of 0.5 W m^{-2} in ~1365 W m^{-2}) although in some estimates it is as large as 0.12% (see Table 2.10). Accounting for the 11-year cycle, which has also increased in amplitude from the Maunder Minimum to the present, the total irradiance increase is 0.08%. From 1750 to the present there is a net 0.05% increase in total solar irradiance, according to the 11-year smoothed total solar irradiance time series of Wang et al. (2005), shown in Figure 2.21. This corresponds to a RF of 0.12 W m^{-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, there is a 0.12% irradiance increase since 1750, for which the RF is 0.3 W m^{-2}. The lower limit of the irradiance increase from 1750 to the present is 0.026% 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 2.21 HERE]

2.7.1.3 Indirect effects of solar variability

Approximately 1% of the Sun's radiant energy is in the ultraviolet portion of the spectrum at wavelengths below ~300 nm, which the Earth's atmosphere absorbs. Although of considerably smaller absolute energy than the total irradiance, solar UV radiation is fractionally more variable 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), creates and modifies the ozone layer, but is not considered as a direct climate forcing because it does not reach the troposphere. Since TAR, new studies have confirmed and advanced the plausibility of indirect effects involving the modification of the stratosphere by solar UV irradiance variations (and possibly by solar-induced variations in the overlying mesosphere and lower thermosphere), with subsequent dynamical and radiative coupling to the troposphere. Whether solar wind fluctuations (Boberg and Lundstedt, 2002) or solar-induced heliospheric modulation of galactic cosmic rays (Marsh and Svensmark, 2000b) also contribute indirect forcings 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 lower stratospheric response (Hood, 2003), which may have a dynamical origin caused by changes in temperature affecting planetary wave propagation, but it is not currently reproduced by models.

Increased stratospheric ozone and temperature during solar cycle maxima alters both incoming solar radiation and outgoing IR radiation, producing latitudinal and altitudinal 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 manifest in, for example, tropical rainfall (Kodera*,* 2004) and Atlantic storm tracks (Haigh*,* 2001; Haigh et al., 2004). Both analysis and modelling suggest that solar variability influences annular-like patterns of wintertime climate anomalies (Annular Modes) that extend from sea level to the top of the stratosphere (Ruzmaikin and Feynman, 2002; Shindell et al., 2003). Equatorial winds (QBO) 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; Gray et al., 2004; Ruzmaiken et al., 2005). 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 (Gray et al., 2005).

When solar activity is high, the more complex magnetic configuration of the heliosphere reduces the flux of galactic cosmic rays in the Earth's atmosphere. Various scenarios have been proposed whereby solar-induced galactic cosmic ray fluctuations might influence climate (as surveyed by Gray et al., 2005). Carslaw et al. (2002) suggest that 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 affect thunderstorm electrification. By altering the population of cloud condensation nuclei and hence microphysical cloud properties (droplet number and concentration) cosmic rays may also induce processes analogous to the indirect effect of tropospheric aerosols. The presence of ions, such as produced 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. 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 galactic cosmic ray-induced changes in aerosol and cloud formation have not been reached.

Empirical associations have been reported between globally averaged low-level cloud cover and cosmic ray fluxes during 1984–1990 (Marsh and Svensmark, 2000a, 2000b). Hypothesized to result from changing ionization of the atmosphere from solar-modulated cosmic ray fluxes, this empirical association of cloud cover variations and the solar cycle remains controversial because of uncertainties about 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. In particular, the cosmic ray time series does not correspond to global total cloud cover after 1991 or to global low-level cloud cover after 1994 (Kristjánsson and Kristiansen, 2000; Sun and Bradley, 2002). Furthermore, the correlation is significant with low-level cloud cover based only on infrared (not visible) detection. Nor do multidecadal (1952–1997) time series of cloud cover from ship synoptic reports exhibit a relationship to cosmic ray flux. But cloud cover anomalies from 1900–1987 over the United States do have a signal at 11-years that is in phase (rather than anit-phased, as the GCRs are) with solar variability (Udelhofen and Cess, 2001). In this case the cloud variations are hypothesized to result from modulation of the atmospheric circulation by variations of the solar-UV-ozone-induced heating of the atmosphere.

An unequivocal determination of specific mechanisms – whether direct or indirect - that involve solar variability and climate has yet to be accomplished. Alternate explanations are often proffered for common empirical evidence. For example, the apparent relationship between solar variability and cloud cover has been interpreted to result not only from changing cosmic ray fluxes modulated by solar activity in the heliosphere (Usoskin et al., 2004) and solar induced changes in ozone (Udelhofen and Cess, 2001), but also as a result of sea surface temperatures altered directly by changing total solar irradiance (Kristjánsson et al., 2002) and internal variability by ENSO (Kernthaler et al., 1999). In reality, different direct and indirect physical processes may operate simultaneously.

2.7.2 Explosive Volcanic Activity

2.7.2.1 Radiative effects of volcanic aerosols

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 (Bluth et al., 1992; Read et al., 1993). The e-folding time (by mass) for sedimentation of sulfate aerosols is about 12-14 months as observed for Pinatubo aerosol cloud (Lambert et al., 1993; Baran and Foot, 1994; Bluth et al., 1997; Barns and Hoffman, 1997). Also emitted directly during an eruption are volcanic ash particulates (siliceous material). These are particles usually larger than 2 μm that sediment out of the stratosphere fairly rapidly due to gravity (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. The stratospheric aerosol data incorporated for climate change simulations and analyses tends to be mostly that of the sulphates (Sato et al., 1993; Stenchikov et al., 1998; Ramachandran et al., 2000; Hansen et al., 2002; Ammann et al., 2003). As noted in SAR and TAR, explosive volcanic events are episodic, but the stratospheric aerosols resulting from them yield substantial transitory perturbations to the radiative energy balance of the planet, with both solar and longwave radiative effects sensitive to the microphysical characteristics of the aerosols (e.g., size distribution).

Long-term ground-based and balloonborne instrumental observations have been invaluable for understanding the optical effects and microphysical evolution of volcanic aerosols (Deshler et al., 2003; Hofmann et al., 2003). Important ground-based observations of aerosol characteristics from pre-satellite era spectral extinction measurements have been analyzed by Stothers (2001a,b), unfortunately they do not provide a global coverage. Global observations of stratospheric aerosol over the last 25 years has been possible owing to a number of satellite platforms e.g., TOMS and TOVS have been used to estimates of SO2 loadings from volcanic eruptions (Krueger et al., 2000; Prata et al., 2003). The Stratospheric Aerosol and Gas Experiment (SAGE) and Stratospheric Aerosol Measurement (SAM) projects (e.g., McCormick et al., 1987) have provided vertically resolved stratospheric aerosol spectral extinction data for over 20 years, the longest such record. However, this dataset too has limitations e.g., a significant gap in coverage at the time of the El Chichón eruption in 1982 (the second most important in the 20th century after Mt. Pinatubo), and inability to do remote sensing when the aerosol cloud is dense; then, lidar and field campaigns are useful in partially filling the gap (e.g., Antuña et al., 2003).

Volcanic aerosols transported in the atmosphere to polar regions are preserved in the ice sheets, thus recording the history of the Earth's volcanism for thousands of years (Yalcin et al., 2002; Mosley-Thompson et al., 2003). An improved statistical technique for "volcanic peak recognition" in the ice cores was proposed by Naveau et al. (2003) to identify the timing and magnitude of the past eruptions. A new, partly reconstructed and partly hypothesized, climate forcing time series for 500 year that includes greenhouse gas and volcanic effects was developed by Robertson et al. (2001). However, the atmospheric loadings calculated using volcanic time series from high-latitude ice records suffer from uncertainties due to poor quantitative knowledge of transport and deposition mechanisms.

The best documented explosive volcanic event to-date, by way of reliable and accurate observations, is the 1991 eruption of Mt. Pinatubo. The growth and decay of aerosols resulting from this eruption have provided the basis for modeling the radiative forcing due to explosive volcanoes. There have been no explosive and climatically-relevant volcanic events since Pinatubo. As pointed out in TAR, stratospheric aerosol concentrations are now at the lowest concentrations since the satellite era and global coverage began in late 1970s. Altitude-dependent stratospheric optical observations at a few wavelengths, together with columnar optical and physical measurements, have been used to construct the time-dependent global field of stratospheric aerosol size distribution formed in the aftermath of volcanic events. 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 plus longwave) perturbations due to stratospheric aerosol variations.

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 wavelengths and column-average effective radii. Stenchikov et al. (2006) introduced a slight variation to this dataset employing UARS observations to modify the effective radii relative to Hansen et al. (2002), which accounts for variations with altitude. Ammann et al. (2003) have developed a similar dataset 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. Ammann et al. (2003) used a fixed effective radius of 0.42 μm for calculating aerosol optical properties. Ammann et al (1993) also have a different aerosol spatial distribution in comparison with Sato et al. (1993); in general, they infer higher values of optical depth than Sato et al. (1993). The above datasets have essentially provided the bases for the volcanic aerosols implemented in virtually all of the climate models that have performed the 20th century climate integrations (Stenchikov et al., 2006). Figure 2.22 compares the global-average volcanic aerosol visible optical depths of Sato et al. (1993) and Ammann et al. (2003). The Ammann et al. (2003) estimate yields a larger value of the optical depth, 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.

[INSERT FIGURE 2.22 HERE]

The global-mean RF calculated using the Sato et al. (1993) prescription is about –3 W m^{-2} for the strong (rated in terms of emitted SO_2) 1860 and 1991 eruptions of Krakatau and Pinatubo, respectively. The value is about –2 W m^{-2} for the relatively less intense El Chichón and Agung eruptions (Hansen et al., 2002). As expected from the arguments above, Ammann's RF is roughly 20–30% larger than Sato's RF.

Not all features of the aerosols are well quantified, and extending and improving the available aerosol data sets remains an important and active area of research. This includes improved estimate of the stratospheric aerosol size distribution parameters (Bingen et al., 2004), a new approach for calculating aerosol optical characteristics using SAGE and UARS data (Bauman et al., 2003), and intercomparison of data from different satellites and combining them to fill gaps (Randall et al., 2001). While the aerosol characteristics are better constrained for the Pinatubo eruption, and to some extent for El Chichón and Agung eruptions, the reliability degrades for aerosols from explosive volcanic events further back in time as there are little, if any, observational constraints on their optical depth and size evolution.

The radiative effects due to volcanic aerosols from major eruptions are manifest in 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. 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). In the case of the Mt. Pinatubo eruption, with a global visible optical depth maximizing at about 0.15, simulations yield a large negative perturbation as mentioned above, about –3 W m^{-2} (Ramachandran et al., 2000; Hansen et al., 2002) (see also Chapter 9). This modelled estimate of reflected solar radiation by Pinatubo aerosols compares well with the ERBS observations (Minnis et al., 1993). However, the ERBS observations were for a relatively short period of time and the model-observation comparisons are likely affected by differing cloud effects in simulations and measurements. It is interesting to note that, in the Pinatubo case, the GISS models that use Sato et al. (1993) data set yield an even greater solar reflection than the NCAR model which uses the larger (Ammann et al., 2003) optical depth value (Stenchikov et al., 2006). Thus, at present, differences in model responses yield additional uncertainties to that in the basic volcanic aerosol optical properties.

2.7.2.2 Thermal, dynamic and chemistry perturbations forced by volcanic aerosols

At least four distinct mechanisms have been invoked with regards to the climate response to natural RF (volcanic and solar) activity. First, these forcings can directly affect the Earth's radiative balance and thus alter surface temperature. Second, they introduce horizontal and vertical heating gradients; these can alter the stratospheric circulation, in turn affecting the troposphere and providing a 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). Fourth, volcanic aerosols provide surfaces for heterogeneous chemistry affecting global stratospheric ozone distributions (Chipperfield et al., 2003) and perturbing other trace gases for a considerable period following an eruption. Each of the above mechanisms has its own spatial and temporal response pattern. Also, the mechanisms could 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 interact with each other.

The complexity of radiative-dynamical response forced by volcanic impacts suggests that it is important to calculate aerosol radiative effect interactively within the model rather than prescribe them (Andronova et al., 1999; Broccoli et al., 2003). Despite differences in volcanic aerosol parameters employed, models computing the aerosol radiative effects interactively yield tropical and global-mean lower stratospheric warming that are fairly consistent with each other and with observations (Yang and Schlesinger, 2002; Stenchikov et al., 2004; Ramachandran et al., 2000; Hansen et al., 2002; Ramaswamy et al., 2006a), however, there is a considerable range of responses in the polar stratosphere and troposphere. The global-mean warming of the lower stratosphere is due mainly to aerosol effects in the longwave spectrum, in contrast to the flux changes at the top-of-the-atmosphere that are essentially due to aerosol effects in the solar spectrum. The net radiative effects of volcanic aerosols on the thermal and hydrologic balance (e.g., surface temperature and moisture) has been highlighted by recent studies (Free and Angell, 2002; Jones et al., 2003; see Chapter 6; see Chapter 9 for significance of the simulated responses, and model-observation comparisons for 20th century eruptions). A mechanism closely linked to the optical depth perturbation and ensuing warming of the tropical lower stratosphere is the potential increase in the cross-tropopause water vapour flux (Joshi and Shine, 2003, see 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 have yet to be thoroughly examined.

Anomalies in the volcanic aerosol-induced global radiative heating distribution can force significant changes in atmospheric circulation. Tropical eruptions set up a transient radiative warming of the low-latitude lower stratosphere which perturbs the equator-to-pole heating gradient, with the magnitude dependent on the state of the climate system at the time of the eruption (Ramaswamy et al., 2006b). Further, the surface-troposphere system in lower latitudes experiences a reduction in radiative flux relative to the middle and polar latitudes (Robock, 2000). Since TAR, there is a better understanding of the regional effects of volcanic forcing on the high-latitude temperature and circulation (see Chapter 9). 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 (Shindell et al., 2003b, Stenchikov et al., 2002, 2004, 2006; Miller et al., 2006; Perlwitz and Graf, 2001; Perlwitz and Harnik, 2003; Rind et al., 2005). In contrast, high-latitude volcanic eruptions are not as effective in forcing stratosphere-troposphere interactions because of insufficient solar heating of volcanic cloud in winter season (Oman et al., 2005).

Stratospheric aerosols affect the chemical processes in the stratosphere by serving as surfaces for heterogeneous reactions, liberating anthropogenic chlorine and resulting in ozone depletion (Brasseur and Granier, 1992; Tie et al., 1994; Solomon et al., 1996; Chipperfield et al., 2003). Stenchikov et al. (2002) demonstrate a climate link between ozone depletion and AO response; this is a secondary radiative mechanism induced by volcanic aerosols through stratospheric chemistry. Stratospheric cooling in the polar region associated with a stronger polar vortex initiated by volcanic effects could 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 (Tabazadeh et al., 2002). The above studies indicate effects on the ozone layer in the wake of a volcanic eruption and in a stratosphere with enhanced anthropogenic halogen loading. Interactive microphysics-chemistry-climate models (Rozanov et al., 2002, 2005; Timmreck et al., 2003; Shindell et al., 2003b; Dameris et al., 2005) generally show that aerosol-induced stratospheric heating affects the dispersion of the aerosol cloud, thus affecting the spatial RF. However the models' simplified treatment of aerosol microphysics introduces biases; further, they usually overestimate the mixing at the tropopause level and intensity of meridional transport in the stratosphere (Schoeberl et al., 2003; Douglass et al., 2003). For present climate studies, it is practical to utilize simpler approaches that are reliably constrained by aerosol observations.

2.8 Utility of Radiative Forcing

The TAR and other assessments have concluded that RF is a useful tool for estimating, to a first order, the relative global climate impacts of differing climate-change mechanisms (Ramaswamy et al., 2001; Jacob et al., 2005). In particular RF can be used to estimate the relative equilibrium globally averaged surface temperature change due to different forcing agents. However, RF is not a measure of transient climate change or the role of emissions (see Sections 2.2 and 2.10). Previous GCM studies have 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 ozone changes at some altitudes 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), Ramaswamy et al. (2001) and Jacob et al. (2005) concluded that RF is the most simple and straightforward measure for the quantitative assessment of climate change mechanisms, and especially for the LLGHGs. This section discusses the several studies since TAR that have examined the relationship between RF and climate response.

2.8.1 Vertical Forcing Patterns and Surface Energy Balance Changes

The vertical structure of a forcing agent is important both for efficacy (see 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 sensible and latent heat fluxes as well as solar and longwave irradiance, therefore 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 RF and surface forcing should not be directly compared nor should the surface forcing be considered in isolation for evaluating the climate response (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 Sections 2.9.4 and 2.9.5).

2.8.2 Spatial Patterns of Radiative Forcing

Each RF agent has a unique spatial pattern (see Ramaswamy et al., 2001, Figure 6.7 for examples). When combining RF agents it not just the global-mean RF that needs to be considered, even with a net global mean RF of zero, significant regional RFs can be present and these can affect the global mean temperature response (see Section 2.8.5). RF spatial patterns also affect the pattern of climate response. However, note also 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. This 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, patterns of RF are presented as a diagnostic in Section 2.9.5.

2.8.3 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 (Jacob et al., (2005) refer to these mechanisms as non-radiative forcings, see also Section 2.2). Alternatives to the standard stratospherically-adjusted RF definition have been proposed that may help account for these processes. Since TAR several studies have employed GCMs to diagnose the zero-surface-temperature-change RF (see Figure 2.2 and Section 2.2).

These studies have used a number of different methodologies. Shine et al., (2003) fixed both land and sea surface temperatures globally and calculated a radiative energy imbalance: this technique is only feasible in GCMs with relatively simple land-surface parameterizations. Hansen et al. (2005) fixed sea-surface temperatures and calculated a RF by adding an extra term to the radiative imbalance that takes into account how much the land surface temperatures had responded. Sokolov (2006) diagnosed this RF by computing surface-only and atmospheric-only components of climate feedback separately in a slab model and then modifying the stratospherically adjusted RF by the atmospheric-only feedback component. Gregory et al. (2004) (see also Hansen et al., (2005) and Forster and Taylor (2006)) used a regression method with a globally-averaged-temperature-change ordinate to diagnose this RF: this method had the largest uncertainties. Shine et al. (2003), Hansen et al. (2005) and Sokolov (2005) all find that that the fixed-surface-temperature RF is a better predictor of the equilibrium global mean surface temperature response that the stratospherically adjusted RF. Further, it was a particularly useful diagnostic for changes in absorbing aerosol where the stratospherically adjusted RF could fail as a predictor of the surface temperature response (see Section 2.8.5.5). The difference between the zero-surface-temperature-change RF and the stratospheric-adjusted RF essentially can be interpreted as a semi-direct effect and for most mechanisms, aside from the case of absorbing aerosols, it appears small (Shine et al., 2003; Hansen et al., 2005; Sokolov, 2005). These calculations also remove problems associated with defining the tropopause in the stratospherically-adjusted RF definition (Shine et al., 2003; Hansen et al., 2005). However, stratospheric-adjusted RF has the advantage that it does not depend on relatively uncertain components of a GCMs response, such as cloud changes. For the LLGHGs the stratospheric-adjusted RF also has the advantage that it is also readily calculated in detailed off-line radiation codes. For these reasons the stratospherically adjusted RF is retained as the measure of comparison used in this chapter (see Section 2.2). However, to first order all methods are comparable and all provide useful diagnostics for understanding climate response.

2.8.4 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; Gillett et al., 2004; Matthews et al., 2004; Meehl et al., 2004). Two of these studies also examined realistic changes in many other forcing agents without finding evidence of a non-linear response (Meehl et al., 2004; Matthews et al., 2004). In all four studies even the regional changes typically added linearly. However, Meehl et al., (2003; 2004) observed that neither precipitation changes nor all regional temperature changes were linearly additive. This linear relationship also breaks down for global mean temperatures when aerosol-cloud interactions are included in GCMs (Feichter et al., 2004; see also Lohmann and Feichter, 2005; Rotstayn and Penner, 2001). Studies which include these effects modify clouds in their models producing an additional radiative imbalance. Rotstayn and Penner (2001) find that if these aerosol-cloud effects are accounted for as additional forcing terms, the inference of linearity can be restored (see Sections 2.8.3 and 2.8.5). Studies also find non-linearities for large negative RFs, where static stability changes in the upper troposphere affect the climate feedback (e.g., Hansen et al., 2005). For the magnitude and range of realistic RFs discussed it this chapter, and excluding cloud-aerosol interaction effects, there is high confidence of a linear relationship between global mean RF and global mean surface temperature 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). It is also related to the linearity of the forcing-response relationship (see Section 2.8.4). The “efficacy” terminology is new to IPCC reports. Each type of RF multiplied by its appropriate efficacy (E) would give the effective RF (E-RF) that can be directly compared to a RF from CO_2. For this effective RF the climate sensitivity parameter is independent of the mechanism, so comparing this forcing is equivalent to comparing the equilibrium global mean surface temperature change. That is, $\Delta T_s = \lambda_{CO2}(\text{E-RF})$, where λ_{CO2} is the climate sensitivity parameter for a carbon dioxide change. Preliminary studies have found that efficacy values for a number of forcing agents 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 the zero-surface-temperature change RF, which has efficacies closer to unity, may be another way of achieving similar goals (see Section 2.8.3). This section assesses the efficacy associated with stratospherically-adjusted RF, as this is the definition of RF adopted in this chapter (see Section 2.2). The findings presented in this section are from an assessment of all the studies referenced in the caption of Figure 2.23, 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 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 2.23 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 compared to 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 (Govindasamy et al., 2001b; Joshi et al., 2003; Sokolov, 2005). It is therefore difficult to draw generic conclusions.

[INSERT FIGURE 2.23 HERE]

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 2.23). Further, the most recent result with the NCAR CCM3 GCM (Govindasamy et al., 2001b) 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 solar RF from total solar irradiance changes, any indirect effects (see 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 increases. 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 and 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. 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. However, in these studies efficacies were calculated assuming that cloud-aerosol lifetime effects are an additional RF term. In the formulation of this chapter these cloud lifetime effects should be included as part of the variation in climate sensitivity/efficacy term (see Section 2.2). Using the cloud-albedo RF and cloud-lifetime effect estimates from Section 2.6, the efficacy range would be ~0.9–2.5.

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; Penner 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 (see Chapter 7, Section 7.5; 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 black carbon 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 Chapter 7, Section 7.5). 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 effect (Cook and Highwood, 2004) but its magnitude is very uncertain (see Chapter 7, Section 7.5) and dependant on aspects of cloud parameterizations within GCMs (Johnson, 2005). Two studies using realistic vertical and horizontal distributions of black carbon 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 black carbon changes finds an efficacy of 1.3 (Sokolov, 2005). Further Penner et al. (2006) also model black carbon changes and find efficacies both very much larger and very much smaller than 1.0 for biomass and fossil fuel carbon respectively (Hansen et al. (2005) find similar efficacies for biomass and fossil fuel carbon). In summary there is no consensus as to black carbon efficacy and this may represent problems with the stratospherically adjusted definition of RF (see Section 2.8.3).

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 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). The results of Hansen et al. (2005) and Forster and Shine (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 (see Section 2.8.5). We have increased confidence that stratospherically-adjusted RF is a predictor of global mean temperature response, to within 25% for most realistic RF mechanisms aside from aerosol and stratospheric ozone changes. Further, zero-surface temperature change RFs appear to have efficacies closer to 1.0 for all mechanisms. It should be noted that efficacies have only been evaluated in GCMs and actual climate efficacies could be different than those quoted in Section 2.8.5.

Most GCMs already have at least some representation of aerosol-cloud interactions (Section 2.8.5.6) although many may still poorly represent them. By comparing stratospheric-adjusted RF and zero-surface-temperature-change RF (Figure 2.2 and Section 2.8.3) the magnitude of these aerosol-cloud interaction effects can be gauged. Hansen et al. (2005) evaluated both these forcings for many different mechanisms. It is clear, at least in their model, that all mechanisms exhibit some semi-direct effect but it is only appreciable (>20% of the initial RF) for ozone, absorbing aerosol and the aerosol cloud albedo effect (if the cloud lifetime forcing is associated with efficacy). As well as any aerosol-cloud interaction term the estimate of efficacy will include an additional component associated with the surface temperature response. By attaching efficacies to the RF the cloud interaction terms are accounted for, but they are not separated out as explicit forcings.

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.

RFs are calculated in various ways depending on the agent: from changes in emissions and/or changes in concentrations; from observations and other knowledge of climate change drivers. Current RF depends on present day concentrations of a forcing agent, which in turn depends on the past history of emissions. Some climate response to these forcings can have been expected to have already occurred. Additionally, as RF is a measure of equilibrium climate change and the Earth's climate is not in an equilibrium state, additional climate change in the future is also expected from present day RFs (see Section 2.2. and Chapter 10). As previously stated in Sections 2.2 and 2.8, RF is not alone a suitable metric for weighting emissions, for this purpose the lifetime of the forcing agent also needs to be considered (see Section 2.10).

RFs are considered external to the climate system (see Section 2.2). Aside from the natural RFs (solar, volcanoes and cosmic rays), the other RFs are considered to de directly attributable to humans. For the LLGHGs it is assumed that all changes in their concentrations since preindustrial times are human-induced (either directly through emissions or from land-use changes): these concentration changes are used to calculate the RF. Likewise stratospheric ozone changes are also taken from satellite observations and these are generally solely attributed to Montreal Protocol controlled gases (although there maybe a climate feedback contribution to these trends, see Section 2.3.4.1). For the other RFs, human emissions and/or human-induced land-use changes are used in conjunction with Chemical Transport Models and/or GCMs to estimate the anthropogenic RF.

2.9.1 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 (see Box TS-1 on uncertainty in the Technical Summary). 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. Strong evidence implys that observations have verified the existence of the RF and that there is a physical model to explain the RF. "Consensus" is assessed by assigning a number between 1–3, where 1 implies a good deal of consensus and 3 insufficient consensus. This ranks how well studies agree on quantifying the RF, and especially how well observation based studies agree with models. The "Evidence" and "Consensus" factors are combined to give a level of 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 (see Section 2.4). The quoted uncertainty range is roughly equivalent to a one-sigma value uncertainty from available results. Table 2.11 summarises the key certainties/uncertainties and 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 2.11 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 also excluded from this analysis as they are deemed to be part of the climate response (see Section 2.2). 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 2.11 HERE]

2.9.2 Global Mean Radiative Forcing

The RFs discussed in this chapter, their uncertainty ranges, and efficacies are summarized in Figure 2.24 and Table 2.12. 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 (see Section 2.4.5). Table 2.1 gives a further component breakdown of RF for the LLGHGs. RFs are the stratospherically –adjusted RF, they have not been multiplied by efficacies (see Sections 2.2 and 2.8).

[INSERT FIGURE 2.24 HERE]

[INSERT TABLE 2.12 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) and Section 2.8 still apply. However, several previous studies now have added RFs together (e.g., Schwartz and Andreae, 1996; Boucher and Haywood, 2001). Also as efficacies are now better understood and quantified (see Section 2.8.5) and as the linear-additivity assumption has been more thoroughly tested (see Section 2.8.4) it becomes scientifically justifiable for RFs from different mechanisms to be combined, with certain exceptions as noted below. Adding the anthropogenic RF values shown in the upper panel of Figure 2.24 and combining individual uncertainties results in the probability density function of RF shown in the bottom panel of Figure 2.24. Two probability density functions are shown: the combined effects of RFs with high and medium levels of understanding (LLGHGs and ozone); and one combining all anthropogenic RFs shown in the top panel of Figure 2.24: the solar RF is not included. The probability density functions are generated by combining the 1σ uncertainty estimates independently for the RFs.

These summations imply that since 1750 humans have *very likely* exerted a net warming influence on climate. Global mean net RF for the better understood anthropogenic RF agents (LLGHGs and ozone) is estimated to be 2.9 ± 0.3 W m^{-2}. Anthropogenic RF drivers that are less well understood (e.g., aerosol direct and cloud-albedo RFs; land-use albedo RF) are likely to contribute a negative global mean RF. However, the net RF for all anthropogenic drivers taken together is likely to be positive. Variation in efficacy and hence the semi-direct and cloud-lifetime effects are not accounted for. These add the largest uncertainty to the resulting probability density function for all anthropogenic RFs (other missing RFs agents are likely to be small). Accounting for efficacy would generally broaden the probability density function and a large cloud-lifetime effect (if that becomes realistic) could reduce central estimate.

2.9.3 Global-Mean Radiative Forcing by Emission Precursor

The RF due to changes in the concentration of a single forcing agent can have contributions from emissions of other compounds (Shindell et al., 2005). The RF of methane, for example, is affected by direct methane emissions, as well as NO_x emissions. The methane RF quoted in Table 2.12 and shown in Figure 2.24 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 current RF caused by 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 section assessed the RF terms associated with each principal emission, with the results shown in Figure 2.25. Each principal component can have a direct RF and one or more indirect RFs related to perturbations of other forcing agents. The following indirect forcing mechanisms are considered.

- Fossil carbon from non-CO_2 gaseous compounds, which eventually increase CO_2 in the atmosphere (from CO, CH_4, and NMVOCs emissions)
- Changes in stratospheric ozone (from N_2O and halocarbon (CFCs, HCFC, halons etc.) emissions)
- Changes in tropospheric ozone (from CH_4, NO_x, CO, and NMVOCs emissions)
- Changes in OH affecting the lifetime of CH_4 (from CH_4, CO, NOx, and NMVOCs emissions)
- Changing nitrate and sulphate aerosols through changes in NO_x and SO_2 emissions, respectively.

For some of the principal RFs (e.g., BC, land-use and mineral dust) there is not enough quantitative information available to asses their indirect effects, thus their RFs are the same as those presented in Table 2.12. Table 2.5 gives the total (fossil and biomass burning) direct RFs for black carbon and organic carbon aerosols that are used in Figure 2.25 (taking an average of the AEROCOM and non-AEROCOM models). Table 2.13 summarizes the direct and indirect RFs presented in Figure 2.25, including the methods used for estimating the RFs and the associated uncertainty. Note that for indirect effects through changes in chemically active gases (e.g., OH or ozone) the emission based RF is not uniquely defined since the effect of one precursor will be affected by the levels of the other precursors. The RFs of indirect effects on methane and ozone by NOx, CO and VOC emissions are estimated by removing anthropogenic emissions of one precursor at a time. Sensitivity analysis by Shindell et al. (2005) indicates that the effect of non-linear chemistry on RF is of the order of 10% or less. Very uncertain indirect effects are not included in Table 2.13

and Figure 2.25. These include ozone changes due to solar effects, changes in secondary organic carbon aerosols through changes in the O_3/OH ratio, and cloud albedo changes caused by non-sulphate aerosols.

[INSERT FIGURE 2.25 HERE]

[INSERT TABLE 2.13 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 from in-situ measurements over the last few decades and extending further back using firn and ice-core data (see Section 2.3, Figure 2.4 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 Ozone Depleting Substances (ODS) appears to have peaked 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 2.1). The halocarbon RF in this report, as shown in Table 2.12, is lower than in TAR; this is due to a re-evaluation -the trend in halocarbon RF since TAR has been positive (see Table 2.1).

RF timeseries for the natural forcings are reasonably well known for the past 25 years (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 them. Several time evolution datasets for these and other RFs exist in the literature. (e.g., TAR, Myhre et al., 2001; Hansen et al., 2002). GCMs also employ their own time-dependent histories of many of the forcing agents. As an example of the time history of the RF and surface forcing due to the principal agents, the global-and-annual-mean time series, as implemented in one particularl model, the MIROC AOGCM (Nozawa et al., 2005; Takemura et al., 2005), is illustrated in Figure 2.26 (see also Chapter 10). Although there tends to be differences amongst the various models with regards to their RF reconstructions and their respective present-day RF values, they typically have qualitatively 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 TAR and Figure 2.26; however, uncertainties at the global-mean level are likely of a smaller magnitude than at smaller spatial scales e.g., continental scales.

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). While LLGHGs contribute the most to the present RF (refer also to Figure 2.24 and 2.4), they have also exceeded the contributions due to other agents in the 20th century except for the model-simulated aerosol ‘cloud albedo’ effect and the episodic effects due to explosive volcanic events. The LLGHG RF history especially over the 20th century is one of a steady increase, and more rapidly so over the last 20 years of the 20th century. None of the time evolution of other agents, especially the anthropogenic agents, exhibits the feature of a rapid rise in the present decades. In fact, even the aerosol ’cloud albedo’ effect over the end of the 20th century, which is estimated to have a considerable magnitude, has more of a linear change, in contrast to the LLGHGs over the same period. The volcanic forcing, though transitory in nature, can enable a relatively large magnitude in the natural forcing because of the large value of the transient perturbation e.g., the two explosive eruptions in the last two decades of the 20th century. Chapter 9 employs the available knowledge of the time evolution of the RF over the 20th century, particularly the principal features distinguishing the time evolution of LLGHGs from the other agents.

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. (2006) and Forster and Taylor (2006) for further discussion on uncertainties in GCMs calculation of RF and surface forcing). Again, as for RF, it is difficult to specify uncertainties in the time evolution, as emissions and concentrations for all but the LLGHGs are not well to poorly constrained.

[INSERT FIGURE 2.26 HERE]

2.9.5 Spatial Patterns of Radiative Forcing and Surface Forcing

Chapter 6, 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 Chapter 6, 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 the instantaneous flux change at the tropopause, due to the sum of the estimates in the respective models of the various natural plus anthropogenic agents discussed in the chapter, is illustrated in Figure 2.27 (although this is the instantaneous flux change, it is essentially similar to RF, considering that the species for whom the stratospheric adjustment does matter e.g., stratospheric ozone depletion, do not contribute much to the total forcing). The MIROC AOGCM includes an aerosol cloud-albedo effect and the GFDL AOGCM does not. The generic features are that the RF over most of the globe is positive and is dominated by the LLGHGs. This is more so for the southern than the northern hemisphere, owing to the pronounced aerosol presence in the northern hemisphere. The regions of significant aerosol RF over especially continental areas is clearly manifest (see also Figure 2.14 for another illustrative example of direct aerosol effects on surface forcing, particularly over the continental areas where sources are concentated), although there are some differences in the distributions of these 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 2.27 HERE]

The instantaneous spatial pattern of surface forcing (also shown in Figure 2.27) between the year 2000 and 1860 is computed as for the tropopause. It shows a pronounced deficit relative to preindustrial times where aerosol RF dominates, particularly the Northern Hemisphere. It is essentially because of the aerosol influence on the reduction of the shortwave radiation at the surface that there is a net (sum of solar and longwave) negative forcing. If, for example, there were no aerosol influence, LLGHGs would have increased the longwave emission to the surface and this would have increased the net radiative flux at the surface. 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 instantaneous tropopause and surface forcing imply differences in the Northern Hemisphere equator-to-pole gradients for the surface and tropopause. This, in turn, implies a difference in the amount of energy absorbed by the troposphere from the low to high latitudes. The aerosol influences are also manifest in the difference between Northern and Southern hemisphere, in terms of instantaneous surface and tropopause flux changes, These features are of relevance to the climate response discussed in other chapters (see especially Chapters 8, 9 and 10).

2.10 GWPs and Other Metrics for Comparing Different Emissions

2.10.1 Definition of an emission Metric and the GWP

Multi-component abatement strategies to limit anthropogenic climate change need a framework and numerical values for the trade-off between emissions of different forcing agents (gases and aerosols). The Global Warming Potential (GWP) or other emission metrics provides the necessary tool to operationalize comprehensive and cost-effective policies (Article 3 of the United Nations Framework Convention on Climate Change (UNFCCC)) in a decentralised manner so that multi-gas emitters (nations, industries) can compose cost-effective mitigation measures according to a specified target by allowing for substitution between different climate agents. The metric formulation depends on whether a long-term target to comply with the UNFCCC goal of avoiding dangerous climate change is set (either by a cost-benefit analysis or by a more political judgement), or if we are concerned about reducing the impacts of climate change, but so far have not agreed on any specific long-term target (as in the Kyoto Protocol). In both cases the metric formulation requires knowledge of the contribution to climate change from emissions of various components over time, i.e., their radiative efficiency (see definition below) and atmospheric residence time. In addition, both formulations also involve input from economics. Economists have argued that, ideally, the metric should be the outcome of an analysis that minimizes the discounted present value of damages and mitigation costs (e.g., Manne and Richels, 2001). If a climate forcing reduction trajectory is formulated to achieve a long-term target the proper trade-off between gases is then their relative contribution to that trajectory, that is, the ratio of the shadow prices. The shadow price of gas g is the reduced cost of meeting the desired policy if we were allowed to emit one extra unit of gas *i* at time *t*. Otherwise, if a long-term target is not set, the proper trade-off is the relative contribution of various gases to the impacts, that is, the ratio of the marginal damage costs. Substitution of gases within an international climate policy with a long-term target and including economic factors is discussed in Chapter 3, Sections 3.3 and 3.6 of IPCC WGIII AR4, and metrics based on this approach will not be discussed further in this report.

A very general formulation of an emission metric can be given by (e.g., Kandlikar*,* 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 (damage and benefit) of change in climate (ΔC) at time *t*. The expression $g(t)$ is a weighting function over time (e.g., $g(t) = e^{-kt}$ as a simple discounting giving short-term impacts more weight) (Heal*,* 1997; Nordhaus*,* 1997; IPCC WGIII AR4 Chapter 3, Section 3.6). The subscript *r* refers to a baseline emission path. For two emission perturbations *i* and *j* the absolute metric values $AM_{\rm i}$ and $AM_{\rm j}$ 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 in the case of GWP).

There are several problematic issues related to defining a metric based on the general formulation given above (Fuglestvedt et al., 2003). A major problem is to define appropriate impact functions, although there have been some initial 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 their inherent uncertainties (Shine et al., 2005b). Other problematic issues include the definition of the weighting function *g(t)* and the baseline emission scenarios.

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 compound (*i*) relative to that of 1 kg of the reference gas CO_2 was developed (IPCC, 1990) and adopted for use in the Kyoto Protocol. The GWP of component *i* is defined by

$$GWP_i \equiv \frac{\int_0^{TH} RF_i(t)dt}{\int_0^{TH} RF_r(t)dt} = \frac{\int_0^{TH} a_i \cdot [C_i(t)]dt}{\int_0^{TH} a_r \cdot [C_r(t)]dt} \qquad 37$$

Where TH is the time horizon, RF_i is the global mean RF by component *i*, a_i is the RF per unit mass increase in atmospheric abundance of component *i* (radiative efficiency), $[C_i(t)]$ is the time-dependent decay in abundance of *i*, and the corresponding quantities for the reference gas in the denominator. The numerator and denominator are called the absolute global warming potential (AGWP) of *i* and *r* respectively. All GWPs given in this report use CO_2 as the reference gas. The simplifications made to derive the standard GWP index include, set $g(t) = 1$ (i.e., no discounting) up until the time-horizon (TH), and then $g(t)=0$ thereafter, the choice of a 1 kg pulse emission, the definition of the impact function, $I(\Delta C)$ as the global mean RF, the assumption that the climate response is equal for all RF mechanisms, and the evaluation of the impact relative to a baseline equal to current concentrations (i.e., setting $I(\Delta C_r(t)) = 0$). The criticism of the GWP metric have focused on all of these simplifications (e.g., Smith and Wigley, 2000, O'Neill, 2000; Bradford, 2001; Godal, 2003). However, as long as there is no consensus on what is the relevant impact function ($I(\Delta C)$) and temporal weighting function to use (both involve value judgements), it is difficult to assess the implications of the simplifications objectively (O'Neill, 2000; Fuglestvedt et al., 2003).

The adequacy of the GWP concept has been widely debated since its introduction (O'Neill, 2000; Fuglestvedt et al., 2003). By its definition, two sets of emissions that are equal in terms of their total GWP weighted emissions, will not give equivalence in terms of temporal evolution of the climate response (Smith and Wigley, 2000; Fuglestvedt et al., 2000). Using a 100 year time horizon as in the Kyoto Protocol, the effect of current emissions reductions (e.g., during the first commitment period under the Kyoto Protocol) that contain a significant fraction of short-lived species (e.g., methane) will give less temperature reductions towards the end of the time horizon compared to reductions of CO_2 emissions only. GWPs can really only be expected to produce identical changes in one measure of climate change – integrated temperature change following emissions impulses – and only under a particular set of assumptions (O'Neill, 2000). The GTP metric (see Section 2.10.6.2) provides an alternative approach by comparing global mean temperature change at the end of a given time horizon. Compared to the GWP, the GTP gives equivalent climate response at a chosen time, while putting much less emphasis on near term climate fluctuations caused by emissions of short-lived species (e.g., methane). However, as long as it has not been determined, neither scientifically, economically nor politically, what is the proper time horizon for evaluating "dangerous climate change", the lack of temporal equivalence does not invalidate the GWP concept or provide a guidance to replace it. O'Neill (2003) have argued that the disadvantages of GWPs are likely to be out-weighed by the advantages. This can be done by showing that the cost difference between a multi-gas strategy and a CO_2-only strategy is likely to be much larger than the difference between a GWP-based multi-gas strategy and a cost-optimal strategy (accounting for damage and mitigations costs). Thus although it has several known short comings, the GWP remains the recommended metric to compare future climate impact of emissions of long lived climate gases.

Global averaged GWPs can be calculated for short lived species (e.g., ozone precursors and aerosols). On a global level the mean metric values can be used to give an indication of the total potential of mitigating climate change by including a certain forcing agent in climate policy (cf. Figure 2.28). As discussed by Hansen and Sato (2004) and Rypdal et al. (2005) there might be a potential for more effective climate mitigation strategies if climate mitigation and air quality issues are viewed together. Assessing the climate impact of key species affecting air quality is therefore needed. However, the metric values for short-lived compounds vary significantly by region and time so that for operationalization on a decentralized level, robust regionally varying GWPs must be established and agreed upon. Improved scientific understanding of ozone chemistry and the climate effects of aerosols is needed before this can be established, with the possible exception of carbonmonoxide (Berntsen et al., 2005). A more fundamental question related to the application of GWPs for short lived species is whether the more short-term climate fluctuations caused by pulse emissions of these components should be weighted equally to long-term climate warming by long lived gases, as is implicitly assumed through application of the GWP concept. However, as long as there is no consensus on what constitutes "dangerous anthropogenic interference with the climate system" there is no clear conclusion to this question. A more long term perspective, e.g., by calculating the contribution from current emissions to climate change at a time (or time interval) when global warming is predicted to reach a given threshold value would lead to reduced emphasis on the short lived species.

2.10.2 Direct GWPs

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 about 378 ppm. The radiative efficiency per kilogram CO_2 has been calculated using the same expressions as in IPCC (2001), but with an updated background CO_2 mixing ratio of 378 ppm. For a small perturbation from 378 ppm the RF is 0.01413 W m^{-2} ppm^{-1} (8.7% lower than the TAR value). The CO_2 response function (see Table 2.14) is based on an updated version of the Bern carbon-cycle model, using a background CO_2 concentration of 378 ppm. The increased background concentrations of CO_2 means that the airborne fraction of emitted CO_2 (cf. Chapter 7, Section 7.3) is enhanced, contributing to an increase in the AGWP for CO_2. The AGWP values for CO_2 for 20, 100, and 500 years time horizons are 2.47×10^{-14}, 8.69×10^{-14}, and 28.6×10^{-14} W m^{-2} yr $(kg(CO_2))^{-1}$. The uncertainty in the AGWP for CO_2 is estimated to be ±15%, with equal contribution from the CO_2 response function and the RF calculation.

Updated radiative efficiencies for well mixed-greenhouse gases are given in Table 2.14. Since the TAR radiative efficiencies have been reviewed by Montzka et al. (2003) and Velders et al. (2005). Gohar et al.

(2004) and Forster et al. (2005) have investigated HFC compounds with up to 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 Ramaswamy et al. (2001) (see TAR, Chapter 6, Table 6.2). The perturbation lifetime for CH_4 has been reduced since the TAR from 12 to 10.8 years (see Chapter 7, Section 7.4). Table 2.14 gives GWP values for time horizons of 20, 100 and 500 years. The species in Table 2.14 are those for which either significant level or trends in concentrations are observed, or a clear potential for future emissions have been identified. The uncertainties of these direct GWPs are taken to be ±20%, as in the TAR (the TAR value of ±35% is 2-σ uncertainty).

[INSERT TABLE 2.14 HERE]

2.10.3 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. Direct effects of degradation products of greenhouse gases are not considered to be important (WMO*,* 2003). The indirect effects discussed here are linked to ozone formation or destruction, enhancement of stratospheric water vapour, changes concentrations of the OH radical with the main effect of changing the lifetime of methane, and secondary aerosol formation. 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. For some species (e.g., NOx) the indirect effects can be of opposite sign, further increasing the uncertainty of the net GWP. This can be because background levels of reactive species (e.g., NO_x) can affect the chemical response non-linearly, and/or because the lifetime or the radiative effects of short lived secondary species formed can be regionally dependent. Thus the usefulness of the global mean GWPs to inform policy decisions can be limited. However, they are readily calculable and give an indication of the total potential of mitigating climate change by including a certain forcing agent in climate policy. Following the approach taken by the SAR and TAR, we do not include CO_2 produced from oxidation of CH_4, CO and NMVOCs of fossil origin in the GWP estimates since this carbon should be included in the national CO_2 inventories

2.10.3.1 Methane

Four indirect radiative effects of methane emissions have been identified (cf. Prather et al. (2001) and Section 6.6 of Ramaswamy et al. (2001)). 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. The GWP given in Table 2.14 includes the first three of these effects. The lifetime effect is included through the revised perturbation lifetime of 10.8 years. The effect of ozone production is still uncertain, and as in TAR it is included by enhancing the net of the direct and the lifetime effect by 25%. The estimate of forcing caused by increase in stratospheric water vapour has been increased significantly since the TAR (see Section 2.3.8). This has also been taken into account in the GWP estimate for methane by increasing the enhancement factor from 5% (TAR) to 15%. Due to counteracting effects of reduced perturbation lifetime and enhanced effect of stratospheric water vapour, the net GWP_{100} for methane is still 23 as in the TAR.

2.10.3.2 Carbon monoxide

The indirect effects of carbon monoxide (CO) occur through reduced OH levels (leading to enhanced concentrations of methane) and enhancement of ozone. The TAR gave a range of 1.0–3.0 for GWP_{100}. Since the TAR Collins et al., (2002) and Berntsen et al. (2005) have calculated GWPs for CO emissions, which range between 1.6 and 2.0, depending on the location of the emissions. Berntsen et al. (2005) found that emissions of CO from Asia had a 25% higher GWP, compared to European emissions. Averaging over the TAR values and the new estimates give a mean of 1.9 for the GWP_{100} for CO, which is used for the GWP-weighted emissions in Figure 2.28.

2.10.3.3 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 (through changes in OH), 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 2.15. Weighting these GWPs by the emissions of the respective compounds gives a weighted average GWP_{100} of 3.4 which is used for Figure 2.28. 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. (2002) estimate an uncertainty range of –50% to +100%.

[INSERT TABLE 2.15 HERE]

2.10.3.4 NO_x

The short lifetime and complex non-linear chemistry which cause two opposing indirect effects through ozone enhancements and methane reductions makes calculations of GWP for NO_x emissions very uncertain (Shine et al., 2005b). In addition, the effect formation of nitrate aerosols (see Section 2.4.5.5) which has not yet been included in model studies calculating GWPs for NOx, can be significant. 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 (daily, seasonal) of the emissions (Fuglestvedt et al., 1999; Derwent et al., 2001; Wild et al., 2001; Stevenson et al., 2004; Berntsen et al., 2005; Berntsen et al., 2006). Due to the lack of agreement even on the sign of the global mean GWP for NOx among the different studies and the omission of the nitrate aerosol effect, a central estimate for the GWP_{100} for NOx is therefore not presented.

2.10.3.5 Halocarbons

Chlorine and bromine containing halocarbons lead to ozone depletion 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 the physical characteristics of the CFC warming effect and ozone cooling effect were very different from each other.

2.10.3.6 Hydrogen (H_2)

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 and thus also affects 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 a indirect GWP_{100} for the tropospheric effects of H_2 of 5.8, which includes the effects of methane lifetime and tropospheric ozone.

2.10.4 Aerosols and aerosol precursors

Previous IPCC reports have not given GWPs values for aerosols or aerosol precursors. As discussed above (see Section 2.10.1) there are many important limitations to applying GWPs for short-lived species like aerosols. Since the TAR significant progress has been made in the understanding of the radiative effects of aerosols (see Section 2.4). AGWPs for SO_2, BC and OC, including only the direct aerosol effect, are calculated based on the results from AEROCOM experiment presented in Tables 2.5, and 2.6.

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 Table 2.5 in this report. Using the averaged modelled global anthropogenic sulphate loading of 2.15 mg $(SO_4)\ m^{-2}$, and all sky RF of $-0.35\ W\ m^{-2}$, an oxidation efficiency of 52% from SO_2 to SO_4 (Berglen et al., 2004) and a residence time of 4.1 days, an AGWP value for SO_2 of $-5.5 \times 10^{-12}\ W\ m^{-2} yr\ (kg(SO_2))^{-1}$ is derived (independent of time horizon due to the very short atmospheric residence time for SO_4). GWP values of –220, –64, and –19 are estimated for time horizons of 20, 100 and 500 years respectively. Based on model variability the uncertainty is estimated to be ±50%.

Similarly global mean GWP for BC and OC have been derived based on the AEROCOM results. For BC using an averaged modelled global anthropogenic loading of 0.25 mg m^{-2}, and all sky RF of 0.29 Wm^{-2} and a residence time of 7.1 days, an AGWP value of 4.4×10^{-11} W m^{-2} yr $(kg(BC))^{-1}$ is derived. GWP values for BC of 1800, 510, and 160 are estimated for time horizons of 20, 100 and 500 years respectively. For OC the averaged modelled global anthropogenic loading of 0.50 mg m^2 (anthropogenic fraction 53%, POM/OC = 1.4) and all sky RF of –0.18 W m^{-2} and a residence time of 6.46 days, an AGWP value of -1.25×10^{-11} W m^{-2} yr $(kg(OC))^{-1}$ is derived. GWP_{100} values for OC of –510, –140, and –44 are estimated for time horizons of 20, 100 and 500 years respectively. Bond and Sun (2005) have estimated the uncertainty in the GWP for BC to be –70%/+100%. Based on the inter-model variance in Table 2.5., an equal uncertainty is estimated for OC.

The main sources for the relatively large uncertainties are model assumptions about transport and removal of particles and optical properties.

2.10.5 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_{100} values to obtain equivalent CO_2 emissions. This is consistent with the Kyoto Protocol through its adoption of GWPs with 100 years time horizon. Figure 2.28 shows the equivalent CO_2 emissions for all climate agents (or groups of agents) considered in this report. The same caveats as discussed by Velders et al. (2005) in terms of direct versus indirect GWPs for halocarbons applies when comparing GWPs of the other indirect effects and is probably even more important for the GWP from short lived species. These caveats and the GWP uncertainties must be kept in mind when the contributions to CO_2-equivalent emissions shown in Figure 2.28 are compared. Given the ambiguity of the GWP-concept (Section 2.10.1 and O'Neill, 2000) and our improved understanding of forcing-response relationships (Section 2.8), comparing the CO_2-equivalent emissions in Figure 2.28 give an indication of the future climate impact of current emissions of the different climate agents.

It should be noted that the compounds with long lifetimes (in particular CO_2) tend to contribute more to the total with this "forward looking" perspective than in the standard "IPCC RF bar-chart diagram" (Figure 2.24). 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. Again the 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 2.28 HERE]

2.10.6 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 problematic as discussed above for handling short-lived gases or aerosols (e.g., NO_x or black carbon aerosols). One alternative, the radiative forcing index (RFI) introduced by IPCC (1999), should not be used as an emission metric since it does not account for the different residence times of different forcing agents.

2.10.6.1 Revised GWP Formulations

2.10.6.1.2 Including the climate efficacy in the GWP

As discussed in Section 2.8.5 the climate efficacy can vary between different forcing agents (within 25% for most realistic RFs). Fuglestvedt et al. (2003) have proposed a revised GWP concept including the efficacy of a forcing agent in the GWP expression. Berntsen et al. (2005) have calculated GWP values in this way for NO_x and CO emissions in Europe and in South East Asia. The efficacies are less uncertain than climate sensitivities. However, Berntsen et al. (2005) showed that for ozone produced by NO_x emissions the climate efficacies will also depend on the location of the emissions.

2.10.6.2 The Global-Temperature Potential

Shine et al. (2005a) 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 (e.g., 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. The GTP's do not require simulations with AOGCMs, but are given as transparent and simple formulaes with a small number of input paraters required for calculation. 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 use 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 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. (2005a) 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.

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

Greenhouse-Gases

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). These gases accumulate in the atmosphere, causing concentrations to increase with time. Significant increases in all of these gases have occurred in the industrial era (see Question 2.1 Figure 1). All of the increases are all attributable to human activities.

- *Carbon dioxide* has increased from fossil fuel use in transportation, building heating and cooling, and the manufacture of cement and other goods. Deforestation has also increased carbon dioxide by releasing carbon dioxide and reducing the total uptake of carbon dioxide by plants.
- *Methane* has increased in the past as a result of human activities related to agriculture, natural gas distribution, and landfills. Methane is also released in natural processes. Currently methane abundances are decreasing in the atmosphere.
- *Nitrous oxide* is also emitted by both human activities and natural processes.
- *Halocarbon gases* are released in human activities, which account for most atmospheric abundances. Natural processes are also a significant source. Principal halocarbons include the chlorofluorocarbons (e.g., CFC-11 and CFC-12), which were used extensively as refrigeration agents and in other uses before their presence in the atmosphere was found to contribute to stratospheric ozone depletion.

[INSERT QUESTION 2.1, FIGURE 1 HERE]

Ozone

Ozone is a greenhouse gas that is continually produced and destroyed in the atmosphere by chemical reactions. In the troposphere, human activities have increased ozone through the release of gases such as carbon monoxide, hydrocarbon and nitrogen oxide gases, which chemically react to produce ozone. Other released gases, namely the halocarbons, destroy ozone in the upper atmosphere (stratosphere). The accumulation of halocarbons in the atmosphere has reduced stratospheric ozone layer over the globe, with the most notable example being the ozone hole over the Antarctic continent.

Water vapour

Water vapour is the most abundant and important greenhouse gas in the atmosphere. However, human activities have only a small *direct* influence on the amount of atmospheric water vapour. *Indirectly*, humans can change water vapour significantly by changing climate. For example, a warmer atmosphere is expected to contain more water vapour. Human activities do influence water vapour through methane emissions, because methane undergoes chemical destruction in the stratosphere to produce a small amount of water vapour.

Aerosol particles

Aerosol particles are 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 have increased aerosols containing sulphur compounds, organic compounds, and black carbon. Human activities such as surface mining and industrial processes have increased dust in the atmosphere. Natural sources include mineral dust released from the surface, biogenic emissions from the land and oceans, and volcanic eruptions.

[START OF QUESTION 2.1, BOX 1]

What is a radiative forcing? The influence of a climate change agent, such as a greenhouse gas, on Earth's climate is often evaluated using the concept of radiative forcing. Radiative forcing is a measure of how the energy present in the global Earth-atmosphere system will change due to the presence of a climate change agent. The word *radiative* arises because agents change the balance between incoming solar radiation and outgoing infrared radiation involving Earth's surface and atmosphere. The term *forcing* arises because the presence of a climate change agent in the atmosphere causes an ongoing long-term or short-term change in this radiative balance. Radiative forcing is often quantified as the 'rate of energy change per unit area of the globe as measured at the top of the atmosphere', is expressed in units of 'Watts per square meter', and is abbreviated as $W\ m^{-2}$ (see Question 2.1 Figure 2). When radiative forcing from an agent or group of agents is evaluated as *positive*, the energy of the Earth-atmosphere system is *increasing* and the consequence is expected to be an average *warming* of the system. In contrast, for a *negative* radiative forcing, the energy is *decreasing* and the consequence is expected to be an average *cooling* of the system. One challenge for climate scientists is to identify all climate agents, to quantify the radiative forcing of each agent, and to evaluate the total radiative forcing from the group of agents.

[END OF QUESTION 2.1, BOX 1]

Radiative Forcing of climate change agents affected by human activities
The contributions to radiative forcing from each of the agents influenced by human activities are shown in Question 2.1 Figure 2. The values reflect the total forcing relative to the start of the industrial era (about 1750). The forcings for all greenhouse gas increases are positive because the gases absorb outgoing infrared radiation in the atmosphere. Carbon dioxide increases have caused the largest forcing. Tropospheric ozone increases have led to a warming and stratospheric ozone decreases have led to a cooling.

Aerosol particles influence radiative forcing *directly* through reflection and absorption of solar and infrared radiation in the atmosphere. Some aerosols cause a positive forcing while others cause a negative forcing. The direct radiative forcing summed over all aerosol types is slightly negative. Aerosols also cause radiative forcing *indirectly* through the changes they cause in cloud properties throughout the atmosphere. The indirect effects combine to produce a negative forcing.

Human activities since the industrial era have altered the nature of land cover over the globe, principally through changes in croplands, pastures, and forests. They also have modified the reflective properties of ice and snow. Overall, more solar radiation is now being reflected from Earth's surface (increased surface albedo) as a result of our activities. This change results in a negative forcing.

[INSERT QUESTION 2.1, FIGURE 2 HERE]

Aircraft produce persistent contrails in regions that have suitably low temperatures and high humidity. Contrails are a form of cirrus cloud that reflects solar radiation and absorbs infrared radiation. Persistent contrails also spread, thereby increasing the total amount of cirrus cloudiness. Global aircraft operations are estimated to have increased Earth's cloudiness, which causes a small positive radiative forcing.

Radiative Forcing from Natural Changes
The largest changes in natural forcings in the industrial era are solar changes and explosive volcanic eruptions. Solar output has increased gradually in the industrial era, causing a positive radiative forcing (see Question 2.1, Figure 2). 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 temporary increases that occur in sulphate aerosol in the stratosphere. The stratosphere is currently free of volcanic aerosol because the last major volcanic eruption was Mt. Pinatubo in 1991. As a result volcanic forcing is not included in Question 2.1 Figure 2. The radiative forcings from solar changes and volcanoes in the industrial era are both small compared to the sum of those that result from human activities.

Tables

Table 2.1. Present day concentrations and RF for the measured LLGHGs. The changes since TAR are also shown. Note: we will try to update these for 2005 concentrations in final version

	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 ± 0.4 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 ESRL and SIO networks (see Section 2.3.1).
CH_4 measurements are combined data from the ESRL and AGAGE networks (see Section 2.3.2).
Halocarbon measurements are average of ESRL and AGAGE networks. UEA and PSU measurements were also used (see 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 2.14
(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 2.2. 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-modal 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) 2-channel using 0. 64 µm and 0.83 µm gives $\tau_{\lambda=0.55}$ and Å over ocean assuming a bimodal aerosol size distribution (Higurashi et al., 1999)
TOMS[a] (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 the 8 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 2003–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 Tanré, 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 Nakajima et al., 20001998).
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)	1998-present		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	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 (Veefkind et al., 1998;

Radiometer)				Holzer-Popp et al. 2002).
SeaWiFS (Sea-Viewing Wide Field-of-View Sensor)	1997-present	0.765 and 0.865 µm	τ_{aer}, Å	2-channel using 0. 765 µm and 0.856 µm gives $\tau_{\lambda=0.856}$ and Å over ocean. Bi-modal aerosol size distribution assumed (Wang et al., 2005)

Notes:

(a) TOMS followed up by OMI instrument on EOS-Aura satellite, launched July 2004**.**

Table 2.3. The Direct aerosol 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 ($W\ m^{-2}$) (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 or from NOAA-NESDIS algorithm used to estimate the direct radiative effect.	–3.8 (NESDIS) to –5.5 (MODIS)
Remer and Kaufman (2005)	MODIS	Aug 2001–Dec 2003	Best prescribed aerosol model fitted to MODIS data. τ_{aer} from fine mode fraction.	–5.7 ± 0.4
Zhang et al. (2005); Christopher and Zhang (2004)	CERES/ MODIS	Nov 2000–Aug 2001	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	Nov 1996– Jun 1997	Best prescribed aerosol model fitted to POLDER data	–5.2
Loeb and Kato (2002)	CERES/ VIRS	Jan 1998–Aug 1998, Mar 2000	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)	SeaWiFSs	1998	Radiative transfer calculations with SeaWiFS AOD and prescribed optical properties	–5.4
Boucher and Tanré (2000)	POLDER	Nov 1996–Jun 1997	Best prescribed aerosol model fitted to POLDER data	–5 to –6
Mean (standard deviation)				–5.3 (0.8)

Table 2.4. The direct radiative forcing for sulphate aerosol derived from models published since TAR and from the AeroCom simulations using identical emissions. Load and aerosol optical depth (τ_{aer}) refer to the anthropogenic sulphate, $\tau_{aer\ ant}$; fraction of anthropogenic sulfate to total sulfate τ_{aer} of present day, NRFM; normalized RF by mass, NRF; normalized RF per unit τ_{aer} .

No	**Model**	**LOAD [mg SO$_4$ m^{-2}]**	t_{aer} **(0.55 μm)**	$t_{aer\ ant}$ **[%]**	**RF [W m^{-2}]**	**NRFM [W g^{-1}]**	**NRF [W m^{-2} t_{aer}^{-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	SPRINT	1.55	0.015	72%	–0.21	–135	–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	2.64	0.020	58%	–0.58	–220	–28	(Liu and Penner, 2002)
N	UIO_CTM	1.70	0.019	57%	–0.35	–208	–19	(Myhre et al., 2003)
O	LOA	3.64	0.035	64%	–0.49	–136	–14	(Reddy and Boucher, 2004)
P	LSCE	3.01	0.023	59%	–0.42	–138	–18	(Textor et al. 2005)
Q	MPI_HAM	2.47	0.016	60%	–0.46	–186	–29	(Stier et al., 2005)
R	GISS	1.34	0.006	41%	–0.19	–139	–31	(Koch, 2001)
S	UIO_GCM	1.72	0.012	59%	–0.25	–145	–21	(Iversen and Seland, 2002; Kirkevag and Iversen, 2002)
T	SPRINT	1.19	0.013	59%	–0.16	–137	–13	(Takemura et al., 2005)
U	ULAQ	1.62	0.020	42%	–0.22	–136	–11	(Pitari et al., 2002)
Average A-L		2.80	0.024		–0.46	–176	–17	
Average M-U		2.15	0.018	55%	–0.35	–161	–20	
Minimum A-U		1.19	0.006	41%	–0.96	–293	–32	
Maximum A-U		5.50	0.042	72%	–0.16	–72	–8	
Stddev A-L		1.18	0.010		0.20	75	9	
Stddev M-U		0.83	0.008	8%	0.15	34	7	

Table 2.5. Recent estimates of anthropogenic carbonaceous aerosol forcing. BB=biomass burning sources included, FFBC=fossil fuel black carbon, FFPOM= fossil fuel particulate organic matter. Lines A-K: recently published / Lines L-T: Models used AeroCom emissions

No	Model	LOAD POM [mgPOM m^{-2}]	AOD POM	AOD POM ant	LOAD BC	RF BCPOM [W m^{-2}]	RF POM [W m^{-2}]	RF BC [W m^{-2}]	RF FFPOM [W m^{-2}]	RF FFBC [W m^{-2}]	RF BB [W m^{-2}]	Reference
A	SPRINT					0.12	–0.24	0.36	–0.05	0.15	–0.01	(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[a]	0.18[a]	–0.05[a]	(Koch, 2001)
E	GISS	2.39			0.39	0.32	–0.18	0.50	–0.05[a]	0.25[a]	0.12[a]	(Chuang and Seinfeld., 2002)
F	GISS	2.49			0.43	0.30	–0.23	0.53	–0.06[a]	0.27[a]	0.09[a]	(Liao and Seinfeld, 2005)
G	SPRINT	2.67	0.029	82%	0.53	0.15	–0.27	0.42	–0.07[a]	0.21[a]	0.01[a]	(Takemura et al., 2005)
H	GATORG				0.45	0.47	–0.06	0.53	–0.01[a]	0.27[a]	0.22[a]	(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)
L	UMI	1.16	0.0060	53%	0.19	0.02	–0.23	0.25	–0.06[a]	0.12[a]	–0.01	(Liu and Penner, 2002)
M	UIO_CTM	1.12	0.0058	55%	0.19	0.02	–0.09[e]	0.10[e]	–0.04	0.11	–0.05	(Myhre et al., 2003)
N	LOA	1.41	0.0085	52%	0.25	0.14[b]	–0.16[b]	0.27[b]	–0.04[a]	0.13[a]	0.05[a]	(Reddy and Boucher, 2004)
O	LSCE	1.50	0.0079	46%	0.25	0.13	–0.17	0.30	–0.04[a]	0.15[a]	0.02[a]	(Textor et al., 2005)
P	ECHAM	1.06[c]	0.0028[c]	55%[c]	0.18[c]	0.34[b]	–0.39[b]	0.64[b]	–0.05[c]	0.16[c]	0.23[a]	(Stier et al., 2005)
Q	GISS	1.22	0.0060	51%	0.24	0.08	–0.14	0.22	–0.03[a]	0.11[a]	0.01[a]	(Koch, 2001)
R	UIO_GCM	0.88	0.0046	59%	0.19	0.24	–0.27	0.36	–0.07[a]	0.18[a]	0.13[a]	(Iversen and Seland, 2002)
S	SPRINT	1.84	0.0200	49%	0.37	0.22	–0.10	0.32	–0.01	0.13	0.06	(Takemura et al., 2005)
T	ULAQ	1.71	0.0075	58%	0.38	0.10[d]	–0.07	0.16	–0.02[a]	0.08[a]	0.03[a]	(Pitari et al., 2002)
Average A-K		2.38	0.019		0.38	0.26	–0.24	0.44	–0.06	0.25	0.07	
Average L-T		1.32	0.008	53%	0.25	0.14	–0.18	0.29	–0.04	0.13	0.05	
Stddev A-K		0.42	0.006		0.08	0.14	0.08	0.13	0.04	0.11	0.09	
Stddev L-T		0.32	0.005	4%	0.08	0.11	0.10	0.15	0.02	0.03	0.08	

Notes:

(a) Models A-C are used to provide a split in sources derived from total POM and total BC ; FFPOM=POM*0.25 ; FFBC=BC*.5 ; BB=(BCPOM)-(FFPOM+FFBC)

(b) Models L,O,Q-T are used to provide a split in components : BCPOM=Total aerosol - SO4 ; POM=BCPOM*-1.15 ; BC=BCPOM*1.9

(c) MPI_HAM fossil fuel carbonaceous forcing of –0.11 W m^{-2}, POMant=POM*0.55; Bcant=BC*0.8

(d) ULAQ all sky values=clear-sky*0.3 ; GISS absorption coefficient assumed to equal that of AeroCom models (7.4 m^2 g^{-1})

(e) Estimated using the fractions used in calculation of the optical properties

Table 2.6. Recent estimates of anthropogenic aerosol load (LOAD), anthropogenic aerosol optical depth (τ_{aer}), its fraction of present day total aerosol optical depth ($\tau_{aer\ ant}$), cloud cover in aerosol model, total aerosol direct radiative forcing (RF), surface forcing and atmospheric forcing.

No	Model	LOAD	τ_{aer}	$\tau_{aer\ ant}$	Cloud Cover	RF top clear sky	RF top all sky	Surface Forcing all sky	Atmospheric Forcing all sky	Reference
		[mg m^{-2}]	[ ]	[%]	[%]	[W m^{-2}]	[W m^{-2}]	[W m^{-2}]	[W m^{-2}]	
Published since IPCC, 2001										
A	GISS	5.0			79%		-0.39^{a} $+0.01^{b}$	-1.98^{a} -2.42^{b}	1.59^{a} 2.43^{b}	(Liao and Seinfeld, 2005)
B	LOA	6.0	0.049	34%	70%	–0.53	–0.09			(Reddy and Boucher, 2004)
C	SPRINT	4.8	0.044	50%	63%	–0.77	–0.06	–1.92	1.86	(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	4.0	0.028	25%	63%	–0.80	–0.41	–1.24	0.84	(Liu and Penner, 2002)
I	UIO_CTM	3.0	0.026	19%	70%	–0.85	–0.34	–0.95	0.61	(Myhre et al., 2003)
J	LOA	5.3	0.046	28%	70%	–0.80	–0.35	–1.49	1.03	(Reddy and Boucher, 2004)
K	LSCE	4.8	0.033	40%	62%	–0.94	–0.28	–0.93	0.66	(Textor et al., 2005)
L	MPI_HAM	4.3	0.042	30%	62%	–0.50	–0.12	–1.07	0.95	(Stier et al., 2005)
M	GISS	2.8	0.014	11%	57%	–0.29	–0.11	–0.81	0.79	(Koch, 2001)
N	UIO_GCM	2.8	0.017	11%	57%		–0.01	–0.84	0.84	(Kirkevag and Iversen, 2002)
O	SPRINT	3.2	0.036	44%	62%	–0.35	+0.04	–0.91	0.96	(Takemura et al., 2005)
P	ULAQ	3.7	0.030	23%		–0.25	–0.08			(Pitari et al., 2002)
Average A-G		5.1	0.046	42%	67%	–0.73	–0.23	–1.95	1.73	
Average H-P		3.8	0.030	26%	63%	–0.60	–0.18	–1.03	0.84	
Stddev A-G		1.4	0.004			0.18	0.21			
Stddev H-P		0.9	0.011	11%	5%	0.28	0.16	0.23	0.15	
Average A-P		4.3	0.035	29%	64%	–0.63	–0.21	–1.21	1.01	
Stddev A-P		1.3	0.012	13%	7%	0.26	0.18	0.44	0.40	
Minimum A-P		2.7	0.014	11%	57%	–0.94	–0.63	–1.98	0.61	
Maximum A-P		6.7	0.049	50%	79%	–0.25	0.04	–0.81	1.86	

Notes:
(a) External mixture
(b) Internal mixture

Table 2.7. Published model studies of the RF due to cloud albedo listing some relevant modeling details

Model	Model Type	Aerosol Types[3]	Aerosol Mixtures[4]	Cloud Types for Indirect Effect	Microphysics	**Cloud Albedo Radiative Forcing (W/m^2)**
Lohmann et al. (2000)	AGCM + sulfur cycle (ECHAM4)	S, OC, BC, SS, D	I ----- E	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)** ----- –1.5 (total)
Jones et al. (2001)	AGCM + sulfur cycle fixed SST (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. (2001)	GCM with slab ocean + sulfur cycle (Hadley) --------------- AGCM fixed 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)	AGCM (CSIRO) fixed SST and sulfur loading -----------	S	-	warm and mixed phase	Rotstayn (1997); Rotstayn et al. (2000)	**–1.39 (albedo)**
----------------------- Rotstayn and Liu (2003)	Interactive sulfur cycle				---------------------------- Inclusion of dispersion	------------------------- 12–35% decrease **–1.12 (albedo, mid valued decrease)**
Ghan et al. (2001)	AGCM (PNNL) + chemistry (MIRAGE) fixed SST	S, OC, BC, SS, N, D	E (for different modes) I (within modes)	warm and mixed phase	droplet number conc. and LWC, crystal concentration and IWC different processes affecting the various modes	–1.7 (total) **–0.85 (albedo)**

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[3] S; sulphate, SS: sea salt, D: mineral dust, BC: black carbon, OC: organic carbon, N:nitrate

[4] E: external, I: internal

Chuang et al. (2002)	CCM1 (NCAR)+ chemistry (GRANTOUR) fixed SST	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. (2002)	GCM (GISS) + sulfur cycle fixed 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) fixed SST	S, OC, BC, SS, D	E (for nucleation mode and fossil fuel BC) I (for accumulation mode)	warm and mixed phase	Rasch and Kristjánsson (1998) stratiform and detraining convective clouds	–1.82 (total) **–1.35 (albedo)**
Suzuki et al. (2004)	AGCM (Japan) fixed SST	S, OC, BC, SS	E	stratiform	Berry (1967), Sundqvist (1978), L&F (1997)	**–0.54 (albedo)**
Quaas et al. (2004b)	AGCM (LMDZ) + interactive sulfur cycle fixed SST	S	-	warm and mixed phase	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995); Boucher et al. (1995)	**–1.3 (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, 2004)	**–0.77 (albedo)**
Kristjansson et al. (2005)	CCM3 (NCAR)+ sulfur and carbon cycles slab ocean	S, OC, BC, SS, D	E (for nucleation mode and fossil fuel BC) I (for accumulation mode)	warm and mixed phase	Kristjansson (2002) stratiform and detraining convective clouds	–1.15 (total, at the surface)
Quaas and Boucher (2005)	AGCM (LMDZ) + interactive sulfur cycle fixed SST	S, OC, BC, SS, D	E	warm and mixed phase	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995); Boucher et al. (1995) control run -----------------	**–0.9 (albedo)** -----------------

					fit to POLDER data fit to MODIS data	–0.5 (albedo)[a] –0.3 (albedo)[a]
Quaas et al. (2005)	AGCM (LMDZ and ECHAM4)	S, OC, BC, SS, D	E	warm and mixed phase	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995), control runs (ctl) --------- aerosol mass and cloud droplet number concentration fitted to MODIS data	–0.84 (total LMDZ-ctl) –1.54 (total (ECHAM4-ctl) -------------- 0.53 (total LMDZ)[a] –0.29 (total (ECHAM4)[a]
Dufresne et al. (2005)	AGCM (LMDZ) + interactive sulfur cycle fixed SST	S	-	warm	Aerosol mass and cloud droplet number concentration, Boucher and Lohmann, (1995), fitted to POLDER data	–0.22 (albedo)[a]
Takemura et al. (2005)	AGCM + SPIRNTARS slab ocean	S, OC, BC, SS, D	E (50% BC from fossil fuel) I (for OC and BC)	warm	Activation based on Kohler theory and updraft velocity	–0.94 (total) **–0.52 (albedo)**
Ming et al. (2006c)	AGCM (GFDL) (prescribed SST and sulfur loading)	S	-	warm	Rotstayn et al (2000), Khainroutdinov and Kogan (2000) (aerosols offline)	–2.3 (total) **–1.4 (albedo)**

Notes:

The shaded boxes in the first column indicate the studies that have been included in Figure 2.16, where only the bold numbers in the last column are used.

(a) These simulations have been constrained by satellite observations, using the same empirical fit to relate aerosol mass and cloud droplet number concentration

Table 2.8. Estimates of forest area, contribution to CO_2 rise from anthropogenic land cover change, and RF due to land-cover-induced CO_2 rise and surface albedo change, relative to pre-industrial and Potential Natural Vegetation (PNV). Carbon emissions from land cover change for the 1980s and 1990s are discussed in Chapter 7 (see Section 7.3, Table 7.3.2)

Main Source of Land Cover Data	**Forest Area PNV** 10^6 km^2	**Forest Area circa 1700** 10^6 km^2	**Forest Area circa 1990** 10^6 km^2	**Contribution to CO_2 Rise 1000–2000 (ppmv)**	**CO_2 RF[a] (W m^{-2})**	**Albedo RF vs PNV (W m^{-2})**	**Albedo RF vs 1750 (W m^{-2})**
Ramankutty and Foley (1999)	55.27	52.77[n]	43.97[b]	24[c]	–0.40	–0.24[d] –0.29 - +0.02[e] –0.2[f]	–0.18[d] –0.14[f,g] –0.15[g,h] –0.15[o]
Klein Goldewijk (2001)	58.6	54.4	41.5	22[c]	0.36	–0.66 - +0.1[e]	–0.50 - +0.08[q]
Houghton (1983, 2003)[m]		62.15	50.53[i]	43[c]	0.71		
MODIS (Schaaf et al., 2002)						–0.09[r]	–0.07[q]
Wilson and Henderson-Sellers (1985)						–0.2[j] –0.29[e]	–0.15[q] –0.22[q]
SARB[p]						–0.11 - –0.55[e]	–0.08 - –0.41[q]
Matthews (1983)						–0.12[e] –0.4[k] –0.08[l]	–0.09[q] –0.3[q] –0.06[q]

Notes:
(a) CO_2 RFs calculated as fraction of land use change contribution to the total increase in CO_2
(b) 1992 forest area
(c) Brovkin et al. (2004)
(d) Betts et al. (2006)
(e) Myhre and Myhre (2003). Range of estimate for each land cover dataset arises from use of different albedo values
(f) Model of Goosse et al. (2005) in Brovkin et al. (2006)
(g) Estimate relative to 1700
(h) Matthews et al. (2003)
(i) 1980 forest area
(j) Betts (2001)
(k) Hansen et al. (1997)
(l) Govindasamy et al. (2001)
(m) cited in Richards (1990)
(n) 1750 forest cover reported as 51.85
(o) Hansen et al. (2000)
(p) http://www-surf.larc.nasa.gov/surf/
(q) RF relative to 1750 estimated as 0.75 of RF relative to PNV following Betts et al. (2006) and Brovkin et al. (2006)
(r) Myhre et al. (2005)

Table 2.9. 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 (0.006–0.015)	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 2.10. Compared are estimates of the increase in RF from the seventeenth century Maunder Minimum (MM) to contemporary solar minima.

Reference	Assumptions and Technique	RF from the Maunder Minimum to contemporary minima (W m^{-2})
Schatten and Orosz (1990)	extrapolation of the 11-year irradiance cycle to the MM, using the sunspot record.	~ 0
Lean, Skumanich, and White (1992)	no spots, plage or network in Ca images assumed during MM	0.26
Lean, Skumanich, and White (1992)	no spots, plage or network and reduced basal emission in cell centres in Ca images non-cycling stars assumed during MM	0.45
Hoyt and Schatten (1993)*	convective restructuring implied by changes in sunspot umbra/penumbra ratios from MM to present	0.65
Lean, Beer, and Bradley (1995)	reduced brightness of non-cycling stars, relative to those with active cycles, assumed typical of MM	0.45
Fligge and Solanki (2000)*	combinations of above	0.72
Lean (2000)	reduced brightness of non-cycling stars (revised solar-stellar calibration) assumed typical of MM	0.38
Foster (2004) Model	non magnetic sun estimates by removing bright features from MDI images assumed for MM – similar approach to Lean et al (1992).	0.28
Solanki and Krivova (2005)	accumulation of bright sources estimated from simple parameterization of flux emergence and decay from MM to present	0.38
Wang, Lean, and Sheeley (2005)*	flux transport simulations of total magnetic flux evolution from MM to present	0.09
Dziembowski and Goode (2005)	helioseismic observations of solar interior oscillations suggest that the historical Sun cannot be any dimmer than current activity minima	~ 0

Notes:
The RF is the irradiance change divided by 4 (geometry) and multiplied by 0.7 (albedo).
The solar activity cycle, which was negligible during the Maunder Minimum and is of order 1 W m^{-2} (peak-to-peak) during recent cycles, is superimposed on the irradiance changes at cycle minima, and increases the net radiative forcing in the table by an additional 0.09 W m^{-2}. .
Reconstruction identified by * extend only to 1713, the end of the Maunder Minimum.

Table 2.11. Uncertainty assessment of RFs discussed in this report. Uncertainties are in approximate order of importance with first-order uncertainties listed first.

	Evidence	Consensus	LOSU	Certainties	Uncertainties	Basis of 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 observed 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 lightning; 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 lightning
Stratospheric water vapour from methane	B	2	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 aerosol RFs	A	3	Low	Ground-based and satellite observations; some source regions and modelling	Emission sources and their history vertical structure of aerosol, optical properties, mixing and separation from natural background aerosol;	Range of published model results with allowances made for comparisons with satellite data
Cloud albedo aerosol RF	B	3	V. Low	Observed in case studies – e.g., ship tracks; GCMs model one	Lack of direct 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 and model-satellite comparison (see Chapter 7)	Lack of observations, inadequately represented in global models; little consensus,	Not given
Semi direct effect	B	3	V. Low	Cloud resolving models and GCM model exhibit an effect	Lack of observations; inadequately represented in global models; lack of agreement on sign of global semi direct effect	Not given

Contrails	A	3	Low	Cirrus radiative and microphysical properties; aviation emissions; contrail coverage in certain regions	Global contrail coverage and optical properties	Best estimate based on recent work and range from published model results
Aviation Induced Cirrus	B	3	V. Low	Cirrus radiative and microphysical properties; aviation emissions; contrail coverage in certain regions	Aviation's effect on cirrus clouds; transformation of contrails to cirrus	Not given
Solar	A	3	Low	Measurements over last 25 years; proxy indicators of solar activity	Relationship between proxy data and TSI; indirect ozone effects	Range from available reconstructions of TSI and their qualitative assessment
Cosmic rays	C	3	V. Low	Some empirical evidence and some observations as well as microphysical models suggest link to clouds	General lack/doubt regarding physical mechanism; dependence on correlation studies	Not given
land-use surface albedo	A	3	Low	Some quantification of deforestation and desertification.	Separation of anthropogenic changes from natural;	Based on range of published estimates and published uncertainty analyses.
Black-carbon aerosol on snow (albedo)	B	3	V. Low	Estimates of black carbon aerosol on snow; Some model studies suggest link	Separation of anthropogenic changes from natural; mixing of snow and black carbon aerosol ;quantification of RF	Estimates based of few model studies
Other surface effects	C	3	V. Low	Some model studies suggest link and some evidence of relevant processes	quantification of RF and interpretation of results in forcing feedback context difficult.	Not Evaluated
Volcanic aerosol	A	3	Low	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	Past constructions/estimates of explosive volcanoes and observations of Mt. Pinatubo aerosol

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 a level of scientific understanding is determined (LOSU, quoted in the 4th Column).

Table 2.12. Global mean radiative forcings

	Global mean radiative forcing (1750–2004) (W m^{-2}) [Uncertainty]			
	SAR	TAR	AR4	Summary comments on changes since TAR
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 ± 0.16); CH_4 (0.48 ± 0.05); N_2O (0.15 ± 0.02); Halocarbons (0.33 ± 0.03)}**	**Total increase in RF, due to upward trends, particularly in CO_2. Halocarbon RF- re-evaluated to be lower (trend is positive)**
Stratospheric O_3	**–0.1 [2X]**	**–0.15 [67%]**	**–0.03 ± 0.07**	**Re-evaluated to be weaker**
Tropospheric O_3	**+0.40 [50%]**	**+0.35 [43%]**	**+0.35 [+ 0.15/–0.10]**	**Better constrained**
Total direct aerosol	**Not evaluated**	**Not evaluated**	**–0.50 ± 0.40**	**Combination of models and observations**
Direct sulphate aerosols	*–0.40 [2X]*	*–0.40 [2X]*	*–0.40 ± 0.20*	*Better constrained*
Direct nitrate aerosols	*Not evaluated*	*Not evaluated*	*–0.10 ± 0.10*	*Newly evaluated*
Direct biomass burning aerosols	*–0.20 [3X]*	*–0.20 [3X]*	*+0.0 ± 0.10*	*Re-evaluated to be smaller. Response affected by semi-direct effects*
Direct FF aerosols (BC)	*+0.10 [3X]*	*+0.20 [3X]*	*+0.20 ± 0.10*	*Re-evaluated to be slightly larger. Response affected by semi-direct effects*
Direct FF aerosols (OC)	*Not evaluated*	*–0.10 [3X]*	*-0.1.0 ± 0.10*	*Similar to TAR evaluation*
Direct mineral dust aerosols	*Not evaluated*	*–0.60 to +0.40*	*–0.10± 0.20*	*Re-evaluated to have a smaller anthropogenic fraction*
Cloud albedo indirect aerosol effect	**0 to –1.5 (sulphate only)**	**0.0 to –2.0 (all aerosols)**	**–0.90 ± 0.50 (all aerosols)**	**best estimate now possible**
Linear contrails Aviation-induced cirrus	**Not evaluated**	**0.02 [3.5 X]** 0 to +0.04	**0.01 [2X]** 0.02 to 0.10	Re-evaluated linear contrail RF is smaller and better constrained
Land surface albedo	**Not evaluated**	**Not evaluated**	**–0.10 ± 0.3**	**Combination of opposing RFs**
Land-use (albedo)	Not evaluated	–0.20 [100%]	–0.20 ±0.30	Additional studies
Black-Carbon on snow (albedo)	*Not evaluated*	*Not evaluated*	*+0.10 [3X]*	*Not evaluated in TAR*
Stratospheric water vapour from CH_4	**Not evaluated**	**+0.01 to +0.03**	**+0.07 ± 0.05**	**Higher than TAR evaluation**
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 2.24

For the AR4 column 1σ value uncertainties appear in brackets [2x] refers to a factor of two 1σ uncertainty and a log-normal distribution of likely RFs. Uncertainties in SAR and TAR had a similar basis, but their evaluation was more subjective.

Table. 2.13. Emission based radiative forcings for emitted components with radiative effects other than through changes in their atmospheric abundance. Minor effects where the estimated RF is less that 0.01 W m^{-2} are not included. Effects on sulphate aerosols are not included since SO_2 emission is the only significant factor effecting sulphate aerosols. Method of calculation and uncertainty ranges are given in the footnotes.

		Atmospheric or Surface Change Directly Causing Radiative Forcing											
		CO_2	CH_4	CFC/ HCFC	N_2O	HFC/ PFC /SF6		BC snow albedo	OC	O_3(T)	O_3(S)	H_2O(S)	Nitrate aerosols
Component Emitted	CO_2	1.56[a]											
	CH_4	0.016[a]	0.41[c]							0.164[e]		0.07[d]	
	CFC/HCFC			0.32[i]							–0.065[b]		
	N_2O				0.15[i]						–0.035[b]		
	HFC/PFC/ SF_6					0.014[i]							
	CO	0.036[a]	0.13[c]							0.036[e]			
	NOx		–0.10[c]							0.135[e]			–0.15[f]
	VOC	0.021[a]	0.05[c]							0.015[e]			
	BC						0.54[g]	0.1[h]					
	OC								–0.3[g]				

Notes:

(a) RF of the observed CO_2 change (Table 2.12), and then subtracted the contributions from CH_4, CO and VOC emissions from fossil sources. Historic emissions of CH_4, CO and VOCs from EDGAR-HYDE (Van Aardenne et al. (2001)), CO_2 contribution from these sources calculated by with CO_2 model described by Joos et al. (1996).

(b) 65% of RF from observed ozone depletion in the stratosphere (Table 2.12) is attributed to CFCs/HFCs, remaining 35% to N_2O (Isaksen et al., 1986)

(c) RF of the observed CH_4 change (Table 2.12), and then subtracted the contributions through lifetime changes through OH changes caused by emissions of NOx, CO and VOC. The effects of NOx, CO and VOCs have been calculated using the linear response relations for OH derived in TAR (page 269). There are significant uncertainties related to these relations indicated by the range in model results. Following Shindell et al., (2005) we estimate the uncertainty to be ±20%.

(d) All the radiative forcing from changes in stratospheric water vapour is attributed to methane emissions (see Section 2.3.8 and Table 2.12)

(e) RF of tropospheric ozone changes (see Section 2.3.7 and Table 2.12) is attributed to the ozone precursors NOx, CH_4, CO, and VOCs according the relative contribution calculated by the linear ozone response relation given in Ramaswamy et al. (2001) (page 269). Uncertainties are ±50% (Shindell et al., 2005)

(f) RF cf. Table 2.12, Uncertainty ±0.15 W m^{-2}

(g) Mean of all studies in Table 2.5, includes FF, biofuel and biomass burning, Uncertainty (1-σ) 0.32 W m^{-2} (BC) and 0.18 (OC) based on range of reported values in Table 2.5.

(h) RF cf. Table 2.12, Uncertainty [2X]

(i) RF calculated based on observed concentration change, cf. Table 2.12 and Section 2.3

Table 2.14. Lifetimes, radiative efficiencies, and direct (except for methane) global warming potentials (GWP) relative to carbon dioxide. For ozone depleting substances and their replacements data are taken from IPCC/TEAP (2005) unless otherwise indicated.

Industrial Designation or Common Name	**Chemical Formula**	**Other Name**	**Lifetime (years)**	**Radiative Efficiency ($W\ m^{-2}\ ppb^{-1}$)**	**Global Warming Potential for Given Time Horizon (years)**			
					SAR (100)	**20**	**100**	**500**
Carbon dioxide	CO_2		See below[a]	See below[b]	1	1	1	1
Methane[c]	CH_4		10.8[c]	3.7×10^{-4}	21	67	23	6.9
Nitrous oxide	N_2O		114[c]	3.1×10^{-3}	310	291	298	153
Substances controlled by the Montreal Protocol								
CFC-11	CCl_3F	Trichlorofluoromethane	45	0.25	3800	6700	4750	1620
CFC-12	CCl_2F_2	Dichlorodifluoromethane	100	0.32	8100	11000	10800	5200
CFC-113	CCl_2FCClF_2	1,1,2-Trichlorotrifluoroethane	85	0.3	4800	6540	6130	2700
CFC-114	$CClF_2CClF_2$	Dichlorotetrafluoroethane	300	0.31		8040	10000	8700
CFC-115	$CClF_2CF_3$	Monochloropentafluoroethane	1700	0.18		5310	7370	10000
Halon-1301	$CBrF_3$	Bromotrifluoromethane	65	0.32	5400	8480	7140	2760
Halon-1211	$CBrClF_2$	Bromochlorodifluoromethane	16	0.3		4750	1890	575
Halon-2402	$CBrF_2CBrF_2$	1,2-Dibromotettrafluoroethane	20	0.33		3680	1640	500
Carbon tetrachloride	CCl_4	(Halon-104)	26	0.13	1400	2700	1400	435
Methyl bromide	CH_3Br	(Halon-1001)	0.7	0.01		17	5	1
Bromochloromethane	CH_2BrCl	(Halon-1011)	0.37					
Methyl chloroform	CH_3CCl_3	1,1,1-Trichloroethane	5	0.06		510	146	45
HCFC-22	$CHClF_2$	Chlorodifluoromethane	12	0.2	1500	5200	1800	550
HCFC-123	$CHCl_2CF_3$	Dichlorotrifluoroethane	1.3	0.14	90	270	77	24
HCFC-124	$CHClFCF_3$	Chlorotetrafluoroethane	5.8	0.22	470	2070	610	185
HCFC-141b	CH_3CCl_2F	Dichlorofluoroethane	9.3	0.14		2250	730	220
HCFC-142b	CH_3CClF_2	Chlorodifluoroethane	17.9	0.2	1800	5500	2300	705
HCFC-225ca	$CHCl_2CF_2CF_3$	Dichloropentafluoropropane	1.9	0.2		430	120	37
HCFC-225cb	$CHClFCF_2CClF_2$	Dichloropentafluoropropane	5.8	0.32		2030	600	180
Hydrofluorocarbons								
HFC-23	CHF_3	Trifluoromethane	270	0.19	11700	12000	14800	12200
HFC-32	CH_2F_2	Difluoromethane	4.9	0.11	650	2330	675	205
HFC-125	CHF_2CF_3	Pentafluoroethane	29	0.23	2800	6350	3500	1100
HFC-134a	CH_2FCF_3	1,1,1,2-Tetrafluoroethane	14	0.16	1300	3830	1430	435
HFC-143a	CH_3CF_3	1,1,1-Trifluoroethane	52	0.13	3800	5890	4470	1590
HFC-152a	CH_3CHF_2	1,1-Difluoroethane	1.4	0.09	140	437	124	38
HFC-227ea	CF_3CHFCF_3	1,1,1,2,3,3,3-Heptafluoropropane	34.2	0.26	2900	5300	3200	1000

HFC-236fa	$CF_3CH_2CF_3$	1,1,1,3,3,3-Hexafluoropropane	240	0.28	6300	8100	9800	7660
HFC-245fa	$CHF_2CH_2CF_3$	1,1,1,3,3-Pentafluoropropane	7.6	0.28		3400	1030	310
HFC-365mfc	$CH_3CF_2CH_2CF_3$	1,1,1,3,3-Pentafluorobutane	8.6	0.21		2500	790	240
HFC-43-10mee	$CF_3CHFCHFCF_2CF_3$	1,1,1,2,2,3,4,5,5,5-Decafluoropentane	15.9	0.4	1300	4100	1640	500
Perfluorocarbons								
	SF_6	Sulfur hexafluoride	3200	0.52	23900	16300	22800	32600
	NF_3	Nitrogen trifluoride	740	0.13		8300	11200	13200
PFC-14	CF_4	Carbon tetrafluoride	50000	0.10[f]	6500	4200	5900	8950
PFC-116	C_2F_6	Perfluoroethane	10000	0.26	9200	8600	12200	18200
PFC-218	C_3F_8	Perfluoropropane	2600	0.26	7000	6300	8800	12500
PFC-318	$c\text{-}C_4F_8$	Perfluorocyclobutane	3200	0.32	8700	7300	10300	14700
PFC-3-1-10	C_4F_{10}	Perfluorobutane	2600	0.33	7000	6300	8900	12500
	C_5F_{12}	Perflouropentane	4100	0.41		6500	9200	13300
PFC-5-1-14	C_6F_{14}	Perfluorohexane	3200	0.49	7400	6600	9300	13300
	$C_{10}F_{18}$	Perfluorodecalin	>1000[d]	0.56		>5500	>7500[d]	>9500
	SF_5CF_3	Trifluoromethyl sulfur pentafluoride	>1000[e]	0.57		>13000	>18000[e]	>23000
Fluorinated ethers								
HFE-125	CF_3OCHF_2		150	0.44		13900	15400	9300
HFE-134	CHF_2OCHF_2		26.2	0.45		12200	6360	1980
HFE-143a	CH_3OCF_3		4.4	0.27		3420	1000	305
HCFE-235da	$CF_3CHClOCHF_2$		2.6	0.38		1230	350	106
HFE-245cb2	$CF_3CF_2OCH_3$		4.3	0.32		2080	597	182
HFE-245fa2	$CF_3CH_2OCHF_2$		4.4	0.31		2060	592	180
HFE-254cb2	$CHF_2CF_2OCH_3$		0.22	0.28		107	30	9
HFE-347mcc3	$CF_3CF_2CF_2OCH_3$		4.5	0.34		1730	500	150
HFE-347pcf2	$CF_3CH_2OCF_2CHF_2$		7.1	0.25		1900	580	175
HFE-356pcf3	$CHF_2CF_2CH_2OCHF_2$		3.2	0.39		1570	450	140
HFE-449sl	$CH_3O(CF_2)_3CF_3$	(HFE-7100)	5	0.31		1390	404	123
HFE-569sf2	$CH_3CH_2O(CF_2)_3CF_3$	(HFE-7200)	0.77	0.3		200	57	17
H-Galden 1040x	$CHF_2OCF_2OC_2F_4OCHF_2$		6.3	1.37		6300	1900	570
HG-10	$CHF_2OCF_2OCHF_2$		12.1	0.66		8000	2800	860
HG-01	$CHF_2OCF_2CF_2OCHF_2$		6.2	0.87		5100	1500	460
Hydrocarbons and other compounds *- Direct Effects*								
Dimethylether	CH_3OCH_3		0.015	0.02		1	1	<<1
Methylenechloride	CH_2Cl_2	(Freon-40) Dichloromethane	0.38	0.03		31	8.7	2.7
Methyl chloride	CH_3Cl	(Freon-30) Chloromethane	1.3	0.01		59	17	5.1

Notes:

(a) The CO_2 response function used in this report is based on the revised version of the Bern Carbon cycle model used in Chapter 8 of this report using a background CO_2 concentrations 378 ppm. The decay of a pulse of CO_2 with time t is given by

$$a_0 + \sum_{i=1}^{3} a_i \cdot e^{-t/\tau_i}$$

, where a_0=0.217, a_1=0.259, a_2=0.338, a_3=0.186, τ_1=172.9 years, τ_2=18.51 years, and τ_3=1.186 years.

(b) The radiative efficiency of CO_2 is calculated by the expression RF=α ln(C/C_0), where α=5.35 (IPCC, 2001) and C_0=378 ppm. The radiative efficiency of CO_2 is then 1.4×10^{-5} W m^{-2} ppb^{-1}.

(c) The adjustment time for methane has been reduced from 12 years since the TAR (cf. Chapter 7, Section 7.4.1). The GWP for methane includes indirect effects from enhancements of ozone and stratospheric water vapor (see Section 2.10.3.1)

(d) Shine et al. (2005c), updated by the revised AGWP for CO_2. The assumed lifetime of 1000 years is a lower limit.

(e) Sturges et al. (2000), updated by the revised AGWP for CO_2. The assumed lifetime of 1000 years is a lower limit.

(f) Hurley et al. (2005)

Table 2.15. 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